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LIST OF ABSTRACTS

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Plenary.1 ID:4598 INVITED 09:30

Chemistry and Climate

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The Earth has not warmed as much as it would have in the absence of increases in aerosol levels, making aerosols unsung warriors in the fight against global warming. Uncertainty in aerosol radiative forcing is the largest contribution to uncertainty in global climate sensitivity. With continuing increases in greenhouse gases, however, the exact amount of aerosol radiative forcing will eventually assume less importance to global radiative forcing. Many important effects of aerosol dynamics and atmospheric chemistry on climate, nonetheless, will continue to play a key role. These include: feedbacks associated with black carbon mitigation, aerosol-cloud interactions and associated feedbacks, gas/aerosol/climate coupling, and regional climate effects of aerosol forcing.

Chemistry-Climate 1.1 ID:4241 10:15

A multi-model assessment of Intercontinental Source-Receptor relationships for ozone pollution in the 21st century.

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We present results from two climate-chemistry transport models for present-day and future climate using the SRES A2 climate forcing scenario, in support of the Task Force on Hemispheric Transport of Air Pollution (HTAP). The “climate penalty” - whereby the effect of the change in climate on surface ozone concentrations partly offsets the effect of the emission controls, such that stricter emission controls are required to attain a given air quality standard in the future- is clearly shown in high emission regions. In these highly polluted regions, enhanced water vapour concentrations in the 2090s decade lead to surface ozone increases of 2-6 ppbv compared to the 2000s decade. In remote regions, the climate effect of increased water vapour leads to ozone destruction. Ozone concentrations in southern mid-latitude regions also increase due to enhanced stratosphere-troposphere exchange (STE) in this future climate. Due to the strong temperature–dependent lifetime of Peroxyacetylnitrate (PAN), tropospheric PAN burdens decrease by 32-45% in the 2090s compared to 2000s. However, tropospheric ozone burden changes vary in sign between the two models depending on whether the effect of higher water vapour in unpolluted regions, or enhanced STE dominates.

Using fixed emissions at present-day levels, we find that 20% NOx, CO and NMVOCs emission reductions over major source regions decreases surface ozone concentrations by ~0.5-3 ppbv in the source region in the 2090s climate. These surface ozone reductions in the source and receptor regions fall within multi-model ranges of ozone responses to the same emission controls for present-day, found in a previous HTAP study. Comparing the effect of these emission reductions under the 2000s and 2090s climate, we see a greater impact of these controls on surface ozone concentrations in the source region than in the downwind region under the future climate, which we attribute to PAN lifetime changes.

Chemistry-Climate 2.1 ID:4455 INVITED 11:00

In search of regional signatures of anthropogenic aerosol emission changes in climate forcing and response.

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Aerosols have been recognized to be a potent forcing agent in the climate system for decades. They interact with the climate system in many ways, including their direct effects of scattering and absorbing solar radiation, and their indirect role as Cloud Condensation Nuclei and Ice Nuclei. Frequently, modelers have explored the role of aerosols by simulating the forcing and response of the climate system to changes in global emissions of aerosols and their precursors. Occasionally modelers have explored the role of emissions from a particular region on climate change. Frequently those studies focused on one class of forcing (for example the “direct” aerosol effect), or treated the indirect effects very simply, by introducing a dependence in cloud fraction on soluble aerosol, or ignoring the role of aerosols on ice nucleation. In this study we attempt to use a somewhat more elaborate formulation for aerosol cloud interactions to include more of the physical underpinnings for the direct and indirect effect, and contrast the signatures in forcing and response from separate and combined emission changes in a number of the major source regions for aerosols.

Chemistry-Climate 2.2 ID:4269 11:30

Assessing an NO_x mitigation technique: Chemical and climatic consequences of rising methane and NO_x concentration levels in the troposphere

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Methane (CH_4) is a green house gas, and its global warming potential in a 20 year time horizon is 72 times that of CO_2. According to the IPCC report, there is possibility for a 3-fold increase in CH_4 emissions by the end of this century. Besides, there is a huge deposit of frozen CH_4 as clathrates in seabed, whose seepage is underway. The positive feedback between CH_4 seepage from clathrates and climate warming has raised concern about a catastrophic emission of CH_4 to the atmosphere. One countermeasure could be emitting NO_x to accelerate the CH_4 oxidation in the atmosphere. But with increased NO_x, warming effects from increased O_3 production and possible cloudiness change due to aerosol formation may render this NO_x -mitigation technique questionable.

Aiming to assess the applicability of this NO_x -mitigation technique, we carried out a series of model analysis. A column model, SOSA, was used to run four month long simulations for scenarios where tropospheric concentrations of CH_4 and NO_x were increased up to 20 times of today’s level. The CH_4 lifetime increases by 73% and troposphere O_3 concentration increases by 7% when CH_4 concentration is 10 times of today’s level; the numbers then change to -8% and 19% when NO_x concentration is doubled. Radiative forcings of CH_4 and O_3 for different scenarios were calculated by the global climate model ECHAM-5 and the Line By Line Radiative Transfer Model (LBLRTM). Cloud droplet number concentration (CDNC) variation due to the changes in OH, O_3, NO_3 and resulting effects on cloud radiative forcing are further studied by the global model ECHAM-5 and GLOMAP. Finally, the energy demand for implementing the technique was roughly estimated.

Chemistry-Climate 2.3 ID:4125 **11:45**

The vertical distribution of tropospheric ozone instantaneous radiative forcing from satellite and chemistry climate models

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Tropospheric ozone radiative forcing is primarily driven by longwave absorption, and its accurate estimation is contingent on the ability of models to simulate the spatial and vertical distribution of ozone within the entire troposphere. We evaluate the instantaneous radiative forcing (IRF) of tropospheric ozone predicted by four state-of-the- art global chemistry climate models (AM2-Chem, CAM-Chem, ECHAM5-MOZ and GISS-PUCCINI) against ozone distribution observed from the NASA Tropospheric Emission Spectrometer (TES) during August 2006. The change in the top-of-the- atmosphere radiative flux to vertical and spatial discrepancies between model and TES ozone was calculated through all-sky instantaneous radiative forcing kernels from TES. Through this technique, we find total tropospheric IRF biases from -0.4 to +0.7 W/m^2 over large regions within the tropics and mid-latitudes, due to ozone differences over the region in the lower and middle troposphere, enhanced by persistent bias in the upper- troposphere lower-stratospheric region. The zonal mean biases also range from -30 to +40 milli-Watts/m^2 for the models. However, the ensemble mean total tropospheric IRF bias is less than 0.2 W/m^2 within the entire troposphere. The IRFK present a new and consistent observationally-based method for evaluating the instantaneous radiative impact of ozone from several chemistry climate models.

Chemistry-Climate 3.1 ID:4596 **INVITED 13:30**

Air Quality and Climate Change - considerations for the Australian region

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We will begin the presentation with a brief overview of the current state of air quality in Australia (population of 22.3 million). Across the country, air pollution is estimated to result in over 1000 premature deaths per year and to lead to health costs in excess of $A1billion/annum. Primary particulate matter (PM) and photochemical smog including a secondary PM component) are considered to be the most significant pollutant groups with respect to health impacts. Within urban areas the dominant sources of PM are motor vehicles and domestic wood combustion. However the most significant individual PM episodes are caused by wild fires, fire management burns and dust storms. Considering photochemical smog, peak ozone concentrations are observed to approach or exceed air quality standards in most Australian capital cities (with Sydney experiencing up to 15 exceedences per year of the 4 hour- 80 ppb standard). Motor vehicles, biogenic emissions and smoke from wild fires are considered to be the major sources of photochemical smog. The frequency of both the PM and photochemical smog episodes generally peak during El Nino years when the environment is dry, and the frequency of hot days, wild fires and dust storms are highest.

The presentation will then go on to explore the likely response of air pollution in Australian airsheds to climate change. In this regard, the coupling between non-urban sources of air pollution and population health

impacts suggests that the air quality in Australia may be quite sensitive to climate change-particularly a trend towards a warmer and drier climate (as projected for major regions of Australia). In this regard, we have explored the sensitivity of ozone pollution in Sydney to climate change using a dynamical downscaling system forced by a global A2 GCM climatology, and estimated that hospital admissions in Sydney resulting from ozone exposure would increase by 40% and 200% respectively in 2025 and 2055 as a result of climate change alone. Another area of concern is the projected increase in the frequency of high and extreme fire-weather events-potentially leading to an increased frequency of wild fires and greater population exposure to both PM and ozone. An increase in high fire risk weather is also likely to be associated with an increase in fire management burning, potentially leading to a challenging environmental management problem.

The presentation will be completed with a brief discussion about the need to consider any potential air quality implications when developing climate mitigation or adaptation strategies.

Chemistry-Climate 3.2 ID:4265 13:50

Assessment of aerosol-cloud interactions employing parametrisations of various complexities

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Atmospheric aerosols play an important role in the global climate system through modifications of the global radiation budget: directly, by scattering and absorption of radiation and indirectly, by the modification of cloud properties and abundance with impacts on the hydrological cycle. In particular the indirect aerosol effects on clouds and precipitation are subject to large uncertainties.

State of the art global aerosol-cloud climate models allow estimates of aerosol-cloud interactions. However, limitations in the underlying cloud and aerosol microphysics and in particular the requirement to reduce their complexity for the implementation in global climate models introduce significant uncertainties. Previous comparisons of conceptually different approaches were often of limited explanatory power due to the usage of varying models and setups.

In this study we investigate uncertainties in estimates of indirect aerosol effects through studies with the ECHAM5-HAM2 aerosol-climate model. We employ parametrisations of the aerosol-cloud interactions of various complexities in an identical model setup to quantify the contribution of the process parametrisation to the uncertainty in the simulated aerosol-cloud interactions. The consideration of the direct and semi-direct effects as well as the indirect cloud albedo and lifetime effects allows for a comprehensive investigation of the aerosol effects on precipitation.

A detailed evaluation of the results with satellite observations of aerosol and cloud parameters will provide observational constraints on the simulated aerosol-cloud interactions in the different model setups. Our results help to understand and quantify uncertainties in estimates of aerosol-cloud interactions and yield valuable information about the necessary level of detail of the process representation in global climate models.

Chemistry-Climate 3.3 ID:4256 14:05

A new approach to an observationally-based estimation of aerosol-cloud radiative forcing

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Uncertainty in IPCC AR4 radiative forcing estimates of the albedo (Twomey) effect is high and constitutes one of the largest uncertainties in climate predictions. Estimates of aerosol effects on cloud microphysics from observations are highly uncertain because of the difficulty of remotely measuring both the aerosol and cloud properties in the same location, and the fact that it is very difficult to identify cloud-free regions in the vicinity of clouds. The satellite observations of aerosol-cloud interaction metrics that are used in climate models result in the weakest of all IPCC forcing estimates. Using large eddy simulation, and detailed microphysical models as a proxy data set, we systematically address the factors that bias satellite observations of aerosol-cloud interactions. These include failure to sort data by cloud liquid water, averaging over large spatial scales, and large physical separation between aerosol and cloud measurements. We propose a new consistent approach that removes the biases. The method relies on surface measurements of joint probability distribution functions (pdfs) of liquid water path and updraft velocity, and pdfs of the cloud-active aerosol, at the cloud scale. Within specific cloud regimes, the pdf method is applied along with a simple cloud model to derive a climate-model grid (order 100 km) representation of aerosol effects on cloud microphysics, and radiative forcing.

Chemistry-Climate 3.4 ID:4518 14:20

Black carbon effects on clouds: Implications for mitigation

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Black carbon (BC) absorption of radiation in the atmosphere and in snow contributes to global warming. However BC also influences clouds in ways that could potentially result in cooling. First, since BC affects the cloud condensation nuclei (CCN) population, it influences the aerosol indirect effect. Second, BC perturbs the thermal structure of the atmosphere, and influences cloud distributions, typically called ‘the semi-direct effect’. We discuss both of these BC-cloud effects, uncertainties, and implications for potential future BC reductions.

To study BC indirect effects, we conducted BC-reduction experiments with six global models that simulate aerosol microphysical responses to BC changes and indirect effects (ECHAM5, CAM-Oslo, CAM-PNNL, GISS, LSCE and SPRINTARS). The models performed three experiments, removing: fossil fuel BC; biofuel BC and organic carbon (OC); and diesel BC and OC. Results are compared to the full-emission simulations. BC affects the CCN population by increasing the primary particle number; however as secondary species condense on BC, it also diverts them from forming pure easily activated particles. Therefore the models typically show regional variability in their cloud responses, with some having net negative and some positive BC indirect effect.

The effects of BC absorption on cloud cover (semi-direct effect) depend upon the altitude of BC relative to cloud and meteorological conditions including atmospheric stability, relative humidity and cloud type. We organize and synthesize cases from the literature in which aerosol absorption increases or decreases cloud cover. Many global climate model studies indicate a net negative semi-direct effect due to increased low-

level and/or decreased high-level clouds. We present results from a GISS climate simulation in which BC reduction resulted in fewer clouds and minimal climate cooling.

We discuss uncertainties in models and observations and recommendations for future research.

Observations 1.1 ID:4248 14:35

Results from 6 years of operation of the Ozone Monitoring Instrument (OMI)

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The Ozone Monitoring Instrument (OMI) is a Dutch-Finnish built instrument that will complete 6 years of operation in orbit this year on NASA's Aura spacecraft. Though several UV/VIS backscatter instruments have flown in space in the past 4 decades, OMI is the first instrument of this type that has been optimized to track small-scale variabilities in tropospheric traces gases and UV-absorbing aerosols. The Aura spacecraft has 4 instruments dedicated to atmospheric chemistry and air quality research and the spacecraft itself is apart of a novel multi-spacecraft constellation called the A-train. This allows synergistic use of data from multiple instruments. In this talk we will focus on the study of inter- annual variability of tropospheric trace gases and aerosols derived from 6 years of OMI data and will discuss several multi-instrument products that have been successfully produced by combining OMI data with data from other A-train instruments. The presentation will include discussion of uncertainties in these products, particular those related to the effects of clouds on satellite retrieval of boundary layer trace gases, and our ongoing efforts in reducing these uncertainties. Finally, we will discuss how we are incorporating lessons learned from these studies in designing more advanced sensors to study air quality from space.

Observations 1.2 ID:4551 14:50

Global correlation patterns of ozone and CO derived from TES observations and model simulations

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Simultaneous measurements of ozone and CO taken over 5 years by the Tropospheric Emission Spectrometer (TES) provide a unique opportunity to examine global correlation patterns and the processes controlling those patterns. Here, we use measurements and output from four chemistry-climate and chemistry-transport models (GISS, ECHAM5-MOZ, UKCA, p-TOMCAT), to examine how mid-tropospheric ozone and CO correlate in different regions and seasons. We find a remarkable agreement between the correlation patters in the observations and in the models. A prominent feature is a contrast between much of the Northern Hemisphere and the Southern Hemisphere, with mostly negative correlations in the former and positive correlations in the latter. An exception to this is the Northern Pacific, where the correlations are largely positive and high. By examining the results of model sensitivity experiments, we determine the drivers of these correlation patterns. Processes affecting tracer abundances, like long-range transport, stratosphere-troposphere exchange, the influence of clouds and emissions can be investigated using this methodology. Model deficits discovered will be tackled in further model development.

Observations 1.3 ID:4572 **15:05**

Characterizing differences between AOD trends derived from MODIS-Terra and MODIS-Aqua

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Aerosols are integral to Earth's energy balance, and affect climate and human health. NASA's MODerate resolution Imaging Spectroradiometers (MODIS) on the Terra and Aqua platforms retrieve global, clear-sky, aerosol optical depth (AOD) products at 10 x 10 km. Both MODIS instruments employ identical, aerosol retrieval algorithms that have been validated to retrieve AOD to within ±(0.04 + 5%) over ocean and ±(0.05 + 15%) over land. As we pass the ten-year anniversary of these sensors, we assess the MODIS-observed aerosol trends.

Both sensors report statistically significant (>95% level) global increase of ~0.01/decade over ocean, although Terra's average AOD is larger than Aqua's by ~0.015 (~10%). Instead of diurnal differences, we believe the 10% offset in AOD to be due to <2% difference in relative calibration. When we look at "regional" trends, we find general consistency that there are increases in some regions and decreases in others. Increases over the vast southern oceans seem to drive the global ocean trend.

While both derive similar averages over land, Terra shows decrease of ~0.02/decade, while Aqua shows increase of ~0.005/decade. Neither sensor's trend is statistically significant over land. When we look regionally, the trends do not generally agree. We believe that the major contributor to these trend discrepancies land are tiny drifts in relative calibration between the two sensors, especially in channel bands most susceptible to long-term degradation. We look to leverage other long-term data, such as AERONET or air quality networks, as well as model outputs, to help interpret MODIS's regional and global aerosol record.

To complicate matters, the two instruments view the Earth from different orbits, and experience diurnal differences in cloud distributions. This means that the differences in the sampling of clear-sky aerosol must also be considered. As we are determining global and regional aerosol trends, we also are assessing different methods of data aggregation that will properly account for issues of sampling.

Observations 1.4 ID:4311 **15:20**

Investigating atmospheric composition using solar occultation: the Atmospheric Chemistry Experiment (ACE) and beyond

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Reliable chemical composition measurements are required to understand changes occurring in the Earth's

atmosphere due to climate change and air pollution. Within this data set, altitude-resolved data are required because many processes occur at specific altitudes or over limited vertical length scales. Solar occultation has proven to be an effective technique for measuring profiles of atmospheric trace gases and aerosols. This paper will discuss the results from the Canadian-led Atmospheric Chemistry Experiment (ACE), a satellite mission using the solar occultation technique, and the need to continue measurements of this type for atmospheric monitoring and scientific studies.

Launched on 13 August 2003, the ACE satellite uses infrared and UV-visible spectroscopy to investigate the chemical composition of the Earth's atmosphere. The primary instrument on-board, the ACE Fourier Transform Spectrometer (ACE-FTS) is a high-resolution (0.02 cm^{-1}) FTS operating between 750 and 4400 cm^{-1}. It also contains two filtered imagers (0.525 and 1.02 microns) to measure atmospheric extinction due to clouds and aerosols. The second instrument is a dual UV-visible-NIR spectrophotometer called ACE-MAESTRO (Measurements of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation), which extends the ACE wavelength coverage to the 280-1030 nm spectral region. The ACE-FTS and ACE-MAESTRO have been making regular solar occultation measurements for nearly 6.5 years and, from these measurements, altitude profiles of over 30 different atmospheric trace gas species, aerosols and temperature are obtained.

P-Sources.1 ID:4521 15:35

Can we reconcile reported emissions of SF6 with atmospheric observations?

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SF6 is one of the most potent greenhouse gases known to date. As such it is one of the six gases covered by the Kyoto Protocol. NOAA ESRL has been measuring SF6 from a global network of sampling locations since 1997. The analysis of two air archives, Cape Grim and Niwot Ridge, provide data back from 1978 and 1986 respectively. SF6 natural background levels are very low. Due to its extreme stability, SF6 has been used since the 1950s in various industrial applications, the most important one being electricity transmission and distribution. Due to its very long lifetime (>1000 years), its atmospheric mean mixing ratio has been increasing from 0.03ppt in 1970 to 6.8ppt in 2009. These atmospheric measurements show that global source of SF6 is significantly greater than the total emissions reported to the United Nations Framework Convention on Climate Change (UNFCCC) by Annex II countries, and the difference can not be made up by inclusion of emissions from rapidly developing countries. We show the evolution of the SF6 mixing ratio at different locations and compare the rate of increase with bottom-up inventories (such as EDGAR) and with emissions estimates reported to the UN. To understand how the global distribution of emissions affect the regional and global distribution of SF6 and what kind of observing network would be needed to verify emissions at the continental scale, several emissions scenarios have been run in the CAM global atmospheric circulation model from 1970 onward.

P-Sources.2 ID:4201 15:35

East Asian Emissions of Anthropogenic Halocarbons estimated from In-situ Measurements at Gosan, Jeju Island (Korea)

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National emissions have been estimated mainly by bottom-up approaches. To verify and supplement those bottom-up inventories, top-down approaches based on observation are required. In this study, high-frequency in-situ measurements of a wide range of Halocarbon compounds including chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs), have been made at Gosan station (Jeju Island, Korea). We have been estimated the emission ratios of CFCs, HCFCs and HFCs during 2008 from East Asia including China, Japan, and Korea. The air masses from every country were identified by Particle dispersion model. We chosen the reference compound in individual country under the assumption: (1) To have good linearity with all other compound, (2) To have long-lifetime, (3) emission rate of reference compound is correct. The emission rate of reference compounds for 2008 were estimated by inverse modeling. Our results confirm the signs of successful phase-out of CFCs, along with substantial emissions of replacement HCFCs in East Asia. The emission rate of CFCs, HCFCs and HFCs in East Asia accounts for 14%, 28%, 15% of global emission, respectively.

P-Sources.3 ID:4384 15:35

European emissions and trends of ozone depleting substances from continuous measurements at a European mountain site.

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Chlorofluorocarbons (CFCs), methyl chloroform, carbon tetrachloride, methyl bromide and halons are the principal ozone depleting substances. In response to the UNEP Montreal Protocol on Substances That Deplete the Ozone Layer and subsequent amendments, production and consumption of these gases have been prohibited in developed (non-A5) countries since 1996, with the exception of Methyl Bromide, for which the Protocol asked for a complete phase-out in 2005. The Protocol also regulates the hydrochlorofluorocar-bons (HCFCs), i.e. low-ozone-depletion potential substitutes classified under the Protocol as transitional substitutes to be used during the time it took to commercialize new ozone-safe alternatives and replacements. HCFCs are being progressively phased out in non-A5 countries meanwhile in A5 countries they will not be regulated before 2016. In the absence of regulations in de-veloping countries the demand for HCFCs in many applications is expected to increase

Continuous high-frequency measurements of a wide range of ozone depleting substances (ODSs) have been made at the CNR atmospheric research station of Monte Cimone on the highest peak of the Italian Northern Apennines since 2001, using an automated gas chromatographic-mass spectrometric (GC-MS) system. The activity has been carried out within the EU funded project SOGE (System for Observation of halogenated Greenhouse gases in Europe). Long time series are therefore available, showing different trends according to their different lifetimes.

However, for methyl chloroform and halons differently than expected in a phase out re-gime, elevations

from the baseline are recorded, indicating that ODSs are still being re-leased even if not reported. Elevations above the baseline together with air mass back trajectories analysis allow the identification of source regions.

P-Sources.4 ID:4517 **15:35**

Hydration of the lower stratosphere by ice crystal geysers over land convective systems

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The impact of deep convective overshooting over land on stratospheric composition has been explored by a series of simultaneous balloon soundings of water vapour, particles and ozone in the lower stratosphere next to Mesoscale Convective Systems during the monsoon season over West Africa. The water vapour measurements were carried out using a fast response FLASH-B Lyman-alpha hygrometer. The high vertical resolution observations of the instrument show the presence of enhanced water vapour layers between the tropopause at 370 K and the 420 K level. Most of these moist layers are shown connected with overshooting events occurring upwind as identified from satellite IR images over which the air mass probed by the sondes passed during the three previous days. In the case of a local overshoot identified by the collocated MIT C-band radar, tight coincidence was found between enhanced water vapour, ice crystal and ozone dip layers indicative of fast uplift of tropospheric air across the tropopause. The presence of such layers strongly suggests hydration of the lower stratosphere by geyser-like injection of ice particles over overshooting turrets. The pile-like increase of water vapour up to 19 km seen by the high-resolution hygrometer during the season of maximum temperature of the tropopause, suggests that the above hydration mechanism may contribute to the summer maximum moisture in the lower stratosphere. If this interpretation is correct, hydration by ice geysers across the tropopause might be an important contributor to the stratospheric water vapour budget.

P-Sources.5 ID:4369 **15:35**

Trace gas emission from sea surfaces.

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The mechanisms giving rise to emission of trace gas species (organics and halogenated compounds) due to photochemical or oxidative chemical processes at the air-sea interface are poorly understood. We have initiated a series of laboratory experiments which probe interfacial chemistry via surface-sensitive spectroscopy and gas phase product analysis. I will describe the experiments and our results on trace gas formation and evolution from the interface. Of particular importance in this chemistry is the role of the marine microlayer, composed of biologically active compounds, on the surface photochemistry.

P-Sources.6 ID:4307 15:35

Transport of Greenhouse Gases in Trees

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Emission of the greenhouse gas methane (CH4) has been measured in cultivated and natural regions, quantifying emissions for croplands, wetlands, and forests. However, segregation between soil and plant emissions is less clear, and the dynamics behind each respective emission type differs. A global composite of greenhouse gas emissions must include all relevant sources, and current inventories are potentially missing large plant-mediated emissions of CH4 from wetland and upland systems. Better defined plant transport mechanisms will yield more accurate determination of CH4 flux, contributing to a comprehensive theory quantifying greenhouse gas emissions globally. While the mechanisms of CH4 emission from rice have not been fully identified, for trees these mechanisms are virtually unknown. CH4 emission from several tree species (Alnus rubra, Populus trichocarpa, Thuja plicata, Fraxinus latifolia) native to the Pacific Northwest has been measured. To identify mechanisms of gas transport, correlation between emissions and stomatal conductance, transpiration, and photosynthesis is tested. A synthesis between plant physiological data and emissions is sought to elucidate the role plant physiology plays in the production and transport of CH4. Extrapolation of plant-level emissions to climate-level CH4 fluxes will combines the results of greenhouse experiments with remotely-sensed vegetation parameters from space-borne instruments.

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P-Sources.7 ID:4475 15:35

Soil properties effects on carbon loss of mixed deciduous forest soil

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Carbon loss of forest soil in term of CO2 emission was investigated in natural forest (mixed deciduous forest) at the Siridhorn International Environmental Park (SIEP), Phetch-buri province, Thailand. The study was undertaken in November 2008 to November 2009. Soil and air samples were collected by using dark static chamber sizing 30*15*15 (height) cm. In order to keep the system closed throughout the sample collection, the chamber was placed at a depth of 5 cm in the ground. Sampling of gas from the chambers was collected in 3 collection periods, morning (8.00-8.30 a.m.), afternoon (12.00-12.30 p.m.) and evening (16.00-16.30 p.m.). The result showed that carbon loss of study site in rainy season was 67% higher than the rate in dry season. The result can be concluded that amount of rainfed, soil moisture, soil organic carbon effect soil carbon loss in term of CO2 emission from mixed deciduous forest soil.

P-Sources.8 ID:4511 15:35

Soil carbon loss and stock of eucalyptus forest soil

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The study aims to estimate carbon stock and loss from Eucalyptus forest soil, included considers influencing parameters on carbon loss as carbon-dioxide and methane to atmosphere from Eucalyptus forest soil. The experiment was investigated Eucalyptus forest at Kasetsart University at Kamphaeng Saen campus, Nakornpathom province, Thailand. The study was undertaken in November 2008 to August 2009. Air and soil samples were collected every month. The dark static chamber method was used to collect air sample and to be analyzed for carbon loss by Gas Chromatography. Soil samples were collected and to be analyzed to achieve soil properties included soil carbon loss and stock. Average carbon-dioxide emissions rate for 12 months from study site was 66.47 g CO2/m2. Highest emission rate was found in July 2009, which was 120.88 g CO2/m2. Lowest emission rate was 21.97 g CO2/m2, which was found in December 2008. However, methane emission from study area was not found. Soil carbon stock in study site was 21.33 g/m2. Soil moisture, soil organic carbon was major influencing factors effect soil carbon loss and stock in study site.

P-Sources.9 ID:4205 **15:35**

Improvement and evaluation of simulated global biogenic soil NO emissions in an AC-GCM

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15 years ago, based on very few measurements, Yienger and Levy (1995) published their algorithm to calculate soil biogenic emissions of NO (SNOx), which is still the most widely applied in global models. Since then numerous new measurements had been published; based on this, we have built up a database of world wide conducted measurements in the field covering the period from 1978 to 2009, including 108 publications with 560 measurements.

SNOx plays an important direct and indirect role in chemical processes of the troposphere. Recently, several satellite based top-down approaches, which recalculated the different emissions sources of NOx (fossil fuel, biomass burning, soil and lightning), have shown an underestimation of SNOx by the algorithm of Yienger and Levy (1995). Nevertheless, to our knowledge no general improvements of this algorithm have yet been published.

Here we present major improvements to the algorithm, which should help to optimize the representation of SNOx in atmospheric-chemistry global climate models, without modifying the underlying mathematical equations. The changes include:

1) Using a new up to date land cover map, with twice the number of land cover classes, and using annually varying fertilizer application rates;
2) Adopting the fraction of SNOx induced by fertilizer application based on our database;
3) Switching from soil water column to volumetric soil moisture, to distinguish between the wet and dry state;
4) Tuning the emission factors to reproduce the measured emissions in our database and calculate the emissions based on their mean value.

These steps lead us to increased global yearly SNOx, and our total SNOx source ends up being close to one of the top-down approaches. In some geographical regions the new results agree better with the top-down approach, but there are also distinct differences in other regions. This suggests that a combination of both top-down and bottom-up approaches could be combined in a future attempt to provide an even better

calculation of SNOx.

P-Sources.10 ID:4415 15:35

How to access global and regional burnt biomass from satellite observations to derive gases and particle emission inventories?

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Calculating biomass burning emissions requires a knowledge of appropriate emission factors together with spatial and temporal distributions of burnt biomass. Several satellite products are currently used for the latter, among them, the distributions of active fires (AF) and of burnt areas (BA). Though progress has been achieved by individual groups using these new satellite-based products, there still are substantial differences in the results obtained and methods used. In this context, BBSO1 and BBSO2 workshops were jointly organized in Toulouse, France in 2005 and 2009 between users and producers of burnt biomass estimates within the ACCENT network and the GEIA/AIMES/IGBP project, with an intercomparison exercise focused on CO emission estimates for 2003. In this talk, results for this exercise will be shown, dealing with the origin of differences between emission estimates by regions, land cover types... Three datasets using a bottom-up approach (Seiler and Crutzen 1980) and satellite-based biomass burned estimates (VGT, ASTR and MODIS) were compared and one dataset derived from a top-down approach from satellite (NASA-MOPITT) concentration data. This work will also be compared to FRP-based emission inventory (Kaiser et al., 2010) and GFED inventories. Results confirm the high variability of spatial and temporal patterns of CO emissions, with the greatest discrepancies occurring in the forest classes of the Northern Hemisphere, while Africa shows the best agreement among inventories, both in terms of total annual amounts and seasonality. Finally these results, discussed in the BBSO2 workshop allowed to yield regional recommendations for users in terms of satellite product uses and also to focus on the need of international works, both experimental and algorithmic.

P-Sources.11 ID:4458 15:35

Results from a large, multi-platform study of trace gas and particle emissions from biomass burning

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We report preliminary results from a large, multicomponent study focused on North American biomass burning that measured both initial emissions and post-emission processing. In the laboratory component,

vegetation commonly managed with prescribed fires was collected from the southeastern and southwestern US and burned under “realistic” conditions at the USFS Fire Sciences Laboratory in Missoula, MT. The smoke emissions were measured with a comprehensive suite of state-of-the-art trace gas and particle instruments. The University of Montana open-path FTIR system measured many trace gases and additional instruments (some deployed in smoke for the first time) allowed measurements of over 150 additional VOC. A subsequent, extensive field campaign featured ground-based sampling of smoldering emissions in North Carolina with FTIR and airborne measurements of initial emissions and plume aging in California, Arizona, and North Carolina on a Twin Otter aircraft outfitted with airborne FTIR, AMS, SP2, nephelometry, and WAS from November 2009 through March 2010. The lab fire studies revealed the presence of large amounts of HONO and high molecular weight gas-phase NMOCs in the initial emissions. Our airborne measurements confirmed the lab observations of large initial emissions of HONO (~10-20% of NOx) and “heavy” NMOC from many of the fires. High OH from rapid HONO photolysis likely contributed to the fast formation of PAN, organic acids, ozone, and aerosol and rapid loss of several precursors observed in our downwind measurements. In one isolated plume we measured the post emission chemical changes for about 4 hours of smoke aging. Over this aging period, the light scattering to CO ratio increased by a factor of 2.3 implying significant formation of secondary aerosol. Large growth factors for ozone, PAN and organic acids were also observed. The data we collected will be used to construct improved emissions inventories and to improve detailed photochemical models.

P-Sources.12 ID:4230 15:35

Historical (1850-2000) and future (2000-2100 gridded anthropogenic and biomass burning emissions of reactive gases and aerosols for IPCC AR5

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We present and discuss the recently created emission datasets for reactive gases and aerosols anthropogenic and biomass burning emissions that are being used for the chemistry simulations in the IPCC AR5. Among others, the dataset includes aircraft and shipping emissions, aerosols, aerosol precursors and ozone precursors. For 2000-2100, the emissions are based on 4 scenarios by integrated assessment models, selected for climate model simulations for IPCC (called Representative Concentration Pathways). In this contribution, we describe the process through which (1) historical emissions were created (using a variety of existing global datasets) and (2) future emissions were constructed to be in full agreement base year emissions and the underlying scenarios targeting specific radiative forcing targets. Considerable attention has been given on creating the most up-to-date database for 2000 emissions and for 1850-2000 (combination of a variety of inventories). For all gases, information has been generated for a range of sources and regions. All the emissions available at 0.5 x 0.5 degrees every decade. We will also present results of the first simulations for historical and future conditions using several chemistry-climate models.

The IPCC Emission team consists of: J.-F. Lamarque, T. C. Bond, V. Eyring, C. Granier, A. Heil, Z. Klimont, D. Lee, C. Liousse, A. Mieville, B. Owen, M. G. Schultz, D. Shindell, S. J. Smith, E. Stehfest, J. Van Aardenne, O. R. Cooper, M. Kainuma, V. Naik, K. Riahi and D. P. van Vuuren

P-Sources.13 ID:4586 15:35

Linking smoke plume injection height to fire characteristics and weather conditions in the continental United States using CALIPSO data and back trajectory calculations

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We use CALIPSO observations of smoke for 2006-2009, and back trajectory calculations, to build a database linking biomass plume injection heights to atmospheric conditions and fire behavior in the continental United States. We consider fires from agricultural and range lands, representing widely different land surface characteristics, atmospheric conditions, and fuel loadings. Fire parameters used include ecosystem type, and estimates of fire radiative power, area burned, and fuel consumed, based on GOES, MODIS and ASTER satellite products. Back trajectory calculations are used to link distant smoke plumes to their fire sources. The database offers the potential for improving representation of fire emissions in the National Emissions Inventory, for use in regional air quality and global chemical transport models.

P-Sources.14 ID:4331 15:35

Distribution of Polycyclic Aromatic Hydrocarbons on Different Sizes of Particulate Matters Emitted During Open Biomass Burning

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Open biomass burning has been one of the issues that affects the regional atmospheric pollution in South East Asia, and potentially contributes to a long-range or intercontinental transport of air pollution. One of the key source regions is in Northern Thailand, where the haze problem occurs severely in the dry season each year (around February-April). Although forest fires have been prevalent during the dry season, the burning of agricultural residues to prepare land for next-cycle plantation in irrigation area has added considerably to the fine particulate emission (253,854 tons in year 2005), and escalates the problem. Concentrations of Polycyclic Aromatic Hydrocarbons (PAHs) were measured in different size classes of fine particulate matter to find the source signatures of open biomass burning in Lampang, Northern Thailand. Field samples were collected with an 8-stage cascade impactor, and PAH analysis was made with a GC-MS. It was found that the particulates from rice straw burning were mostly in the size range 0.4–0.7 micron, and likely to contain PAHs with 4–6 aromatic rings. By contrast, particulates collected in the ambient air during the haze episodes were bimodal, with the majority of particles in the size class smaller than 0.4 micron. Particles in the larger mode (size range 4.7–5.4 microns) were associated with PAHs with 3 aromatic rings. This research points out that the source signatures of particulates in ambient air can be determined from the ratio of Fluorene/Fluorene+Pyrene to trace the contribution of rice straw burning, and the ratio of Benzo(a)Anthrazene/Chrysene to trace the contribution of deciduous forest fire burning. The information is crucial for identifying the sources and impacts of open biomass burning, and the aerosol size range data will be used further in a regional climate model study.

P-Sources.15 ID:4489 15:35

Determination of ionic species in aerosols emitted from biomass used over delhi

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The study on chemical properties of aerosol emitted from biomass/bio-fuels used over Delhi has been initiated to estimate their concentration and role in changes of ambient air quality. In rural India the main source of energy for cooking and heating is the combustion of bio-fuels as major percentage of energy requirement is met by this source. The emission concentration of ionic species from rural consumption of biofuels have been determined and discussed in this paper. The cow dung cake, crop residue and fuel wood samples were collected district-wise from the rural sector of Delhi, burnt in natural condition and analyzed their water soluble ionic species using ion chromatography. Simultaneously the ambient air quality of different places of Delhi has been monitored to correlate/linkage of these emissions (from bio-fuels) with ambient air quality of Delhi. We observed that in most of the aerosol samples the water soluble ions i.e., NH4+, SO42-, NO3-, Na+, K+, Ca2+ and Mg2+ were present and concentration varied significantly on parent bio-fuels characteristics. In anionic species the average concentration of Cl- is highest followed by PO42->SO42->NO3->F->NO2->Br- and in cationic species the average concentration of K+ is highest followed by NH4+>Na+>Ca2+>Mg2+ from biofuels used over Delhi.

P-Sources.16 ID:3465 15:35

Characterization of Particulate Matter Emissions from a Modern Wood Burner and Determination of Wood Smoke Emission Factors

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Many urban areas in New Zealand are subject to air particulate matter pollution episodes due to particulate matter during winter. Air pollution monitoring, receptor modelling and emissions inventories all point to emissions from solid fuel fires used for domestic heating as the primary source of the particulate matter pollution. Reliable emission factors are required for emission inventories so that any air quality management options and policy interventions can be assessed for effectiveness. Samples of wood combustion emissions were collected from a wood burner typical of those that would be used for home heating in New Zealand. Particulate matter emissions associated with the combustion of three wood species commonly burned in New Zealand were measured and the concentrations of particle-phase polycyclic aromatic hydrocarbons (PAHs) and inorganic elements were determined under different operating conditions (start-up, high burn and low burn). Samples were analysed for elemental content by Ion Beam Analysis techniques, black carbon was determined by light reflectance, total carbon was determined by GC isotope ratio mass spectrometry and particle-phase PAH concentrations were determined by thermal desorption GC/MS. Scanning Electron Microscopy – Energy Dispersive Spectroscopy (SEM-EDS) was used to investigate particle morphology, size distribution and individual particle elemental composition. The analysis revealed black carbon and organic carbon comprised the majority of particulate matter mass emissions, with approximately 4 % of

emissions made up of inorganic fly ash. PAH emission factors were found to vary greatly with a maximum total PAH emission factor of 452 mg/kg wood burned under one set of operating conditions. Wood type and burner operating condition were found to have a significant effect on the concentration of particle-bound PAHs released in wood smoke. Analysis of individual particle composition by SEM-EDS confirmed high carbon content in wood combustion emissions with operating condition and wood type playing significant roles in particle distribution and morphology. The results obtained provide reliable emission factors for future air quality management options and policy interventions to be assessed.

P-Sources.17 ID:4371 15:35

Interannual variability of Nitrogen compounds emission and deposition in West and Central Africa (2002-2007)

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In this study, a first attempt at estimating the interannual variability of the nitrogen emission and deposition fluxes for the years 2002 to 2007 is made, through measurements and simulations at seven stations of the IDAF (IGAC-DEBITS-Africa) network situated in dry savanna, wet savanna and forest ecosystems. The purpose of this study is to estimate the impact of the interannual variability in precipitations on the interannual variability in emission and deposition of nitrogen compounds. The interannual variability of rains between 2002 and 2007 is responsible for changes in emission (both anthropogenic and biogenic) and deposition fluxes, due to changes in soil moisture and temperature, LAI, and turbulence, and also occurrence of fires, linked to meteorological conditions. Dry and wet deposition fluxes are calculated from gas measurements of NO2, HNO3 and NH3 concentrations (from passive samplers), and from nitrate and ammonium concentrations in rain. Deposition velocities are simulated with the surface model ISBA. Organic nitrogen species are not taken into account.Emission fluxes are evaluated including NO biogenic emission from soils (simulated with ISBA), emissions of NOx and NH3 from domestic and biomass fires (calculated from satellite data), and volatilization of NH3 from animal excreta (statistical data). Meteorological conditions are provided by the forcing developed in ALMIP (AMMA Land surface Model Intercomparison Project). Long range transport of products from fires occurring in the southern hemisphere is also quantified.In dry savanna ecosystems, emission and deposition fluxes increase at the beginning of the rainy season because of large emissions of biogenic NO (pulse events), and emission of NH3 is dominated by the process of volatilization from animal excreta. Biomass burning emissions are more important in wet savanna and forest sites. This study uses original and unique data from remote and hardly-ever-explored regions.

P-Sources.18 ID:4206 15:35

Application of satellite observations for timely updates to bottom-up NOx emission inventories.

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Anthropogenic NOx emissions can change rapidly due to economic growth or control measures. Bottom-up emissions require years to compile and can become quickly outdated. We use satellite observations of tropospheric NO2 columns and the relationship between NO2 columns and NOx emissions from a GEOS-Chem model simulation to estimate the annual change in bottom-up NOx emissions. Comparison between

bottom-up and the emissions trend predicted from the SCIAMACHY observations offer consistent results demonstrating decrease in NOx emissions in western Europe and the United States and substantial increase in eastern China. Bottom-up and predicted emissions differ by < 0.3TgN/Yr (<8%) over North America, Europe, and East Asia. The emission updates offer an improved estimates of NOx that are critical to our understanding of air quality, acid deposition, and climate change.

P-Sources.19 ID:4584 **15:35**

Regional NOx Emission Inversion based on OMI observations

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The adjoint version of the community multiscale air quality (CMAQ) model of the USEPA is used for variational inverse modeling of NOx emissions using OMI observations. Emission inversion will be performed for the summer of 2007 with emphasis on the period of intensive measurements during the BAQS-Met campaign. OMI retrievals together with surface observations are used to evaluate how various sources of information would impact the inversion process. The current adjoint of CMAQ which only includes gas-phase processes is augmented by the addition of N2O5 hydrolysis to better represent NOx chemistry. Approximated lightning NOx emissions are added to CMAQ emissions to achieve better representation of tropospheric NO2. Boundary conditions are dynamically provided from GEOS-Chem to minimize the impact of boundaries on source inversion. The modeling domain covers continental US and much of Canada at a horizontal resolution of 32 km which is comparable with the resolution from OMI. Observational operators (and their adjoints) are developed for horizontal and vertical mapping of CMAQ concentration fields onto the OMI grid. Emission inversion results in posterior emission scaling factors that are applied to a priori emission estimates. Overall emission scaling factors consist of superimposed monthly and hourly emission factors where separate scaling factors are used for weekdays and weekend/holidays. This variational approach results in spatial maps of monthly and hourly NOx emission scaling factors.

P-Sources.20 ID:4234 **15:35**

Seasonal and spatial variation in nitrous oxide sources over the United States

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Nitrous oxide (N_2O) is the single-most important species depleting the stratospheric ozone layer, and it has the third-largest radiative forcing of any non-CO_2 greenhouse gas. However, nitrous oxide sources, predominantly from agriculture, are poorly constrained. STILT, the Stochastic Time-Inverted Lagrangian Transport Model, is a high resolution, particle-following model capable of assessing regional scale trace gas sources. We combine STILT with an ensemble of tall tower and aircraft measurements along with inverse

modeling techniques to quantify both the magnitude and spatial variation of nitrous oxide sources over multiple seasons. Model results show that commonly used emissions inventories may underestimate nitrous oxide sources over the United States by more than a factor of three.

P-Sources.21 ID:4294 15:35

Source attribution of the recent increase in atmospheric methane

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Atmospheric methane (CH4) is a major greenhouse gas also playing a key role in the tropospheric production of Ozone. After a decade of stagnation, the growth rate of CH4 has started to increase again after 2006.

We analyse this increase in terms of variations of the CH4 sources and sinks, using both a process-based model of wetland emissions and the inversion of atmospheric transport and chemistry. The focus is put on wetland emissions as a major contributor to the interanual variability of CH4 emissions.

Top-down atmospheric inversions allow to quantify the contribution of the different CH4 sources and their uncertainties. Bottom-up land-surface model ORCHIDEE provides insights on the processes causing the variations of wetland emissions.

In this presentation, we show the dominant contribution of tropical wetland emissions to the 2007-2008 atmospheric increase, and the significant contribution of boreal ecosystems, especially in Eurasia in 2007. Precipitations in the tropics and temperature in the northern latitudes appear to be key elements to explain the changes in CH4 emissions from wetlands in 2007-2008. We present the latitudinal and regional partition of emissions explaining the 2007-2008 changes in atmospheric CH4. The sensitivity to the inversion setup and to the meteorological forcings used in the ORCHIDEE model provide an estimation of the robustness of these results.

P-Sources.22 ID:4359 15:35

Characteristics of greenhouse gas flux from various ecosystem surface: field measurements of CH4, CO2, N2O emission from a cultivated cabbage field in Korea

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Intensively managed and cultivated soils have been known as a dominant source of atmospheric N2O through increase in use of nitrogen fertilizer and soil microbial processes, contributing to about 57% (9Tg y-1) of total N2O annual global emission. Organic carbons in soil and wetland sediment affect the CO2 and CH4 emission in such environments depending on physicochemical conditions. Major GHG (i.e. N2O, CH4, CO2) emission measurements were conducted from a cabbage cultivated field at Kunsan (35o56’23’’ N, 126o43’14’’ E), Korea by using closed static chamber during period from October 2009 to January 2010. Hourly GHG emissions mostly from 10:00 LST to 18:00 LST were measured in each measurement day

(total 18 days) during the experimental period. As soon as set-up the static chamber onto agricultural soil, 50 ml of air was drawn from inside of the chamber by syringe 3 times with 15 minute interval (e.g. 0min, 15min, 30min). GHG emission is calculated by use of a formula derived from mass balance of GHG between in and out of the chamber. Time change of the GHG gas concentration must be required to calculate the gas emission. In addition, soil parameters (e.g. soil moisture, soil temperature, soil pH, total organic in soil, soil N and C) were monitored and measured at the same site. The gas samples in syringes were analysed by gas chromatography (equipped with ECD/FID). The average soil pH during the experimental period was ~pH5.7 at the plot. The average fluxes and ranges of GHG during the experimental period were 0.002±0.09 mg-m-2 hr-1 (-0.036 ~ 0.045 mg-m-2 hr-1) for CH4, 23.854±170.61 mg-m-2 hr-1 (-73.109 ~ 139.379 mg-m-2 hr-1) for CO2, and 0.317±0.48 mg-m-2 hr-1 (-0.082 ~ 1.144 mg-m-2 hr-1) for N2O, respectively. Monthly base flux measurement results revealed that monthly mean GHG flux during October was significantly higher than those during November and January when soil temperatures during these months were significantly low. Soil temperature during the experimental periods were ranged from 0.2 to 18.8oC (17.5±1.2oC for October; 12.4±2.4oC for November; 1.1±0.7oC for January). It seems that soil temperature has significant role in controlling the soil GHG emission, and low temperature during November and January could weaken microbial activity in soil and now turn soil GHG emission decrease. Other soil parameters were also correlated with GHG emissions and discussed. Negative gas fluxes for both CH4 and CO2 were observed during these measurements, but not for N2O. CH4 and CO2 gases seem to be emitted or deposited depending on other factors such as background concentrations and physicochemical soil conditions. However, still there are uncertainties in their determination. Improvement of measurement techniques and well-understanding of relationships between gas emission and controlling factors in such environments need to be required.

P-Sources.23 ID:4585 15:35

Methane emission from natural wetlands: what are wetlands and where are they?

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Natural wetlands are the world's largest source of methane. Their emission dynamics are strongly influenced by inter-annual variations in climate and, together with wild fires, control inter-annual variations in the growth rate of atmospheric methane concentrations. About half of the world's wetlands are north 50°N where temperature, permafrost, and peaty soils dominate. Another ~40% is in low latitudes governed primarily by large-scale riverine flooding in forested environments. Wetlands display exceptional variability in vegetation, hydrological regime and seasonality which means that defining wetlands is not straightforward. In fact, there is no consensus on what a wetland is for methane or any other studies. Existing classification systems reflect regional complexes and are not globally applicable. Meaningful distinctions among wetlands, rivers and lakes are impossible in some regions and seasons which further complicates defining, mapping, monitoring and modeling wetlands.

Models simulating methane dynamics in natural wetlands, developed since the mid-1990s, are usually applied to an independent wetland data set due to difficulties in modeling the distribution of wetlands themselves although researchers are now attempting to predict methane-producing wetland distributions from climate variables.

While models evolve, wetlands remain undefined. We found that distributions and areas of wetland data sets primarily reflect the strengths and weaknesses of the instruments and approaches employed to develop them. Therefore, apparent discrepancies among data sources mostly represent methodological differences rather than true uncertainties. Nevertheless, we found that spatial patterns and strengths of simulated methane emissions are closely tied to their underlying wetland distributions strongly suggesting that uncertainties in estimating methane emission from wetlands can be reduced by improving information on methane-relevant wetland distributions. We outline development of a novel methane-centric wetland-classification system, and associated data set, designed to comprehensively characterize methane-relevant wetland ecosystems for global modeling studies.

P-Sources.24 ID:4390 15:35

Methane emission from wetlands and its role in present day and paleo climate

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Methane is the 2nd most important green house gas after CO2. At present day, among all the natural and anthropogenic sources of methane, wetland emission is the single largest source of methane and represents ~ 20-45% of total emissions (~500 Tg). Even under preindustrial conditions methane emission from wetland contributed majorly to a large share of atmospheric methane concentration. Despite their importance considerable uncertainties exist in the quantification of current wetland emissions as well as their distribution on the global scale. The wide spectrum of vegetation covers and hydrological regimes in the wetland characteristics make it difficult even to define wetlands unanimously. In order to specify wetland emission in a chemistry climate model like ECHAM5-MOZ that would also be used in paleo-simulations, a simplified parameterization is needed. Here we present ongoing work on the development of such a wetland emission model. In the first step, high resolution land cover data of vegetation, terrain slope gradient and soil moisture storage capacity along with precipitation areas are used to identify potential wetlands in the present day climate. For each land cover type, threshold values favorable for wetland formation are identified after comparison with available wetland maps. This rapprochement leads to a well constrained potential global wetland map for the present day as well for paleo climate modeling where the input data are available in coarser resolution. In the next step, heterotrophic respiration (a function of soil temperature and available decomposable carbon) is calculated for the potential wetland areas and used to parameterize the strength of methane emissions. The new wetland emission model will be used to obtain the atmospheric concentration of methane in present and paleo climate in a series of comprehensive chemistry climate simulations with ECHAM5-MOZ.

P-Sources.25 ID:4541 15:35

Estimating Australian wetland methane fluxes using train-borne data interpreted using an ensemble Kalman filter

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We estimate Australian wetland emissions using methane concentration measurements from an in situ gas analyzer on board the Ghan train that runs between Adelaide (34.9S, 138.6E) and Darwin (12.5S, 130.9E). To date, five measurement campaigns have been held with the instrument, which is also capable of measuring CO2, N2O, CO, and δ13CO2. The measurements near Darwin suggest local emissions from

wetlands that are not well characterized in bottom-up emissions inventories. Our initial focus is on the three measurement campaigns that took place in February, March, and September 2008, sampling both the wet and dry seasons near Darwin.

We will present comparisons between the GEOS-Chem global 3-D transport model and measurements along the train route, as well as between the model and a ground-based Fourier transform spectrometer installed at Darwin as part of the Total Carbon Column Observing Network (TCCON). We will also present surface flux estimates of methane inferred from the model in conjunction with an ensemble Kalman filter.

P-Sources.26 ID:4481 15:35

The sensitivity of Canada's atmospheric methane observational program to detect Canadian wetland sources.

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The long lifetime and rapid mixing of CH4 in the atmosphere provides a large scale integration of surface fluxes. With sufficient measurement precision and surface coverage (measurement sites), a signature of individual surface source or sink regions can be detected and quantified. For example, preliminary results will be shown from a study to quantify Canada's wetland CH4 budget using atmospheric CH4 measurements and 4DVAR modelling [applying prior gridded source patterns, an atmospheric transport model (TM5) and analyzed wind fields (ECMWF)]. The global analysis was done using CH4 data from NOAA's global air sampling network in addition to continuous CH4 measurements from 4 of Environment Canada's 12 observational sites. The figures show gridded wetland fluxes for 2004 (~ 7.5 Tg) and 2006 (~12Tg). The presentation will also include annual results for the period of 2003 to 2008, thus demonstrating the potential to track wetland source patterns for variations in size and distribution. Possible correlations/links to climate indicators such as temperature and precipitation will be explored as well.

P-Sources.27 ID:4434 15:35

Spatial and temporal variations of atmospheric methane and carbon dioxide observed in Environment Canada's GHG measurement network.

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Environment Canada
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Environment Canada conducts high quality ground-based atmospheric measurements of GHGs from coastal, interior and Arctic regions in Canada. The current network of 12 stations is strategically located to catch the regional atmospheric signals of carbon fluxes from major ecosystems over Canada and reflect the long-range transport of GHGs into and out of Canada. These measurements document the spatial and temporal distributions of GHGs in Canada, providing essential constraints to our understanding of Canada's natural and anthropogenic sources and sinks of GHGs.

An analysis of the spatial distribution of atmospheric CO2 and CH4 extent utilizing backward trajectory calculations is demonstrated. The results show that methane and carbon dioxide sources have the potential to

affect regions covering several hundred kilometers. Temporally (synoptic, seasonal and inter-annual variations), atmospheric observations of CO2 and CH4 clearly provide information on controlling processes in important ecosystems such as the boreal forest and wetland regions in Canada and their possible response to changes in climate.

P-Sources.28 ID:4507 **15:35**

ECCAD : Emissions of Atmospheric Compounds and Compilation of Ancillary Data

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There is considerable uncertainty in estimating surface emissions of atmospheric compounds for the recent past and up to now, no single data set exists which would describe the geographical and temporal distribution of emissions for all species relevant to air quality, atmospheric composition change and climate change in a comprehensive and consistent manner. We will describe the ECCAD project, which has two major goals. The first goal is to provide scientific users with an easy access to a large number of existing datasets on surface emissions of atmospheric compounds at the global and regional scales. We are also providing access to ancillary data required to quantify surface emissions. The other goal is to provide data manipulation tools as well as statistical information over the different regions (climatic regions, continents, oceans, OECD regions, etc.). These tools allow an easy identification of each dataset characteristics and differences between the datasets. Visualization tools are also provided, and the users can download the data of interest. The data in ECCAD are currently used in different international projects. The most recent addition to the database include the emissions developed in support of the IPCC AR5 report, and the data used in the MACC and CITYZEN European projects. The emissions are provided as gridded data at a 0.5 or 1° spatial resolution. Time periodicity and temporal coverage periods vary among the datasets from one year to up to 150 years. The database will be described, and examples of the tools will be presented. We will also provide information on the access to the datasets.

P-Sources.29 ID:4190 **15:35**

The CIERA US Initiative on Emissions

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While emission inventories at a variety of spatial and temporal scales are critical inputs to the understanding

and prediction of air quality and climate, inconsistencies in the methodology and structure of these inventories have hindered research progress. Systematic inventory evaluations and quantification of emission uncertainties and their impacts are crucial to establish confidence in these datasets.

We will present the Community Initiative for Emissions Research and Applications (CIERA), which is under development in the USA. The goals of the work planned within this multi-agency project are to harmonize global and regional emission inventory development and to improve the exchange of emission inventory data. CIERA will facilitate the evaluation of inventories produced using a variety of methods, the investigation of the impacts of emission uncertainties and changes, and the use of these findings by the community.

We will discuss the motivation for organizing this collaborative initiative and the first steps taken. We will present plans for developing the CIERA web-based distributed data system. We will also encourage the international community to join the CIERA effort and describe how CIERA is already working with the Global Emissions Inventory Activity (GEIA).

P-Sources.30 ID:4222 15:35

The Global Emissions Inventory Activity (GEIA)

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The objective of the GEIA project is to bring together people, analyses, data, and tools to quantify the anthropogenic emissions and natural exchanges of trace gases and aerosols that drive earth system changes and to facilitate use of this information by the research, assessment and policy communities. This presentation will provide an overview of the current activities of GEIA.

We will present the GEIA network, which currently includes over 1000 people around the globe, and the plans to extend this network to different communities working on environmental changes issues. We will also present our plans to develop within the GEIA Center (www.geiacenter.org) a new database of scientific papers as well as national and international reports dealing with emissions issues. The GEIA conference on emissions took place in October 2009 in Oslo, Norway; we will present the main conclusion of the conference.

Given the differences often found among data sets, we will also discuss the formation of a GEIA working group composed of emission developers and modelers who will compare data sets and implications for modeling. We will discuss how this activity will include groups using chemistry-transport and chemistry-climate models, in order to demonstrate the implications of the differences.

It has been recognized that consistent information on emissions at the global and regional scale is required, as well as an accurate quantification of emissions in the different megacities of the world. In order to develop these perspectives, GEIA will begin strengthening links with different key regions: we will discuss the plans for developing regional centers through corporation with other entities and individuals working in these regions.

P-Sources.31 ID:4456 15:35

Towards regional-scale modelling of industrial CO2 emissions

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Anthropogenic and biogenic fluxes of CO2 are essential factors for the evolution of atmospheric CO2 concentrations on both short- and long-term scale. A possible approach to study of the surface fluxes is to use inverse modelling techniques together with chemical transport models. A regional-scale modelling system can bring new insights into the relations between surface fluxes and atmospheric CO2 concentrations with more complex physics and much finer time scale than usual global models used for climatological studies.

A regional-scale modelling system is currently being developed for simulating transport of greenhouse gases in Northern Alberta. The system will directly relate both ground- and satellite-based observations with regional surface fluxes, using both forward and inverse modelling. One of the goals of inverse modelling is to evaluate the feasibility of the system to estimate major anthropogenic point sources of CO2 associated with heavy industrial development in Northern Alberta.

P-Sources.32 ID:4211 15:35

Spatial distribution of emission factor and budget of carbonaceous aerosols from domestic bio fuel over Indo Gangetic plain

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Biomass is a major source of energy over rural sector of south-east Asia. On global basis the major source of carbonaceous aerosols (organic (OC) and elemental carbon (EC)) is biomass burning. As a part of determining emission factor of trace gases and aerosol from domestic fuels, bio fuel samples are collected along with information on socio-economic parameters at district level over Indo-Gangetic plain (IGP). Based on this information, domestic bio fuel consumption patterns have been determined at district level over IGP of India. The study reveals that the dung cake is the major biomass used as domestic fuel for cooking purpose followed by fuel wood and crop residue at the rural sector of India. Using these consumption pattern and crop production data, refined consumption value of crop residue as bio fuel has been estimated. To determine the emission factor we have simulated the burning using the collected bio fuel samples. The emission factor of EC and OC from dung cake, fuel wood and crop residue over Uttar Pradesh and Delhi are computed as [(0.093 ± 0.07, 4.77 ± 1.37g/kg), (0.132±0.132, 1.51±1.52 g/kg), (0.114±0.091, 2.32±1.69 g/kg)] and [(~0.07, ~0.51 g/kg), (~0.07, ~ 1.07 g/kg), (~0.1, ~4.48 g/kg)] respectively. Similarly for Punjab and Haryana the emission factor of EC and OC from fuel wood and crop residue are computed as [(0.201±0.09, 0.39±0.11 g/kg), (0.201±0.02, 3±2.4 g/kg)] and [(0.18±0.04, 0.69±0.05 g/kg), (0.14±0.05, 1.15±0.1 g/kg)]. The budget for EC and OC from dung cake, fuel wood and crop residue of Uttar Pradesh is determined as [(3.46, 177 Gg/Y), (2.19, 25.06 Gg/Y), (0.09, 1.74 Gg/Y)] and for Punjab and Haryana budget of EC and OC from fuel wood and crop residue are determined as [(0.38, 0.74 Gg/Y), (0.15, 2.25 Gg/Y)] and [(0.31, 1.17 Gg/Y), (0.13, 1.10 Gg/Y)] respectively. Similarly other parts of Indo-Gangetic plain will be presented.

P-Sources.33 ID:4569 **15:35**

Inverted Terrestrial CO2 Fluxes and Their Sensitivity to Climatic Conditions

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On the basis of our previous inversion using GlobalView-CO2 data for 30 regions of the North America and 20 regions for the rest of the globe, we improved the inversion results through considering the diurnal variations of both the atmospheric boundary layer and the CO2 flux of the terrestrial ecosystem. 7-year (2001-2007) CO2 concentration observations have been used to infer the monthly CO2 flux with a time-dependent Bayesian synthesis inversion scheme. Preliminary inversion results of CO2 fluxes of these 50 regions show considerable temporal variations among years. These variations are found to be closely related to climate variations, indicating that atmospheric inversion is an effective tool in assessing the interannual variation of the terrestrial carbon flux at regional scale. The appropriate responses of the inverted terrestrial carbon fluxes to the seasonal climatic conditions imply that considering the seasonal climatic conditions rather than the annual means in a terrestrial carbon model and a coupled carbon-climate model could improve our ability to simulate and predict the feedbacks of the terrestrial carbon cycle to climate change.

P-Sources.34 ID:4325 **15:35**

Constraints on CO2 flux emissions: reconstructions of in-situ measurements from Lagrangian stochastic inversion

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Uncertainties in the CO2 fluxes limit the validation of emission mitigation policies and of process level understanding of the carbon cycle science. We have performed diffusive Lagrangian reconstructions of in-situ CO2 measurements in order to obtain constraints of CO2 fuxes. Data has been collected during the CONTRAIL aircraft campaign and from ground/tower sites, nearby the CO2 emission hot spot of the Tokyo Bay Area. Advective transport based on analyzed meteorological winds is characterized by the sensitivity/transition probability (Green's) function of transport allowing direct comparison with observations via the reconstruction of the volume mixing ratio of CO2. Sensitivity to different simplifed boundary layer representations, turbulent mixing representations and meteorological felds is firstly studied and applied to the assessment of published inventory data. We employing the WRF- Chem model for high-resolution meteorological data products as well as Eulerian reconstruction of the CO observations.

P-Sources.35 ID:4409 **15:35**

Inverse modeling of atmospheric CO_2 and CH_4 using a fixed-lag Kalman smoother and an atmospheric transport model

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An inverse modeling system consisting of a fixed-lag Kalman smoother and a NIES off-line atmospheric transport model, was developed in order to perform sequential estimations of regional CO_2 and CH_4 surface fluxes. It utilizes several months of the observational data to estimate monthly regional fluxes, which in turn are incorporated into the background states of tracers at the next time step (Bruhwiler et al., 2005). Thus, the system allows us to estimate long-term fluxes, with considerably reduced computational costs as compared to batch inversions with pre-calculated regional response functions. We tested the system for CO_2 with a basic inversion setup for 22 terrestrial and oceanic regions based on TransCom experiment (e.g. Gurney et al., 2004). Our results show that estimated fluxes falls within the priors' range of uncertainty, in most of the months and the regions such as the northern hemispheric summer and the southern hemispheric land regions. The estimated Eurasian fluxes in winter were increased beyond the prior uncertainties, which indicated the need to improve constraints for these regions. The ocean regions were generally heightened their seasonal amplitudes. In general the estimated fluxes were reasonable when comparing them with estimations of TransCom experiments. We apply the system to estimate CO_2 and CH_4 fluxes from the 1990's to the present using analyzed data set of GLOBALVIEW-2009. CO_2 fluxes for 66 regions are estimated by using interannually varying fossil fuel emissions. For CH_4 we target flux estimation of 44 terrestrial regions, owing to the dominance of CH_4 sources. Simple parameterizations of chemistry to represent atmospheric CH_4 sink were included in the model. A preliminary trial of the flux inversions with GOSAT XCO_2 observations for the year 2009 will be discussed.

P-Sources.36 ID:4396 15:35

The effect of monthly anthropogenic CO2 emissions on the seasonal cycle of atmospheric CO2

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A previous study (Erickson et al. 2008) approximated the monthly global emission estimates of anthropogenic CO2 by applying a 2-harmonic Fourier expansion with coefficients as a function of latitude to annual CO2 flux estimates derived from United States data (Blasing et al. 2005) that were extrapolated globally. These monthly anthropogenic CO2 flux estimates were used to model atmospheric concentrations using the NASA GEOS-4 data assimilation system. Local variability in the amplitude of the simulated CO2 seasonal cycle were found to be on the order of 2-6 ppmv. Here we used the same Fourier expansion to seasonally adjust the global annual fossil fuel CO2 emissions from the SRES A2 scenario. For a total of four simulations, both the annual and seasonalized fluxes were advected in two configurations of the NCAR Community Atmosphere Model (CAM) used in the Carbon-Land Model Intercomparison Project (C-LAMP). One configuration used the NCAR Community Land Model (CLM) coupled with the CASA´ (carbon only) biogeochemistry model and the other used CLM coupled with the CN (coupled carbon and nitrogen cycles) biogeochemistry model. All four simulations were forced with observed sea surface temperatures and sea ice concentrations from the Hadley Centre and a prescribed transient atmospheric CO2 concentration for the radiation and land forcing over the 20th century. The model results exhibit differences in the seasonal cycle of CO2 between the seasonally corrected and uncorrected simulations. Moreover, because of differing energy and water feedbacks between the atmosphere model and the two land biogeochemistry models, features of the CO2 seasonal cycle were different between these two model configurations. This study reinforces previous findings that suggest that regional near-surface atmospheric CO2 concentrations depend strongly on the natural sources and sinks of CO2, but also on the strength of local anthropogenic CO2 emissions and geographic position. This work further attests to the need for remotely sensed CO2 observations from space.

P-Sources.37 ID:4246 **15:35**

Quantifying the impact of model errors on top-down estimates of carbon monoxide emissions using satellite observations

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Inverse modeling is a powerful tool for combining observations of atmospheric composition with knowledge of atmospheric processes to estimate trace gas emissions. In this context, there has been much effort on inverse modeling of atmospheric carbon monoxide (CO). However, significant discrepancies exist in the inferred “top-down” emission estimates of CO, reflecting the sensitivity of the inversion analyses to the inversion configuration employed in the analyses and to biases in the atmospheric models. In this work, we examine the potential impact of four different types of systematic model errors on the inferred CO sources. We assess the impact of aggregation errors on the source estimates, associated with conducting the inversion at a lower resolution than the atmospheric model. We also quantify the impact of errors in atmospheric transport and in the atmospheric OH abundance in the model on the source estimates. Inversion analyses of CO typically linearize the CO chemistry by specifying a fixed distribution of OH to minimize the computation cost of the inversion. We show that if the imposed OH field is inconsistent with the atmospheric forward model, the linearization error in the chemistry can bias the source estimates. Atmospheric CO is emitted directly to the atmosphere as a by-product of combustion and it is produced from the oxidation of methane (CH4) and nonmethane volatile organic compounds (NMVOCs). We also show that the inferred top-down source estimates are sensitivity to how these chemical sources of CO are specified in the inversion.

P-Sources.38 ID:4251 **15:35**

Evaluation of dynamic processes in the tropical troposphere in assimilated meteorological fields using carbon monoxide data from the Aura satellite and GEOS-Chem model

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We use the GEOS-Chem model to interpret the spatial and temporal variations of tropical tropospheric CO observed by the Microwave Limb Sounder (MLS) and the Tropospheric Emission Spectrometer (TES). We apply these data to diagnose and evaluate transport in the GEOS-4 and GEOS-5 assimilated meteorological fields that drive the model. Our analysis focuses on the biomass burning seasons, and on the influence of vertical mixing at the start of the wet season. Over South America, both models reproduce the timing of the observed CO maximum in the lower troposphere (LT). In the upper troposphere (UT), the model's seasonal maximum of CO with GEOS-4 occurs about 1 month late, and with GEOS-5 it occurs even later; in both it is too broad compared to MLS data. Our analysis suggests that these deficiencies are caused by two factors: deep convection decays at too low an altitude, and the source of CO from isoprene in the model is too large early in the wet season. The lag in GEOS-5 is greater in part because convection decays at a lower altitude, and in part because convection moves southward later than in GEOS-4. Over southern Africa, model simulations driven by GEOS-4 and GEOS-5 match the phase of the observed CO variation throughout the troposphere fairly well, reflecting reasonable meteorological patterns. Isoprene does not contribute significantly to the seasonality of CO in the UT in this region. Over northern Africa, both models match the phase and magnitude of the CO variation throughout the troposphere except at 215 hPa, where MLS does not show a strong seasonal cycle, while it is pronounced in the models. The MLS data imply that the upward

transport of CO to the UT is too strong over the Gulf of Guinea in December and January.

P-Sources.39 ID:4166 15:35

Interannual variation of VOCs oxidation sources of atmospheric carbon monoxide: Joint inversion approach using concentration and isotopic ratio information

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We have simulated atmospheric carbon monoxide and its isotopes with MOZART 4 for 1997 through 2004. The sources of CO was optimized for each year using NOAA GMD CO ground measurement. Plus, in 1997, 1998 and 2004, the sources of CO were jointly constrained by concentration and isotopic ratio information. Usually, isotope ratio measurements provide an additional constraint for the inversion analyses since each source of CO has different isotopic source signature. Thus, this enables us to reduce the uncertainties of the source estimates and to verify concentration-only inversion results.

In this study, we focused on the interannual change of VOCs oxidation source of CO which directly affected by the climate change such as ENSO event. We found that VOCs emissions, especially for the isoprene which takes more than 70% of the VOCs oxidation source, are closely connected with ENSO events and the Earth surface temperature change. Moreover, our inversion analyses directed the interannual variation of VOCs emissions are larger than the previous estimates.

P-Sources.40 ID:4353 15:35

Improving the global tropospheric methanol budget through inverse modelling of spaceborne IASI methanol columns and in situ data

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New insights into our understanding of methanol sources and sinks in the global troposphere are brought forward by recent measurements of tropospheric columns of methanol retrieved from the IASI satellite sensor. These data are expected to reduce the uncertainties in the knowledge of the methanol distribution in the troposphere, thanks to the unprecedented global spatiotemporal coverage offered by the IASI instrument. According to current estimates, the ocean biosphere and the terrestrial growth source represent two thirds of the total methanol source, whereas plant decay, atmospheric production, anthropogenic and biomass burning sources account for the remainder. In this study we use the IMAGESv2 global chemical transport model and its adjoint module, in order to interpret the new dataset and derive updated methanol source strengths which bring the model predictions closer to the observed abundances. The resulting fluxes are evaluated against an extensive compilation of air- and ground-based methanol measurements, and the implications for the atmospheric chemistry are discussed.

P-Sources.41 ID:4533 15:35

CARIBIC passenger aircraft measurements of the distribution, seasonal cycle, and variability of acetone in the UTLS

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The CARIBIC platform uses a Lufthansa Airlines Airbus A340-600 that is equipped with dedicated air inlet system. Equipment in the monthly deployed 1.5 ton measurement container records aerosol sizes and ~100 trace gases at 10-12 km cruising altitude. At the same time aerosol and air samples are collected in flight. This poster presents the recently constructed budget of acetone (measured by the PTRMS system) in the upper troposphere and lower stratosphere. More than 100 measurement flights resulted in the largest airborne dataset of acetone from the UTLS. The following findings are described: • A strong seasonal variation of acetone occurs at the mid-latitude tropopause with maxima of ~900 pptv in summer and minima of ~200 pptv in mid-winter. • This seasonality propagates into the LMS with rapidly decreasing concentrations and increasing phase shift reaching ~6 weeks 2 km above the tropopause. • Probability density functions (PDFs) and the course of the seasonal variation of acetone relative to the tropopause are interpreted regarding the in-mixing and subsequent dispersion of acetone in the LS. • The production of HOx due to the photolysis of acetone is compared with the one following the photolysis of ozone and subsequent reaction of O(1D) with water vapour.

P-Sources.42 ID:4318 15:35

Evaluation of isoprene flux and its impact on oxidants and inorganic aerosols in East Asia

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As a major precursor of the ozone and SOAs (secondary organic aerosols) formations, biogenic species are of primary importance in the atmospheric chemistry. Isoprene influences the level of inorganic aerosols (i.e. sulfate and nitrate) by controlling OH radicals. However, isoprene fluxes are highly uncertain in East Asia. While isoprene fluxes from the GEIA (Global Emissions Inventory Activity) and POET (Precursors of Ozone and their Effects in the Troposphere) inventories estimate approximately 20 Tg yr^{-1} in East Asia, those from the MEGAN (Model of Emissions of Gases and Aerosols from Nature) and MOHYCAN (MOdel for Hydrocarbon emissions by the CANopy) estimate approximately half of the GEIA and POET inventories. In order to evaluate and/or quantify the magnitude of the isoprene fluxes over East Asia, the HCHO columns obtained from the GOME (Global Ozone Monitoring Experiment) observations were compared with the HCHO columns from the CMAQ (Community Multi-scale Air Quality) simulations over East Asia. In this study, US EPA Models-3/CMAQ v4.5 modeling using the ACE-ASIA (Asia Pacific Regional Aerosol Characterization Experiment) emission inventory for anthropogenic pollutants and GEIA, POET, MEGAN, and MOHYCAN emission inventories for biogenic species was carried out in conjunction with the Meteorological fields generated from the PSU/NCAR MM5 (Pennsylvania state University/National Center for Atmospheric Research Meso-scale Model 5) model for the summer episodes of the year 2002. In addition to an evaluation of the isoprene fluxes, we investigated the impact of the uncertainty in biogenic emission inventory on inorganic aerosol formations and variations of oxidants (OH, O_3, and H_2O_2) in East Asia. The results show that isoprene fluxes from the GEIA, POET, and MEGAN are overestimated, particularly over South China. Also, differences in biogenic emission fluxes lead to changes

in the levels of sulfate and nitrates by changing the OH radical concentrations.

P-Sources.43 ID:4185 **15:35**

The Relationship Between 0.25-2.5 um Aerosol Emission and CO2 Emission Above the City of Stockholm

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The effects of aerosols on climate and health are currently subjects of intense research in environmental science. In particular urban aerosol is a topic of current interest, as cities act as net sources for aerosols. Very few studies have presented direct emission measurements in cities using micrometeorological methods like the Eddy correlation. Such measurements provide important quantitative information about the sources of the particles (Dorsey et al., 2002; Mårtensson et al. 2006; Schmidt, and Klemm, 2008). In this study we present direct measurements of non-exhaust particles in order to better understand the processes governing the emissions. Size-resolved vertical aerosol number fluxes of particles with diameters of 0.25-2.5 µm were measured with the eddy covariance method from a 118 meter high communication tower over the city Stockholm, Sweden. Based on simultaneous measurements of the flux of CO2, we have estimated emission factors in relation to in relation to the vehicle fuel consumption. If fleet composition and other factors are similar, this can be extrapolated to other cities since fuel statistics is usually available. If we assume the mean fuel consumption and considering the vehicle fleet mix in Stockholm (mainly gasoline vehicles), and a particle density of 1500 g cm-3 we obtain an annual mass emission factor for PM2.5 is with 0.025 [g veh-1·km-1]. This is similar to the value 0.027[g veh-1 km-1] derived for PM2.5-0.6 based on road tunnel measurements in Stockholm by Kristensson et al., (2004). It is somewhat lower than the value 0.046 [g veh-1 km-1] estimated for Hornsgatan based on gravimetric PM2.5 measurements in the street canyon and at roof using NOx as tracer in Stockholm presented by Ketzel et al., (2007). The higher value for Hornsgatan compared to the fleet average seen by the eddy covariance measurements is expected due to the different road and traffic conditions.

P-Sources.44 ID:4423 **15:35**

Modelling combustion aerosols using updated fossil fuel and biofuel emission inventories for 2005 and 2030.

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African regional fossil fuel/biofuel emission inventory for particulates has been developed at resolution of 0.25° x 0.25° for 2005. The original database of Junker and Liousse (2008) was used after modification with updated regional fuel consumption and emission factors. Consumption data were corrected after direct inquiries over Africa, including a new emitter category i.e. two-wheel vehicles (“zemidjans”) and a new activity sector i.e. power plants, both not being considered in the previous Junker and Liousse (2008) emission inventory. Emission factors were measured during the 2005 AMMA campaign (Guinot et al., 2010) and combustion chamber experiments. Prospective inventories for 2030 are based on this new regional inventory and energy consumption forecasts by POLES model (Criqui, 2001) for a reference scenario, with no emission controls after 2003, and for a "clean" scenario where possible and planned political measures for emission control are assumed effective. BC and OCp emission budgets will be discussed and compared to previous global dataset and new IPCC inventories (Lamarque et al., 2010). These new inventories associated with most recent biomass burning inventory (Liousse et al., 2010) were tested in the ORISAM-TM5 global chemistry-climate model with a focus over Africa at a resolution of 1° x 1° resolution. For this purpose, a global simulation was performed for 2005 and for 2030, including a 6-bin sectional aerosol module, comprising BC and primary OC. Modelled particulate concentrations were compared to available measurements in African cities (e.g. Bamako, Cotonou and Dakar) together with modelled aerosol optical depth values with Aeronet and PARASOL and MODIS satellite data. Major trends observed over African towns are better reproduced by our global model supplemented with our regional inventory. Sensitivity tests driven by different emission scenarios will be presented.

P-Sources.45 ID:4212 15:35

Regional aerosol pollution potential of major population centers

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Emissions from major population centers (MPCs), including megacities, are becoming more and more important for the global burden of air pollutants. Once emitted to the atmosphere, aerosols from MPCs are expected to show differences in their atmospheric dispersion, depending on the aerosol size and the geographical location of the MPC, which will result in differences in their potential to pollute the surface and the atmosphere locally and at downwind locations.

Several simulations of monodisperse passive aerosol tracers with sizes of 0.1, 1.0, 2.5, and 10.0 µm are conducted with the global chemistry circulation model EMAC (ECHAM5-MESSy-Atmospheric-Chemistry). Several MPCs around the world were chosen as point sources with the same total, constant emission flux which allows us to compare how different source locations pollute the atmosphere and the surface on different horizontal scales. All tracers undergo transport, dry as well as wet deposition; sensitivity simulations with different assumptions about the aerosol solubility are performed.

The analysis focuses on major transport pathways, either to nearby or remote lower atmospheric levels, or to the upper troposphere, and on the deposition location, strength, and kind of deposition. Larger particles tend to more effectively result in local pollution buildup and to be deposited in greater amounts near their sources, whereas smaller particles are transported more effectively to remote locations, at both low and high altitudes. The 0.1 and 1.0 µm radius aerosols tend to behave similarly in terms of transport and deposition, but show differences concerning the aerosol solubility, while the 2.5 µm aerosols represent a transition between the behaviour of the 1.0 and 10 µm size class.

The analysis presented here provides an effective comparison of how individual MPCs pollute themselves and their environment, particularly in light of differences in regional geographical and meteorological characteristics.

P-Sources.46 ID:4531 **15:35**

Assimilation Of Air-borne Aerosol Measurements In Central Brazil During The 2007 Burning Season Using The Regional CCATT-BRAMS Model

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In Central Brazil every year large areas of forest, cerrado and pasture land are burned emitting primary carbonaceous aerosols into the atmosphere. These interfere with the radiative budget, affecting directly the amount of solar radiation that is reflected and scattered back to space, and partly absorbed by the atmosphere. The long wave terrestrial radiation is also affected. Aerosols also affect cloud microphysics acting as cloud condensation nuclei (CCN), modifying the precipitation pattern and the cloud albedo. During the South American burning season, aerosols have a significant impact on local and regional air quality affecting visibility and human health by particle inhalation causing pulmonary diseases. The recognition of atmospheric aerosol's importance led to the necessity of quantifying and optimizing its sources, distributions and sinks in the troposphere to improve simulations with atmospheric chemical models. An improvement of the aerosol representation in such models can be achieved through data assimilation methods including available aerosol observations. In this study an assimilation system was used based on the three dimensional variational data assimilation method (3D-VAR) using the Coupled Chemistry-Aerosol-Tracer Transport – Brazilian Regional Modeling System (CCATT-BRAMS). The assimilated observations are mass concentration (MC) of particular matter smaller than 2.5 µm (PM2.5). The observations were collected in October 2007, during the Cloud-Aerosol Interaction Measurements (CLAIM) campaign, which took place in the region of the city Alta Floresta in the state of Mato Grosso, Brazil. The measurements of PM2.5 were collected by a DATARAM instrument aboard the aircraft that collected information with 10s or 30s frequency. During the campaign 17 flights were carried out, 13 of which could be used in this study. In this paper we present the data and its assimilation results, evaluate its improvement of the model simulation and discuss its general impact on the chemical state of atmosphere.

P-Sources.47 ID:4422 **15:35**

Variations in the atmospheric elemental carbon concentrations from ~1100 to 2005 AD

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Elemental or black carbon (EC or BC) aerosols or soot absorbs solar radiation causing heating at the top of the atmosphere and dimming of light at the surface. Currently, global atmospheric burdens of EC are estimated from models which use energy consumptions and emission factors for various technologies use in burning fossil fuel, natural forest fires, or biofuel. These models have large uncertainties due to limited information on both the inventory of the fuel consumed and the degree of incomplete combustion which produces EC. Large uncertainty also exists in the contribution from forest fires and biomass burning. Field data needed to verify the models are sparse. We report here a method we have developed to retrieve atmospheric EC concentrations, [EC]atm, for the past ~1000 years using bottom lake sediments. EC-bearing aerosols emitted into the atmosphere are deposited into the lake sediments via wet and dry deposition. It can be shown that for a given lake the EC concentration in the sediment, [EC]sed, is related to [EC]atm, by the

expression, [EC]sed = K[EC], where K is a constant. In the absence of necessary information for several variables K can not be calculated from first principles. We have measured monthly [EC]atm at Whiteface Mountain, NY, from July 1978 to December 2005. Furthermore, we collected sediment cores from two nearby lakes, sectioned cores in thin slices, freeze dried, and determined the time of deposition or ages of various sections by 210Pb dating technique. This technique provides the ages and sedimentation rates accurately for about 150 years. Assuming a constant sedimentation rate we extended the ages to around 1100 AD. The value of K was determined from the [EC]atm and [EC]sed measurements for the 1978 -2005 period. This K was used to determine the [EC]atm from [EC]sed. The data for the ~1900 to 2005 period was compared with the model calculations of Novakov et al [2003] {GRL 30, doi 10.1029/2002GLO16345}. From around 1100 to 1850 the [EC]atm showed small variations. This data can be used to deduce information on forest fires. Data for the 1850 to 2005 is generally consistent with the industrialization.

P-Sources.48 ID:4442 15:35

Intercomparison of measurement methods for elemental carbon in atmospheric aerosol samples at an urban site in Havana Cuba

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There is no generally accepted standard method for the measurement of black carbon (BC) or Elemental carbon (EC). The EC and BC concentrations are method specific and can differ widely (Schmid et al., 2001, ten Brink et al., 2004). In this study measurement methods (both thermal and optical) for EC (BC) in atmospheric aerosol were compared at an urban site in the city of Havana Cuba. Filter samples were collected on quartz microfiber filters and were analyzed for elementac carbon by thermal methods: a modified Cachier method (Cachier et al., 1989), a thermal-optical method (Schmid et al., 2001), for BC with an optical method; integrating sphere method (Hitzenberger et al., 1996) and total carbon (TC) with a combustion method (Puxbaum and Rendl et al., 1983). There is no big difference in the EC concentration measured by Sunset and Cachier method; however TOT gives relatively 20-60 % higher values than sunset and Cachier methods. The brown carbon (measured by Integrating sphere method) is also a major reason in variation of EC values.

Key words: Elemental carbon, brown carbon, atmospheric aerosol measurement

P-Sources.49 ID:4281 15:35

Decreases in elemental carbon and fine particle mass in the United States

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Reductions in black carbon can be an important policy tool for rapid reductions in radiative forcing. Observations at IMPROVE sites, mostly remote locations, show that average elemental carbon and fine particle mass concentrations in the United States both significantly decreased between 1990 and 2005. The internal consistency of the IMPROVE network data will be examined to understand the confidence in these trends. These trends show that emissions controls for black carbon can be effective but that the room for future decreases may be smaller than commonly assumed. Despite the reductions in black carbon, the decreases in non-absorbing particles probably imply a less negative aerosol radiative forcing from US emissions in 2005 than in 1990.

P-Sources.50 ID:4149 15:35

Bacteria in the global atmosphere: estimation of sources and concentrations

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Bacteria are ubiquitous in the atmosphere. It has been proposed that their presence in the atmosphere may impact cloud development, atmospheric chemistry, and microbial biogeography. We present the major results of two recently published papers on airborne bacteria and compare existing approaches to bioaerosol emissions parameterizations.

In Burrows et. al (2009a), we review published observations of total bacteria concentrations in different ecosystems. We find that observations are lacking for some ecosystems and discuss some of the measurement issues that make the interpretation of many published studies difficult.

In Burrows et al (2009b), we simulate the global transport of bacteria, represented as 1 μm and 3 μm diameter spherical solid particle tracers in an atmospheric general circulation model including parameterized deposition processes. By combining simulated transport and emissions, we estimate the source strength of bacterial emissions to the atmosphere, as well as the uncertainty resulting from lack of constraining data.

Other recent studies also make initial steps towards inclusion of bioaerosols into global atmospheric models. Heald and Spracklen (2009) estimate emissions of fungal spores as a function of leaf area index and atmospheric water vapor concentrations. Jacobson and Streets (2009) parameterize bioaerosol emissions as functions of turbulent kinetic energy, snow cover, and relative humidity (for fungal spores). We will present a comparison of these different approaches to bioaerosol emission parameterizations.

P-Sources.51 ID:4328 15:35

Comparison of the distirbution of mineral dust calculated by a regional chemical transport model with the Mie scattering Lidar observations in East Asia

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We are now conducting forecast calculation of chemical species by using a regional chemical transport model WRF (weather research and forecasting) /Chem and a global chemical transport model CHASER (chemical atmospheric general circulation model for study of atmospheric environment and radiative forcing) every day. The lateral boundary and the initial condition of the WRF/Chem is updated every 3 hours from the CHASER's output. An experimental phase for this model system began in July of 2006 and has provided 15-hour forecasts of the distribution of ozone concentration over the Kanto region four times daily.

We are now trying to include mineral dust aerosols in the model, because mineral dusts have a large impact on the radiation in East Asia in spring. In the present study we use WRF/ARW (Advanced Research WRF) version 3.1.1, and the gaseous and aerosol chemistry is based on RADM2 and MADE/SORGAM, respectively. Dust flux is estimated based on Shaw et al. (2008), and slightly modified to adapt to the East Asia following to Uno et al. (2004). Gravitational settlement of mineral dust is based on GOCART (Goddard Chemistry Aerosol Radiation and Transport) model. Landuse is based on the MODIS land-use data. For the evaluation of the model, we conducted one month calculation for May 2007. The model well captured the dust event during 25-26 May observed by the Mie scattering Lidar at Fukue in Japan.

P-Sources.52 ID:4347 **15:35**

Long-range transport of dust aerosols over Indian region – A study using satellite data and mesoscale model

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Dust, which is a common aerosol type over the deserts, emitted by wind erosion in arid and semiarid areas, is considered to be one of the major sources of tropospheric aerosol loading. Desert dust can be transported by the mean wind to thousands of kilometers away from the source regions; this transport plays an important role in the regional and global radiative balance both at the top of the atmosphere (TOA) and at the surface. In the present study a wintertime intense dust storm occurred during the period of 19-24 February 2008 with stronger intensity on 22 February in south Asia (covering Persian Gulf, Arabian Sea and western India), was investigated via remote-sensing observations and ground-based measurements. Several satellite observations from sensors such as MODIS on board Terra and Aqua, OMI on board AURA, CALIPSO and KALPANA-1 VHRR were used to detect the main source regions of the dust outbreak and monitor its spatio-temporal distribution. Additionally, AERONET measurements over six stations in south Asia as well as MICROTOPS-II and MFRSR measurements over Hyderabad, India, accomplished and partially validated the satellite observations. Finally, MM5 model was used to provide the meteorological conditions (wind speed and direction) favoring the dust erosion and transport. The results showed that strong winds, both in surface and middle troposphere, favored the exposure, uplift and long-range transport of desert-dust aerosols covering an extended area of the Persian Gulf, AS, Pakistan and western India. Both MODIS and OMI observations identified aerosols in Pakistan, where the dust plume presented its highest intensity. Lidar measurements over Hyderabad revealed an elevated dust aerosol layer (~3 km), above the thick boundary aerosol layer, suggesting long-range transport of dust aerosols. The dust presence over Hyderabad in the afternoon hours of 22 February caused ~6% reduction in ground reaching solar irradiance.

P-Sources.53 ID:4350 15:35

An unusual dust event over North-eastern India and its association with extreme climatic conditions – A study using satellite data

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Dust storms are atmospheric phenomena most frequently occurs over deserts and regions of dry soil, where particles are loosely bound to the surface, which has implications for local weather. In the present study, we have analysed the unusual dust event occurred in the northeast region of India on 17th March, 2009 using multi-satellite data sets. The study region of Guwahati is characterized by a tropical climate and generally heavy rainfall, which do not favor the erosion of mineral dust. An intense thick layer of dust/haze could be clearly seen over the region on 17th March, 2009. According to local newspaper reports, high intensity dust storms are rare over the region but the current storm's intensity was greater than usual because of drier-than-normal weather conditions prevailed during last three months (January-March) prior to the event. The analysis of NCEP temperature/relative humidity (RH) anomalies variations showed ~0.60C increase in surface air temperature and ~ -4% reduction in RH during March, 2009, which resulted in dry conditions over the region. AIRS derived RH / Temperature values at different altitudes and TRMM accumulated rainfall data showed reduction in March, 2009 compared to March, 2008. The dry soil conditions became susceptible for dust loading during high winds resulting in dust storm observed on 17th March, 2009. A very high value of Terra MODIS AOD550 (~ 1.3) along with lower value of Angstrom exponent 'α' (0.78) were observed on 17th march, 2009 suggesting dominance of coarse mode dust aerosol particles over the region due to dust event.

P-Sources.54 ID:4376 15:35

Dust modelling in the UK Chemistry aerosol model

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Windblown dust emitted from arid regions is one of the most important aerosol types; it contributes significantly to the aerosol mass burden and is an important component of the total aerosol radiative forcing. Because dust has been a feature of the Earth's climate system for thousands of years, and because it is not directly emitted from human activity, it is generally seen as a "natural" climate forcing. However over the years, changes in land use have altered the distribution of dust sources throughout the globe. Dust emissions that arise from these anthropogenic changes can themselves be considered to be anthropogenic, and thus should be considered in the calculation of anthropogenic radiative forcing. However, although estimates of the natural / anthropogenic dust loadings exist, there remains considerable uncertainty as to how much of today's dust loading can be attributed to human activity.

In this paper, we use the newly developed UK Chemistry Aerosol mode (implemented in the UK Met Offices' general circulation model) to simulate the global dust burden under different conditions. We use output from the JULES vegetation model to examine the sensitivity of the simulated dust burden to changes in vegetation. We consider the anthropogenic dust loading in term of the importance for aerosol optical depth, total radiative forcing and contribution to exceedance of air quality standards.

P-Sources.55 ID:4221 15:35

An Urban emissions inventory for South America and its application in numerical modeling of atmospheric chemical composition: Impact on local and regional scales.

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This work describes the development of an urban vehicle emissions inventory for South America (SA), based on the analysis and aggregation of available inventories for major cities, with emphasis on its application in regional atmospheric chemistry modeling, for the study of chemical composition modification on SA and its impact on the regional climate change. This database integrates information from local inventories of vehicle emissions into existing global databases for the South American continent. Due to the limited number of available local inventories, urban emissions were extrapolated based on the correlation between city vehicle density and 2004 mobile source emissions of carbon monoxide (CO) and nitrogen oxides (NOx). Emissions were geographically distributed using a methodology that delimits urban areas using high spatial resolution remote sensing products. This numerical algorithm enabled a more precise representation of urban centers. The derived regional inventory was evaluated by analyzing the performance of a chemical weather forecast model in relation to observations of CO, NOx and Ozone in two different urban areas, São Paulo and Belo Horizonte, both important Brazilian metropolitan areas. The gas mixing ratios simulated using the proposed regional inventory show good agreement with observations, consistently representing their hourly and daily variability. These results show that the integration of municipal inventories in a regional emissions map and their precise distribution in fine scale resolutions are important tools in regional atmospheric chemistry modeling. In the next phase of this project, we are performing simulations for recent years in order to study the regional impact of the large urban areas in chemistry composition of South America.

P-Sources.56 ID:4147 15:35

Using inverse modeling techniques to evaluate the new on-road emissions inventory of the Metropolitan Area of Buenos Aires, Argentina

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Being the 10th megacity of the world and the 3rd of Latin America, with an average population density of 4,600 inhabitants/km2 and a fleet of 2.4 million vehicles, the Metropolitan Area of Buenos Aires (MABA), Argentina represents a concentrated site of on-road mobile emissions: 815 ton of CO/ km2 at city level vs. 0.7 country-wise for the year 2000. In order to better analyze this situation within the SAEMC 4-year regional project we have developed annual emission inventories of criteria pollutants and greenhouse gases from on-road mobile sources for the year 2006 assessing the level of uncertainty associated with the inventories as well as the impact of emissions factors from different data sources on the computed emissions. Key features of these inventories include (i) the compilation of a regional emissions factors database, that includes emission factors measured in dynamometers and circulating vehicles in Argentina, Brazil, Chile and Colombia, and (ii) a better characterization of emissions from the local fleet through the incorporation of local driving conditions and the dynamic composition and technology distribution of the local fleet, obtained using an innovative approach. The accuracy of the carbon monoxide (CO) inventory was improved by data assimilation, comparing modeled CO concentrations and observations from the air quality monitoring network present in MABA. Polyphemus direct chemistry transport model was used to estimate ambient CO concentrations. Main inputs to this model were: weather fields generated by the MM5 model, and spatially disaggregated emissions, according to vehicle type and direct and indirect indicators of traffic activity. Finally the inventory was optimized by optimal interpolation with the aim to minimize the observations and emissions errors employing the Best Linear Unbiased Estimation (BLUE).

P-Sources.57 ID:4273 15:35

Empirical evaluation of vehicle emissions of carbon monoxide and nitrogen oxides in Santiago, Chile and Buenos Aires, Argentina

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We use concurrent observations of carbon monoxide (CO) and nitrogen oxides (NOx) during the morning rush hour to evaluate CO and NOx emissions estimates from mobile sources in Santiago, Chile and Buenos Aires, Argentina. The data for Santiago considers three stations and an eight year period (2000-2008). For Buenos Aires only a year between October 2008 and November 2009at two stations. Further, we compare the emissions from these cities based on available air quality observations, and traffic activity and technology data collected in Santiago and Buenos Aires using a common methodology, the so-called International Vehicle Emissions (IVE) model. A preliminary analysis of the data, and results from previous work using inverse modeling techniques for CO, suggest that NOx emissions in Santiago are probably overestimated by a factor 2 to 3. This stresses the need of improving the methodologies for estimating traffic related emissions. Our results also underline the need of local knowledge to assess traffic emissions: for instance the older gasoline fleet and the use of natural gas in light duty vehicles in Buenos Aires, produce much higer CO and VOCs emissions than in Santiago, in spite of traffic activity is only 20% larger.

P-Sources.58 ID:4309 15:35

Comparison of Traffic Impacts on Particle Pollution in Micro-environments around Commuters in Taiwan Metropolitans

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Traffic emission is the most predominated particle source in urban environments, especially in Taipei metropolitan area with high population and traffic densities (9600 person/km2 and 6500 vehicle/km2). There are 271 cars and 412 motorcycles per 1000 persons in Taipei city and 230 cars and 577 motorcycles per 1000 persons in Taipei County, respectively. The annual PM10 were 65.4, 47.8, 21.8μg/m3 in traffic, urban, and mountain sites in Taiwan EPA stations in 2009, respectively. Exposure to pollutants from traffic emission such as particulate matters (PM) and particulate polycyclic aromatic hydrocarbons (pPAHs) can

result in acute and chronic health effects. Thus, it is important to assess the traffic impacts on PM2.5, PM10 and associated pPAHs levels in micro-environments around commuters (i.e. commuters' exposures) in Taiwan. We assessed commuters' exposures during morning, noon, and evening hours with Personal Environmental Monitors (PEM). Subjects were commuting with the same starting and ending locations via three different modes, namely motorcycling, driving, and taking subway. One real-time Grimm PM monitor was accompanied with motorcyclist sampling. Results showed that PM2.5 exposures of commuters of motorcycling, driving, and taking subway were 52-268, 32-264, 57-294µg/m3 in the first 5-day sampling period, respectively. The corresponding concentrations in the second 5-day period were 85-460, 42-149, and 45-153µg/m3. The mean exposures of pPAHs (sum of sixteen analyzed PAH species) of motorcyclists and car drivers were 3.2 and 12.1ng/m3, respectively, while pPAH exposures of subway riders were all under detection limits. On average, commuters riding motorcycles in the morning had the highest PM and pPAH exposure compared to others. In addition, peak PM exposure of motorcyclists; which occurred while waiting for traffic lights, can be as high as 5 times the mean exposure concentrations. Vehicle emission apparently was the major PM exposure source of motorcyclists and car drivers; while crowd was an important PM exposure source of subway riders.

P-Interfaces.1 ID:4095 15:35

Les connexions mondiales de fer entre la poussière du désert, la biogéochimie des océans et du climat.

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Les conditions environnementales de la Terre, y compris le climat, sont déterminées par des agents physiques, interactions chimiques, biologiques et humains qui transforment et le transport de matériaux et d'énergie. C'est le système de la Terre: une entité très complexe, caractérisé par des réponses multiples non linéaires et des seuils, avec des liens entre les éléments disparates. Une partie importante de ce système est le cycle de fer, dans lequel le fer contenant de la poussière du sol est transporté de la terre à travers l'atmosphère vers les océans, affectant la biogéochimie des océans et d'avoir donc des effets de rétroaction sur le climat et la production de poussière. Ici, nous passons en revue les principales composantes de ce cycle, en identifiant les incertitudes majeures et les priorités pour la recherche future.

P-Interfaces.2 ID:3438 15:35

INTERRACTIONS BETWEEN ATMOSPHERE, ECOSYSTEM AND MARINE ENVIRONMENT IN NIGERIA

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Over the past decades we have witnessed extra-ordinary natural and anthropogenic ally-driven changes in Ocean Biogeochemical composition. Most of atmospheric and oceanic climatic variability have been related to interaction between Ecosystem, Atmosphere and Marine environment.

Between 2004-2006 we studied the Interconnection and Teleconnection between ecosystem, atmosphere and marine environment. We noted that critical input of nutrients by Riverine and increase in Green house gases caused significant changes in Biogeochemical properties of ocean around Lagos area of Nigeria. In turn, the feedback to local communities has resulted in changes in their economies and diets. More practical issues will be presented

P-Interfaces.3 ID:4459 15:35

Chlorine-chemistry and Polar Stratospheric Cloud evolution over the Antarctic Polar Vortex derived from MLS observations

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When very low temperatures occur in the Southern hemisphere winter stratospheric polar vortex, type I-II Polar Stratospheric Clouds (PSC) formation occurs above the Antarctic. An enhanced number of PSC particles are observed in a highly activated Antarctic Polar vortex (APV) where heterogeneous ozone-chemistry reactions occur on the surface of these particles. Chlorine activation processes, a set of chemical reaction cycles which involve the conversion of chlorine reservoir species into more reactive chlorine forms, are an unambiguous sign of the presence of sunlit PSC. We present in this study an examination of the spatial and temporal evolution of type I-II PSC inferred from temperature variations in the lower-middle atmosphere over the APV, derived from MLS satellite observations and NCEP/NCAR reanalysis. We then utilize MLS chemical concentration observations to correlate the role of temperature variations on PSC formation with stratospheric chlorine partitioning.

In particular, we present our latest results on the analysis of HCl, an important chlorine reservoir in the stratosphere, and daytime ClO change signal calculations as a way to unambiguously determine PSC formation and evaporation temperatures together with an assessment of the likely periods of the year where PSC and chlorine activation events occur. Preliminary results are used to derive the time lags between chlorine activation events and initial suggestions of PSC based on temperature and measured when formation temperature thresholds are crossed. Finally, we present a set of results using Antarctic vortex edge calculations to determine how dynamics affects PSC formation and thereby stratospheric chlorine-chemistry over the Antarctic region.

P-Interfaces.4 ID:4398 15:35

Identification of aerosol transport pathways in the Arctic region in early spring from semi-lagrangian tracer advection in Polar-WRF model

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In the framework of POLARCAT campaign carried out in 2008 in the Arctic region, we used a mesoscale model for high resolution case studies of pollutant transport and aerosol radiative effects. The dynamical part of the model is based on the Polar-WRF model, developed by NOAA. An aerosol module based on NCAR climate model schemes has been implemented in the model at PNNL. The WRF chemistry version allows the simulation of trace species. We present here the meteorological conditions and the main transport pathways of aerosols advected from mid-latitude sources in the Arctic in early spring. This study is based on semi-lagrangian tracer advection in the model to include the dynamical sub-grid processes impact on aerosols and trace gases vertical transport. Comparisons with the high resolution measurements obtained

during POLARCAT (airborne lidar observations, CALIOP vertical profiles…) and particle dispersion model (FLEXPART) initialized by ECMWF are conducted to retrieve the critical processes responsible of the horizontal and vertical transport. This turns out to be also a first step to calculate the radiative fluxes of the aerosol plumes in the Arctic region.

P-Interfaces.5 ID:4315 15:35

Comparison of Snow-pit and Atmospheric Concentrations of Sulphate, Nitrate, MSA and Black Carbon in the High Arctic

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Ice-cores and snow-pits hold a wealth of historical information on the composition of the atmospheric aerosol that has deposited to the snow surface. A comparison between the aerosol measurements in the atmosphere and the snow-pit can provide knowledge about rates of aerosol deposition and potentially provide information on historical concentrations in the atmosphere. We have a unique data-set of major ion and black carbon concentrations in snow-pit samples collected at Mt. Oxford ice-cap by Natural Resources Canada (Northern Ellesmere Island, 82°11'N, 73°02'W, 1782 m above sea level) and in the atmosphere at Alert (about 150 km northeast of Mt. Oxford, 230 m above sea level). Alert is a WMO-GAW measurement station operated by Environment Canada since 1979 for the atmospheric aerosol measurements. It is estimated that snow-pit samples at the depth of 10.4 m correspond to the atmospheric deposition from 1979. A preliminary comparison of the snow-pit concentrations of sulphate, nitrate, methane-sulphonic acid (MSA) and black carbon with measurements of the same aerosol quantities in the air at Alert from 1980 to 2009 will be shown. Despite the difference in elevation between the two sites, vertical profiles in Arctic haze from several studies (AGASPII, 1986; ISDAC, 2008) suggest that differences in the mean aerosol concentrations at both elevations are insufficient to explain the differences between the atmospheric and snow-pit measurements. If that is true, then the atmospheric aerosol concentration may not be a dominant factor for determining the level of aerosol deposition in this region. Net deposition rates will also be determined for sulphate, nitrate, MSA and black carbon.

P-Interfaces.6 ID:4277 15:35

Nitrogen oxides in the firn air at Summit, Greenland: an interannual comparison.

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Previous studies have shown that the sunlit polar snowpack is an active reactor, where previously deposited chemical species can undergo photochemical processes that subsequently result in an efflux of trace gases to the overlying atmosphere, altering its oxidative capacity. Among these species, nitrate has been widely recognized as the substrate for nitrogen oxides (NOx) fluxes observed at a number of different remote polar sites, as well in mid-latitude snow-covered environments. However, the extent and the impact of these cryosphere-atmosphere interactions on tropospheric chemistry cannot yet be adequately quantified due to scarcity of long-term field measurements. Such measurements are prerequisite to the development of

parameterizations that can be applied in atmospheric chemistry models. In an effort to fill this gap, we performed continuous measurements of nitrogen oxides and ozone in the firn air and above the snowpack at Summit, Greenland from summer 2008 to summer 2010. These observations have shown strong diurnal and seasonal patterns for NOx, with evidence of a very different behavior and magnitude in concentrations for NO and NO2. Diurnal cycles are observed in and above the snowpack, as high as 9 m, showing the influence of snowpack emissions on the overlying atmosphere. These new chemical observations and supporting micrometeorological variables are now being applied to develop a more mechanistic representation of cryosphere-atmosphere reactive trace gas exchange in a chemistry-climate model. This will allow us to assess the significance of cryosphere-atmosphere chemical exchange in tropospheric chemistry under climate and global change.

P-Interfaces.7 ID:4155 15:35

A network of autonomous surface ozone monitors in Antarctica: technical description and first results.

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Concentrations of surface ozone in polar regions cannot be derived from satellite data, so must be measured by ground-based sensors. To understand the regional picture a carefully designed network of sensors is required. Here we report on a network of 10 autonomous ozone monitors, deployed in the Weddell Sea sector of coastal Antarctica plus inland towards the Antarctic Plateau. Measurements were made for a full year. Each ozone monitor measured successfully within its predefined duty cycle throughout the year, with some differences in performance dependent on power availability and air temperatures, which we discuss. Preliminary results show that on several occasions the same ozone depletion event was observed by many monitors covering much of the region. The results also allow us to explore regional differences in ozone enhancements due to photolysis of nitrate in snow during early summer, and in ozone loss that might be related to the supply of halogens during late summer.

P-Interfaces.8 ID:4464 15:35

Uptake and reactivity of hydroxyl radicals on ice

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Ice surfaces are important substrates for atmospheric chemistry. Reactions in ice are expected to occur primarily in the disordered region near the air-ice interface known as the quasi-liquid layer (QLL). Both the physical properties of the QLL and its role in reaction mechanisms and kinetics are poorly understood. Although there are some similarities between the QLL and liquid water, reaction kinetics often differ greatly in the two environments. Using a surface-sensitive spectroscopic probe, we measured kinetics of reactions between hydroxyl radicals and aromatic hydrocarbons in aqueous solution and at air-ice interfaces. Reactions proceeded as expected in aqueous solution, but were significantly suppressed on ice, both when OH was formed from the photolysis of precursor compounds present at the air-ice interface, and when it was introduced from the gas phase. To better understand this observed lack of reactivity, we studied the adsorption of hydroxyl radicals to air-ice and air-water interfaces using molecular dynamics.

P-Interfaces.9 ID:4349 **15:35**

Modelling of polar emissions of NOx from snowpack and its impact on polar bromine

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Measurements in recent years show that several species, important for the oxidizing capacity in the troposphere, are emitted by snow in polar regions and other snow covered areas of the world. We have used the Cambridge chemical transport model, p-TOMCAT to study the impact of emission of NOx from the snowpack. A parametrization of the production of NOx by photolysis of snowpack nitrate has been developed and used with an assumed global nitrate inventory. Multiannual integrations show that NOx emissions from snow have a small impact globally but represent a significant source of NOx in the polar regions.

It is well established that 'bromine explosion' events occur regularly in the polar regions with large areas of elevated BrO observed around the sea-ice. In previous work, we have also developed parametrizations for the heterogeneous production of bromine from snow-sourced sea-salt and snow that can reproduce observations in model simulations using p-TOMCAT. In this paper we report on work in which the impact of the NOx release from snow on the polar bromine chemistry is assessed, particularly with respect to the formation and distribution of BrONO2 and how it alters the ozone budget.

P-Interfaces.10 ID:4495 **15:35**

Bromine Cycling in Snow, Firn Air, and the Atmospheric Boundary Layer at Summit, Greenland During the GSHOx Field Campaign

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Sunlit snow and ice are known to play an important role in determining the chemical composition of the Arctic boundary layer, for example ice chemistry is important for boundary layer ozone depletion events at polar sunrise. In remote Arctic regions pollutants originating mainly from lower latitudes react with various atmospheric radical species potentially impacting the ozone budget and oxidizing elemental mercury. Despite the important role of atmospheric chemistry in the Arctic, our understanding of the underlying chemical transformations in the remote snow pack and its impact on the fate of air pollutants is incomplete.

Motivated by indirect evidence that reactive halogens are present on the Greenland ice sheet, two field campaigns have been conducted at Summit, Greenland in 2007 and 2008 to study air and snow properties. Our measurements identified BrO mixing ratios of up to 3ppt with typical levels in the range of 1 - 2ppt. Peaks in soluble gaseous bromine generally occurred 1-3 days after free troposphere air above Summit penetrated the stable surface boundary layer which resulted in increased concentrations of bromide in surface snow.

We have also developed a new vertically resolved model for snow physics and chemistry in order to understand the chemical and physical processes occurring during these field experiments. Both measurements and modeling suggest efficient photochemical activation of trace concentrations of bromide in the snow that results in significant gas phase bromine release to the overlying air. In addition, both observations and modeling results show the strong impact of snow and firn on the chemistry of NOx, reactive bromine, and the speciation of Hg in the near surface air above the Greenland ice sheet.

P-Interfaces.11 ID:4457 15:35

Synoptic-scale meteorological control on reactive bromine production and ozone depletion in the Arctic boundary layer: 3-D simulation with the GEM-AQ model

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Episodes of high bromine levels and surface ozone depletion in the springtime Arctic are simulated by an on-line air-quality transport model, GEM-AQ, implemented with gas-phase and heterogeneous reactions of inorganic bromine species and with a simplistic scheme of air-snowpack chemical interactions. Snowpack on sea ice is assumed to be the only source of bromine to the atmosphere and capable of converting relatively stable bromine species to photolabile Br_2. Bromine explosion, by which bromide in the snowpack is autocatalytically released to the atmosphere as a result of dry deposition of HOBr and $BrONO_2$, is assumed to occur on first-year sea ice, whereas the snowpack on multi-year sea ice and land surface is assumed only to recycle a part of bromine lost via dry deposition back to Br_2. A trigger of the bromine explosion is assumed to occur only on the first-year sea ice with Br_2 production tentatively associated with dry deposition of ozone under sunlight. The model often captures the synoptic-scale evolution of enhanced BrO column amounts as seen from space. The results strongly suggest the ubiquitous source of reactive bromine on the first-year sea ice during the Arctic springtime while the timing and location of bromine release are largely controlled by meteorological forcing on the transport of ozone to the near-surface air. Also, if indeed reactive bromine in the Arctic boundary layer is supplied predominantly from the snowpack, it should be capable of releasing reactive bromine at temperatures nearly as high as −10°C, particularly on the ice off the Siberian coast, as indicated from sensitivity experiments with varying a temperature threshold below which bromine activation is assumed to occur in the snowpack. The present air-snowpack interaction scheme yields a major part of atmospheric bromine input via dry deposition of ozone to snowpack, pointing to the role of chemistry in the interstitial air of sunlit snowpack.

P-Interfaces.12 ID:4150 15:35

Strong evidence for BrO in the free troposphere during enhancements in Antarctica, in contrast to IO.

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Tropospheric BrO and IO were measured by a ground-based remote-sensing spectrometer at Halley in Antarctica, and BrO and IO were measured by remote-sensing spectrometers in space using similar spectral regions and DOAS analyses. Surface BrO was simultaneously measured locally by CIMS, and in an earlier year surface BrO and IO were measured over a long path by a DOAS spectrometer. During enhancement

episodes, total amounts of tropospheric BrO from the ground-based remote-sensor were similar to those from space, but if we assume that the BrO was confined to the boundary layer they were very much larger than values measured by either surface technique. Amounts observed by the ground-based remote sensor at different elevation angles also suggest much of the BrO was often in the free troposphere. By contrast, if we assume that the IO was confined to the boundary layer then amounts of IO from the remote sensors were similar to surface measurements. This contrast is explored in terms of what we know about sources and sinks of tropospheric BrO and IO during polar enhancements, and the situation during Arctic enhancements is discussed.

P-Interfaces.13 ID:4132 15:35

Tropospheric halogen multiphase chemistry: Mechanism development and modeling

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Halogens play a key role in marine chemistry. There has been a great effort to understand halogen chemistry in the atmosphere, either by field experiments, laboratory or modeling studies.

In the present study a comprehensive halogen module (HM2) was constructed for the use with the multiphase mechanism RACM-MIM2ext/CAPRAM 3.0i. Reactions of inorganic chlorine, bromine, and iodine species are treated in the gas the aqueous phase as well as their interactions with VOCs and inorganic species in either phase. In contrast to most other modeling studies a fully explicit gas-phase oxidation scheme of alkyl halides has been included. The HM2 totals to 205 halogen species taking part in 597 reactions and 37 phase transfer processes.

The mechanism was designed for the simulation of a great variety of different conditions, e.g. remote marine, or mixed marine and urban air masses. First simulations under marine remote conditions including non-permanent clouds were performed with the model SPACCIM. Sensitivity studies were carried out omitting the iodine and both the iodine and the bromine chemistry to investigate the influence of the different halogen subsystems.

The model results reflect the typical concentration profiles of the halogen species among other trace gases, such as O_3 or OH, measured in field experiments. For the first time detailed time-resolved analyses of chemical source and sink fluxes were performed, which revealed new reaction cycles, e.g. the accumulation of bromine and iodine species in cloud droplets during cloud events. On the other hand, clouds can suppress halogen chemistry such as in the interstitial gas-phase. Further calculations concern the relative contributions of alkyl iodides to the inorganic iodine budget. Photolysis has proven to be the dominant pathway. Connections between the different halogen subsystems have been investigated and show the importance of many interactions, e.g. the ability of iodine chemistry to trigger chlorine chemistry.

P-Interfaces.14 ID:4148 15:35

Multiphase bromine cycling over the eastern tropical North Atlantic during spring

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Multiphase halogen chemistry impacts important, interrelated chemical processes in marine air. Bromine activation chemistry leads to catalytic ozone destruction and modification of oxidation processes including HOx and NOx cycling. Spatiotemporal variability in many reactants, products and reaction pathways are poorly characterized, rendering uncertain the global significance of tropospheric halogen chemistry.

During spring 2007, constituents of marine air were measured at the Cape Verde Atmospheric Observatory on the windward shore of São Vicente Island (16.8 N, 24.9 W). Total aerosol Na and Br and inorganic gaseous Br were sampled over 3- to 4-hour intervals with filter packs; volatile acids were sampled in parallel over 2-hour intervals with mist chambers.

Sodium concentrations ranged from 49 to 334 (median = 143) nmol per standard cubic meter. The presence of volatile acids at detectable mixing ratios (HCl ranged from 29 to 613 pmol per mol) indicates that aerosols were either acidic or rapidly acidified throughout the campaign. The median enrichment factor of aerosol Br relative to seawater composition was 0.40 with a range from 0.05 to 1.08. Concentrations of inorganic gaseous Br were typically about double those of aerosol Br (medians of 0.22 and 0.11 nmol per standard cubic meter, respectively). Systematic diel variability was evident in neither aerosol Br enrichment factor nor inorganic gaseous Br.

Observations will be compared with model predictions based on CAABA (Chemistry As A Boxmodel Application) with the chemistry code MECCA (Model Efficiently Calculating Chemistry of the Atmosphere). In addition to the standard chemistry of HOx, NOx, and methane, the model explicitly treats gas-phase as well as aqueous-phase chemical reactions of halogen species. Results will be presented and discussed.

P-Interfaces.15 ID:4548 15:35

Size distributions and chemical properties of aerosols generated by the bubble bursting of seawater samples with phytoplankton

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Oceanic surface waters contain a large amount of organic substances produced by marine biota, which is transferred to the atmosphere as primary marine aerosols (PMA) by the bubble bursting process. The organics are potentially important because they probably influence on the physico-chemical properties of PMA, including the cloud condensation nuclei (CCN) activity. However, our current knowledge about the relationships of organics in PMA to the marine primary production and to the particle properties is limited despite recent laboratory/field studies on PMA. To better characterize the flux, size distribution and chemical composition of PMA associated with organics from phytoplankton, we performed laboratory-based bubble bursting experiments using seawater samples in which marine prymnesiophyceae *Isocrysis Galbana* were cultured. The glass made aerosol generator filled with 6 L of seawater sample was used to generate PMA. Compressed dry air was bubbled through the glass ball filter positioned at 5-90 cm below the air-water interface. The size distributions of generated sub-micrometer aerosol particles and the mass concentrations of organics therein were measured using a scanning mobility particle sizer and a high resolution time-of-flight aerosol mass spectrometer, respectively. The log-normal distributions fitted to the size distributions have the mode diameters at 80-90 nm. When the bubble path length increased from 5 to 90 cm, the particle number flux decreased by a factor of 10, whereas the organic mass to total mass ratio showed an increasing trend. The result supports the hypothesis that organics are absorbed on the surfaces of bubbles during the rise toward the air-water interface. The number flux of PMA generated from seawater with phytoplankton was up to twice that generated from seawater without them. The difference suggests that organics produced by phytoplankton alter the properties of bursting bubble surfaces and result in the increase in the particle number flux.

P-Interfaces.16 ID:4324 **15:35**

Size distributions and chemical characterization of water-soluble organic aerosols from marine biological sources over the remote western North Pacific

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Size-segregated aerosol samples were collected over the remote western North Pacific in summer 2008 to investigate the relative contribution of marine biological origin to organic aerosols. They were analysed for organic carbon (OC), water-soluble organic carbon (WSOC), and water-soluble organic compounds, including methanesulfonic acid (MSA), low molecular weight organic acids and α-dicarbonyls as well as major ionic species. The average concentrations of OC and oxalic acid are about twice more abundant in more marine biologically influenced aerosols, defined by the concentrations of MSA and azelaic acid, than in less influenced aerosols. WSOC, which can serve as a proxy for marine biogenic secondary organic aerosols (SOA), accounted for 15-24% of the total mass concentrations determined in the submicrometer range of more biologically influenced aerosols. Substantial fractions of oxalic acid and other organic acids were found in the submicrometer range over the region. The results indicate preferential net production of oxalic acid and WSOC from precursors in the oceanic region with higher biological productivity. On the other hand, insignificant difference between the two types of aerosols was found for mass concentrations and size distributions of glyoxylic acid and methylglyoxal, implying that the abundance of isoprene-derived SOA from marine sources is small over the region. Evidence for marine biological sources of organic nitrogen aerosols and their contribution to organic aerosols will be also presented.

P-Interfaces.17 ID:4171 **15:35**

Evidence of a marine biogenic source of oxalic acid

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Oxalic acid has been often observed in marine aerosol, nevertheless, given the ubiquitous character and the high concentrations found in polluted environments, its origin has always been attributed to continental sources or precursors. Here, we present new results suggesting a biogenic global source of aerosol oxalic acid over the oceans. Oxalic acid has been detected in strictly controlled clean marine aerosol samples at Mace Head (53°20'N, 9°54'W), during the year 2006, and at Amsterdam Island (37°48'S, 77°34'E), from 2003 to 2007, in concentrations ranging from 2.7 to 39 ng m^{-3} and from 0.31 to 17 ng m^{-3}, respectively. In both hemispheres, oxalic acid concentration showed a clear seasonal trend, with maxima in spring-summer and minima in the fall-winter period, in analogy with other marine biogenic aerosol components (e.g., MSA). At both sites, oxalic acid mass size distribution presented a peak in the 1-2.5 µm size range, while in continentally influenced samples it was mainly present in the submicron mode. Although this difference can

be explained by a different partitioning of oxalic acid due to the acid-base properties of marine aerosol (acidic in the submicron fraction, mildly alkaline above 1 µm), it can also suggest the existence of a peculiar oxalic acid formation pathway over the oceans. The most likely oxalic acid formation pathways in the marine boundary layer are the photochemical oxidation of precursor compounds, produced in the atmosphere by the oxidation of biogenic unsaturated fatty acids [1], or in-cloud oxidation of glyoxal [2]. Interestingly, a source of gas-phase glyoxal over remote marine regions, with a seasonal trend similar to that of aerosol oxalic acid at mid latitudes, has recently been documented [3].

[1] Kawamura & Sakaguchi, J. Geophys. Res., 104, D3, 1999 [2] Warneck, Atmos. Environ., 37, 2003 [3] Myriokefalitakis et al., Atmos. Chem. Phys., 8, 2008

P-Interfaces.18 ID:4524 15:35

Investigation of mineral dust aerosol-chemistry interactions in marine environments

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Mineral dust aerosols play an important role in atmospheric chemistry through photolysis and heterogeneous uptake. Both mechanisms strongly depend on the size and composition of dust aerosols. Because of the complex nature of dust, however, chemistry modeling commonly relies on simplified assumptions about the properties of dust particles relevant to physiochemical processes. The goal of this study is to investigate the impact of size-resolved composition of dust aerosols on atmospheric photochemistry. The relative importance of dust characteristics in photolysis and heterogeneous loss and the relative roles of the two mechanisms on atmospheric photochemistry are investigated.

A new block of spectral aerosol optical properties was developed and incorporated into the tropospheric ultraviolet and visible radiative transfer code, NCAR TUV, in order to calculate spectral actinic fluxes and photolysis rates, J-values. The Fuchs-Sutugin approximation was employed to compute mass transfer from gas to dust species and heterogeneous loss rates, $k_{loss,j}$. The J-values and $k_{loss,j}$ were incorporated into a one dimensional photochemistry model to simulate the diurnal cycle of a vertical profile of photochemical species. Several cases of dust loading were considered in the remote marine and polluted atmospheric conditions. A size-resolved mineralogical composition was constructed by selecting a range of the mass fraction of the three main mineral species, specifically considering iron oxide-containing clay minerals and alkalinity from carbonate-containing species. We used the spectral refractive indices from the Library of Atmospheric Aerosol Refractive Indices and the uptake coefficients of individual minerals and dust samples from recent laboratory studies. Several size distributions reported from recent field experiments and/or used in prior modeling studies were considered. This work demonstrates that the most important factor controlling the photochemistry field is the dust size distribution, followed by the amount of mineral dust species with high uptake coefficients, and the amount of iron oxide-clay aggregates.

P-Interfaces.19 ID:4135 15:35

Water adsorption on MgCl2×6H2O salt surface

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Sea-salt particles emitted by the oceans in the form of concentrated solution droplets and transported into relatively dry inland regions can lose water and take it up again depending on atmospheric humidity conditions. The kinetic and thermodynamic information on this salt/water interaction is of great importance since the surface adsorbed water may alter aerosol composition, radiative and visibility effects, particle deposition as well as the kinetics and mechanisms of the processes on the salt surface. One of the most studied phenomena is adsorption of water on NaCl: crystals, films and nanometer aerosols. The motivation of this interest was clear: sodium chloride is the main constituent of sea salt. Minor components are Na2SO4, KCl, CaCl2, MgCl2, NaBr etc. Recent investigations of segregation phenomenon of NaBr and MgCl2 in NaCl crystal doped with NaBr and MgCl2 showed that minor constituent concentration in near surface area could be much higher than the bulk one. This observation indicates that minor sea salt components may play an important role in the chemistry of sea salt particles and emphasizes the importance of the investigation of their processing with atmospheric trace gases and water in particular. Present work reports the results of the experimental study of the interaction of water vapor with dry solid films of MgCl2×6H2O as a function of partial pressure of water and over the temperature range 240 to 340 K. Experiments were carried out using a flow reactor coupled to a modulated molecular beam mass spectrometer. The adsorption data are well described by Freundlich isotherm with the heterogeneity parameter close to 0.5. The isosteric heat of adsorption was found to be -(44.7 +/- 1.2) kJ mol-1 independent of the salt surface coverage in the range (0.8 – 30) × 1015 molecule cm-2. An empirical equation is proposed for the amount of water adsorbed on MgCl2×6H2O as a function of relative humidity. The observed results suggest that under atmospheric conditions sea salt particles are probably enveloped by a MgCl2×6H2O brine. This work was financially supported Russian Federal Science Agency grant № 02.740.11.5176.

P-Interfaces.20 ID:4229 15:35

Infrared detection and modeling of an interfacial liquid layer at the ice-Ge interface

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The surfaces and near-surface regions of ice and snow are known to contribute to chemistry that affects the composition of the atmosphere. Much is known about ice under conditions relevant to the cold upper troposphere and stratosphere. However, there is a dearth of information about the surface chemistry of ice at temperatures near its melting point where the properties of the ice surface change drastically. Attenuated total reflection infrared spectroscopy (ATR-IR) was employed to study the near-surface region of ice at warmer temperatures. This method allows ice films on a Ge crystal to be probed to a depth of about 1 μm without interference from water vapor. This penetration depth is varied with the angle of incidence of the infrared radiation. We present evidence for the formation of an interfacial liquid layer at the ice/Ge interface of frozen aqueous NaCl solutions (0 to 0.5 M NaCl). The morphology of the films and depth of the interfacial layer as a function of salt concentration is described. Interpretation of infrared spectra is supported by spectra of varying morphology modeled from the Fresnel equations for internal reflection. Relevance of these results to atmospheric chemistry and glaciology will be discussed.

P-Interfaces.21 ID:4500 15:35

Accelerated atmospheric conversion of calcite to nitrate by nitrogen dioxide in the presence of H2O: a Raman microscopic study

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Conversion of mineral dust in dust plume to nitrate by nitric acid changes the morphology and hygroscopic properties of mineral dust, thus impacts on climate and ecosystems, while the conversion by nitrogen dioxide (NO2) is believed insignificant. To study the heterogeneous reaction on single particles in the presence of H2O, we developed a novel Raman microscopy technology to map changes of chemical compositions of individual micrometer-size particles during the NO2-CaCO3-H2O ternary reaction. We found the reaction uptake coefficient of NO2 on the internal mixed CaCO3-Ca(NO3)2-H2O particles in the level of 10-5, this is about 10,000 times faster than previous reported, suggesting NO2 could play an important role in the atmosphere in converting mineral dust to nitrate and producing nitrous acid, an important source of hydroxyl radicals. Detail information about reaction mechanism was also revealed by the novel Raman microscopy methodology we developed.

P-Interfaces.22 ID:4291 15:35

Morphological characterization of carbonaceous aerosols and its implications on climate and health.

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The carbonaceous particles, when present in atmosphere, are capable of significantly altering the atmospheric radiative balance (absorption of solar ultraviolet and visible radiation) and increase the global warming. This trait counters the expected cooling of surrounding atmosphere, as expected due to the scattering of light (e.g. non-carbonaceous aerosols). In addition to the direct emission of carbonaceous aerosols (particles) from inefficient combustion they also arise from the conversion of emitted volatile organic compounds in the atmosphere involving photochemical reactions (Secondary Organic Aerosols). Given the multiple affects of carbonaceous aerosols on the local, regional and global climate regime, it is important to undertake systematic investigations of relevant aspects of carbonaceous aerosols. In the study we have characterize carbonaceous aerosols (soot) using scanning electron microscope obtained from different fuels. Then after image processing and other analysis diesel soot was found to have structure that can provide site to other toxic environmental contaminates. In addition we tried to found out particle based scattering and absorption cross-section of these soot particles. The approach allowed us to discriminate and rank carbonaceous aggregates, derived from different sources, and their ability to absorb light.

P-Interfaces.23 ID:4570 15:35

Atmospheric deposition of nitrate, sulphate and ammonium on the Brazilian Territory: what do we know?

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In this work nitrate, sulphate and ammonium deposition through rainwater over the Brazilian territory is presented. Depositions were estimated based on the existing rainfall chemical data and on rainfall climatologic normal over the whole territory. The estimated monthly deposition variations are discussed based on the rainfall amount and on its control on the deposited amount. The diverse level of deposition in a range of sites from higher urbanized such as São Paulo city to Central Amazon is examined on the light of possible sources. Finally it is put forward how much is need to build up on such deposition levels knowledge

to improve our ability to identify the actual deposition levels and impacts, and how they can influence on our policy and ecosystem management needs.

P-Interfaces.24 ID:4446 15:35

Gas-particle partitioning and bi-directional flux of atmospheric ammonia

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Gas phase ammonia mixing ratios are affected both by emissions from and deposition to the land surface and by uptake to and volatilization from atmospheric particles. The partitioning between gas phase ammonia and solid or aqueous ammonium is strongly influenced by temperature, humidity, pH and other physical and chemical variables. During the Border Air Quality and Meteorology Study (BAQS-Met) in the summer of 2007, measurements of both gas phase ammonia and particulate ammonium were made for three weeks at a ground site near Harrow Ontario. The sampling location was in an agricultural region in southwestern Ontario, frequently downwind of industrial and transportation emission sources. Gas phase ammonia was measured by quantum cascade tunable infrared laser differential absorption spectroscopy (QC-TILDAS) and the chemical composition of particles with diameters less than 1 μm (PM1) was measured by time of flight aerosol mass spectrometry (C-ToF-AMS). The gas-particle partitioning of ammonia is investigated as a function of meteorological parameters and aerosol composition. While the fraction of total ammonia (NHx) observed in the gas phase peaks between 0.2 and 0.8, the chemical transport model AURAMS (A Unified Regional Air-quality Modelling System), tends to predict fractions of either less than 0.1 or greater than 0.8. The model frequently predicted acidic aerosol, however observations of NHx always exceeded the observed equivalents of sulphate. One explanation for our observations is that the net flux of ammonia from the land surface increases when aerosol sulphate is present, effectively buffering the mixing ratio of gas phase ammonia. We explore the impact of a bi-directional flux parameterization on the predicted gas-particle partitioning of atmospheric ammonia.

P-Interfaces.25 ID:4587 15:35

Understanding aerosol mixing state impacts on CCN activity using particle-resolved aerosol simulations

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The hygroscopic behavior of aerosol particles depends on their size, and also on their chemical composition. Observations have shown that initially hydrophobic particles, e.g. soot, can be turned hydrophilic by adding a coating of soluble substances. Therefore the composition of individual particles, i.e. their mixing state, is an important factor for determining whether an aerosol particle can act as cloud condensation nucleus (CCN). However, tracking the mixing state in conventional aerosol models requires treating a multidimensional size distribution, which is computationally prohibitive. Thus current models usually assume an internal mixture within one mode or size section. The uncertainties associated with this assumption regarding CCN properties are not well quantified.

In this study we address this problem and present a new approach for aerosol modeling, the stochastic

particle-resolved model PartMC-MOSAIC. It explicitly resolves the composition of individual particles in a given population of different types of aerosol particles, and accurately tracks their evolution due to emission, dilution, condensation and coagulation. We use PartMC-MOSAIC as a tool to investigate the impact of mixing state on CCN spectra. We simulate the evolution of urban aerosols of different source types over the course of several days in an idealized urban plume scenario. We then use the calculated aerosol populations as input for an adiabatic cloud parcel model. The cloud parcel model explicitly predicts activation and condensational growth of the particles as the parcel undergoes adiabatic cooling and the particles compete for water vapor. We quantify the differences between particle-resolved calculations and merely size-resolved calculations for a range of different environmental conditions and separate the effect of composition-averaging from the effect of size-averaging.

P-Interfaces.26 ID:4290 **15:35**

Observations of particle chemical composition and influence on the phase of cold clouds

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This paper studies the influence of particle chemical composition on the phase of cold clouds observed during two intensive measurement periods of the Cloud and Aerosol Characterization Experiments conducted at the Jungfraujoch site (Switzerland). Cloud droplets and particles were sampled simultaneously using a suite of optical, chemical, and microphysical instruments. Sulphate and organic matter were the most abundant semivolatile species observed in the particulate phase during both campaigns. Periods of relatively large loadings of organic and inorganic species were also accompanied by enhancement of light-absorbing aerosol concentrations. The cloud phase exhibited sharp transitions, alternating between highly glaciated and liquid phases over a few seconds within the same cloud event. It was also observed that conditions of elevated pollution were accompanied by an increase in occurrence of glaciated periods. A 24-hour cloud event investigated on the 8 March 2004 was in the mixed phase for approximately 260 minutes, in the glaciated phase for approximately 64 minutes and in the liquid phase for the remainder of the time. On the 23 March 2004 another 24-hour cloud event was captured in which the number of minutes as mixed-phase and glaciated cloud was shorter (196 and 31, respectively). The loadings of black carbon as well as organic and inorganic species were larger during the first period. The investigation was extended for the whole data set, and a statistical analysis was performed across the chemical data. The amount of organic and inorganic material found in liquid and glaciated clouds was statistically different, with organic and inorganic material as well as black carbon being enriched in glaciated conditions. The case studies and the statistical analysis together suggest an influence of the particle chemical composition on the cloud phase, which may be important in perturbing precipitation production, cloud lifespan and precipitation patterns in polluted regions.

P-Interfaces.27 ID:4490 **15:35**

Cloud-aerosol interactions impacts on cloud phase: a case study.

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Cloud phase is a key parameter in cloud microphysics. The vertical profile of cloud phase has a major role in precipitation formation processes and cloud electrification. Active sensors, such as Lidar, can be used to derive cloud phase vertical profile, but these measurements are limited due to the narrow spatial coverage and low repetition time. Passive sensors, such as MSG-SEVIRI, have much better spatial coverage and temporal resolution but retrieve phase information only from cloud tops. Here we present a methodology to gain information on cloud phase vertical profile of cloud system from MSG data. The 15 minutes cycle of MSG is used to monitor vertical phase development providing additional information on precipitation and electrification. An application is presented to lucid the usage of the cloud phase vertical profile. Clouds growing over polluted air mass are shown to have more soopercooled water tops and wider mixed phase region, while full glacition take place at extreme temperatures.

P-Interfaces.28 ID:4363 15:35

Cloud Condensation Nuclei Measurements and Closure Study on high Arctic Aerosol during the ASCOS Field Campaign 2008

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A fog / low-level cloud layer is often observed in the high Arctic planetary boundary layer during summertime, which potentially has a large impact on the radiative balance of the atmosphere and thus on regional climate. However, the meteorological processes and aerosol-cloud interactions involved in this phenomenon are not fully understood yet. This includes the physico-chemical properties of the aerosols and their ability to act as cloud condensation nuclei (CCN). During the Arctic Summer Cloud Ocean Study (ASCOS) aerosol physical and chemical properties were measured with various instruments in the high Arctic (north of 80° N) on the Swedish icebreaker Oden. The goal was to study Arctic summertime clouds. The campaign lasted six weeks in summer 2008. Here we present CCN properties of the ambient Arctic aerosol. CCN number concentrations and CCN activated fractions were obtained from in-situ measurements using a continuous-flow streamwise thermal-gradient CCN counter and a condensation particle counter. Five different supersaturations were scanned for the polydisperse aerosols. Furthermore, a CCN closure study was performed. This includes a comparison of the directly measured CCN number concentration with the number concentration gained by using bulk aerosol mass data from an aerosol mass spectrometer and size distributions obtained from a differential mobility particle sizer. Koehler theory and an internally mixed aerosol were assumed. Sensitivity studies on the closure were carried out, varying the assumed hygroscopic properties of the considered compounds as well as their chemical composition. Closure could be achieved with different sets of parameters for most of the data from the six week time period.

P-Interfaces.29 ID:4554 15:35

Hygroscopicity of ambient organic aerosol from CCN measurements in relation to degree of aerosol oxygenation

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During the spring of 2007, ambient concentrations of cloud condensation nuclei (CCN) were measured at Egbert, Ontario and compared to concentrations predicted using κ Köhler theory from the aerosol chemical composition and size distribution. This study looks specifically at the hygroscopicity of the organic component of the aerosol, and characterizes it using two methods, both of which are based on its degree of oxygenation, as measured by an aerosol mass spectrometer. The first method uses a statistical technique, positive matrix factorization, to characterize the organic component into oxygenated and unoxygenated factors, while the second method uses the mole ratio of atomic oxygen to atomic carbon (O/C). By assuming that the unoxygenated factor is non-hygroscopic, we find that the oxygenated factor has a hygroscopicity parameter of 0.22 ± 0.04. Similarly, by assuming a simple linear relationship between the hygroscopicity of the organic component (κ_{org}) and the O/C, we find that $\kappa_{org} = (0.29 \pm 0.05) \times (O/C)$ for the limited range of O/C measured in this study (0.3 to 0.6).

P-Interfaces.30 ID:4151 15:35

Aerosol properties affecting CCN ability of Mixed Organics in Aqueous Aerosol Mimics

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Glyoxal (G) and methylglyoxal (MG) are both gas-phase oxidation products of many volatile organic compounds (VOCs). They have also been shown to be potential precursors to the formation of secondary organic aerosol (SOA) material via heterogeneous processes. Most previous studies of SOA formation by G and MG have focused on either species separately, however G and MG are unlikely to exist in isolation in the troposphere. We studied the formation of SOA material in aqueous aerosol mimics containing mixtures of MG and G with ammonium sulfate. Light-absorbing secondary organic products were characterized using UV-Vis spectrophotometry. We found that this process can be described using models for the formation of light-absorbing products by G and MG in isolation operating in parallel. Pendant drop tensiometry measurements show surface tension depression in these solutions, which can be modeled as a linear combination of the effects of G and MG in isolation. Product species were identified using chemical ionization mass spectrometry with a volatilization flow tube inlet (Aerosol-CIMS). Most peaks could be attributed to self-reactions of G or MG, but some peaks were consistent with G-MG cross reaction products. The effect of SOA formation by MG and G on aerosol CCN activity will be discussed. We conclude that cross reactions may increase SOA mass yield from uptake of G and MG, but they do not influence the effects of this process on aerosol surface tension or light absorption.

P-Interfaces.31 ID:4120 15:35

Aerosol Coupling in the Earth System (ACES): The formation of secondary organic aerosol from real plant emissions.

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Biogenic volatile organic compounds (BVOC) can undergo photo-oxidation reactions in the atmosphere. The products of these reactions may condense onto atmospheric particles, or nucleate in the absence of pre-existing seed, to form secondary organic aerosol (SOA). The ability of BVOC to form SOA is particularly important in remote tropical locations where almost half of all reactive BVOC are emitted. However, the ease with which individual BVOC, or their products, partition into the particle phase is the subject of some debate.

This study characterised the BVOC emissions from three south-east Asian tropical plant species (*Ficus cyathistipula, Ficus benjamina* and *Caryota millis*) and, in a series of smog chamber experiments, examined their ability to contribute to SOA formation under atmospherically relevant conditions. All three species were found to be high isoprene, low monoterpene and sesquiterpene emitters. Under simulated tropical rainforest environmental conditions, we found that these predominantly isoprene emitting plants did not contribute to the formation of SOA. However, analysis of gas phase GC-MS and PTR-MS data indicated that isoprene epoxide and hydroxyhydroperoxide, both suggested to be isoprene gas phase aerosol precursors, were produced. In contrast, experiments under the same conditions using common silver birch (*Betula Pendula*), a high monoterpene and sesquiterpene, but low isoprene emitter, resulted in SOA formation via both fresh nucleation and condensation onto pre-existing seed. Under these experimental conditions, our data suggest that isoprene may contribute only indirectly to the formation of SOA via reactions involving second and third generation oxidation products.

P-Interfaces.32 ID:4432 15:35

The Photo-reduction of Oxidized Mercury Species with Selected Thiols at the Air/Water Interfaces

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Mercury is an environmental contaminant of global concern. The reduction of oxidized mercury species (Hg(II)) to elemental mercury (Hg(0)) affects the global distribution of mercury and competes for methylation processes of mercury in aquatic environment. This study focused on the kinetics and mechanistic studies of the reduction of Hg(II) by thiols. The reaction kinetics was studied using cold vapor atomic fluorescence spectroscopy (CVAFS), the formation of Hg(II)-thiol complexes (Hg(SR)2) were confirmed by UV-visible spectrometry and Atmospheric Pressure Chemical Ionization-Mass Spectrometry (APCI-MS), and the reaction products were analyzed using Electron Impact-Mass Spectrometry (EI-MS) and Solid Phase Microextraction coupled with Gas Chromatography-Mass Spectrometry (SPME/GC-MS). The reaction rates were obtained for 1-propanethiol, 1-butanethiol, and 1-pentanethiol at environmental relevant conditions. The effects of light, pH, dissolved oxygen and chloride ion on reaction rates were also investigated. Our results indicated that the reaction intermediate, i.e. (Hg(SR)2), could absorb photons with wavelengths longer than 290 nm (the sunlight limit) and subsequently be photo-reduced to Hg(0). The identified products were Hg(0) and disulfides (R-S-S-R). The potential environmental implications are herein discussed.

P-Interfaces.33 ID:4189 15:35

Determination of Formaldehyde in Ambient Air Using Solid Phase Microextraction (SPME)

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Formaldehyde is the highest concentration aldehyde in the atmosphere with typical concentration ranging from 0.3 to 2 ppbv in Canada. It is an important intermediate in the gas-phase methane oxidation chain and plays an important role in the chemistry of troposphere by influencing its hydroxyl radical (HOx) budget, and thus its oxidative capacity. It can be produced anthropogenically and biogenically and can also be transferred from the ocean. Weather the ocean is the source or sink of the formaldehyde is of interest to scientists in order to completely understand the chemical processes happening in the ocean/air interface. We develop a method for determining formaldehyde in ambient air using solid phase microextraction method (SPME), a facile, low-cost, versatile technique. The formaldehyde is derivatized on the SPME fiber to its pentafluorobenzyl oxime using 1, 2, 3, 4, 5-pentafluorobenzylhydroxylamine (PFBHA) and then analyzed with gas chromatography with flame ionization detector (GC-FID). We herein present the methodology developed, the calibration curves, and some results from outdoor/indoor samplings. We also present an inter-comparison of this method with DNPH derivatization with HPLC method.

P-Interfaces.34 ID:4346 15:35

Effects of coupling and relative humidity on HONO mixing ratios in a spruce forest

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We made HONO measurements using two long path absorption photometers (LOPAPs) at a field site located in the Fichtelgebirge mountains in north-eastern Bavaria, Germany (50°09’N, 11°52’E, 775m above sea level) within the framework of the project “ExchanGE processes in mountainous Regions” (EGER). HONO was measured simultaneously close to the forest floor (0.5 m above ground) and at 24.5 m height (just above the spruce forest canopy top) from 13 to 26 September 2007 using two long path absorption photometers (LOPAPs). Diel cycles of HONO and mixing ratio differences between above canopy and close to the forest floor are discussed in view of the coupling between canopy and the air above and are related to the dependency of HONO mixing ratios on relative humidity. Since HONO is rapidly photolyzed during daytime, it typically featured a pronounced diel cycle with low levels (50 to 100 ppt) around noon and higher levels (up to 500 ppt) at nighttime during a dry period (20-25 Sep.). The shading from the forest canopy reduces photolysis in the trunk space to about 4-10% of the values measured above the canopy. Thus, the HONO lifetime ranges from 70 to 200 minutes below the canopy and is about 10 minutes above the canopy around noontime on a clear sky day. Despite this, the differences of the measured HONO mixing ratios are about zero (± 20 ppt) from 8:00 to 13:00 CET. This is attributed to an enhanced vertical mixing within the forest canopy during daytime as indicated by the coupling regimes calculated from turbulence measurements of horizontal and vertical wind components and sonic temperature using sonic anemometers according to Thomas and Foken [1]. In the early afternoon, starting about 13:00 CET, a trend towards higher HONO mixing ratios in the trunk space was detected, which considerably increases at 17:00 CET one hour before sunset, when the coupling regime changes. Coinciding with the increasing HONO mixing ratios, the relative humidity increases below the canopy.

[1] C. Thomas and T. Foken; Flux contribution of coherent structures and its implications for the exchange of energy and matter in a tall spruce canopy; Boundary-Layer Meteorology;123; 2007; 317-337

P-Interfaces.35 ID:4410 **15:35**

IMPACT OF TROPOSPHERIC OZONE ON VEGETATION AS SIMULATED BY THE CHIMERE-ORCHIDEE COUPLED MODEL

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The impact of ozone on different European forest ecosystems is largely under-investigated, despite huge areas of Europe are exposed to high ozone levels (> 40 ppb). We investigated the potential effects of ozone on photosynthesis and leaf area index using a land surface model (ORCHIDEE) fully coupled with a chemistry transport model (CHIMERE) for the whole year 2002. Results shown that the effect of tropospheric ozone on vegetation lead to a reduction in yearly gross primary production (GPP) of about 22%, as well as a reduction in leaf area index (LAI) of 15-20%. Larger impacts have been found during summer, when ozone reach the higher concentrations. During these months the maximum GPP decrease is up 4 gC m-2 d-1, and the LAI reduction is up to 0.7. In general we found that crops are the most sensitive to the ozone, while the deciduous trees exhibit a larger reduction in photosynthesis with respect to needle-leaves species. Since CHIMERE uses the LAI computed by ORCHIDEE to account for the biogenic emissions, a LAI reduction has severe implications on the simulated atmosphere chemistry. Specifically, we found a significant reduction in ozone precursors that lead to a change of both tropospheric and stratospheric ozone concentrations, and to large changes in surface NO2 concentration.

P-Interfaces.36 ID:4602 **15:35**

Ozone pollution during stratosphere – troposphere exchange events over equatorial Africa

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Both natural factors such as thunderstorm events and anthropogenic activities contribute to very high ozone production. On the flight route from Johannesburg to Vienna enhanced ozone and relative humidity spikes were observed by MOZAIC (Measurements of Ozone by Airbus In service airCraft). MOZAIC recorded high resolution in-situ ozone and relative humidity at a flying altitude of 250-200 hPa at equatorial Africa. This area is one of lightning hot spot regions of the world. We report introduction of enhanced ozone of stratospheric origin into the troposphere during two events and the resulting pollution. Vertical cross-section of potential vorticity over the region of interest showed high PV intrusion below the tropopause level. Both OLR and vertical wind have indicated presence of strong convection. Cloud water content transport and high latent heat have confirmed the existence of thunderstorm activity coupled to PV intrusion. The two distinct events are characterized by very low ozone within the thunderstorm cloud and very high ozone of stratospheric origin outside the thunderstorm cloud. The events have produced ambient air pollution.

P-Chemistry Climate.1 ID:3354 **15:35**

ASSESSMENT OF FACTORS RESPONSIBLE FOR CLIMATE CHANGE AND HUMAN HEALTH

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Weather and climate play important roles in determining patterns of air quality over multiple scales in time and space. Air quality is strongly dependent on weather and is therefore sensitive to climate change. There is growing recognition that development of optimal control strategies for key pollutants like fine particles now requires assessment of potential future climate conditions and their influence on the attainment of air quality objectives. Climate change induced by anthropogenic warming of the earth's atmosphere is a daunting problem. In addition, other air contaminants of relevance to human health, including smoke from wildfires and airborne pollens and molds, may be influenced by climate change. While further research is needed, climate change coupled with air pollutant exposures may have potentially serious adverse consequences for human health in urban and polluted regions. Climate change producing alterations in: food webs, lipid dynamics, ice and snow melt, and organic carbon cycling could result in increased PMs level in air. In this study, the focus is on the ways in which health-relevant measures of air quality, including particulate matter, and aeroallergens, may be affected by climate variability and change. The small but growing literature focusing on climate impacts on air quality, how these influences may play out in future decades, and the implications for human health is reviewed. Keywords: Air pollution, Climate change, health effects,

P-Chemistry Climate.2 ID:4556 **15:35**

The feedbacks of short-term emission control and annual emission changes on the regional climate during the 2008 Beijing Olympic Games

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As the host of the 2008 Summer Olympic Games, Beijing drew international attention for its severe air pollutions. To improve air quality and maintain clean air throughout the event, systematic long- and short-term emission reduction measures and regulations were implemented. However, there has been a debate going on whether the good air quality during the Olympic Games was resulted from the emission control and/or the “favorable” meteorological conditions. For example, the precipitation data taken from the United Precipitation Project of the NOAA Climate Prediction Center have shown that from 8 August to 17 September, in Beijing 225 mm rain accumulated in 15 rainy days, substantially more than 73 and 91 mm in 6 and 7 rainy days for the same period in 2006 and 2007 respectively [Mijling et al., 2009]. Therefore, this raises the question of whether the feedbacks of the reduced emissions (long-term reduction and short-term control) on the local and regional weather system itself resulted in a weather system that was more “favorable” to the pollutant scavenging and dilution in Beijing. Did the aerosol direct and indirect effects arising from the emission reductions lead to more precipitation and increased frequency of stronger northerly wind? To gain insight into this aspect, numerical simulation experiments were carried out with the Weather Research and Forecasting Model with fully coupled “on-line” chemistry model (WRF-Chem [Grell et al., 2005]) for the Olympic period to exam the air quality, regional climate, and aerosol-cloud-precipitation interactions in the Beijing and its surrounding region. Different emission scenarios were applied and the detailed analyses about the feedbacks of short-term emission control and annual emission changes on the regional climate, especially on the precipitation are presented.

P-Chemistry Climate.3 ID:4103 15:35

Integrated Modeling System of Environmental Risks in the Metropolitan Area of São Paulo, Brazil

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Urban areas concentrated more than 50% of global population and the projection is of increasing participation. In Brazil, nowadays, approximately 80% of the population is living in urban areas. Large urban areas as megacities have a significant contribution to emissions of aerosols, primary gases and greenhouse gases, playing a great impact on climate and human health. In the Metropolitan Area of São Paulo (MASP) pollution events characterized by high concentration of particles and ozone are aggravated by meteorological conditions that are unfavorable to dispersion, which frequently occur during wintertime. Several scientific studies have shown an increase in hospital admissions associated with these events, especially among children and the elderly, due to respiratory and cardiovascular diseases. In the MASP, bad air quality is, mainly due to standards violations by ozone and PM10. Vehicles contribute for more than 90% of primary gases pollutants emissions. The Weather Regional Forecast Chemical model (WRF/chem) has been used to study the regional air quality of São Paulo megacity and their implications in human health. Simulations were performed using the model WRF/Chem for two periods: from September 05 to September 10 of 2004 and from October 29, 2006 to November 01, 2006. Temperature, humidity, CO, NOx, SO2, PM10, O3 data were used to validate the model. Also the integrated air quality model was applied to estimate risks of mortality and morbidity due to ozone and PM10 concentration levels. The preliminary results indicate that the integrated model is able to represent the behavior of ozone in ground surface and also in vertical layers, although over estimates the peak of ozone. The results concerning the aerosol also indicate that the model can represent the diurnal variability but with less agreement between simulated and observed values. The risk of mortality and cardio-respiratory diseases for all ages in the MASP due to ozone concentrations indicate higher risks outside the downtown of region, because of the transport of its precursors. In addition, the risk of mortality associated to PM10 was higher than to ozone mainly for respiratory diseases up to 2% of increasing.

P-Chemistry Climate.4 ID:4417 15:35

A study of the impact of land use change in Borneo on atmospheric composition using a global model

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A high resolution version of the Cambridge p-TOMCAT model has been used to analyze results from the recent NERC-funded OP3 measurement campaign in Borneo. By using the chemical transport model at 50km resolution, we have begun to understand the impact of emissions and other physical processes on regional chemistry. In particular, we have run several model scenarios looking at the potential impact of land use change of forest to oil palm in Borneo, and the corresponding change in isoprene emissions, on local and regional atmospheric composition. Oil palm is one of the World's most rapidly expanding equatorial crops, with Indonesia and Malaysia being the two largest producing countries. Several model emission scenarios are run for the OP3 measurement period, including emissions from global datasets and local flux measurements. Isoprene fluxes observed during OP3 at a forest site were considerably less than fluxes based

on the global MEGAN dataset. Using the OP3 observed fluxes in the model substantially improved the comparison between modeled and observed isoprene mixing ratios, and had a significant impact on modeled O3 and OH over Borneo. Further model scenarios performed show that replacing forest with oil palm has the potential to significantly alter the atmospheric oxidizing capacity over Borneo.

P-Chemistry Climate.5 ID:4430 15:35

Biogenic VOCs in the future: Can atmospheric CO2 and land-use change shift drastically isoprene emission estimates?

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Isoprene is the first volatile organic compound (VOC) emitted by the terrestrial biosphere with emission estimates ranging so far between 400 and 600 TgC/yr. As a reactive species involved in both gas phase reactions (ozone cycle, atmosphere oxidising capacity…) and secondary organic aerosol formation, isoprene is of high interest for atmospheric chemistry processes. Environmental conditions of temperature, radiation, vegetation type or even atmospheric chemical composition drive the level and distribution of isoprene emissions. Indeed recent works underline that the evolution of carbon dioxide concentration strongly impacts the isoprene emission capacity of plants. In the future, changes in climate, land-use or atmospheric CO2 concentrations could therefore affect significantly biogenic isoprene emissions and, consequently, impact atmospheric concentrations of key compounds such as ozone or nitrogen oxides.

The objective of our work is to examine the evolution of biogenic emission of isoprene in the future together with the consequences on atmospheric chemistry. The ORCHIDEE global vegetation model, including parameterisations for BVOC emissions, as well as the global chemistry-transport model LMDz-INCA are used for this study. A parameterisation based on recent publications is included in the ORCHIDEE model to take into account the impact of atmospheric CO2 concentrations on plant isoprene emission capacity. Several scenarios of changes in climate and land-use are considered and their related impacts on emissions and atmospheric chemistry are compared.

P-Chemistry Climate.6 ID:4388 15:35

Climate Feedback of Methane Emissions From Rice Fields.

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Rice fields are known to be an important source of anthropogenic methane to the atmosphere and have contributed to its increase over the last century as well as causing some of the slowdown of trends that has been observed in recent times. But as the Earth’s climate warms, these emissions will change since the underlying biological process depend on temperature. Moreover, since the natural wetlands emit methane by similar mechanisms, there could be a substantial feedback from global warming. The actual response is affected by two competing effects: The increase of temperature adds to the production of methane by methanogenic bacteria, but at the same time it may increase the consumption of methane by methanotropic bacteria. Further, the transport process that affect the net emissions are also dependent on ambient temperature and may increase causing greater emissions. The combination of these mechanism can either increase, stabilize or decrease emissions depending on the environmental circumstances and the sensitivity of the biological system to temperature changes. Our work examines the relationships between these processes and provides estimates of the effect of these feedbacks on future emissions. If these ideas are extended to natural wetlands the positive feedback effect can add to current calculations of future methane concentrations.

P-Chemistry Climate.7 ID:4581 15:35

Sea of Scenarios: what do we know about projected methane emissions?

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Climate-chemistry model experiments for the 21st century have relied almost exclusively on a restricted set of methane (and other) emission projections primarily from economic-centered models. In baseline scenarios, emissions commonly rise continuously for several decades while those reflecting implementation of mitigation technologies often exhibit deep emission reductions by 2030 and later. Lack of transparency makes it extremely difficult to determine drivers of the emissions and which technology mixes underpin the high levels of mitigation reported. In contrast to the baseline-and-mitigation approach, the IPCC SRES family of projections encompassed multiple economic, demographic, environmental etc. trajectories quantified by several models. However, the wide range of SRES future emissions is equally difficult to understand or evaluate. Lastly, the primary focus of most emission projections has been on carbon dioxide, with later and modest inclusion of methane.

The largely black-box nature of emission projections used in climate-chemistry experiments and policy analyses has received almost no attention despite the substantial influence exerted by these inputs. This lack of transparency makes assessing the likelihood, or even plausibility, of projected emissions all but impossible. Thus modeled climate-chemistry interactions with future emissions are more like sensitivity experiments than predictions, while policy discussions lack the practical understanding needed to decide on mitigation strategies. Plans for the next IPCC assessment include additional scenario development. We propose further analyzing existing projections and mitigation options with the goal of reducing, not increasing, the suite of scenarios for climate studies.

We have developed new, transparent methane projections for the major methane sources. We report on work to assess the plausibility of assumptions and data underlying the new and existing projections that includes analysis against historical trends, and development and testing of plausibility criteria.

P-Chemistry Climate.8 ID:4403 15:35

Ozone and Methane budgets in the ACCMIP models

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I will present a preliminary analysis of ozone and methane budgets from the ACCMIP model integrations. These span from pre-industrial times up to the present-day and then follow a variety of future scenarios (the RCPs, or representative concentration pathways) agreed for the forthcoming IPCC AR5. Several models have submitted (and will be submitting) results, allowing inter-model uncertainty to be quantified. Findings

will be compared to those reported in IPCC AR4 and from previous intercomparisons (e.g. ACCENT, HTAP).

P-Chemistry Climate.9 ID:4337 15:35

Potential climate feedbacks involving the methane cycle

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Natural emissions of methane may increase in a warmer climate due to potential feedbacks in the Earth System related to wetlands, permafrost, and marine hydrates. As a result, atmospheric methane concentrations could continue to rise over the next century despite efforts to reduce anthropogenic emissions.

In this study, the atmospheric response to methane pulse emissions is investigated and the sensitivity of the response to the size, location, and season of release is also explored. A particular focus of the atmospheric response is on methane itself, methane lifetime, tropospheric ozone, stratospheric water vapor, radiative forcing, and the 100-year global warming potential. The impact of methane emissions on the direct and indirect effects of sulfate and nitrate aerosol is also examined.

On the basis of these experiments, the forcing resulting from a potential methane release from wetlands and/or permafrost is estimated for a range of climate change scenarios.

P-Chemistry Climate.10 ID:4404 15:35

Does the location of aircraft nitrogen oxide emissions affect their climate impact?

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We present results from 112 one-year global chemistry-transport model integrations: a base case, then variants with extra aircraft nitrogen oxide (NOx) emissions added to specific regions in the first month (July). The NOx stimulates ozone (O3) production and methane (CH4) destruction. Responses vary spatially: low background NOx regions are most sensitive. Integrated (100-year time horizon) radiative forcings (IRF) are calculated. Net (O3 + CH4) IRFs for July aviation NOx are generally negative: the global average, weighted by emissions, is -1.9 mW m-2 yr(Tg NO2)-1. The positive IRF associated with the short-term O3 increase (4.1 mW m-2 yr(Tg NO2)-1) is overwhelmed by the effects of the long-term CH4 decrease. Aircraft NOx net IRFs are spatially variable, with July values over the remote Pacific approximately balancing the IRF associated with aviation CO2 emissions (28 mW m-2 yr(Tg NO2)-1). The overall climate impact of global aviation is often represented by a simple multiplier for CO2 emissions. These results show that this is inappropriate.

P-Chemistry Climate.11 ID:4228 15:35

A Reassessment of the net climate impact of aviation

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Aircraft emissions can affect climate change through increasing CO_2 but also a host of other short-lived non-CO_2 effects that are complex, involve impacts that are both warming and cooling and are unique to this sector. The most important effects involve ozone, sulfate, black carbon (soot), methane, water vapor and indirect impacts on cloudiness. Combined, the non- CO_2 climate impacts outweigh those of CO_2 on short timescales (20-30 years). Accounting for non-CO_2 impacts in cross- sector emissions trading schemes is controversial, but some have proposed using a simple multiplicative factor between 2 and 4 to be applied to aviation CO_2 emissions. This factor is based on previous assessments of aviation climate impacts by the Intergovernmental Panel on Climate Change and updates. Those assessments have used a segmented approach whereby each effect was calculated separately and the effects summed. However, more comprehensive integrated approaches that represent coupled heterogeneous interactions between gas and aerosol particles in the exhaust and background atmosphere are now possible and give markedly different results. Here, application of a fully coupled global chemistry-aerosol-climate model that includes both tropospheric and stratospheric chemistry leads to a lower ozone radiative forcing and higher direct aerosol radiative forcing from aviation emissions than previously estimated. The new results do not support the use of any multiplicative factor on short timescales in climate policy. The future evolution of the aviation climate impact is compared to those from a broad range of other key economic sectors over the next century. The climate consequences of future growth in aviation emissions and potential mitigation scenarios are explored.

P-Chemistry Climate.12 ID:4535 15:35

Importance of distinct organic aerosol sources on gas-phase chemistry and climate

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Organic aerosols, a ubiquitous component of atmospheric particulate matter, are still poorly constrained in global models. New sources and production mechanisms have been discovered in the last few years, including primary and secondary sources from the ocean, important new terrestrial secondary organic aerosol precursors, and cloud processing. Most global models to date include few, if any, of these new sources. Although their importance on gas-phase chemistry and climate is expected to be very significant, many models still include very simple organic aerosol calculations, treating both the primary and secondary components as non-volatile. In order to quantify the role organic aerosols play in gas-phase chemistry and climate, we studied their interactions with solar radiation. Aerosols affect gas-phase chemistry via their light extinction potential, affecting the photolysis rates of trace gases. As a result, both oxidant levels and the chemical composition of the atmosphere in general are altered, feeding back to secondary organic aerosol production. In addition, changes in organic aerosol distribution, both temporal and spatial, affect climate via aerosol direct and indirect effects. The total impact of organic aerosols on these processes will be studied. The importance of new aerosol sources like their production from isoprene in addition to terpenes, the oceanic source of primary as well as secondary organic aerosol precursors will be evaluated. The calculated changes in aerosol distribution and impacts due to the inclusion of semi-volatile secondary organic aerosols will be examined. The importance of organic aerosols on both direct and indirect effect will be calculated. Organic aerosol distributions will be evaluated against measurements.

P-Chemistry Climate.13 ID:4249 15:35

Composition, Size Distributions, and Elemental Source Markers in Aerosols Over Snow and Ice - Comparing Greenland and Mid-Latitude Mountain Environments to Improve Global Simulation of Aerosol-Cryosphere Interactions.

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Aerosol deposition is believed to significantly alter the albedo, and thus melt rates, of snow and ice in polar and montane environments, with important consequences for planetary radiation balance, regional hydrology, and water resources. Accurate modeling of aerosol-snow interactions requires detailed information on the physical and chemical characteristics of the aerosols. Analysis of in-situ measurements of aerosols over snow and ice provides insights into the nature and sources of the aerosols and permits evaluating the quality of model simulations at local to global scales. We report analyses of more than 2000 Daily size-resolved aerosol samples collected from 2003 through 2006 at the summit of the Greenland Ice Cap and at high altitude sites in the Sierra-Cascade Range in western North America. These data permit comparison of aerosols over snow at hemispheric, regional, and local scales. We find significant differences in the nature of aerosol - cryosphere relationships at all three scales.

P-Chemistry Climate.14 ID:4208 15:35

Model evaluation for air quality prediction in a changing climate

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Climate change may have a significant impact on the dynamics and chemistry of the atmosphere and may thus affect air quality. To date, little is known about the effects of climate change on the concentrations of ozone and particulate matter. Especially for particulate matter the few available studies do not provide a consistent result, including the sign of the effect. The regional chemistry transport model LOTOS-EUROS has a long tradition in modelling ozone and particulate matter, with transport driven by ECMWF analysis meteorology. Recently, the model was coupled to the regional climate model RACMO. We have investigated the performance of the combination LOTOS-EUROS/RACMO for climate modelling. To this end, air quality simulations driven by analysed meteorology were compared to two present day climate simulations downscaling ECHAM and MYROC GCM results. All simulations were compared to (climatological) observations. In this way, biases in the RACMO climate meteorology and their impact on modelled air quality can be quantified. Thorough knowledge on biases and their impacts is crucial for the assessment of scenarios for a future climate.

P-Chemistry Climate.15 ID:4463 15:35

Ensemble Analyses of the Effects of Global Change on US Air Quality

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Global change will clearly have a significant impact on the environment. In this study, we examined the effect of changes in chemical boundary conditions and continental US emissions on US air quality using a multi-scale modeling framework. Meteorological fields were downscaled from results of CCSM3 and ECHAM5 global climate models using WRF. Two nested domains were employed, one covering most of the Northern Hemisphere from eastern Asia to North America at 220-km horizontal resolution (hemispheric domain) and one covering the continental US at 36-km resolution. Meteorological results from WRF were used to drive MEGAN biogenic emissions model, SMOKE emissions processing tool, and CMAQ chemical transport model to predict ozone and aerosol concentrations for the current (1995-2004) and future decade (2045-2054). MEGAN model was used to calculate biogenic emissions for both domains. For the current decade and hemispheric domain, year 2000 global emissions of ozone precursors from anthropogenic, natural, and biomass burning sources from POET and EDGAR emission inventories were used. Global emissions inventories for black and organic carbon from Bond et al (2004) were applied. For the future decade, these emissions were projected to 2050 following the Intergovernmental Panel for Climate Change A1B emission scenario. WRF and CMAQ results from the hemispheric domain simulations provided the boundary conditions for the US continental simulations. For the continental simulations, US anthropogenic emissions for the current decade were based on the 2002 National Emissions Inventory prepared by the Environmental Protection Agency. For the future decade simulation, these emissions were projected to 2050 using growth factors from the U.S. EPA MARKAL database and energy system model following a scenario that assumes baseline criteria pollutant policies, including the application of the Clean Air Interstate Rule (CAIR). We present results showing the changes in US air quality due to changes in future global emissions and US anthropogenic emissions.

P-Chemistry Climate.16 ID:4395 **15:35**

Particulate matter in global climate and air quality policies: co-benefits and trade-offs

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Policies to reduce emissions of traditional pollutants may increase or decrease emissions of greenhouse gases. Likewise, policies to reduce greenhouse gases can have positive or negative effects on air pollution. Moreover, abatement measures which lead to regional emission reductions on the short term might not be beneficial on the long term and at global scale, instead emissions may merely have been moved outside the polluted area, where in the worst-case net increases in emissions may occur. An integrated approach aimed at designing robust and synergistic abatement policies is highly desirable and should be based on a scientific understanding of the multi-component greenhouse gas-air pollutant system, considering co-benefits and trade-offs resulting from both structural and end-of-pipe abatement strategies. For this study we apply consistent emission scenarios for greenhouse gases and air pollutants in a global chemical transport model (TM5). We will discuss the co-benefits of a 2° target climate policy scenario towards air quality in different world regions, and evaluate which sectors are preferably targetted in order to optimally improve air quality in combination with a reduction of the positive radiative forcing caused by short-lived warming agents (soot aerosols, tropospheric ozone.) The focus will be on particulate matter which plays a pivotal role in both environmental (health) impact and the atmosphere's radiative balance.

P-Chemistry Climate.17 ID:4299 15:35

Correlations between fine particulate matter (PM2.5) and meteorological variables in the United States: implications for the sensitivity of PM2.5 to climate change

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We applied a multiple linear regression (MLR) model to quantify the relationships of total PM2.5 and its components with meteorological variables using an 11-year (1998-2008) observational record over the scale of the contiguous US. The data were deseasonalized and detrended to focus on synoptic-scale correlations. We find that daily variation in meteorology as described by the MLR model can explain up to 50% of PM2.5 variability, with temperature and precipitation being the most significant predictors. Temperature is positively correlated with sulfate, organic carbon (OC) and elemental carbon (EC) almost everywhere. The nitrate-temperature correlation is mostly negative but is positive in California and the northern US. Relative humidity is positively correlated with sulfate and nitrate, but negatively with OC and EC. Precipitation is strongly negatively correlated with all PM2.5 components. We find that PM2.5 concentrations are on average 2.6 μg m-3 higher on stagnant vs. non-stagnant days, and more than 5 μg m-3 higher in some areas. Extrapolation of current stagnation trends suggests that summer mean PM2.5 could increase by up to 1 μg m-3 by 2050 in some areas as a result of increased stagnation. Our observed climatological correlations provide a test for chemical transport models (CTMs) used to simulate the sensitivity of PM2.5 to climate change. They point to the importance of representing the temperature dependence of agricultural, biogenic and fire emissions in CTMs. Application to the GEOS-Chem CTM will be presented.

P-Chemistry Climate.18 ID:4552 15:35

Aerosol Hygroscopicity Distribution and Mixing State Determined by Cloud Condensation Nuclei(CCN) Measurements

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This paper presents, firstly the concept of hygroscopicity distribution and its application in the analysis of cloud condensation nuclei (CCN) measurement data. The cumulative particle hygroscopicity distribution function H(κ) is defined as the number fraction of particles with a hygroscopicity parameter, κ, smaller than a certain value of κ. Since the measured CCN (at supersaturation S) can be considered as those particles with κ larger than a certain value, the CCN efficiency spectra (activation curve) can be easily converted to H(κ) distributions. Unlike studies calculating only one hygroscopicity parameter from a CCN activation curve, the concept of H(κ) shows the usefulness of all points on the activation curve. Modeling studies of three assumed H(κ) distributions are used to illustrate the new concept H(κ) and how it is related to the size-resolved CCN measurements. Secondly, we discuss the aerosol mixing state information that can be obtained from the shape of H(κ). A case study is performed based on the CCN measurements during the CAREBEIJING 2006 campaign. In the campaign-averaged H(κ) distribution, most particles (>80%) lie in a mode with a geometric mean κ around 0.2-0.4, and an increasing trend in the mean κ is found as particle size increases. There seems to be another less hygroscopic mode but the κ resolution (depending on the size resolution) in the campaign is not high enough to interpret it. It is also clear that H(κ) is not a monodisperse distribution (implying an internal mixture of the aerosols). The dispersion parameter σg,κ, which is the geometric standard deviation of H(κ), can be used as an indicator for the aerosol mixing state. The indicator σg,κ shows good agreement with the soot mixing state measured by a volatility tandem differential mobility

analyzer (VTDMA) during the CAREBEIJING 2006 campaign. The concept of H(κ) can be widely used to study aerosol mixing states, especially in the lab experiment where a high κ resolution can be achieved. Since many cloud models are treating aerosols as internally mixed, it is worthwhile to carry out sensitivity studies based on the H(κ) measured for real atmospheric aerosols.

P-Chemistry Climate.19 ID:4288 15:35

Changing anthropogenic emissions and their regional scale impacts on aerosol characteristics

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Effects of aerosols on climate and human health depend on their size as well as their chemical composition. These properties are governed by the history of the aerosols within the atmosphere. We investigate the ability to simulate ambient aerosols characteristics on a regional scale, using a comprehensive Eulerian chemistry transport model (CTM). The aim is to quantify contributions to aerosol composition made by different emission source categories. We are then looking into the effects that proposed emission regulations would have on the characteristics of ambient aerosols. The weather prediction model COSMO has recently been extended by detailed, online-coupled gas-phase chemistry and aerosol description. This framework is called COSMO-ART. Our simulation domain covers Europe at approx. 10 km horizontal resolution and 40 levels in the vertical. The model is forced by IFS analyses from ECMWF. TNO (Netherlands) provided a very detailed, spatially high-resolved (approx. 8 km) anthropogenic emissions inventory. Reanalyses of the global CTM IFS-MOZART serve as lateral forcing for chemical tracers. Both, a winter and a summer period of 2 weeks were chosen to investigate the accuracy of the model simulations under different meteorological conditions. Detailed comparisons against long-term station observations (EMEP), other model simulations (MM5/CAMx), satellite observations (NO_2 columns from OMI, AOD estimates from Meteosat SEVIRI/MODIS) and aerosol mass spectrometer (AMS) measurement campaigns are made to assess the quality of the model results. Sensitivity studies with changing total emissions from different source categories like industry or traffic are conducted to investigate their contributions to the total amount and composition of ambient aerosols. Further, the introduction of new policies (e.g. EURO6 regulations for car emissions), and overall future scenarios (IIASA projections) are also simulated. With our work we can deliver detailed advice to air quality policy makers, as well as supply improved input data for better constrained climate projections.

P-Chemistry Climate.20 ID:4278 15:35

First results from the implementation of the piecewise log-normal approximation for aerosols in GEM-AQ

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The computationally efficient piecewise log-normal approximation (von Salzen, 2006) is employed in the Global Environmental Multiscale Air Quality (GEM-AQ) model (Kaminski et al., 2008). GEM-AQ is based on the Canadian weather forcast model (GEM - Global Environmental Multiscale). GEM-AQ includes tropospheric and stratospheric chemistry. Aerosols are included but are computationally expensive so we have evaluated the aerosol scheme using the piecewise log-normal approximation of von Salzen (2006). We

have included sea salt, sulphate, nitrate, organic carbon, black carbon and mineral dust and results will be discussed. With the goal of optimizing the size spectrum resolution to process and aerosol mode dependency, column model sensitivity tests have been performed to determine the response of dry and wet deposition and assumed mass and number distributions to the size partitioning of the distribution. The results from GEM-AQ using this optimum size-partitioning for dry and wet deposition will be presented.

P-Chemistry Climate.21 ID:4574 15:35

Simulations of Aerosol Microphysics in the NASA GEOS-5 Model

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Aerosol-cloud-chemistry interactions have potentially large but uncertain impacts on Earth's climate. One path to addressing these uncertainties is to construct models that incorporate various components of the Earth system and to test these models against data. To that end, we have previously incorporated the Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) module online in the NASA Goddard Earth Observing System model (GEOS-5). GEOS-5 provides a platform for Earth system modeling, incorporating atmospheric and ocean general circulation models, a land surface model, a data assimilation system, and treatments of atmospheric chemistry and hydrologic cycle. Including GOCART online in this framework has provided a path for interactive aerosol-climate studies; however, GOCART only tracks the mass of aerosols as external mixtures and does not include the detailed treatments of aerosol size distribution and composition (internal mixtures) needed for aerosol-cloud-chemistry-climate studies. To address that need we have incorporated the Community Aerosol and Radiation Model for Atmospheres (CARMA) online in GEOS-5. CARMA is a sectional aerosol-cloud microphysical model, capable of treating both aerosol size and composition explicitly be resolving the aerosol distribution into a variable number of size and composition groupings. Here we present first simulations of dust, sea salt, and smoke aerosols in GEOS-5 as treated by CARMA. These simulations are compared to available aerosol satellite, ground, and aircraft data and as well compared to the simulated distributions in our current GOCART based system.

P-Chemistry Climate.22 ID:4226 15:35

Implications of climatological versus fully interactive aerosol distributions on direct and semi-direct aerosol forcing and response in the NASA GEOS-5 atmospheric general circulation model

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Absorbing aerosols such as dust and black carbon reduce the amount of radiation reaching Earth's surface (aerosol direct effect), and contribute to a radiative heating in the atmosphere (aerosol semi-direct effect). This re-partitioning of energy between the surface and the atmosphere has consequences for regional dynamics and the hydrologic cycle. We have implemented a version of the Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) model online in the NASA Goddard Earth Observing System model version 5 (GEOS-5). GOCART includes representations of the sources, sinks, and chemical transformation

of externally mixed dust, sea salt, sulfate, and carbonaceous aerosols. GEOS-5 is an integrated Earth simulation modeling system that includes components for atmospheric and oceanic general circulation and physics, atmospheric chemistry, oceanic biogeochemistry, land surface modeling, and data assimilation. With the GEOS-5/GOCART system, we have an atmospheric general circulation model (AGCM) in which aerosol and trace gas species are dynamically and radiatively interactive with meteorology. Here we assess the sensitivity of global and regional climates and the hydrologic cycle to the direct and semi-direct effects of aerosols by considering a series of simulations in the GEOS-5/GOCART atmospheric general circulation (AGCM) model forced by observed sea surface temperatures. In the control simulation, there is no radiative forcing by aerosols. In another simulation, we have direct and semi-direct forcing from prescribed aerosols only (i.e. aerosol distributions are decoupled from AGCM meteorology). In the final simulation aerosol species are dynamically and radiatively interactive with model meteorology. Each simulation's aerosol distributions are evaluated for their representativeness with comparisons to observations from satellite (MODIS, MISR) and in-situ observations (AERONET). The response of each simulation to aerosol forcing is examined in comparison to the control simulation.

P-Chemistry Climate.23 ID:4527 15:35

Investigating the climate response to the direct and indirect effects of anthropogenic aerosols over South-Asia using an improved version of the coupled atmosphere-mixed layer ocean model SM2.1

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For almost two decades now, aerosols from South Asia has attracted the attention of researchers all across the globe. Investigating the climate response to the direct and indirect effects of anthropogenic aerosols emitted from this region is essential to understand and predict climate change not just in Asia but also globally. This study investigates the equilibrium climate response to the direct and indirect effects of the present day anthropogenic and biomass burning aerosols over South-Asia and monsoon dynamics using an improved version of the coupled atmosphere-mixed layer ocean model SM2.1. Our control experiment includes emission of aerosols from only natural sources such as wind blown dust, sea salt, organics from vegetation, and volcanic aerosols. The successive perturbation experiments include emissions of aerosols from present-day biomass burning and anthropogenic sources (based on IPCC AR5 emission estimates for the year 2000) in addition to the naturally produced aerosols. In all our experiments, we used the 1860 (pre-industrial) forcing data for long-lived green house gases, ozone and land surface properties. The simulations for control and all perturbation experiments have been carried out for 100 model years, and the average results from the last 40 years have been used for further analysis. Anomalies in simulated parameters like precipitation, temperature, atmospheric heating rate, vertical wind velocity, surface pressure, liquid cloud amount, ice cloud amount, SW TOA clear and cloudy sky fluxes etc. between the perturbed cases and the control run will be examined to understand the impact of anthropogenic and biomass burning aerosols on the climate over South-Asia and monsoon dynamics. Our results show that the anthropogenic aerosols over south Asia cools the atmosphere over land and northern Indian ocean, stabilizes the atmosphere over land and weakens the monsoon circulation.

P-Chemistry Climate.24 ID:4557 15:35

Concentration of Nano-particles and Gases in the Environment of Gaborone, Botswana

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This study was intended to carry out continuous and simultaneous monitoring of particles and some of the gases in the environment of Gaborone. Measurements of particulate matter were taken using scanning mobility particle sizer (SMPS) capable of measuring particles with diameters from about 7.5 to 280 nm for about seven months. The concentration of gases CO, CO2, NOx and HC were simultaneously monitored during September 2002 and August 2004 using Horiba gas analyzer APMA360, VIA 510, APNA 360 and APHA 360 respectively. The SMPS and gas analyzers were housed in the Atmospheric Research Laboratory located on the first floor of the building of Department of Physics, University of Botswana. Our measurements show that the average concentrations of carbon monoxide varied from 0.16ppm in March 2004(summer) to 1.48ppm in July 2004(winter). The monthly average concentration of CO2 was seen to vary between about 302ppm to 450ppm. The concentration of NOx was also seen to increase in July. The annual concentrations varied between 0.0025 ppm in March to 0.0636 ppm in July. The concentrations of total hydrocarbons were seen to rise up to 4.8ppmC at certain times of the day while the average concentration was about 1.910ppmC in July (winter). The number concentration of particles vary from 684 cm-3 to 1984 cm-3 from summer (January) to winter(July). It was also observed that the concentration of small particles was high during night and low towards morning. It started increasing during the morning hours and evening when traffic starts to build up. Concentration of the gases was also seen to be quite high during the morning hours and in the evening hours thus showing a good correlation between particle concentration and gas concentration. These results indicates that particles and gases are mainly contributed by the traffic and biomass used for cooking and warming. Particle concentration of aerosols larger than 0.1 µm has its peak during winter months. Some interesting findings will be reported.

P-Chemistry Climate.25 ID:4420 15:35

New particle formation determines climate forcing of regional SO2 emissions

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Anthropogenic emissions of sulphur dioxide exert a cooling influence on the Earth's climate via enhanced scattering of short-wave radiation and by increased cloud droplet number concentrations (leading to brighter, longer-lived clouds). Recent studies (e.g. Merikanto et al, 2009) have demonstrated the large contribution of secondary particle formation from binary H2SO4-H2O nucleation in the free troposphere.

In a recent paper (Manktelow et al, 2009), we showed that the potential of a region's emissions to perturb climate via enhanced cloud droplet concentrations is related to the lofting of the emissions to regions of the atmosphere where new particle formation can take place. Following air quality legislation, SO2 emissions have reduced over North America and Europe in the last decades. In this study we use a global aerosol microphysics model to calculate the climate forcing that results from regional reductions in SO2 emissions and compare against equivalent simulations where new particle formation processes are neglected.

We demonstrate that in order to adequately characterize aerosol influences in climate through the global dimming and brightening period, climate models need to resolve microphysical processes like new particle formation, coagulation and condensation that control the global distribution of CCN number concentration. We also demonstrate the skill of the GLOMAP global aerosol microphysics model used in the study by evaluating against a large compilation of observed CCN concentrations from a wide range of locations.

P-Chemistry Climate.26 ID:4186 15:35

Aerosol-climate interactions over southern Africa: interannual variability and the role of ENSO

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Southern Africa is a region that experiences high interannual climatic variability. It is also a region that, in general is under-developed, has high population growth and is at times politically unstable. As a whole, the region is extremely vulnerable to climatic changes, with a large proportion of the population depending on rain-fed agriculture as a source of income and subsistence. It is well known that the El Niño/Southern Oscillation contributes significantly to climate variability over much of southern Africa. This in turn effects vegetation growth, and as a result the extent of biomass burning in the following dry season; with above-average wet seasons leading to increased burning, and drier than average seasons being followed by less extensive burning.

The savannas of Africa experience some of the most extensive burning in the world, and contribute a significant portion of the aerosol loading over southern Africa during the austral winter season, from June through October. At present, the climatic impact of these aerosols over southern Africa is poorly understood, particularly in terms of their interannual variability. In order to further investigate these impacts, the RegCM3 model is used in conjunction with emissions developed at the Laboratoire d'Aerologie.

Preliminary results indicate that the impacts of the direct aerosol effect on regional temperature, precipitation and circulation patterns vary between El Niño and La Niña years. Surface temperature decreases during the peak biomass burning season in all years, but more significantly in years with increased burning (generally following La Niña years). Climatic signals are also visible away from the major aerosol source regions, in the so-called 'river of smoke', the main exit passage of air from the region out to the Indian Ocean. This outflow pathway shifts northwards during La Niña periods, and southwards during El Niño periods, with the associated climatic impacts shifting correspondingly.

P-Chemistry Climate.27 ID:4245 15:35

A global modeling study on carbonaceous aerosol microphysical characteristics and radiative forcing

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Recently, attention has been drawn towards black carbon aerosols as a short-term climate warming mitigation candidate. However the global and regional impacts of the direct, cloud-indirect and semi-direct forcing effects are highly uncertain, due to the complex nature of aerosol evolution and the way that mixed, aged aerosols interact with clouds and radiation. A detailed aerosol microphysical scheme, MATRIX, embedded within the GISS climate model is used in this study to present a quantitative assessment of the impact of microphysical processes involving black carbon on aerosol cloud activation and radiative forcing. Our best estimate for net direct and indirect aerosol radiative forcing between 1750 and 2000 is -0.56 W/m2. However, the direct and indirect aerosol effects are quite sensitive to the black and organic carbon size distribution and consequential mixing state. The net radiative forcing can vary between -0.32 to -0.75 W/m2 depending on these carbonaceous particle properties at emission. Assuming that sulfates, nitrates and secondary organics form a coating around a black carbon core, rather than forming a uniformly mixed

particle, changes the overall net aerosol radiative forcing from negative to positive. Taking into account internally mixed black carbon particles let us simulate correct aerosol absorption. Black carbon absorption is amplified by sulfate and nitrate coatings, but even more strongly by organic coatings. Black carbon mitigation scenarios generally showed reduced radiative forcing when sources with a large proportion of black carbon, such as diesel, are reduced; however reducing sources with a larger organic carbon component as well, such as bio-fuels, does not necessarily lead to climate benefits.

P-Chemistry Climate.28 ID:4279 **15:35**

Modeling the aerosol optical depth in a global chemistry climate model

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Aerosol particles influence the Earth's radiative balance by the scattering and absorption of solar radiation, and they can contribute to climate change. Aerosol optical depth (AOD) is one of the most important diagnostics for climate forcing. Our new AEROPT module has been developed for use in the ECHAM MESSy Atmospheric Chemistry model (EMAC) to calculate the aerosol optical depth (AOD), and is coupled to the aerosol sub-model GMXe. The AEROPT module calculates the extinction cross section (sigma), single scattering albedo (omega), and asymmetry factor (gamma) from look-up tables, based on the aerosol composition and size distribution simulated by GMXe. The AOD calculation is based on the refractive indices of the different species such as black carbon, particulate organic matter, dust, sea salt, sulfate, ammonium, nitrate and the interactively computed aerosol associated water. We compare the model calculated annual and monthly mean AODs with the corresponding AODs observed by the MODIS satellite instrument and AERONET stations. The model captures both the magnitude and the distribution of AOD quite well, although there are some regions, especially over the tropical oceans, where the AOD is underestimated. We discuss possible causes, partly related to the aerosol hygroscopic growth, based on sensitivity analyses, and suggest further improvements.

P-Chemistry Climate.29 ID:4428 **15:35**

Improved aerosols and radiative budget in the Canadian GEM model

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Since 1995, the Canadian GEM model has only considered background aerosol represented as an annual mean quantity for its direct effect in modulating the radiative fluxes. Owing to significant biases in the model radiative fluxes found to be linked to this crude representation, a revised representation of tropospheric aerosols, that takes into account the emission sources of aerosols, the water vapor uptake on particles size, and the dependence of aerosol optical properties on particle size, composition and wavelength, are introduced in the model. Year-long simulations are performed with the old and the new aerosol and are compared to a reference simulation using AeroCom aerosol. All simulations are constrained by MODIS land surface albedo and are evaluated against observations from CERES, MODIS and MISR.

The new aerosol representation brought a number of improvements in the model, most notably improvements in aerosol optical depth and clear-sky direct effect. The satellite estimates of optical depth are generally larger than the model estimates by 16% over global ocean and 14% over global land in the annual average. However, with a correction for cloud contamination this discrepancy would be reduced. An adequate representation of the aerosol direct effect is found at the top of atmosphere, where the observational uncertainty is smaller. The model estimates over ocean increase from –4.36Wm–2 to –4.58Wm–2 and –5.76Wm–2 respectively for the old, the new and AeroCom representation and gradually approach the satellite estimates. Over land, currently satellite measurements alone cannot provide a measure of aerosol direct effect with good accuracy, and model simulations predict a range of direct effect estimates that is comparable to the range of the satellite estimates. The direct effect at the bottom of the atmosphere is poorly constrained due to lack of reliable measurements of aerosol absorption and single scattering albedo on global scale and difficulty in characterizing land surface reflection. CERES derives ocean surface cooling roughly 35% larger than the cooling at the top of atmosphere and land surface cooling 1.9 times the top of atmosphere cooling; by comparison, other satellite studies suggest stronger aerosol absorption. A comparable uncertainty characterizes the aerosol absorption in the model.

The evaluation of the clear-sky solar energy budget in the model shows an adequate simulation of the top of atmosphere budget. The surface clear-sky budgets, however, show excessive surface insolation in the model by an amount of more than 7 W m–2 in the global and annual average. The revised aerosol representation corrects up to 2 W m–2 of this bias, however significant uncertainties remain in the aerosol representation and more generally in the atmospheric and surface energy budgets that require further examination against more accurate ground bases measurements.

P-Chemistry Climate.30 ID:4322 15:35

Aerosol radiative forcing over an urban location: contribution of absorbing aerosols

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Atmospheric aerosols both from natural and anthropogenic sources are capable of altering Earth's climate by scattering and absorbing the incoming solar and outgoing terrestrial radiation. Aerosols are known to exert a net cooling effect on climate. But, unlike the other aerosol particles, black carbon (BC) aerosols which are produced due to incomplete combustion processes can absorb sunlight, heat the air and contribute to global warming. Therefore it is an important to estimate the amount of BC in the atmosphere and its contribution to the aerosol radiative forcing. Measurements of aerosol optical depth (AOD) at six different wavelengths (0.38, 0.44, 0.50, 0.65, 0.875, 1.02 µm) were conducted over an urban industrialized location Ahmedabad, located in the western India during 2008. Measurements of BC mass, as well as scattering and absorption coefficients were also conducted during the study period. AODs and BC mass concentrations are found to exhibit strong seasonal variability with the highest AOD during summer (0.43 at 0.5 µm) and lowest during post-monsoon (0.33 at 0.5 µm). BC mass is found to vary from 2 µgm-3 during summer to 11 µgm-3 during winter and post-monsoon seasons. Aerosol single scattering albedo calculated using scattering and absorption coefficients are found to be < 0.7 during winter while it increases (>0.8) during summer at 0.55 µm. The spectral variations of AOD are also analyzed to get some important information about the aerosol size distribution. This spectral variation of AOD has been used to separate the contributions of fine and coarse mode aerosols as well as contribution of soot in the measured AOD. Aerosol radiative forcings are estimated using radiative transfer model and the contribution of different aerosol species are obtained. Details of the methodology adopted and the results obtained will be presented and discussed.

P-Chemistry Climate.31 ID:4088 15:35

On the contribution of black carbon radiative forcing to the composite aerosol radiative forcing over an urban environment in India

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This paper discusses the extent of Black Carbon (BC) radiative forcing in total aerosol atmospheric radiative forcing over Pune, an urban site in India. Collocated measurements of aerosol optical properties, chemical composition and BC were carried out for a period of six months (during October 2004 to May 2005) over the site. Chemical composition data sets of water soluble, insoluble and BC components were used in Optical Properties of Aerosols and Clouds (OPAC) to derive aerosol optical properties for composite aerosols. The BC fraction alone was used in OPAC to derive optical properties solely for BC aerosols. The aerosol optical properties for composite aerosols and BC aerosols were separately used in SBDART model to derive aerosol radiative forcing due to composite aerosols and solely due to BC aerosols. The atmospheric radiative forcing for composite aerosols were found to be +35.5, +32.9 and +47.6 Wm-2 during post-monsoon, winter and pre-monsoon seasons respectively. The average BC mass fraction found to be 4.83, 6.33 and 4 µgm-3 during the above seasons contributing around 2.5 to 6% to the total aerosol load. The atmospheric radiative forcing estimated due to BC aerosols was +18.8, + 23.4 and +17.2 Wm-2, respectively during the above seasons. The study suggests that even though BC contributes only 2.2 to 6% to the total aerosols load; it is contributing an average of around 55% to the total lower atmospheric aerosol forcing due to strong radiative absorption, and thus enhancing greenhouse warming.

P-Chemistry Climate.32 ID:4560 15:35

The impact of natural and anthropogenic aerosol particles on radiation and cloud formation during a Saharan dust event over the Mediterranean and Western Europe

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In May 2008 a major dust outbreak occurred. Its origin was in the Sahara and from there mineral dust particles were transported over the western Mediterranean and finally covered large areas of Western Europe. During that episode high aerosol concentration were observed over Europe and a strong increase of ice nuclei was measured by Bingemer et al. (2009) at Kleiner Feldberg, Germany. In addition a strong positive deviation between the fore-casted and the observed temperatures was detected. Operational weather forecast models currently neglect the impact of the actual aerosol load on atmospheric processes. Therefore, we apply the regional scale online coupled model system COSMO-ART (Vogel et al., 2009) to quantify the contributions of the feedback processes between chemistry, aerosols, radiation, and clouds to the observed deviations. Primary and secondary aerosols from natural and anthropogenic sources are accounted for. Our simulations show that the deviation between the fore-casted and the observed temperature can not be explained by the radiative effect only. The very good correlation between the observed ice nuclei and our simulated dust concentration at Kleiner Feldberg indicates that the aerosol cloud feedback is responsible for the overestimation of the temperature. Results on that aspect will be presented at the conference.

P-Chemistry Climate.33 ID:4559 **15:35**

Ice Crystal Number Concentration Sensitivity Heterogeneous Freezing: A Global Modeling Study

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This study presents the sensitivity of ice crystal concentration, N_c, to the competition between homogeneous and heterogeneous freezing using predictions of ice nuclei (IN) concentration derived from theoretical considerations and field campaign data. For this, a physically-based, analytical, ice microphysics parameterization is implemented into the NASA Global Modeling Initiative (GMI) chemical and transport model. When competition between homogeneous and heterogeneous freezing is considered, global mean N_c vary by factor of 20 depending on the heterogeneous freezing spectrum used. IN effects on N_c are highly dependent on local dust and black carbon concentrations and most important under conditions of weak updraft and high temperature. Regardless of the heterogeneous spectrum used, dust is an important contributor of IN over large regions of the northern hemisphere. Black carbon aerosol have an appreciable effect on N_c for freezing fractions greater than 1%. It was found that only cloud formation scenarios where competition between homogeneous and heterogeneous freezing is the dominant feature would result in a global distribution of maximum supersaturation consistent with observations.

P-Chemistry Climate.34 ID:4130 **15:35**

A one dimensional cloud model with trimodal convective outflow

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Convection has a strong impact on the distribution of chemical species, especially in the tropics. In most global models, the effects of convection are parameterized rather than explicitly simulated. The results of a new one-dimensional convective cloud model, intended to be used as a convective parameterization, are summarized. The relationships between penetrative downdrafts, congestus outflow, and stability in the lower tropical troposphere are discussed, as well as some of the implications for the transport of trace species.

P-Chemistry Climate.35 ID:4372 **15:35**

Climatic impact of Saharan dust Aerosol on a west Africa/ tropical east Atlantic region

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We investigate the climatic impact of shortwave and longwave radiative forcing of Saharan dust on the West African monsoon and Sahel precipitation using a regional climate model interactively coupled to a dust model and running for the period 1996-2006. Two competing effects are found. First a reduction of monsoon intensity in the lower troposphere induced by the dust surface cooling causes a reduction of precipitation, and second an ‘elevated heat pump effect’ in the higher troposphere induced by the dust diabatic warming causes an increase of precipitation. The net impact of these effects is a reduction of precipitation over most of the Sahelian region (by about 8 % on average) except over a Northern Sahel - Southern Sahara band, where precipitation increases. This pattern is shown to be very sensitive to dust absorbing properties. Over the Atlantic ocean (dust outflow region), the heat pump effect is predominant resulting in a northward shift

of the rain belt in line with other studies. Beside the average climatic response, the intra-seasonal impact of dust as well as possible dust on simulated African easterly wave are also discussed. Finally, through a set of sensitivity studies, we discuss the dependency of these simulated responses to what is often pointed out as major limitations of regional climate models, namely the presence and the position of lateral boundary forcing as well as the lack of full ocean coupling.

P-Chemistry Climate.36 ID:3400 15:35

Sentinel-4 and -5: GMES Missions Related to Atmospheric Composition

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Global Monitoring for Environment and Security (GMES) is a European initiative for the implementation of a sustained operational capacity for Earth observation. It is the European contribution to GEOSS (Global Earth Observation System of Systems) and aims at providing accurate and timely information services for management of the environment, for a better understanding and mitigation of the effects of climate change and for ensuring civil security.

The space component of GMES is procured by the European Space Agency (ESA) and comprises a series of Earth observing satellite missions called 'Sentinels'. The combined objective of the geostationary mission Sentinel-4 and the low earth orbit mission Sentinel-5 is monitoring of the composition of the Earth's atmosphere.

In this presentation status and timeline of Sentinel-4 and -5 missions and programmatic links with other missions are outlined. The observational and mission requirements of Sentinel-4 and -5 are presented and an overview of related instrument concepts and estimated performances is provided.

P-Chemistry Climate.37 ID:4553 15:35

Aerosol radiative heating as a mechanism for plume rise: evidence from the 2009 Black Saturday Fires

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It is well known that biomass burning is an important global source for absorbing aerosols and that these aerosols affect regional and global radiation balance which in turn affect climate. The Black Saturday bushfires in Australia's Victoria province ignited on 7 Feb 2009, resulted in an aerosol plume detected by satellite instruments at stratospheric altitudes. Evidence from both ground-based and satellite instruments suggest that the pyrogenic plumes were not initially injected into the stratosphere. Results show that this dense smoke plume traveled several thousand kilometers horizontally and more than 10 kilometers vertically from the lower troposphere into the stratosphere over a period of 3 days. Our findings based on radiative transfer simulations suggest that radiative heating played a major role in plume rise as corroborated by observations of strongly absorbing aerosol properties within the plume. By using satellite measurements of aerosols (OMI, GOME2, CALIPSO) in combination with trajectories and ground-based information we describe the transport and height of this plume and its optical properties. This work highlights that absorbing aerosols not only play a role in regional and global radiation balance but can also modify the thermodynamic nature of the atmosphere.

P-Chemistry Climate.38 ID:4197 15:35

Bushfires and air quality under climate change

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Under a changing climate the frequency of bushfires, the duration of the bushfire season and the severity of bushfires are expected to change. Current projections for Southeastern Australia suggest there could be an increase in the frequency of very high and extreme fire days (Hennessey et al., 2007). Annually, biomass burning results in the emission of quantities of trace gases and aerosol to the atmosphere. Biomass burning aerosols influence the radiative balance of the earth-atmosphere system directly through the scattering and absorption of radiation, and indirectly through their influence on cloud microphysical processes, and therefore constitute an important forcing in climate models. They also reduce visibility, influence atmospheric photochemistry and can have a significant effect on human health. Australia experiences bushfires on an annual basis. In most years fires are restricted to the tropical savannah forests of northern Australia, however, there have been many bushfires in southern Australia's history. During December 2006 – February 2007, the State of Victoria in Australia was ravaged by 690 separate bushfires that devastated 1,116,408 Ha of native vegetation over 69 days. On several occasions, thick smoke haze was transported to Melbourne. During this period, detailed atmospheric chemical composition measurements were carried out at an air quality station in Melbourne. This presentation will discuss the likely effects of increased bushfire frequency and severity suggested in climate projections on urban air quality using the observations made at the air quality station as a basis for the discussion.

Hennessey et al. 2007: Australia and New Zealand. Climate Change 2007: Impacts, Adaptation and Vulnerability. Contribution of Working Group II to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, M.L. Parry, O.F. Canziani, J.P. Palutikof, P.J. van der Linden and C.E. Hanson, Eds., Cambridge University Press, Cambridge, UK, 507-540

P-Chemistry Climate.39 ID:4258 15:35

Impacts of intense Siberian forest fire aerosols on meteorology over East Asia in May 2003

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Intense forest fires occurred over Siberia in May 2003 and released high concentrations of biomass burning aerosols into the atmosphere, affecting regional air quality in East Asia. The high fire aerosols loads in East Asia may subsequently affect regional climate through their solar radiation extinction. In this study, we examine the effects of Siberian fire aerosols on regional meteorology using the National Centers for Environmental Prediction-Department of the Energy (NCEP-DOE) reanalysis II data and the NCAR

Community Atmosphere Model (CAM3). The surface cooling over Siberian and downwind regions where large amount of fire aerosols exist in the atmosphere is significantly shown in May 2003. Our analysis of the NCEP-DOE reanalysis data show 99% statistically significant increases in both surface air pressure over Siberia and precipitation over the NW Pacific, possibly associated with significant changes in surface air temperature due to the Siberian fire aerosols, in 2003 relative to the 30 years climatology. We also conducted NCAR CAM3 simulations forced by 3-D daily mean biomass burning aerosols mainly consisting of black and organic carbons and sulfate aerosols from a global chemistry-transport model, GEOS-Chem. We found that the model simulation with the Siberian forest fire aerosols reproduced the observed cooling over Siberia and downwind East Asia and large-scale perturbations in surface pressure and precipitation. The simulated results clearly indicate a significant impact of Siberian fire aerosols on regional meteorology over East Asia, having important implications not only for regional climate but also for synoptic scale weather patterns.

P-Chemistry Climate.40 ID:4379 15:35

Trends in concentrations of short lived components causing radiative forcing

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Historical trends in concentrations of short lived components causing radiative forcing are calculated with the Oslo CTM2 model. These trends will be used to calculate radiative forcing time series which will be used in a Bayesian statistical approach combining observations of temperature change and a simple climate model for constraining the climate sensitivity. Oslo CTM2 is an off-line atmospheric chemistry transport model. It is driven by meteorological data generated by Integrated Forecast System model at the European Centre for Medium-Range Weather Forecasts. The model is used with a tropospheric chemistry scheme, including the ozone-NOx-hydrocarbon cycle, as well as modules for sulphate, nitrate, black carbon, primary organic, and secondary organic aerosols. The resolution of the model used in this study is T42, approximately 2.8 x 2.8 degrees, and 60 vertical layers ranging from the surface and up to 0.1hPa. Time slice simulations are done for the year 1850 and then every 10th year from 1900 until present using meteorological data for the years 2005-2006. The emission data used are the historical emission inventory developed in support of the IPCC AR5. The total anthropogenic emissions have increased since 1850 until the end of the 20th century. The spatial distribution of the emissions has changed over these years. Emissions of relevant species have been reduced in North America and Europe at the end of the 20th century, while emissions in East Asia are increasing. Detailed modeling of the concentration changes of short lived components due to the changes in emissions is done. An improved trend for all short lived components causing radiative forcing is calculated, including tropospheric ozone and sulfate, nitrate, black carbon, primary organic aerosols and secondary organic aerosols.

P-Chemistry Climate.41 ID:4444 15:35

On the radiative impact of chemical constituents using the Environment Canada numerical weather prediction system

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The recent upward extension of the data assimilation system at the Meteorological Service of Canada from 10 hPa to 0.1 hPa has contributed significantly to the improvment of meteorological analyses and forecasts. This development effort has provided the opportunity of generating global analyses of chemical constituents which are used for improving the representation of radiative processes particularly in the upper troposphere lower stratosphere region. Radiative heating in the region depends on ozone absorption in both UV-visible and Infrared spectral regions and has a significant impact on temperature forecasts at the medium range timescale. This study shows the overall benefits of chemical assimilation in the context of NWP studies.

P-Chemistry Climate.42 ID:4285 15:35

Climate Response to 1950-2050 US Aerosol Trends

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United States aerosol concentrations peaked in the 1980s and have been decreasing since. Continued decreases are expected in the future in order to meet air quality standards. The associated change in aerosol radiative forcing could have significant climatic implications. Here we use a general circulation model (GISS GCM 3) with aerosol fields from a global chemical transport model (GEOS-Chem CTM) to study the climatic effects of US aerosol emissions over the 1950-2050 period. GEOS-Chem simulates coupled aerosol and gas-phase chemistry for 1950-2050 with decadal resolution, using historical and projected (IPCC A1B) anthropogenic emissions and either including or excluding US emissions. We conduct ensemble GCM simulations including both direct and indirect aerosol effects to determine the climate sensitivity to these US aerosol trends. Preliminary simulations accounting for only the aerosol direct effect indicate that US aerosol sources cooled the eastern US by 0.5°C in summer 1950-2000, and that the regional pattern of cooling largely follows that of radiative forcing. After 2000 the aerosol decline causes the US to warm 0.4°C. Additional simulations incorporating the aerosol indirect effects on cloud albedo and lifetime will be presented.

P-Chemistry Climate.43 ID:4137 15:35

Potential climate change impacts on air quality; new methodology and simulation

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Climate change affects the air quality in several ways. Air quality is dependent to meteorological parameters including humidity, temperature, wind speed and direction, radiation rate and mixing height which have direct effect on emissions, transport, dilution, chemical transformation, and final deposition of air pollutants in a region. Although the Kyoto Protocol is a great step toward reducing climate change impacts, more serious works is necessary to cut green house gas (GHG) emissions to effectively control global warming impacts. GHG emissions are projected to significantly grow in next decades. It is shown that many methods intended principally to reduce air pollution, decrease GHG emissions as well. On the other hand, many of GHG reduction plans result in reductions of air pollution, too. It is necessary to assess potential future

climate conditions and their potential influence on air pollutants like ozone and fine particles. Modeling results shows that climate change will increase atmospheric ozone amount in urban polluted areas up to 10 ppb by the next decades, specifically in pollution episodes. The effect of climate change on particulate matter (PM) is very complex. Modeling studies found that climate change affects PM concentrations in polluted regions up to 1 μg m−3 in the upcoming decades. Climate change could cause significant increase in PM pollutants. This paper reviews the current literature focusing on modelling tools and methodologies used to study regional climate impacts on air quality and the predicted view in future decades. It addresses the ability of current environmental models to simulate regional air pollution meteorology and its links to climate change and the changes in emissions. This work explains climate change and air quality links and reviews positive effects of implementation of GHG mitigation policies on human health and environmental systems due to improvement in air quality.

P-Chemistry Climate.44 ID:4538 15:35

Towards the attribution of ozone radiative forcing from satellites, assimilation, and modeling

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Tropospheric ozone is at the nexus of chemistry and climate through its role as a greenhouse gas, air pollutant, and regulation of the oxidative capacity of the atmosphere. The IPCC AR4 reports that radiative forcing of tropospheric ozone as the third most uncertain ranging from .25 to .65 W/m^2, the magnitude of which is dominated by preindustrial emissions and spatio-temporal distribution. However, the uncertainties have not substantially changed since the 2nd IPCC report. We show the potential of observations from the Tropospheric Emission Spectrometer (TES) to constrain global tropospheric ozone and its instantaneous radiative forcing through assimilation into the GEOS-Chem chemistry transport model. The assimilation leads to increases of up to 40% in upper tropospheric ozone resulting in imbalances of more than .3 W/m^2 for instantaneous radiative forcing. The vertical dependence of ozone radiative forcing to global precursor distributions and emissions is quantified through an adjoint sensitivity analysis. The implications of this sensitivity to chemistry- climate interactions is explored.

P-Chemistry Climate.45 ID:4119 15:35

Impacts of changing transport and precipitation on pollutant distributions in a future climate

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Air pollution (e.g., ozone and aerosols in surface air) varies with weather and thus is sensitive to climate change, as demonstrated by numerous studies. The sign of regional air quality responses to a warming climate, particularly for aerosols, remains uncertain. Multiple processes that determine ozone and aerosol distributions (emissions, chemistry, transport, deposition) are all likely to change with climate. We use a simple CO-like tracer (COt) with CO emissions and a fixed 25-day lifetime and a soluble version of that tracer (SAt) in the GFDL AM3 chemistry-climate model for 1990s and 2090s conditions to examine climate-

induced transport and precipitation changes and their implications for atmospheric pollutant distributions. In the future climate, convective mass flux out of the boundary layer weakens (-13%), reducing the free tropospheric COt and increasing surface COt. This finding supports prior emphasis on the need for tighter emission regulations to achieve a desired air quality improvement as climate warms. Meanwhile, convection deepens, increasing COt near the tropopause. Stronger stratospheric-tropospheric air mass exchange (+30%) increases the pollutants in the lower stratosphere. These changes in vertical transport largely control the pollutant redistribution (the large scale overturning circulation changes little with <5% in inter-hemispheric exchange, and between the tropics and extra-tropics) and may affect radiative forcing and potentially feed back on climate. The simulated SAt burden and lifetime increase by 12% despite increased total global precipitation (+5%). Changes of pollutants over the northern mid-latitudes and central Africa are sensitive to regional precipitation changes, which are not robust across models. Global precipitation change is a good indicator for soluble pollutant changes. We will focus on North America to further investigate what controls the response of soluble pollutants to warmer climate, including the role of precipitation event frequency.

P-Chemistry Climate.46 ID:4195 15:35

Relationship between the North Atlantic Oscillation and the inter annual variability of ozone and other trace gases: a data analysis and modeling study

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Studying trends and inter annual variability of tropospheric ozone and its main precursors is of great interest when assessing the impact of human activities on atmospheric composition and climate change. Recent studies have shown that the North Atlantic Oscillation (NAO) modulates the distribution of ozone and other trace gases due to changes of the transport pathways and strength over the North Atlantic ocean and to the Arctic. The aim of this study is to better estimate how the NAO modulates the ozone distribution as well as other trace gases, and by how much, over the northern extra tropics.

We use here observation data from the MOZAIC program. Since 1994, this program provides regular ozone vertical profiles over a few European and North American cities as well as a unique ozone climatology in the upper troposphere / lower stratosphere over the North Atlantic ocean. We define the tropopause as a 30 hPa thick layer centered on the surface PV=2 pvu. This tropopause-referenced climatology allows us to study both the upper troposphere and the lower stratosphere regardless of the seasonal variations of the tropopause height. We present here for the first time a detailed analysis of these data sets in relation to the NAO during the winter/spring season. We also investigate the relationship between the natural climate variation and the variability of the ozone-related trace gases over the North Atlantic region using chemistry climate model simulations (CAM-Chem and WACCM) over a fifty year period.

P-Chemistry Climate.47 ID:4504 15:35

Explosive volcanic eruptions as source of carbonaceous and sulfurous aerosol in the lowermost stratosphere

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The influence from explosive volcanic eruptions on the concentrations of sulfurous and carbonaceous aerosol in the lowermost stratosphere are presented. Following the eruptions of Kasatochi in August 2008 (Martinsson et al., 2009), Redoubt (March-April 2009) and Sarychev (June 2009), in situ measurements were undertaken from the CARIBIC platform (Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container) (Brenninkmeijer et al., 2007). Aerosol particles were collected by impaction technique (Nguyen et al., 2006) for subsequent analysis by accelerator-based techniques (Nguyen and Martinsson, 2007) for concentrations of a large number of elements including carbon and sulfur. CARIBIC measurements also include particle size distributions and a large number of trace gases. Measurements show a large influence on the aerosol concentration in the lowermost stratosphere, more than a factor of 3 increase persisted 3 – 4 months after the Kasatochi eruption. Similar and even stronger influence was observed from the Sarychev eruption during the fall of 2009. The composition of volcanic aerosol varies with time after the eruption. The ratio in concentration between carbon and sulfur was 2.6 one week after the Kasatochi eruption. Three to four months later that ratio was 1.2.

P-Chemistry Climate.48 ID:3401 15:35

Projection of the Ozone Quasi-Biennial Oscillation in the Tropical Stratosphere up to Year 2100 as Simulated with the Chemistry-Climate Model of Meteorological Research Institute

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Simulations on the past and future middle atmosphere were made with the chemistry-climate model (CCM) of Meteorological Research Institute (MRI), MRI-CCM. Three runs with slightly different initial conditions were performed for 140 years from 1960 to 2100. The dynamics module of MRI-CCM is a spectral global model of T42 truncation with 68 layers extending from the surface to 0.01 hPa (about 80 km), wherein the vertical spacing is 500m in the stratosphere between 100 hPa and 10 hPa. Hines gravity wave (GW) drag is incorporated with an enhanced GW source in the tropics to spontaneously reproduce the quasi-biennial oscillation (QBO) in zonal wind. The chemistry-transport module treats 36 long-lived species including 7 families, and 15 short-lived species with 80 gas phase reactions, 35 photochemical reactions and 9 heterogeneous reactions. The transport for chemical species is performed with a hybrid semi-Lagrangian scheme, which is formulated to be compatible with the continuity equation. MRI-CCM is integrated with observed forcings of SSTs, sea ice, volcanic aerosols, 11-year solar cycle, greenhouse gases, and halogens, the latter two of which are specified at the surface. It is found that the QBO amplitude in zonal wind in the tropical stratosphere is decreased in future under the global warming due to the greenhouse gas increase and that the ozone QBO is also weakened in the higher chemistry-dominant region above 28 km as well as in the lower transport-dominant region below that level.

P-Chemistry Climate.49 ID:4530 15:35

Influence of stratospheric ozone depletion on tropospheric ozone

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The chemistry-climate model CMAM (Canadian Middle Atmosphere Model) has been extensively used to study the decline and projected recovery, over the course of the 21st century, of stratospheric ozone. A description of tropospheric chemistry has been recently added to CMAM to produce a seamless simulation of atmospheric dynamics and chemistry from the ground to 95 km. An overview of the model and an assessment of the simulation of present-day tropospheric chemistry fields will be presented. Results from a pair of simulations covering 1950 to 2000, one with evolving concentrations of halogen-containing ozone depleting substances (ODSs) and one with constant ODSs, is presented to explore the effects of decreases in stratospheric ozone on ozone in the troposphere.

P-Chemistry Climate.50 ID:4158 15:35

How do future changes in stratospheric ozone and climate influence tropospheric ozone and its budget?

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We assess the impact of future stratospheric ozone recovery and climate change on tropospheric ozone and its budget, separating the effects of climate change and stratospheric ozone recovery. Stratospheric ozone changes between 2000 and 2100 are calculated with a stratospheric chemistry climate model (CCM), and are used to prescribe lower stratospheric ozone in a tropospheric CCM. The calculations show that large increases of stratosphere-troposphere exchange (STE), due to stratospheric ozone and circulation changes – an intensifying Brewer-Dobson circulation – lead to significant increases of ozone in the troposphere, especially at middle and high latitudes. Compared to the climate-change only scenario, stratospheric ozone recovery significantly contributes to increases in Southern Hemisphere surface ozone during austral winter; by 2100, the effect is about equal to that caused by climate change only. In the Northern Hemisphere, however, climate change dominates the changes in surface ozone.

P-Chemistry Climate.51 ID:4400 15:35

Stratospheric background aerosol from COS oxidation

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The modular atmospheric chemistry circulation model EMAC with the aerosol module gmXe has been used for multiyear simulations of stratospheric background aerosol. The model was extended by gas phase stratospheric sulfur chemistry and evaporation of sulfate particles. Lower boundary conditions are observed concentrations of COS and long-lived source gases at the surface and emissions of shorter lived gas and aerosol species. We show that photochemical oxidation of COS explains about 50 to 70% of the observed stratospheric background sulfate aerosol mixing ratios (Junge layer, e.g. SAGE data) and most of the observed profiles of SO2 in the stratosphere. The study will also include a superimposed volcanic eruption and some discussion of radiation effects.

P-Observations 1.1 ID:4227 15:35

The GEMS re-analysis of reactive gases (2003-2008)

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A global six-year data set of atmospheric composition has been produced by assimilating satellite observations into a coupled system which consists of ECMWF's integrated forecast system and the chemical transport model MOZART-3. Ozone retrievals from five instruments, CO total columns retrievals from MOPITT and MODIS aerosol optical depth data were assimilated. The system was built in the GEMS project and is further being developed in the MACC project. The model system is also run every day to produce forecasts and analyses of reactive gases and aerosols at the global scale.

We will focus on tropospheric ozone and CO fields in the GEMS-reanalysis. Ozone total column observations are dominated by stratospheric ozone (about 95 % of the total mass). It will be shown that tropospheric ozone values can be improved if data from multiple instruments are assimilated. The assimilated CO fields showed the biggest improvement in the free troposphere. The values in the boundary layer were less altered because of the weak observed signal and the strong influence of the emissions.

We will use ozone sondes, CO and ozone profiles measured by the MOZAIC programme as well as surface observations of the GAW network to validate the re-analysis. Further, we will present the inter-annual variability of Ozone and CO on the continental scale. We will relate the varying biomass burning emissions (GFEDv2) and meteorological conditions to the identified variability in atmospheric composition.

The re-analysis and forecast data are freely available and can be used for studies of atmospheric composition (http://www.gmes-atmosphere.eu/data/). The provision of boundary conditions for regional air quality models and the support of measurement campaigns (POLARCAT, HIPPO) have been important applications of the data set so far.

P-Observations 1.2 ID:4313 15:35

A re-examination of the use of PTR-MS to identify and quantify VOCs in global background air

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The World Meteorological Organisation Global Atmospheric Watch activity has specified that proton transfer reaction mass spectrometry (PTR-MS) can be used for measurements of some volatile organic compounds (VOCs) in global background air, specifically isoprene, terpenes, acetonitrile, methanol, ethanol, acetone and dimethyl sulfide. As well, the WMO recommends the measurement of benzene and toluene by other methods, and these two compounds can be measured by PTR-MS.

A critical examination of the underlying assumptions that PTR-MS can unequivocally identify and quantify these compounds in global background air is presented here. The factors examined include the range of reagent ions present in the PTR-MS and the distribution of charged product ions resulting from reaction of the reagent ions with the analytes (VOCs). It is the combination of these factors that are used to identify and quantify the VOCs present in air.

The distribution of ions observed in a PTR-MS show that H3O+ and its hydrated product H3O+.H2O, and NO+ and O2+ are all present as reagent ions, the latter generally in lesser amounts than H3O+. A generalised algorithm, which includes the influence of all four ions, representing the quantification of VOCs from PTR-MS measurements is presented.

The fragmentation patterns of 26 common atmospheric VOCs have been measured under a range of conditions of electric field intensity, pressure and temperature are summarised. These and other fragmentation studies are used to build a library of PTR-MS response.

Observations by PTR-MS of VOCs at Cape Grim Tasmania (41°S) and Wagerup, Western Australia (33°S) in clean air are analysed, using the information described above, to ascertain whether the PTR-MS can unequivocally identify and quantify isoprene, terpenes, acetonitrile, methanol, ethanol, acetone and dimethyl sulfide as well as benzene and toluene in global background air. Specific qualifications are presented.

P-Observations 1.3 ID:4564 **15:35**

VOC Observations from GOME-2, GOME, and OMI

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GOME-2 on MetOp-A, the first of a total of three sensors, like its predecessor GOME on ERS-2, and contemporaries SCIAMACHY on Envisat and OMI on EOS Aura, covers the spectral region of the UV and Visible and hence provides observations of the air-quality related volatile organic compounds (VOCs) formaldehyde (H2CO) and glyoxal (C2H2O2). Retrieval of both H2CO and C2H2O2 from space-based instrumentation poses a challenge due to the small optical depth of the absorbers and simultaneous interferences from comparably strong absorbers like O3 and NO2.

Starting with the launch of GOME in 1995, the GOME-2 mission continues to extend the long-term record of H2CO observations, while no equivalent record yet exists for C2H2O2. The necessity for the creation of a consistent data record for both H2CO and C2H2O2 has emerged, to facilitate the use of those products in trend studies and chemical transport models. At a minimum, "consistency" should include a common set of molecular absorption cross-sections to facilitate cross-calibration between platforms, and a common retrieval/fitting approach that minimizes differences between sensors while allowing for instrument ideosyncrasies.

We report on progress in applying our non-linear least squares fitting approach developed for operational H2CO and C2H2O2 retrievals from OMI to GOME-2 and GOME. Recent advancements in OMI retrievals show that a wavelength-dependent air mass factor can greatly improve minor trace gas retrievals where strongly varying interference from O3 over the fitting window is an issue (e.g., BrO and H2CO). Results from GOME and GOME-2 will be compared to the 5-year H2CO and 4-year C2H2O2 records currently available from OMI.

P-Observations 1.4 ID:4381 15:35

Using the CARIBIC observation data set as a constrain to the acetone global budget computed with the LMDz-INCA Chemistry-Transport Model

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In order to correctly represent the radicals and consequently the oxidative chemistry in the upper troposphere (UT), it is essential to simulate precisely the spatial distribution and the temporal variability of acetone. Observations made by the CARIBIC experiment (www.caribic-atmospheric.com) are exploited to evaluate fields of acetone concentrations simulated in the UT by the global chemistry-transport model LMDz-INCA. Acetone mixing ratios are measured onboard the CARIBIC Airbus 340-600 during approximately four long-distance flights per month since 2006, using a PTR-MS instrument. Around 600 h of data is currently available. Most flights considered here were to Asia, namely to the South China Sea in 2006 and 2007, and to South India in 2008. Simulated acetone concentrations are computed by the LMDz-INCA chemistry-transport global model, at a 3.75°x2.5° horizontal resolution and on 19 altitude levels. 30-minute temporal resolution results were either interpolated to the flight tracks either averaged for providing regional maps of daily means. Comparison between measurements and simulations demonstrate that our modelling is able to capture the temporal variability of the manifestation of the South Asian plume along the Chinese South East coast. Temporal variability is important: acetone concentration varies by a factor 2 to 3 from winter season to summer, day-to-day variability can reach 150 ppt, and diurnal cycle 100 ppt. A 20-50% systematic overestimation of the acetone load is generated by the model. Mean atmospheric burden of acetone is 5.1 Tg, with little inter-annual variability. Given a global source/sink strength of 94 Tg/yr, the mean residence time is 20 days. While sources and sinks are balanced over the globe, computation of the budget components by LMDz-INCA indicates that South Asia (5-45°N, 45-150°E) is a strong and systematic net source of acetone, in part due to a strong chemical activity, as 25% of global chemical production of acetone occurs over South Asia. However the fairly constant mean annual acetone content indicates that excess acetone produced over South Asia is transported to other regions.

P-Observations 1.5 ID:4365 15:35

Interpretation of organic compound measurements from the MIPAS-E

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Emissions of anthropogenic pollution result in the injection of a wide range of carbon compounds into the atmosphere. Carbon monoxide (CO), methane (CH_4) and volatile organic compounds (VOCs) are released in significant amounts, affecting both the oxidation capacity of the troposphere and ozone production. It has recently been established that the observation of the global distribution of VOCs in the upper troposphere (UT) can be made by measurements provided by instruments such as the Michelson Interferometer for Passive Atmospheric Sounding onboard ENVISAT (MIPAS-E) or the Atmospheric Chemistry Experiment (ACE) onboard SCISAT-1.

In this work, we show the ability of the MIPAS-E to provide new global measurements of organic compounds (including peroxyacetyl nitrate, acetone, formic acid, acetylene and hydrogen cyanide) in the

UT. The seasonal cycle of PAN is investigated for MIPAS-E full-resolution data in 2003. Data from the MIPAS-E optimised-resolution period are compared to measurements made during the recent POLARCAT campaign over Greenland and the Arctic in the summer of 2008 and INTEX-B over North America in April and May 2006, with the main focus on the quality of peroxyacetyl nitrate. Comparisons are also made with ACE satellite data in the March 2004 period, exploiting the overlap between MIPAS-E full-resolution data and beginning of the SCISAT mission. We finally compare these organic compound data products to output from the TOMCAT model and evaluate the consistency between the measurements and the model. We show, in this work, that the MIPAS-E is providing unprecedented global VOC data with good spatial resolution, providing very important new datasets with which to study the UT. This work on VOCs has the potential to feed into retrieval studies for possible future missions, such as PREMIER.

P-Observations 1.6 ID:4188 15:35

Fog and cloud induced aerosol modification observed by AERONET

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The modification of aerosol optical properties due to the interaction with fog or clouds is examined from measurements made by sun/sky radiometers at several AERONET sites. Almucantar retrievals, which utilize measured sky radiances and spectral aerosol optical depth, are examined for cases where fog and/or clouds have recently dissipated or advected away. Retrieved volume size distributions for cases identified as aerosol modified by cloud show very large median fine mode radius (~0.25-0.42 microns), which often show a bimodal sub-micron distribution. This bimodal accumulation mode distribution may be due to one mode (the larger one) from cloud-processed aerosol and the other from interstitial aerosol, or possibly from two different aerosol species (differing chemical composition) with differing hygroscopic growth factors. The size of the fine mode particles from AERONET retrieved for these cases exceeds the size of sub-micron sized particles retrieved for nearly all other aerosol types, suggesting significant modification of aerosols within the fog or cloud environment. The almucantar retrievals are analyzed from the Kanpur site in the Indo-Gangetic Plain in India (fog in January), Arica on the northern coast of Chile (stratocumulus), Fresno, CA in the San Joaquin Valley (fog in winter), and several other sites with aerosol observations made when clouds dissipate or gaps occur in cloud cover. In-situ measured aerosol size distributions made during fog events are compared to the AERONET retrievals.

P-Observations 1.7 ID:4540 15:35

Development of cloud property retrieval algorithm for nadir measurements.

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We have previously developed a retrieval algorithm to obtain cloud size and composition information from ACE-FTS solar occultation measurements. These measurements, however, cannot resolve the horizontal locations of the clouds, whereas nadir observation can do this. Consequently, we are modifying the ACE retrieval codes for application to nadir measurements. The basic algorithm involves the comparison of the measured spectrum with a library of monodisperse aerosol spectra and extracting the size distribution using a constrained least squares procedure. The development of the nadir retrieval algorithm thus requires that we calculate the angular scattering intensities as a function of solar zenith angle, wavelength, and aerosol properties; create a library of cloud reference spectra from these calculations and develop methods to reduce or account for the gas phase interferences and the noise from surface reflections. In this presentation, we will discuss our approach to the above concepts and report our progress and preliminary results obtained during these developments.

P-Observations 1.8 ID:4387 15:35

UV Aerosol Indices from the satellite-borne SCIAMACHY instrument: AAI, SCI, and cloudUVAI

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The Absorbing Aerosol Index (AAI) is known as a semi-quantitative indicator of UV-absorbing aerosols. AAI is essentially a measure of the contrast of a scene in the UV range, and as such is not very sensitive to surface type. Another advantage of AAI is that – in contrast to most other satellite aerosol retrievals – it can be determined in the presence of clouds (but for quantitative interpretation, the cloud effects need to be corrected). Recently, we introduced a counterpart to AAI: the SCattering Index (SCI). Whereas AAI is most sensitive to elevated layers of UV-absorbing aerosols, SCI can be used to detect aerosols that do not or barely absorb UV radiation ("scattering" aerosols). Low-altitude layers of absorbing aerosols, to which AAI is blind, can also be seen by SCI. Although AAI and SCI (together: UV Aerosol Indices, or UVAI) can be determined in the presence of clouds, the effect of clouds on UVAI can be substantial. High clouds may shield aerosols from view of the satellite and low clouds may cause an increase in apparent surface albedo, leading to an increase of UVAI. A third, often overlooked effect is the own contribution of clouds to UVAI. This effect can be as large as 2 SCI units, and depends mainly on cloud fraction and cloud optical thickness. We have modeled the contributions of clouds to UVAI – the cloudUVAI – and used them to correct measured UVAI from SCIAMACHY for the effects of clouds. We will present modeling studies of AAI, SCI and cloudUVAI, and corroborate our findings with measurements from SCIAMACHY. The good agreement between temporal and spatial patterns of measured UVAI on the one hand, and cloudUVAI that were modeled based on simultaneously measured cloud properties on the other hand, will be presented. In a final step, SCIAMACHY UVAI data will be shown that were corrected with (modeled) cloudUVAI so that only UVAI caused by aerosols remain.

P-Observations 1.9 ID:4611 15:35

Global Estimates of Ambient Fine Particulate Matter Concentrations from Satellite-Based Aerosol Optical Depth: Development and Application

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Long term exposure to $PM_{2.5}$ (fine aerosol with diameter < 2.5 µm) negatively affects human health, yet ground-based monitoring is sparse throughout much of the world. We develop a high-resolution (0.1º x 0.1º) global climatology of $PM_{2.5}$ for 2001-2006 by combining Aerosol Optical Depth (AOD) from two satellite instruments (MODIS and MISR) with aerosol properties from the GEOS-Chem chemical transport model. We find significant agreement with coincident North American $PM_{2.5}$ measurements (r=0.77, slope=1.07, n=1057) and non-coincident global measurements (r=0.83, slope=0.86, n=244). Satellite-derived $PM_{2.5}$ estimates over eastern North America and western Europe are between 10-20 µg/m^3. Parts of northern India and eastern China are found to have annual mean $PM_{2.5}$ concentrations of 35-60 µg/m^3 and 80-100 µg/m^3, respectively, with 35% of the Asian population exposure exceeding the WHO Interim Target 1 of 35 µg/m^3.

P-Observations 1.10 ID:4153 15:35

Physical and chemical properties of biogenic aerosols from Amazonia

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The Amazon basin with more than six million square kilometers contains the world's largest tropical rain forest, and investigations of aerosol characteristics in this large area are important for the understanding of the local and global influence of Amazonia on radiation budget and cloud formation. For the first time in Amazonia, a ground based long-term observations of aerosol properties were performed. The measurements were done at the Cuieiras forest reserve, 60 km NNW of Manaus in Central Amazonia. The site is relatively undisturbed, as the prevailing trade winds blow over vast expanses of intact tropical forest before reaching the measurement tower. Inlet lines run from the measurement level (45m, ~10 m above tree height) to a ground-based lab. A specially designed inlet allows collection of dry aerosol (20-40% RH) with a size cut of 3.5 um. Several measurements are being done: optical scattering (Nephelometer TSI 3665) and absorption (Thermo MAAP 5012), size distribution (SMPS TSI, and Lund DMPS), optical particle size distribution (OPC Lasair II), total particle concentration (CPC 3010 TSI), aerosol mass (TEOM), composition (SFU) among other properties. Trace gases measurements, including O3, CO2 and CH4, were performed as well. Aerosol concentration and properties shows strong seasonal variability. For the wet season, in terms of aerosol mass, PM2.5 is about 2.4 ug/m^3, while coarse mode particles (PM10-PM2.5) is 7.5 ug/m^3. Organic aerosol dominates with 70-80% of aerosol mass. In the wet season, 550 nm light scattering is very low, with an average of 4 Mm^{-1}. Black Carbon concentration varies from 50 to 150 ng/m^3 at 550 nm. Total aerosol particle counts varies between 100 to 300, with a peak mode in the size distribution at about 120 nm. Very few nucleation events were observed. The Aitken mode has been present in most of observed aerosol size distributions, suggesting that new particle formation may be occurring somewhere else above or below the canopy. Aerosol volume size distributions were derived from measured number size distributions assuming spherical particles. Coarse mode particles, possibly from biogenic origin, dominate the volume size spectra, either at dry or wet season. At the dry season, PM10 aerosol mass can reach 30-50 ug/m^3, with black carbon at about 400-2000 ng/m^3 due to biomass burning. All parameters increase by factor of 5 to 10. Single scattering albedo varies between 0.85 to 0.95, with lower values observed at the wet season. Coarse mode

particles, from biogenic origin, dominate the volume size spectra, either at dry or wet season. The volume ratio between coarse and fine mode increases greatly at night, suggesting a nocturnal release of biogenic particles as a result of the ecosystem natural metabolism. Occasionally we observed long range transport soil dust from the Sahara, that occurs mainly from February to May.

P-Observations 1.11 ID:4243 **15:35**

Satellite multispectral observations of carbon monoxide from a geostationary perspective

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The 2007 US National Research Council Decadal Survey calls for a geostationary satellite mission for atmospheric composition and air quality applications (GEO- CAPE, Geostationary Coastal and Air Pollution Events Mission). The requirement includes a multispectral (near- and thermal infrared) measurement of carbon monoxide (CO), a primary pollutant that plays an important role in tropospheric chemistry. CO is also an important precursor of tropospheric ozone and serves as an excellent tracer of transport processes. In this paper we analyze the temporal and spatial variability of CO with the aid of regional models and observations and discuss a role for observing system simulation experiments (OSSEs) in helping to define the CO measurement requirements for GEO-CAPE. Compared with the current capability of low-Earth orbit sensors, CO measurements from GEO-CAPE would have the advantages of increased spatiotemporal resolution and sampling, and enhanced measurement sensitivity to the lowermost troposphere with a multispectral retrieval. We illustrate this with OSSE case studies showing the improvement in surface CO forecasts.

P-Observations 1.12 ID:4254 **15:35**

The Monitoring Atmospheric Composition and Climate MACC Project

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The goal of the MACC (Monitoring Atmospheric Composition and Climate) project is to operate and improve data-analysis and modelling systems for a range of atmospheric constituents that are important for climate, air quality and surface solar radiation. Product lines include data records on atmospheric composition for recent years, data for monitoring present conditions and forecasts of the distribution of key constituents for a few days ahead.

MACC is funded by the European Union and provides the pre-operational atmospheric environmental service of the European Global Monitoring for Environment and Security (GMES). This service complements the weather analysis and forecasting services provided by European and national organisations by addressing the composition of the atmosphere.

The building blocks of MACC are a set of components with specific functions that are grouped into four clusters. The input data cluster acquires the satellite and in-situ observations and carries out preparatory processing of them. It also provides improved estimates of surface emissions of key species, with a particular emphasis on the highly variable emissions from fires. The primary global and regional clusters operate and

refine processing systems that include not only the data assimilation and modelling elements that provide the basic monitoring and forecasting products, but also the elements that provide estimates of climate forcing, inferred corrections to the modelled sources and sinks, and derived products such as UV radiation and resources for solar power generation. An outreach cluster provides the interface to downstream-service providers and other users, runs the service-chain test cases and supports the development of policy for the control of atmospheric pollution.

We will describe the different components of the project, the products currently delivered or planned within MACC, and how the scientific community can have access to the MACC products.

P-Observations 1.13 ID:4354 15:35

SCIAMACHY and GOME: Remote Sensing of Tropospheric O3 precursors, Halogen Oxides and Greenhouse Gases from space

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The SCIAMACHY and GOME series of measurements in the early morning began with the launch of GOME-1 in 1995, followed by the launch of SCIAMACHY in 2002 and GOME-2 in 2006. These instruments all make measurements of the back scattered radiation in the ultraviolet, visible and near infrared. These measurements yield a) the global observation of the ozone, O3, precursors nitrogen dioxide, NO2, and surrogate for the Volatile organic compounds Formaldehyde, HCHO, and Glyoxal, CHO.CHO, b) Bromine oxide, BrO, and iodine oxide, IO, which remove O3 and in the case of IO result in new particle formation, In addition SCIAMACHY makes measurements in the short wave infrared spectral region, which are providing unique measurements of carbon dioxide CO2 and methane CH4. This presentation will focus on the results of the longer term changes of the O3 precursors, the halogen oxides and the greenhouse gases.

P-Observations 1.14 ID:4361 15:35

Dependency of mean satellite NO2 column densities on wind patterns

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We calculated mean NO2 column density distributions separately for different wind directions. The resulting maps reveal clear and consistent patterns of NO2 transport on scales of some tens to hundred km. From the downwind decrease of NO2 and the mean wind speed, mean lifetimes can be estimated for different wind directions and different seasons. The resulting lifetimes are generally of the order of some hours up to a day (in winter). Systematic differences for different wind directions, which generally correlate with different mean meteorological conditions like humidity, can be analyzed.

P-Observations 1.15 ID:4419 15:35

Improving satellite retrievals of NO2 from biomass burning

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Nitrogen Oxides (NOx) are important precursors of tropospheric ozone, which is a pollutant and a greenhouse gas. Remote sensing observations from space are needed in the context of global monitoring and emissions estimates of these sources. However these observations are subject to significant uncertainties related to a priori information and radiative transfer calculations used in the retrievals. Here we examine NO2 retrieved from OMI observation of solar backscatter. In our study we jointly use the LIDORT radiative transfer model and a GEOS-Chem simulation with daily biomass burning emission inventory to assess the impact of fires emissions on the a priori NO2 shape profiles and the scattering weights used in the air mass factor (AMF) calculation, which transforms the spectrally fitted slant column into a vertical column. This modelling approach is supported by the analysis of aircraft in situ data from two major experiments: ARCTAS, which took place in summer 2008 over Canada and California, and DABEX (AMMA), which collected data over western Africa during the winter 2006 dry season. It has been previously proposed that under clear sky conditions and the presence of aerosols in the boundary layer the OMI cloud algorithm correction implicitly takes into account most of the aerosol correction. However, our results show the necessity of including an aerosol correction in the presence of clouds above the aerosol layer, which is a frequent situation over boreal forest fires. Over California and Canada, including the fire emissions in the model results in a mean AMF decrease of 35% from the shape factor modification and a mean AMF increase of 10% from the aerosol correction. Over African biomass burning, a persistent aerosol layer in the mid-troposphere above the high surface NO2 concentrations results in a significant impact of the aerosol correction on the AMF (-20% in average). For the case of African fires, our results suggest a linear relationship between the aerosol optical depth (AOD) and the aerosol correction factor calculated with the radiative transfer model, which makes possible a real-time aerosol correction based on satellite aerosol optical depth retrievals.

P-Observations 1.16 ID:4253 15:35

Ozone profile trends from ground-based and satellite data.

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In view of the upcoming WMO Ozone Assessment, and with four more years in the ozone record, evaluation of the current state of the ozone layer is of interest to the scientific community. The Ozone Assessment in 2006 suggested that the long-term ozone decline over the mid-latitudes had stopped and that ozone had stabilized since 1996. When extended to 2008, the ground-based and satellite data in the middle and upper stratosphere continue to show no significant changes to the ozone layer beyond its natural variability. Moreover, over the northern mid-latitudes, the recent increase in the observed total ozone column is not caused by the expected recovery of upper stratospheric ozone, but rather by changes in the lowermost stratosphere. We will present analyses of upper and low stratospheric ozone changes at northern mid-latitudes with respect to the stratospheric abundance of Ozone Depleted Substances (ODS). We will use the time series of well-established and calibrated ground based Dobson Umkehr instruments, several quality assured European ozone-sounding data, and the SBUV/2 Merged Ozone Data set. We will also discuss the impact of the recent years on the mid-latitude ozone recovery. Analysis will be done by using the Effective Equivalent Stratospheric Chlorine (EESC, A1_2010A WMO scenario) curve fit to the long-term ozone data. A second approach involves the use of the piece-wise linear trend (PWLT) model. Results from both approaches are compared for the slopes before and after the turning point in the EESC curve. Comparisons of the PWLT-determined ozone recovery rates and those predicted by the EESC curve help to identify changes unrelated to declining ODS concentrations.

P-Observations 1.17 ID:4114 15:35

Monitoring the changes in the tropospheric composition over the past two decades using Satellite observations and CTM

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Changes in the atmospheric composition and improvement of air quality are at the top of the agenda of policymakers around the globe. Scientific findings during the last decades have clearly highlighted the need for a comprehensive approach to understand the origin of trends in tropospheric chemical compounds. Observation of atmospheric composition is vital to monitor changes at regional and global scales. Platforms such as aircraft, ground-based in-situ and remote sensing stations, together with earth observation satellite instruments provide essential information on the composition of the atmosphere. Complex chemical transport models have been developed to interpret observations, test our theoretical understanding of atmospheric chemistry, understand changes and forecasting atmospheric composition. The evaluation of numerical models requires accurate information concerning the variability of atmospheric composition for targeted species through detailed comparisons with observations and measurements. We will discuss the changes in anthropogenic emissions of various trace gases (O3, NO2, CO etc) at global and regional scales over the past two decades. We will focus more particularly on rapidly developing regions in Southeast Asia, which showed a continuous increase emissions during the past years, while, in regions like Europe emissions of several compounds have been substantially reduced. In this paper, we provide an overview of changes in the distributions of several compounds as detected from satellite data (SCIAMACHY, IASI and MOPITT), and the analysis of these changes using the MOZART-4 chemical transport model. Targeted species will be CO, NO2 and O3, over selected areas in Europe, Asia and North America, for the 1996-2009 period.

P-Observations 1.18 ID:4215 15:35

Observing tropospheric change from space

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Human activities affect the composition of the troposphere in different ways. The concentrations of many trace species are strongly affected by anthropogenic emissions, mainly as result of fossil fuel combustion but

also from agriculture and biomass burning. In addition to pollutants which are directly emitted into the atmosphere such as NOx, SO2 or CO, other species including ozone and many VOCs are formed through secondary reactions. Changes in land use and agricultural practice as well as climate change can also affect emissions into the troposphere.

In order to understand the changes induced by anthropogenic activities, measurements are needed of key parameters in representative locations over long periods of time. These data sets can then be interpreted directly or used to validate atmospheric models and their predictions. In-situ measurements from the ground and from airborne platforms provide local data with high accuracy and temporal resolution but limited coverage. Satellite observations on the other hand provide the global view and are ideally suited for observation of global change.

In this contribution, long-term data sets derived from satellite observations performed by GOME, SCIAMACHY, and GOME-2 are used to investigate decadal changes in tropospheric composition. From these measurements, tropospheric column amounts of NO_2, SO_2, HCHO, and glyoxal could be retrieved, providing a detailed picture of changes in atmospheric composition. Overall decreasing NO_2 levels in many industrialised countries are observed while at the same time NO_2 levels in China and many large cities throughout the world continue to increase. Sulphur dioxide levels have been increasing in China for many years but this trend was reversed recently while in the US SO_2 amounts have been clearly reduced since the first GOME observations in 1995. Both HCHO and glyoxal have mainly biogenic precursors with some anthropogenic contributions. Nevertheless, there is indication for increasing columns in Asia.

P-Observations 1.18 ID:4589 15:35

Why can we see near-surface tropospheric patterns in OMI and other UV O3 retrievals? How we can we see these better?

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Analysis of tropospheric ozone soundings 2004-2006, primarily for North America, illustrates strong patterns of correlation in the lower troposphere for some sites, mostly eastern and southern, and low correlation for western sites like Trinidad Head, Richland, and Kelowna. Using the sondes, covariances between an upper level that is theoretically well sampled in the UV, e.g., 750 hPa and near-surface ozone yield linear relationships, O3_surface = a + b O3_aloft . Estimates based on "b" can be useful. Regions with recirculation and convection allow modestly accurate estimates of O3_surface to be made, as we show with examples. There are also varying of patterns of corrlation between different levels.

We seek to improve estimation of lower-tropospheric and surface ozone by using the relatively strong correlation of theta (potential temperature) and chi (ozone mixing ratio). Both are conserved for periods of time that regression estimates for chi may be obtained from UV and sonde records. We have found that typically methods with little physical interpretation, neural nets or projection pursuit regression, provide better estimates than geophysical estimates, e.g., based on theta-chi relationships. We will discuss our efforts so far to bridge toward more understandable geophysical statistical modeling.

P-Observations 1.20 ID:4528 **15:35**

Investigating Ozone Chemistry with Measurements of NO2 and HCHO from the Ozone Monitoring Instrument and GEOS-Chem

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Tropospheric NO2 and HCHO vertical column measurements from space-borne sensors can provide constraints on the sources and photochemical processing of NOx and VOC. Quantitative interpretation of the satellite column data requires error characterization and consistency evaluation against other datasets. We evaluate measurements of HCHO from the Ozone Monitoring Instrument (OMI) against an extensive suite of aircraft measurements and concurrent simulated columns from the GEOS-CHEM global chemical transport model. We then interpret OMI measurements of HCHO and NO2 with GEOS-Chem to estimate ozone production efficiency (OPE). We investigate monthly variability in HCHO, NO2, and OPE during April-October over the U.S. for 2005-2007. Both peroxy radical production rate, P(RO2), and NOx loss rate, L(NOx), are strongly correlated with population density. On average OPE > 4 for more than 60% of U.S. land area. Both P(RO2) and L(NOx) increase with population density, resulting in lowest OPE values in urban centers. Our results show that observations of HCHO abundance are an imperfect proxy for VOC reactivity, as local/regional differences in VOC mix induce variability in radical cycling relative to HCHO yield.

P-Observations 1.21 ID:4373 **15:35**

Trend detection in satellite observations of formaldehyde over 1996-2009 using GOME, SCIAMACHY and GOME-2 spectrometers

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Being a short-lived intermediate product in the oxidation of a large number of Non Methane Volatile Organic Compounds (NMVOCs), formaldehyde (H2CO) constitutes a useful indicator of anthropogenic, biogenic and biomass burning hydrocarbon emissions in the troposphere. Over recent years, satellite observations of H2CO have been increasingly used in combination with tropospheric chemistry transport models for constraining emission inventories of NMVOC, through top-down inversion approaches. With satellite measurements of H2CO being available since the launch of GOME/ERS-2 in 1995, the eventual trend in NMVOC emissions might be investigated. Trend analysis based on satellite data sets have been successfully applied for NOx (and more recently SO2) emissions. In this work, consolidated global distributions of formaldehyde columns derived from GOME, SCIAMACHY and GOME-2 instruments are used to analyze the temporal evolution of H2CO over industrialized areas in the period 1996-2009. A linear model with seasonal components has been used to fit the time series, taking into account the errors on the satellite observations as well as uncertainties related to possible biases between the different instruments. Results show that Asia, and more particularly China and India, are the only regions in the world where statistically significant positive trends in H2CO columns are found. The satellite-based (inferred) trends in H2CO columns are compared to reported trends in the REAS bottom-up inventory of NMVOCs emissions in Asia.

P-Observations 1.22 ID:4389 **15:35**

Tropospheric Formaldehyde observations derived from satellite measurements.

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The launch of GOME-1 onboard ERS-2 in 1995 was the beginning of a long term satellite observations, completed with SCIAMACHY in 2002 (onboard ENVISAT) and GOME-2 in 2006 (onboard METOP-A) This provides the opportunity to get information about sources, sinks and trends over specific regions for various trace gases, and especially for Formaldehyde (HCHO) presented in this paper:

The biogenic sources of HCHO are in many cases the strongest HCHO sources (e.g. over evergreen forest). The biomass burning source typically shows pronounced seasonal patterns or is even of sporadic nature. We investigated the time series of monthly mean values for selected regions of the world. Besides the satellite measurements of HCHO, the fire counts (from ATSR) and temperature are taken as proxy for biomass burning events and vegetation activity, respectively. In addition we plan to compare the satellite observations of HCHO also to other trace gases measured from satellite instruments (e.g CO and NO_2 as proxy for biomass burning) as well as the comparison with aerosols scattering index.

Anthropogenic HCHO emissions are shown with a case study linked to shipping emission. The line of enhanced HCHO in the Indian Ocean as seen in the 7-year composite of cloud free GOME-1 observations clearly coincides with the distinct ship track corridor from Sri Lanka to Indonesia. Comparisons with NO_2 values over the shipping route and EMAC model data indicate that direct emissions of HCHO or degradation of emitted NMHC cannot explain the observed enhanced HCHO values. Instead increased CH_4 degradation due to enhanced OH concentrations related to the ship emissions are the most probable reason.

P-Observations 1.23 ID:4111 **15:35**

Seven years' observation of mid-upper tropospheric CH4 from AIRS

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The global distribution of mid-upper tropospheric methane (CH4) has been derived using the space-borne observations by the Atmospheric Infrared Sounder (AIRS) on the EOS/Aqua platform and the IASI on the METOP-1at NOAA/NESDIS/STAR. The products of CH4 for 7 years from AIRS and more than one year from IASI in a 3 degree *3 degree grid are available upon request. Validation of these algorithms versus more than one thousand aircraft profiles demonstrates that the rms errors are less than 1.5%. Here we will present the validation of the retrieved CH4 from AIRS and IASI, comparison of CH4 retrievals from AIRS and IASI, and some finding about the spatiotemporal variation of CH4 based on 7 years' observation from AIRS and its comparison with model simulations, which include (1) some special feathers of the seasonal cycles of CH4 in the high northern hemisphere; (2) CH4 plume over the South Asia associated with the emission from rice paddies. Recent trend of CH4 as viewed from space and its difference from ground-based measurements will be discussed.

P-Observations 1.24 ID:4549 15:35

Consistent Global Long Term (1995-2010) Water Vapor Observations From UV/vis Satellite Sensors

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From UV/vis satellite instruments like GOME-1, SCIAMACHY and GOME-2 it is possible to retrieve the total column precipitable water over both oceans and continents with similar sensitivity. The water vapor analysis is performed with consistent algorithms (similar wavelength ranges and same sets of reference spectra) for all three sensors. Nevertheless, still differences between the three sensors exist including different spatial resolution, different overpass time and different spectral resolution. We investigate and quantify the potential effects of these differences both using simulation studies and experimental data during the overlap periods between the different sensors. As a result we derive a consistent data set ranging from 1995 to 2010, which will be part of the ESA project GlobVapour and will be the official EUMETSAT O3M-SAF product (GOME-2). We also present validation results using SSM/I observations over oceans and radio sonde observations over continents. The combined observations of the three satellite instruments constitute a unique data set, covering not only a strong ENSO event (1997/98) but also a time span of strong temperature increase due to climate change. We determine temporal trends (and their spatial distribution) of total column precipitable water over this period of time.

P-Observations 1.25 ID:4308 15:35

Global distribution and transport of CO2 in upper troposphere and lower stratosphere obtained by commercial aircraft observations

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We analyzed CO2 distributions in the upper troposphere and the lower stratosphere obtained by Comprehensive Observation Network for TRace gases by AirLIner (CONTRAIL). More than 2 million in-situ CO2 data from more than 3000 flights between Japan and Europe, North America, Australia, or Asia were collected during the period from November 2005 to November 2009. Observed mixing ratios were integrated in equivalent latitude and potential temperature grid for a reference year of 2008 to plot climatological distributions of CO2. In the mid-, high latitudinal regions between Japan and Europe or North America, seasonal cycles with summer maximum and spring minimum were observed in lower stratosphere. Tropospheric CO2 in these latitudinal bands showed strong seasonal cycles with springtime maximums and relatively sharp minimums in summer. Longitudinal differences of CO2 mixing ratios in the upper troposphere and lower stratosphere in the mid-, high latitude were relatively small but significant differences were found in July in the upper troposphere; lower CO2 over the southern Eurasian continent and higher mixing ratios over the Northern Pacific. This land-sea difference was gradually reduced after August, suggesting the transport of low CO2 originated from terrestrial sinks to oceanic regions. CO2 cross sections by using the flights between Japan and Australia or Southeast Asia show spread of high mixing rations from the Northern Hemisphere to the Southern Hemisphere from April to June in the upper troposphere. This transport brings a unique seasonal peak in autumn in the Southern Hemisphere.

P-Observations 1.26 ID:4462 15:35

Civil and commercial transportation partnerships for continuous atmospheric monitoring

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Concentrations of trace atmospheric contaminants can exhibit high spatial and temporal variation. In situ sensing is limited to a small portion of the 4D volume, so monitoring strategies must compromise between number of sensors and the sophistication of each device. For example, in the case of atmospheric mercury, modelers rely on a combination of high-cost directed aircraft investigations and established long-term monitoring stations. The former offer targeted measurements and diverse altitudes while the latter provide spatial and temporal continuity. Long-term observations from tropospheric and ocean platforms are a significant gap in sensing capability. Additional measurements of this type could yield benefits both for monitoring and for our understanding of long-range transport.

Commercial and Civil transport networks could address this gap. They could complement existing datasets with diverse locations and altitudes as well as continuous long-term monitoring capability. Partnership with commercial transport offers practical benefits such as access to power, security, and inexpensive transportation that could significantly reduce the cost of mobile installations. Possible platforms include cargo and civil ships, tropospheric short-haul cargo aircraft, and more exotic platforms such as helicopters and airships. We discuss specific technical and organizational challenges that impact both instrument and data system design for these systems. Issues include:

- Ruggedization
- Calibration stability and validation
- Autonomous operation and fault detection
- Data distribution and validation

We will describe some comparative advantages and challenges of different transport options for a mercury sensing application.

P-Observations 1.27 ID:4501 15:35

Equipment extension of the CARIBIC passenger aircraft observatory in 2010

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The present CARIBIC (in operation since 2005, www.caribic- atmospheric.com) system is based on a Lufthansa Airbus 340- 600 permanently equipped with a dedicated inlet system for gas and aerosol in situ analysis, DOAS telescopes for remote sensing and a video camera. The analytical equipment and apparatus for air and aerosol sampling is housed in a large, 1.5 ton, airfreight container. This automated laboratory for ozone, carbon monoxide, carbon dioxide, water vapor, cloud water, certain ovocs, condensation particles, DOAS, NO, NOy, aerosol and air sampling using glass containers, has been operating successfully on a monthly basis using four consecutive long distance flights, and has now been extended significantly. New equipment comprises a cavity ringdown system for measuring the D/H and 18O/16O ratio in water, a similar system for in situ analyses of CO2 and CH4, an Optical Particle Counter, a new high sensitivity DOAS system, new fast and accurate ozone and water analyzers, a convertor for NO2, and finally an 88 sample capacity whole air sampler. At the time of writing this abstract the final stages of certification were being absolved. We will elaborate on our motivation for this extension, and show some possibilities of research using first results.

P-Observations 1.28 ID:4383 15:35

In-service aircraft for a global observation system (IAGOS)

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Understanding the chemistry of our atmosphere and its reaction to human influences is vital in developing global solutions to tackle climate change and air quality. Passenger aircraft provide a unique platform for directly measuring atmospheric composition, particularly in the highly-sensitive tropopause region and for vertical profiles in the troposphere.

The European MOZAIC programme (mozaic.aero.obs-mip.fr), which was initiated in 1993 in cooperation between European Scientific Institutions, Avionic Industry and five European Airlines, used five AIRBUS A340 aircraft operated by Lufthansa, Air France, Austrian and Sabena (later operated by Air Namibia) to monitor atmospheric gases day by day. Starting with measurements of O3 and H2O in 1994, instruments for CO and NOy were added in 2001. MOZAIC has provided data from more than 100 million flight kilometres in the UT/LS and 50,000 vertical profiles in the troposphere. The European project CARIBIC (www.caribic-atmospheric.com) took a different approach by monthly deploying an instrumented cargo container aboard a LTU Boeing 767 and later a Lufthansa A340-600 since 1997. The large set of measurements (18 instruments) comprises those above as well as aerosol particles (number and composition), acetone, acetonitrile, mercury, hydrocarbons, halocarbons, and isotopic composition. A sophisticated inlet system for the above species allows also for remote sensing by differential absorption spectroscopy. MOZAIC and CARIBIC data are available to the scientific community world-wide, and have been used in more than 200 scientific publications.

IAGOS (In-service Aircraft for a Global Observing System, www.iagos.org) prepares the transition of routine aircraft observations from the two individual research projects into a sustainable European infrastructure with enhanced measurement capabilities and global coverage. This is achieved by increased instrument reliability, lighter and smaller instruments, as well as new instrumentation, e.g., for carbon dioxide or aerosol and cloud particles - key unknowns in climate modelling. And important aspect is the development of a suitable maintenance structure for world wide operation and the provision of near-real-time data to centres engaged in air quality forecasting, for example within GMES.

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P-Observations 1.29 ID:4385 15:35

Long term variability of O3 in the UTLS as measured by MOZAIC since 1994 and its link to NAO indices.

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The MOZAIC program measures O3 since August 1994 on board 5 commercial aircraft. Most of the data have been recorded at northern mid-latitudes, between 9 and 12 km altitude, in the upper troposphere – lower stratosphere (UTLS). To better assess the O3 distribution, measurements are scaled to the tropopause altitude. We present a climatology of O3 in the UTLS for the northern mid-latitudes, from Western US to Japan, via North Atlantic and Europe. We focus on the seasonal and regional differences to better highlight the O3 behavior in this critical region. Given the availability of 15 years of data (up to 2008), we aim to further assess the interannual variability and "trends". The first analysis presented in Thouret et al., (2006) showed an increase of O3 of about 1%/year between 1994 and 2003 in both the UT and the LS over a large North Atlantic area. This time period was characterized by the so-called (positive) anomaly 1998-1999. O3 time series and anomalies have been correlated with the atmospheric teleconnections indices (NAO and NAM), showing the coupling between the stratosphere and the troposphere and the role of the variations in large scale dynamics. Later on, Koumoutsaris et al., (2008) have also shown the role of the strong El-Nino event in 1997 in the positive ozone anomaly in 1998-1999 observed at hemispheric scale. In this present study, we aim to go a step further. We will show that recent data actually reveal a leveling off of O3 since 2000 over the US and Europe while it is still increasing over Asia. More over, to further understand the leading processes of such "trends" and to attribute them to various forcing, we will show a more detailed analysis of the links between O3 anomalies and the teleconnections indices.

P-Observations 1.30 ID:4413 15:35

Evaluation of ACE-FTS and OSIRIS satellite retrievals of O3 and HNO3 in the upper troposphere: Application to ozone production efficiency

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Ozone in the upper troposphere has a large radiative forcing of climate. We present an evaluation of tropical upper tropospheric ozone retrieved from two satellite instruments (ACE-FTS, OSIRIS) with in situ ozone

measurements from aircraft campaigns and ozonesondes. Both satellite retrievals generally reproduce broad features in the upper troposphere such as the wave-one pattern. However, these comparisons reveal high biases in both the ACE-FTS (17-23%) and OSIRIS (12-25%) ozone relative to aircraft and ozonesonde observations. We demonstrate that the ozone production efficiency in the upper troposphere can be determined using ACE-FTS O3 and HNO3 measurements. The resulting value of 120±11 mol/mol is in broad agreement with model simulations indicating the capability of satellite observations in the tropical troposphere to provide insight into ozone production.

P-Observations 1.31 ID:4576 15:35

The observing requirements for the prediction of ozone

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Using a photochemical box model and its adjoint, constructed using the Kinetic Pre-Processor, we investigate the impacts of changing satellite observational capacity, observation frequency and quality upon the ability to both understand and predict the nature of peak ozone events within a variety of polluted environments. The model consists of a chemical mechanism based on the Master Chemical Mechanism utilising 171 chemical species and 524 chemical reactions interacting with emissions, dry deposition and mixing schemes. The model was run under a variety of conditions designed to simulate a range of summertime polluted environments spanning a range of NOx and volatile organic compound regimes (VOCs). Using the forward model we were able to generate simulated atmospheric conditions representative of a particular polluted environment, which could in turn be used to generate a set of satellite pseudo observations (with noise) of key photochemical constituents. Next the prediction model was run under a perturbed emission scenario with specified errors in the initial guess of the emissions scaling factors. The prediction model was then forced back to the truth using variational data assimilation and the pseudo observations. Using this described method we assess the optimal time of observation and the diversity of observed chemical species required to provide acceptable forecast estimates of ozone concentrations. Different observing strategies become favourable as the photochemical regime changes according to NOx and VOC concentrations. For instance, under VOC limited photochemical regimes in the presence of model VOC emission uncertainties, observations of CO and NO2 from space borne platforms are insufficient to constrain emission uncertainties and allow successful ozone prediction. In such cases, the residual errors for ozone can be up to 20 ppbv in scenarios that are sensitive to VOC emissions.

P-Observations 1.32 ID:4233 15:35

Source contributions to carbon monoxide, black carbon and ozone distributions in the Arctic

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The Arctic troposphere is heavily polluted in Winter and Spring as a result of long-range transport from northern mid-latitude continents and the lack of photochemical activity needed to cleanse the atmosphere. Massive forest fires in boreal Eurasia and North America impact the Arctic in the Spring and Summer. This talk will integrate fine resolution aircraft campaign measurements with global-scale satellite observations and global chemistry model simulations to examine the sources and impacts of pollution in the Arctic. Aircraft measurements from the NASA ARCTAS, NOAA ARCPAC, POLARCAT and NSF/NCAR

START-08 campaigns during Spring and Summer 2008, along with the NSF TOPSE campaign in Spring 2000, have been analyzed in conjunction with satellite observations and model results. Global observations of carbon monoxide (CO) from the 10-year record of the Terra/MOPITT instrument provide a large-scale and multi-year context for the Arctic aircraft measurements, particularly highlighting the importance of Siberian fire activity during Spring and Summer of 2008. The global chemical transport model MOZART-4 is used to identify the relative importance of anthropogenic pollution and biomass burning emissions from various regions to the Arctic troposphere. Volatile organic compounds (VOCs), a key component of ozone production, are used in correlation with CO to evaluate emission inventories and the model chemistry. The emissions of CO and black carbon are traced to the Arctic, contrasting the contributions of long- and short-lived pollutants. In addition, the ozone produced from emissions in different source regions is quantified to identify the main contributors to the observed Springtime increase in tropospheric ozone.

P-Observations 1.33 ID:4429 15:35

Monitoring of atmospheric composition with the IASI/METOP sounder: CO distributions and trends

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Carbone monoxide (CO) is an important trace gas for understanding of both air quality and atmospheric composition. It is a good tracer of pollution plumes and atmospheric dynamics. In this poster we describe the IASI CO concentration products obtained by the Fast Operational/Optimal Retrievals on Layers for IASI (FORLI) algorithm, developed at the ULB (Belgium), and generated in near real time for further distribution at LATMOS.

The global distribution CO data are validated against similar total column measurements obtained from MOPITT/TERRA, AIRS/AQUA and TES/AURA, also sounding in the thermal infrared spectral range. Specific pollution events will be shown, and vertical information will be discussed. For MOPITT (v3,v4) and IASI, long term trends over specific areas are analyzed in detail and show an excellent agreement. Finally, this poster will also illustrate how the data processed at ULB-LATMOS are currently assimilated in the MACC project to generate the CO pollution forecasts over Europe.

P-Observations 1.34 ID:4448 15:35

Estimated error parameters derived from assimilation residuals in observation space

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The estimation of error statistics is an essential for data assimilation. Traditionally it has been derived from residuals in observation space using innovations or observation-minus-forecasts (OmF) while fitting an horizontal and vertical correlation model – an approach known as the Hollinsgworth-Lonnberg method. Recently Desrozier's et al. have suggested an alternative approach that uses in addition the analysis-minus-forecast (AmF) and observation-minus-analysis (OmA) assimilation residuals in a iterative procedure to determine the error covariance parameters, and without having to fit a correlation model. This new approach has received considerable attention in particular for it ease of implementation, and because it claims being able to determine more error covariance parameters such as cross-channel error covariance in satellite observations.

The general properties of the Desroziers' scheme are first reviewed in light of the assumptions made about the error covariance properties. Then using a dense observation network where observations are provided at each grid point, a spectral analysis of the iterative scheme ca be derived. The specific properties of the scheme are then derived when only one variance is estimated, when both observation and background error variances are estimated, and when a correlation length parameter is also estimated in addition to the error variances. These properties are examined in light of the assumptions made on the correlation length scales.

P-Observations 1.35 ID:4438 15:35

Using long-lived chemical tracers to quantify the stratospheric contribution to tropospheric trace gas distributions

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The interpretation of long-lived trace gas distributions in the troposphere can be quite difficult due to the multiple transport pathways and time scales from source and sink regions. We present a method to use measurements of molecules with photolytic processing in the stratosphere to quantitatively estimate the stratospheric 'overworld' contribution to tropospheric and lowermost stratospheric tracer gradients. With this information the 'age' tracers, such as CO2 and SF6, can reveal transport time scales and surface latitudinal origins of tropospheric and lowermost stratospheric air. We demonstrate these techniques with observations from the Stratosphere-Troposphere Analysis of Regional Transport 2008 (START-08) and HIAPER Pole to Pole Observations (HIPPO) aircraft campaigns, which are uniquely suited to this type of study. Implications of our results for modeling of important greenhouse gases in the troposphere will be discussed.

P-Observations 1.36 ID:4386 15:35

THE EFFECT OF OZONE HOLE ON THE CONCENTRATION AND TRENDS OF STRATOSPHERIC OZONE OVER USHUAIA GAW STATION

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The stratospheric ozone is quite important for the tropospheric systems and crucial for the biological system since this gas absorbs ultraviolet radiation in the UV-A region and part of UV-B. The stratospheric ozone, furthermore, is a source of heat which filters all radiation in the UV-C range affecting the life on Earth. The analysis of the variability and structure of stratospheric ozone is therefore here focused. The Ushuaia Global Atmospheric Watch (GAW) Station is located in Southern Patagonia (54°50' S; 68°18' W ; 18 m.a.s.l) below the middles latitudes. The main advantage lies in the fact that it is possible to monitor events within the stratosphere when the ozone hole occurs. The GAW Station has been carrying out Total Ozone Column (TOC) measurements since 1994 up to the present with a Dobson spectrophotometer. The aim of this paper is to analyse the concentration, variability and trend of the ozone that is influenced by the passage of the ozone hole in the middles latitudes. Every year, It has been observed that at the Ushuaia GAW Station there is a depletion in the concentrations of stratospheric ozone from August to November. The ozone trend and concentrations show a great variability all over these years. This paper will allow a better awareness of the stratospheric process that modulates and affect the ozone trends. Besides establishing a monitoring of the

frequency of the ozone hole that reaches Ushuaia GAW Station and the effect that it produces in the total ozone concentration and trends.

P-Observations 1.37 ID:4542 **15:35**

Sources and seasonal variations of atmospheric hydrogen and carbon monoxide: Tall tower measurements and Lagrangian modeling

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Carbon monoxide (CO) and hydrogen (H2) play important roles in atmospheric chemistry: CO is the dominant global sink for the OH radical and H2 is the most abundant non-methane reactive gas in the atmosphere. Here we present measurement and modeling results from an ongoing field campaign aimed at improving our understanding of these two trace gases. Continuous H2 and CO measurements have been ongoing at the University of Minnesota Trace Gas Observatory (TGO, 200m AGL) since January 2009. TGO is located 24km south of the Twin Cities metropolitan area and at the intersection of the major U.S. ecosystems, providing the opportunity to constrain regional anthropogenic and biogenic sources of CO and H2. We apply here two atmospheric models (STILT and GEOS-Chem) to interpret the measured concentrations, daily-to-seasonal fluctuations, and covariance in terms of present understanding of CO and H2 sources and sinks.

P-Observations 1.38 ID:4378 **15:35**

Monitoring vegetation using DOAS satellite observations

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Vegetation-cycles are of general interest for many applications. Be it for harvest-predictions, global monitoring of climate-change or as input to atmospheric models. From novel spectrally resolving UV/vis satellite instruments (like GOME or SCIAMACHY) the spectral signatures of different types of vegetation can be identified and analysed. Although the spatial resolution of GOME and SCIAMACHY observations is much coarser than those of conventional satellite instruments for vegetation monitoring, our data sets on different vegetation types add new and useful information, not obtainable from other sources.

Common vegetation indices are based on the fact that the difference between Red and Near Infrared reflection is higher than in any other material on Earth's surface. This gives a very high degree of confidence for vegetation-detection. The spectrally resolving data from GOME and SCIAMACHY provide the chance to concentrate on finer spectral features throughout the red and near infrared spectrum. We look at these features using a technique known as Differential Optical Absorption Spectroscopy (DOAS). Although originally developed to retrieve information on trace gases, it can also be used to gain information on vegetation. Another advantage is that this method automatically corrects for atmospheric effects. This renders the vegetation-information easily comparable over long time-spans.

In addition, high-frequency-structures from vegetation also effect the retrieval of tropospheric trace-gases and aerosols.

To optimize vegetation monitoring with DOAS we produce spectrally resolved reference spectra from different vegetation types using our own instrumentation. We analyze the effect of different Pigments on high-frequency-structures of the DOAS Retrieval. Applying these results we investigate how well we can distinguish vegetation types from space.

Plenary 2.1 ID:4362 INVITED 19:00

From Robert Boyle to IGAC: a history of the study of atmospheric composition and chemistry

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Modern studies of atmospheric composition and chemistry commenced when air was no longer seen as an element but as a gaseous mixture being made up of at least two kinds of gases (John Mayow 1645–1679) or "various materials" Robert Boyle (1627–1691). In the 18th century the investigation of atmospheric composition was among the forefront of physical science. Eminent scientists identified nitrogen, oxygen, carbon dioxide, hydrogen and other atmospheric gases. In the 19th century themes emerged including: • Carbon dioxide, oxygen and the carbon cycle • The composition of rainfall and the cycles of nitrogen, sulphur, etc. • Ozone and other oxidants in surface air • Ozone in the upper atmosphere • Air hygene (air pollution) • Airborne particulate matter (aerosol) Study of the organic chemistry of the atmosphere commenced mid 20th century. The drivers of this research have varied widely including: the desire for fundamental knowledge of Cavendish in the late 18th century and Schönbein and Ramsay in the 19th century, the need for understanding of the nutrition of plants and the production of food by de Saussure, Liebig and others in the 19th century, the coupling of urban air pollution and sanitary concerns by Smith in the 19th century and Haagen-Smit in the 20th century, the desire to probe the then inaccessible upper atmosphere by Dobson and Chapman and concerns about continental scale and global pollution by Callendar, Oden, Johnston, Crutzen, Rowland and Molina in the 20th century. Over the same period, the study of atmospheric composition and chemistry passed from mainstream physical research in the 18th century to an adjunct of meteorology in the 19th and early 20th century, to a recognised field of chemistry in the late 20th century, to, today in the 21st century, a core component of Earth systems science, the multidisciplinary study of the Earth in all its facets.

Observations 2.1 ID:4505 **08:30**

10 Years of Pollution Data from the MOPITT Instrument

James Drummond [1], *Merritt Deeter* [2], *David Edwards* [2], *John Gille* [2], *Helen Worden* [2], *Florian Nichitiu* [3], *Jason Zou* [3]
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On 18th December 1999 the Terra platform was launched from the Vandenberg Air Force base carrying the Measurements Of Pollution In The Troposphere (MOPITT) instrument. As of March 3, 2010 MOPITT has completed ten years of operation measuring carbon monoxide (CO) over the planet and the measurements continue. These measurements have demonstrated the changes of CO in both space and time and shown a planet with very large variations in concentrations depending upon events and circumstances. Two of the major conclusions are that CO varies over time and space in such a manner that it is almost impossible to describe an “average year” and that pollution is always a global issue rarely a purely regional one.

During the mission there have been a number of challenges, both scientific and engineering, that have been successfully overcome to bring the instrument to the present time and even after 10 years in orbit significant improvements to the datasets are still being made.

MOPITT was provided to the Terra spacecraft by the Canadian Space Agency and was built by COMDEV of Cambridge, Ontario. Data processing is performed by the MOPITT team at the National Center for Atmospheric Research, Boulder, CO. Instrument control is by the team at the University of Toronto.

Observations 2.2 ID:4510 **08:45**

Next Generation Remote Sensing of Ozone: An Assessment of Tropospheric Sensitivity

Annmarie Eldering [1], *Kelly Chance* [2], *Robert Chatfield* [3], *David Edwards* [4], *Joanna Joiner* [5], *Susan Kulawik* [1], *Thomas Kurosu* [2], *Xiong Liu* [2], *Vijay Natraj* [1], *Ken Pickering* [6], *Robert Spurr* [7], *Helen Worden* [4], *Kevin Bowman* [1]
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We are systematically characterizing the sensitivity that is possible and needed for the next leap forward in remote sensing of ozone, with an emphasis of reaching lower into the troposphere. Space based measurements of ozone have been made for years in the ultraviolet and visible region, and separately, in the infrared. Although we have a long record of measurements, we are still limited in measurements relevant to boundary layer air pollution.

Making measurements from a geostationary satellite, such as GEO-CAPE, would be a huge benefit, allowing measurements on hourly timescales, which would let us capture the rapid diurnal changes of air pollution species. In additional to more frequent measurements, we need ozone measurements that are more sensitive to the lower layers of the atmosphere (lowest 2 kilometers). Previous studies have demonstrated that the

combination of UV- Vis and IR wavelengths can peer into the boundary layer and discriminate ozone from the ozone above. However, these studies have been limited to a few cases and for a few measurement specifications. Our work focuses on systematically quantifying the characteristics of ozone measurements that include wavelengths from the ultraviolet through the infrared, considering a wide range of potential spectral bands. In this work we find that to achieve the needed sensitivity, a combination of bands is required. The exact combinations that meet the sensitivity requirements is dependent on noise, spectral resolution, and the characteristics of the ozone field.

Observations 2.3 ID:4487 09:00

Atmospheric composition from geostationary orbit - Sentinel 4 UVN on Meteosat Third Generation

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The Sentinel 4 UVN instrument is an UV-VIS-NIR imaging spectrometer and part of the geostationary Meteosat Third Generation (MTG) satellite to be launched in 2017/18. UVN is designed to make measurements of the up welling radiation at the top of the atmosphere covering the wavelength range from 305 nm to 500 nm and 750 nm to 775 nm at moderate to high spectral resolution (UV-VIS: 0.5 nm, NIR: 0.12 nm) with a spatial resolution of 8 km x 8 km over Europe. It is designed to yield the total and tropospheric columns of several atmospheric constituents of great significance for European air pollution and air quality applications: the gases Ozone O3, Nitrogen Dioxide, NO2, Sulphur Dioxide, SO2, and Formaldehyde HCHO. In addition, there is the potential to deliver Glyoxal (CHOCHO). Furthermore, aerosol information will be obtained. The presentation will summarise on the expected product quality.

Observations 2.4 ID:4467 09:15

Sensitivity of continental boundary layer chemistry to a new isoprene oxidation mechanism

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We evaluate a new isoprene chemistry mechanism in the GEOS-Chem chemical transport model by simulating observations from the NASA INTEX-A aircraft campaign over the eastern United States and North Atlantic during summer 2004. This new mechanism reflects new constraints on isoprene δ-hydroxy channels under high NO_x ($\equiv NO + NO_2$) conditions, and the formation of dihydroxyepoxides and OH regeneration under low NO_x conditions. Model results are evaluated with observed ozone, HO_x ($\equiv OH + HO_2$), reactive nitrogen species, isoprene, and photochemical tracers. Sensitivity analyses are conducted to investigate the fate of isoprene-produced nitrates, peroxides and carboxylic acids in this new mechanism. We use the observed relationships between isoprene and OH concentrations in the continental boundary layer to evaluate the OH recycling efficacy. We also use the observed relationships between isoprene and formaldehyde (HCHO) columns to evaluate the time-dependent HCHO yield and its implications for interpreting HCHO columns observed from space as a proxy for isoprene emissions. We will discuss the implications of these updates of isoprene chemistry for simulation of ozone, OH, NO_x and HCHO in global tropospheric chemistry models.

Interfaces 1.1 ID:4244 **INVITED 09:30**

Tropospheric aerosol chemistry: challenges, recent progress and unknowns

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The tropospheric chemistry community has identified a number of aerosol heterogeneous and multiphase processes that are likely to be of regional or global importance. The atmospheric effects may arise through loss of a gas-phase reagent, formation of a gas-phase product, or aerosol chemical modification. The challenge to the experimental community is to quantify the rates of these processes using a fundamental approach within the chemical complexity that characterizes the wide array of tropospheric aerosol types. This talk will address recent progress that has been made in tropospheric aerosol chemistry by focusing on examples of specific reaction systems, including: i) halide oxidation, of importance to marine and polar regions, ii) organic oxidation in an aerosol particle, and the impacts that may result on the bulk aerosol properties, and iii) HOx and NOx heterogeneous reactions that modulate global oxidant levels.

Interfaces 1.2 ID:4160 **09:50**

Role of dust alkalinity in atmospheric chemical processing of Asian dust for the North Pacific Ocean fertilization

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Iron (Fe) is an essential nutrient for phytoplankton. Although iron-containing soil dust mobilized from arid regions supplies majority of the iron from the atmosphere to the oceans, the key flux is the amount of soluble or bioavailable iron in terms of the biogeochemical response to atmospheric deposition. Atmospheric chemical processing of mineral aerosols by acid gases may play a key role in transformation of insoluble iron into soluble forms. However, mineral dust particles have a potential of neutralizing the acidic species due to the alkaline buffer ability of carbonate minerals. Here we demonstrate the impact of dust alkalinity on the acid mobilization of iron in a three-dimensional aerosol chemistry transport model, which is incorporated with a mineral dissolution scheme. In our model simulations, most alkaline dusts cannot be entirely consumed by inorganic acids during transport across the North Pacific Ocean. As a result, the inclusion of alkaline dust in aqueous chemistry substantially limits the iron dissolution in aerosol solution during the long-range transport. Over the North Pacific Ocean, insignificant fraction of iron dissolves from hematite in the coarse-mode dust aerosols, which contain internally mixed carbonate minerals. However, if iron-containing minerals are externally mixed with carbonate minerals, significant fraction of iron dissolves from these particles, due to acid mobilization. It may imply that alkaline dust content in minerals is linked to the inverse relationship between aerosol iron solubility and particle size.

Interfaces 1.3 ID:4162 **10:05**

Arctic Aerosols, Springtime Forest Fires, and Climate

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Layers of dense smoke mixed with fossil fuel combustion products were frequently encountered at altitudes from near the surface to 7 km in the Alaskan Arctic in early spring 2008 during NOAA's Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) project. Transport models demonstrate that the smoke originated from agricultural and forest fires in Russia, and was transported over distances >5000 km. These smokes appear to be a frequent occurrence in the Arctic in April-June due to agricultural burning and wildfires near the Kazahkstan border and the Chita/Amur River regions, respectively. Aerosol particles in the Arctic in springtime have been postulated to affect climate through direct radiative forcing and strong feedbacks between deposited absorbing particles and snow physics. However, in 2008 there was little substantive evidence for precipitation scavenging and removal of the smoke particles between the time they were emitted and the time they were observed above the Arctic surface. Given the lack of scavenging, the dense smoke layers aloft may not have contributed substantially to changes in snow albedo. Furthermore, direct radiative forcing from the smoke aerosol is calculated to be small and to cool the surface. Independent of the smoke layers, aerosol concentrations very near the sea-ice surface were reduced, suggesting a deposition process from within this shallow layer to the sea-ice. The relevance of these findings to the "Arctic haze" phenomenon and to climate forcing in the Arctic will be discussed.

Interfaces 1.4 ID:4351 **10:20**

Atmospheric chemistry in MABL of tropical Bay of Bengal: Impact of continental outflow

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The chemical composition of size-segregated ($PM_{2.5}$ and PM_{10}) aerosols, collected from the MABL of Bay of Bengal, has been studied to assess the impact of continental outflow on atmospheric chemistry over the tropical Bay. During the late NE-monsoon (Jan-March), outflow from Indo-Gangetic Plain (IGP) and south-east Asia dominates the widespread dispersal of pollutants (from vehicular emissions and large scale burning of biofuels, agricultural waste) over the Bay region. A noteworthy feature of the data on aerosol chemical composition is reflected in the dominance of nss- SO_4^{2-} (range: 1.3 to 28 $\mu g\ m^{-3}$) in $PM_{2.5}$. The anthropogenic fraction of nss-SO_4^{2-} is 90-100 % and accounts for as much as 60 % of the water-soluble inorganic constituents. The impact of anthropogenic sources is further evident from the widespread depletion of chloride in PM_{10} (range: 40 to 100 %) compared to Cl^-/Na^+ ratio in sea- salts. The carbonaceous species (EC and OC) contribute nearly 25 % to $PM_{2.5}$; and significant linear relationship with K^+ suggests biomass burning as their dominant source (biofuels and agricultural waste). The enhancement in the fractional solubility of aerosol Fe, as assessed in $PM_{2.5}$, re-emphasizes the impact of combustion sources (biomass and fossil-fuel) and chemical processing (of dust) during the long-range transport. The mass-ratio of Ca^{2+}/Ca approaching close to unity provides unequivocal evidence for the chemical uptake of acidic species (SO_4^{2-} and NO_3^-) by $CaCO_3$. The enrichment factors of heavy metals (Pb and Cd) exceeding about 200-400 further demonstrate the influence of pollution sources on the chemistry of MABL. The regional scale transport and chemical transformation processes within the MABL can, thus, have profound impact on the atmospheric chemistry and exchange across the air-sea interface.

Interfaces 2.1 ID:4341 **11:00**

How biospheric cycling affects climate metrics for air quality pollutants.

William Collins [1], *Stephen Sitch* [2], *Olivier Boucher* [1]

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Through their impact on the biosphere and hence the carbon cycle, air quality pollutants can indirectly affect climate. These impacts can include damage of surface ozone to plants, changes to the diffuse/direct fractions of photosynthetically active radiation by aerosols and fertilisation of vegetation through reactive nitrogen deposition. We have included the first two processes interactively in a fully coupled earth system model of climate, chemistry and ecosystems. To quantify the climate forcing we use the changes in global surface temperature due to pulse emissions. These can be formulated as global temperature potential (GTP) metrics. The climate forcing (GTP) from emissions of ozone precursor gases is substantially increased when we take into account the damage the ozone causes to the terrestrial carbon cycle. This is particularly important for NOx. NOx emissions have previously been assumed to cool climate. We now show that for short timescales (around 20 years), by reducing the CO2 uptake of plants, NOx emissions are likely to warm climate. For VOC emissions, including the ozone-biospheric impact doubles their climate effect. Our conclusions are that accounting for biosphere interactions is crucial when assessing the climate impacts of air quality pollutants.

Interfaces 2.2 ID:4427 11:15

Size-resolved CCN measurements in the Eastern Mediterranean: distributions, closure and activation kinetics.

Aikaterini Bougiatioti [1], *Christos Fountoukis* [2], *Nikolaos Kalivitis* [1], *Spyros Pandis* [3], *Athanasios Nenes* [4], *Nikolaos Mihalopoulos* [1]
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During the Finokalia Aerosol Measurements Experiment (FAME07) a study of size-resolved CCN has been conducted from July to August 2007, concurrently to size distribution and chemical composition measurements. Activity and hygroscopicity studies were carried out, with the goal of determinating the nature and mixing state of the aerosols sampled. With supersaturations varying between 0.2 and 0.73%, it has been established that all particles activate at around 0.6% supersaturation. At the lower supersaturation levels, the maximum activated fraction is on average below one, which indicates the presence of a small fraction (~20% on average) of externally mixed CCN-inactive particles with lower hygroscopicity. From the "characteristic" scaled volume fraction (SVF*) data we can see consistently that during the day the fraction of unactivated particles increases from midday until 16.00, a trend which is more evident for the larger particles of the Aitken mode. Concurrently to the decrease of the activated fraction, there is a slight decrease in the scaled volume fraction an hour around local noon. On average the CCN closure is excellent (relative error -1.3±20%), even with grossly-simplifying assumption regarding particle chemical composition over diverse airmasses (with CCN concentrations ranging from less than 200 cm-3 to more than 7,500 cm-3). When the average organic water-soluble fraction is considered in the CCN calculations, the closure error remains very low (+2.8±20%) but the underprediction bias and the scatter is reduced (best fit slope 0.99, R2=0.91). The underprediction bias at the low supersaturations is also substantially reduced and improved at the higher supersaturation.

Interfaces 2.3 ID:4157 11:30

Ultrafine particle growth: Better predictions of atmospheric aerosol number with revised organic condensation schemes

Jeffrey Pierce [1], *Ilona Riipinen* [2], *Taina Yli-Juuti* [2], *Silja Häkkinen* [2], *Mikael Ehn* [2], *Heikki Junninen* [2], *Katrianne Lehtipalo* [2], *Tuukka Petäjä* [2], *Jay Slowick* [3], *Rachel Chang* [3], *Douglas Worsnop* [4], *Spyros Pandis* [5], *Neil Donahue* [5], *Markku Kulmala* [2]

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Atmospheric lifetimes of ultrafine particles are determined by their condensational growth as compared with coagulational scavenging. Sulphuric acid is a key compound in atmospheric nucleation, but its ambient concentrations seem not to be enough to explain observations of particle growth. Organic compounds, on the other hand, represent a large fraction of secondary aerosol mass, and are thus likely to also play an important role in growing freshly formed particles. However, current state-of-the-art organic aerosol partitioning theories do not and cannot describe the observed condensation onto nano-particles. We use observations from two field sites (Hyytiälä, Finland, and Egbert, ON, Canada) to study the growth rates of ultrafine particles. These results are analysed using detailed box modelling to determine the importance of organic condensation onto ultrafine aerosol, and to obtain a semi-empirical approach for describing the ultrafine particle growth in a reasonable manner. The observed particle growth rates can be reproduced with the model if ~50% of the total organic aerosol mass is allowed to dynamically condense on the particle surface area, as opposed to assuming equilibrium partitioning to the pre-existing aerosol mass. A global chemical transport model GEOS-CHEM with online aerosol microphysics is applied to study the sensitivity of global aerosol and CCN concentrations to the organic condensation onto ultrafine particles. The GEOS-CHEM simulations indicate that the representation of organic condensation onto ultrafine particles has a significant impact on predictions of regional and global aerosol and CCN numbers.

Interfaces 2.4 ID:4460 11:45

Improving the representation of tropospheric aerosols over South America in an atmospheric chemistry model by assimilation of satellite and ground-based remote sensing data

Judith Hoelzemann [1], *Karla Longo* [1], *Rafael Fonseca* [2], *Hendrik Elbern* [3], *Saulo Freitas* [1]

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We present results of a case study over South America during the fire season in October 2007, using the aerosol and trace gas data assimilation system of CCATT-BRAMS, the Coupled Chemistry-Aerosol-Tracer Transport model coupled to the Brazilian developments on the Regional Atmospheric Modeling System (BRAMS). Aerosol Optical Depth (AOD) columns from the Moderate Resolution Imaging Spectroradiometer (MODIS), and from ground based AERosol Robotic NETwork (AERONET) sites were assimilated into the CCATT-BRAMS model by applying the three dimensional variational assimilation technique (3D-VAR). Simulation improvements by adding observational information are discussed. In particular, we will address the improvement achieved by introducing inhomogeneous and anisotropic areas of influence to enhance the impact and representativeness of valuable though sparsely distributed AERONET observations. As regionally high aerosol concentrations in South America during this time of the

year are almost exclusively associated with fires, our results may also serve as indication of where fire emission strengths need to be improved in the underlying emission model.

Sources 1.1 ID:4603 INVITED 13:30

Connecting local and global emission inventories: New challenges to improved accuracy and consistency

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Development of emissions inventories must take account of how these data will be exploited. Trying to satisfy both local and global requirements has become very challenging. This presentation will discuss traditional approaches to the construction of emissions inventory methodologies, focusing on the gaps between criteria pollutants and greenhouse gases, urban and global scales, as well as cities/countries with different levels of development. Based on this, I will describe some new approaches to the construction of inventories, using the experience for emissions produced by mobile sources in various cities across the world, where some of these gaps are being considered. Examples that represent some of the main challenges that need to be tackled in the search for greater accuracy and consistency are the use of appropriate sampling techniques from sources, generating dynamic patterns of activity and energy use, producing more sophisticated emission factors, making the modeling processes more flexible for different locations and implementing quality assurance procedures.

Sources 1.2 ID:4473 14:00

Impact of an improved Cuban emissions inventory on air quality simulations

Madeleine Sánchez Gácita [1], *Marcelo Felix Alonso* [1], *Karla Maria Longo* [2], *Saulo Ribeiro De Freitas* [1]
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The energy sector in the Central America and Caribbean regions is primarily fossil fuel based and one of the major sources of air pollution in the region. In Cuba, energy production is responsible for 99% of SO_2 emissions, 98% of NO_X and 94% of CO, with emissions in 2000 of 502.6 Gg, 102.5 Gg and 670.4 Gg, respectively, according to the Cuban National Inventory - CNI. Electric power generation plants, the most important sub-sector, are highlighted as point sources of high emissions. Global inventories are shown to be inaccurate for Cuba. RETRO has non-zero data for just one cell, over the city of Havana. EDGAR has deficiencies in its geographical distribution, with no emissions over the city of Havana, and it distributes emissions nearly equally among all sectors, which is unrealistic according to the CNI. More importantly, emissions are overestimated, with the notable exception of SO_2 and NMVOC. The most important reasons are the particularities of Cuba, including the extensive employ of fossil fuels with little refining and high sulfur content in energy production and industrial processes such as asphalt production, and the use of low efficiency technologies.

This work presents an improved emissions inventory with CNI data and detailed emissions for all major power generation plants. The impact of this improvement was assessed through numerical air quality simulations of the transport and transformation of these emissions from a regional perspective, conducted

with the CCATT-BRAMS 3D atmospheric chemical transport model, developed and maintained by INPE, Brazil. Boundary conditions were supplied by global model MOCAGE with chemistry scheme RELACS. In the coupling with CCATT-BRAMS using chemical mechanism RACM, an unrealistic oscillation near the boundary was observed, which was solved through the incorporation of RELACS in CCATT-BRAMS as part of this work. The behavior of the model was evaluated through sensitivity tests.

Sources 1.3 ID:4408 **14:15**

Constraining NH3 emissions using remote sensing and surface observations

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Uncertainties in NH3 emissions poses challenges to air quality modeling, development of pollution control strategies, and understanding the global transport pathways of reactive nitrogen. We present the first results of inverse modeling of NH3 emissions using new remote sensing observations from TES. Capabilities and limitations for constraining NH3 sources and variability with this data are assessed via inverse modeling tests using pseudo observations (generated by the model). The adjoint-based inverse model is then applied over North America using real observations from the year 2008. The resulting constraints on NH3 inventories are evaluated through cross validation with independent data sets and qualified with calculation of uncertainty reductions. Lastly, the impacts of constraining NH3 emissions on control strategies are determined through novel application of the adjoint model as a sensitivity tool, pinpointing nonattainment to influences from specific emissions locations, sectors, and sources. Benefits of accomplishing these goals are to further our overall knowledge of the environmental impacts of NH3 emissions by affording better estimates of incidents of excessively harmful PM2.5 levels using air quality models as well as reducing uncertainty in quantifying the sources and fate of ecologically disruptive levels of reactive nitrogen. NH3 inventory improvements also enhance calculated emission control efficiencies by more precisely accounting for the response of PM2.5 concentrations to existing or proposed mitigation strategies, thereby targeting emissions controls that minimize risk and cost while maximizing societal benefits.

Sources 1.4 ID:4175 **14:30**

Space-based constraints on global sulfur dioxide emissions

Chulkyu Lee [1], *Randall Martin* [1], *Aaron Van Donkelaar* [1], *Konstantine Vinnikov* [2], *Russell Dickerson* [2], *Nickolay Krotkov* [3], *Andreas Richter* [4]
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Sulfur dioxide (SO2) emissions from anthropogenic and natural sources are oxidized quickly in the atmosphere, leading to aerosol formation and acid deposition. We use satellite observations from SCIAMACHY and OMI to produce top-down constraints on SO2 emissions. We evaluated sulfur dioxide (SO2) columns from two satellite instruments (SCIAMACHY and OMI) with surface SO2 measurements. The surface-level SO2 mixing ratios are inferred by applying local scaling factors from a global chemical

transport model (GEOS-Chem) to SO2 columns retrieved from the satellite instruments. The resulting annual mean surface SO2 mixing ratios for 2006 exhibit a significant spatial correlation (r = 0.86, slope = 0.91 for SCIAMACHY and r = 0.80, slope = 0.79 for OMI) with coincident in situ measurements throughout the United States and Canada from monitoring networks. We evaluated the GEOS-Chem simulation of the SO2 lifetime with that from in situ measurements to verify the applicability of GEOS-Chem for inversion of SO2 columns to emissions. The SO2 lifetime calculated with the GEOS-Chem model over the eastern United States is 13 hours in summer, and 48 hours in winter within 5 hours of that inferred from the in situ measurements. We use SO2 columns from SCIAMACHY and OMI to derive a top-down anthropogenic SO2 emission inventory over land by using the local GEOS-Chem relationship between SO2 columns and emissions. There is little seasonal variation in the top-down emissions (<15%) over most major industrial regions providing confidence in the method. Our global estimate for annual land surface anthropogenic SO2 emissions (52.1 Tg S yr-1 from SCIAMACHY and 47.0 Tg S yr-1 from OMI) closely agrees with the bottom-up emissions (54.6 Tg S yr-1) in the GEOS-Chem model, and exhibits consistency in global distributions with the bottom-up emissions (r = 0.78 for SCIAMACHY and 0.77 for OMI). However there are significant regional differences, including a factor of two underestimate (5 Tg S yr-1) in bottom-up SO2 emissions from Nigeria.

Sources 1.5 ID:4453 14:45

Inverse modelling of CO_2 sources and sinks using Tropospheric Emission Spectrometer (TES) CO_2 observations

Ray Nassar [1], *Dylan Jones* [1], *Susan Kulawik* [2], *Jing Chen* [1], *Parvadha Suntharalingam* [3], *Robert Andres* [4]
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The strength of carbon sources and sinks can be estimated from a set of atmospheric measurements and a model simulation of CO_2 using a Bayesian inversion approach. This approach has traditionally been used with ground-based measurements; however, the capability to retrieve CO_2 from observations by the Tropospheric Emission Spectrometer (TES) on the Aura satellite, has recently been developed. Although the sensitivity of TES CO_2 observations peaks in the mid-troposphere (511 hPa) and TES CO_2 accuracy and precision are lower than those of ground-based in situ measurements, the coverage provided by TES provides an important benefit for flux inversions. Inverse modeling estimates of carbon sources/sinks obtained using TES CO_2 and the GEOS-Chem global 3-D CO_2 simulation with multiple updates (including a chemical source of CO_2 from reduced carbon oxidation), will be compared with inversion results based on the in situ surface network.

Sources 2.1 ID:4447 15:30

Lightning NOx production in midlatitude thunderstorms as observed by OMI

Kenneth Pickering [1], *Eric Bucsela* [2], *James Gleason* [1], *Laura Pan* [3], *Owen Cooper* [4]
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Lightning is responsible for an estimated 15 percent of total global NOx emissions, but is one of the most prominent sources in the upper troposphere over the United States in summer. In this study, we present evidence of lightning-generated NO2 (LNO2) using data from the Ozone Monitoring Instrument (OMI) on the Aura satellite for several case studies over the Central US, Gulf of Mexico, and Western Atlantic. These cases of LNO2 enhancement have been identified through examination of the OMI NO2 products and of FLEXPART lightning-generated NOx (LNOx) model output. Mean OMI NO2 columns from a set of days with little or no lightning in and downwind of the regions of interest are subtracted from the daily data to obtain the presumed LNO2 signal. Model estimates of LNO2 profiles from previous studies in and downwind of convective storms are employed in air mass factor calculations to convert the OMI NO2 slant columns into vertical column amounts. We employ improved off-line processing techniques developed in studies of tropical convective systems to minimize known artifacts in the OMI data. The LNO2 amounts from OMI are converted to LNOx amounts using NASA Global Modeling Initiative (GMI) calculations of [NOx]/[NO2] ratios. Back trajectories at middle and upper tropospheric levels are used to obtain numbers of cloud-to-ground (CG) lightning flashes observed by the National Lightning Detection Network (NLDN). These flash counts are combined with estimates of the climatological IC/CG ratios to obtain the total number of lightning flashes associated with the observed LNOx enhancements. A vertically-weighted average number of upwind flashes was obtained using a typical profile of LNOx mass from a series of midlatitude cloud-resolved storm chemistry simulations. Combining the LNOx results and upwind flash counts, we obtain the production of LNOx per lightning flash in these mid-latitude storms. We compare our results to previous studies of other mid-latitude storms and to studies of tropical and sub-tropical systems. Results yield values in the range obtained for midlatitude storms through other estimation techniques (e.g., aircraft measurements, cloud-resolved models).

Sources 2.2 ID:4550 15:45

Quantification of fossil fuel CO_2 emissions from East Asia using atmospheric observations of $\Delta^{14}CO_2$

Jocelyn Turnbull [1], *Yong Chung* [2], *Jay Gregg* [3], *Scott Lehman* [4], *John Miller* [1], *Pieter Tans* [1]
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Fossil fuel CO_2 emissions are the largest net source of CO_2 to the atmosphere, and accurate quantification of these emissions is essential to furthering our understanding of the global carbon cycle. Currently, fossil fuel CO_2 (CO_2ff) emissions are estimated from bottom-up inventories of fossil fuel use reported by governments and industry. Measurements of the radioactive isotope ^{14}C in CO_2 provide an independent, objective method of constraining CO_2ff emissions, since fossil fuel derived CO_2, unlike other CO_2 sources, is devoid of ^{14}C. These measurements can be used to evaluate bottom-up inventories of CO_2ff, and, using a multi-species approach, also constrain emissions of other trace gases. Emissions from East Asia are of particular interest, since they contribute ~25% of global total CO_2ff emissions, and are growing rapidly by 8-10% per year, but are quite uncertain compared to other regions. We quantitatively determine recently added CO_2ff in samples from the NOAA/ESRL cooperative sampling network site at Tae-Ahn Peninsula, Republic of Korea, which usually sees air which has recently passed over northeastern China and Korea. Samples typically contain a few ppm of CO_2ff from China, and Korea contributes additional CO_2ff in some samples. The observed CO_2 also shows large variability due to biospheric CO_2 exchange, even in winter, so that CO_2 measurements alone cannot accurately estimate CO_2ff. We compare the observational results with estimates of CO_2ff from a Lagrangian particle dispersion model and a prior estimate of CO_2ff emissions, examining our ability to evaluate the reported emissions and determine quantitative uncertainties to these emissions. Measurements

of carbon monoxide (CO) in the same samples provide quantitative top-down estimates of the CO:CO_2ff emission ratio in each sample. Different source regions are identified by their characteristic CO:CO_2ff emission ratios, and these ratios are used to evaluate bottom-up inventories of CO emissions for each source region.

Sources 2.3 ID:4367 16:00

Better constraints on source of carbonaceous aerosol using a combined C14-macro tracer analysis in a rural European background site.

Stefania Gilardoni [1], *Fabrizia Cavalli* [1], *Elisabetta Vignati* [1], *Kristina Stenstrom* [2], *Bo Larsen* [3], *Jean Philippe Putaud* [1], *Frank Dentener* [1]
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Carbonaceous aerosol, described here as the sum of organic and elemental carbon, represents a substantial fraction of atmospheric aerosol, especially in rural locations. Carbonaceous particles are emitted both by anthropogenic activities, such as fossil fuel and biomass combustion, and natural processes, including wildfires, primary and secondary biogenic particles. In order to reduce aerosol concentration, regulators need tools to apportion carbonaceous particle sources. We present the results of a source apportionment study of organic and elemental carbon for a rural site in southern Europe (Ispra) using macro tracers. The Ispra site is part of the European Monitoring and Evaluation Program (EMEP) network. Daily fine aerosol samples (PM2.5), collected continuously during the year 2007, were analyzed for mass, elemental carbon (EC), organic carbon (OC), and water soluble ion concentrations. In addition, the concentration of 14C, arabitol, mannitol, and levoglucosan were determined for a subset composed by 50 samples collected during the same year. 14C measurements indicate the contribution of modern carbon, arabitol and mannitol are tracers of primary biogenic aerosol, and levoglucosan is a tracer of primary biomass burning aerosol. We performed an analysis of the attribution of source contribution to EC/OC using a Monte Carlo analysis of the impact of emissions from biogenic, biomass burning and fossil fuel primary and secondary source categories, using a-priori knowledge on emission factors and their uncertainties. During winter 58% of total carbon (TC) is due to biomass burning and 33% is emitted by fossil fuel burning. In summer 50% of TC is emitted by biogenic sources and 46% is due to fossil fuel use. The source apportionment results agree with measurements reported by previous studies for European rural and background areas.

Sources 2.4 ID:4192 16:15

Evaluation of anthropogenic and natural surface emissions of atmospheric chemical compounds

Claire Granier [1], *Bertrand Bessagnet* [2], *Tami Bond* [3], *Ariela D'Angiola* [1], *Gregory Frost* [4], *Hugo Denier Van Der Gon* [5], *Alex Guenther* [6], *Jean-Francois Lamarque* [6], *Catherine Liousse* [7], *Aude Mieville* [1], *Martin Schultz* [8], *John Vanaardenne* [9]
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The knowledge of the distributions of surface emissions of gases and aerosols is essential for an accurate modeling and analysis of the distribution and evolution of the concentration of gaseous and particulate chemical species. The quantification of surface fluxes by source of origin is furthermore central to the assessment of effects and the development of control measures.

Over the past few years, different ranges of emission fluxes have been proposed by several studies, which have provided emissions at different spatial and temporal scales. We have compared the emissions of a large set of chemical compounds, i.e. carbon monoxide, nitrogen oxides, sulfur dioxide, black and organic carbon, and a large set of volatile organic compounds as provided by global and regional emissions inventories in different regions of the world. When possible, the emissions provided for different sectors (i.e. energy, industries, road traffic, international shipping) have been compared individually, together with their changes during the past few decades.

The presentation will focus on Europe and the United States. Significant differences in the total emissions in these regions have been identified, reaching for example 60% and 35% for anthropogenic emissions of carbon monoxide and nitrogen oxides in both regions, respectively. We will assess the current uncertainties on surface emissions and their recent trends. This analysis is often hindered because of differences in base years and in species considered in the different datasets. We will identify issues related to the speciation of volatile organic compounds and particulate emissions: emissions of PM10 and PM2.5 are provided by regional inventories, while global inventories provide emissions of sulfur, black and organic carbon. Current work aiming at compiling comparable metrics for such species in the regional and global emission sets will be discussed.

Plenary 3 ID:4613 08:30

Particle Health Effects: Understanding and Future Challenges

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In the 1980's particulate air pollution in developed countries was not considered a hazard to the public health. In the early 1990's epidemiologic studies reported acute (daily) and chronic (decades) exposures to particulate air pollution were associated with increased mortality, hospital admissions, and other clinically significant health indicators. The advances in epidemiologic assessment of the health effects of fine particulates were attributable to understanding of the chemical and physical characteristics of these particles and the ability to measure exposures in the community. Nevertheless, ambient particles remain poorly characterized, and the specific characteristics of particles responsible for the observed health effects are not understood. Current epidemiologic research is focused on identifying sub-groups of the population particularly susceptible to the effects of fine particles, mapping the distribution of particles within the community, and identifying the sources and characteristics responsible for the observed associations. Advances in our understanding will require shared insights and partnerships between physical and chemical scientists with biological and medical investigators.

Observations 3.1 ID:4411 09:15

Integrated Focus on West African cities (Cotonou, Bamako, Dakar, Ouagadougou, Abidjan, Niamey): Emissions, Air quality and Health Impact of gases and aerosols

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Fossil fuel and biofuel emissions of gases and particles in Africa are expected to significantly increase in the near future, particularly due to rapid growth of African cities and megacities. Air quality degradation is then expected with important consequences on population health. In the frame of the OMP "Environment and Health" centre and through the AMMA, POLCA, IDAF, MOUSSON programs, we are constructing a new integrated methodology to study the relations between emissions, air quality and health impact. This approach includes: (1) combustion emission characterizations; (2) experimental joint determination of gas concentrations and aerosol chemistry from ultrafine to coarse aerosol size fractions and health factors (toxicology and epidemiology). In the frame of POLCA experiments, some representative individuals (cohort) submitted to long term exposures at the measurement sites have been selected and tested through blood and spirometry analyses; (3) integrated environmental and health modeling. Exposures obtained from our multiscale and multispecies environmental modeling will be associated to a new dedicated aerosol/gas module developed for intake, deposition and clearance of gases and particles in the respiratory tract. In this work, we show the first results illustrating the construction of this methodology: - a new African inventory of fossil fuel and biofuel emissions adapted to regional specificities has been constructed for the years 2000-

2005 and 2030. - an overview of short term intensive campaigns in Cotonou (AMMA, 2005), Bamako and Dakar (POLCA, 2007-2009), Ouagadougou (MOUSSON 2007-2009) will document atmospheric pollution in West African cities. Tentative comparisons between such experimental results, modeling results and satellite data will be displayed. - in the frame of POLCA experiments, in vitro studies have been conducted from sampled filters to correlate aerosol concentrations to oxidative stress and cells inflammatory processes. Preliminary results will be tentatively presented for the Bamako experiment.

Observations 3.2 ID:4515 09:30

Particulate matter (PM10) in Istanbul: Origin, source areas and potential impact on surrounding regions

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Water-soluble ions (Cl-, NO3-, SO42-, C2O42-, Na+, NH4+, K+, Mg2+,Ca2+), water soluble organic carbon (WSOC), organic and elemental carbon (OC, EC) and trace metals (Al, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Cd and Pb) were measured in aerosol PM10 samples above the megacity of Istanbul between November 2007 and June 2009. Source apportionment analysis using Positive Matrix Factorization (PMF) indicates that approximately 80 % of the PM10 is anthropogenic in origin (secondary, refuse incineration, fuel oil and solid fuel combustion and traffic). Crustal and sea salt accounted for 10.2 and 7.5 % of the observed mass, respectively. In general, anthropogenic (except secondary) aerosol shows higher concentrations and contributions in winter. Mean concentration and contribution of crustal source is found to be more important during the transition period due to mineral dust transport from North Africa. During the sampling period, 42 events exceeding the limit value of 50 µg m-3 are identified. A significant percentage (91 %; n=38) of these exceedances is attributed to anthropogenic sources. Potential Source Contribution Function analysis highlighted that Istanbul is affected from distant sources from Balkans and Western Europe during winter and from Eastern Europe during summer. On the other hand, Istanbul sources influence western Black Sea and Eastern Europe during winter and Aegean and Levantine Sea during summer. This is a contribution to the EU funded project CITYZEN (grant 212095).

Observations 3.3 ID:4154 09:45

Trends in tropospheric NO2 over megacities in the Mediterranean and Middle East from GOME and SCIAMACHY

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Since the first years of the 21st century, more than 50% of the Earth's population lives in cities. This development is closely related to highly increased growth rates of megacities. Their resulting high traffic, energy use and industrial production make them hot-spot areas in terms of pollution.

Satellite instruments have proven invaluable to obtain long and consistent time series of atmospheric trace gases with global coverage. They facilitate studying the temporal evolution of atmospheric pollutants, as they allow to apply identical measurement techniques to all investigated regions, yielding comparable results.

In this study, we focus on tropospheric nitrogen dioxide (NO2) as a proxy for atmospheric pollution over megacities. We provide an investigation of tropospheric NO2 trends over megacities, focusing on the Mediterranean and Middle East regions. Our study covers the time period from 1996 until 2009, drawing on a combined dataset spanning the GOME and SCIAMACHY instruments. Improved stratospheric correction and air mass factor databases are employed.

We observe very diverse trends over the considered cities, ranging from decreases of a few percent per year (e.g. some western European cities) to increases of more than 10% per year (e.g. some Arabic and northern African cities), whereas some cities show no clear trend at all.

Notably, our findings show differing inter-annual patterns for the western and eastern Mediterranean regions, suggesting a strong impact of regional meteorological conditions on tropospheric NO2 column densities. Further consideration is given to the seasonality of these trends as well as the possible correlations to population and economic growth.

Observations 3.4 ID:4435 10:00

Modeling of Texas urban, industrial, and power plant plumes observed during TexAQS 2006 field campaign and its implications for NOx and VOC emissions

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Satellite and aircraft observations made during the TexAQS 2006 field mission detected strong urban, industrial and power plant plumes in Texas. We simulated these plumes using the Weather Research and Forecasting – Chemistry (WRF-Chem) model with input from the US EPA's 2005 National Emission Inventory (NEI05). We compared the model results with satellite retrievals of nitrogen dioxide (NO2) columns and airborne observations of several trace gases including nitrogen oxides (NOx), the sum of odd nitrogen species (NOy), and a number of volatile organic compounds (VOC). The model-simulated meteorology, such as wind speed, wind direction, temperature, and boundary layer height, showed excellent agreement with the observations by surface stations, wind profilers, and aircraft on selected days. The model and satellite NO2 columns agree well for regions with large power plants and for urban areas that are dominated by mobile sources, such as Dallas. However, in Houston, where both mobile and industrial sources contribute to NOx emissions, the model NO2 columns are a factor of 2 higher than the satellite columns. The same conclusions are drawn from comparisons of the model with aircraft observations. For Dallas plumes, the model-simulated NOx and NOy showed excellent agreement with aircraft observations. In contrast, the model-simulated NOx and NOy are twice as high as the aircraft observations in Houston plumes that are influenced by industrial sources. These findings may indicate that the NEI05 NOx emissions from industrial sources in Houston are overestimated by about a factor of 2. The model and aircraft observations confirm that reactive VOC emissions originating from industrial sources in Houston are underestimated. The impact of increasing the VOC emissions on the model simulations and the implications for the model - satellite NO2 column comparison will also be addressed.

Observations 3.5 ID:4402 **10:15**

The dependence of ozone production on its precursors in Pearl River Delta and Beijing Area, China

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In China, due to rapid urbanization and motorization, air quality has been deteriorated especially in mega-cities and their surroundings where ozone and fine particle pollution are recognized as major problems. Starting in 2004, the College of Environmental Sciences and Engineering of Peking University took the lead in organizing a series of international field campaigns on regional air pollution in Pearl River Delta and Beijing area (PRiDe and CAREBeijing). These were targeted at both improving the scientific understanding of atmospheric chemistry processes and at providing policy recommendations to local governments. Among those field studies, the data from field campaigns PRiDe2004, PRiDe2006, and CAREBeijing2006 have now been thoroughly processed and analyzed. In this presentation, an overview of the campaigns will be presented with focus on the issues relevant to ozone pollution, temporal and spatial variations of ozone and precursors will be summarized by the data integrated from regional monitoring networks and super-sites, then ozone production processes and its sensitivity in episodes to precursors will be diagnosed by observational-based models and emission-based models, and finally the control strategy on regional ozone pollution will be recommended.

P-Transformation.1 ID:4513 **10:30**

Seasonal and diurnal variations of CH2I2, CH2ClI, CH3I and C2H5I in the marine atmosphere

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Photolysis of iodocarbons emitted from the ocean produces iodine atoms, which decompose tropospheric ozone catalytically, and may contribute to a new particle formation. CH3I is known to be present ubiquitously in the marine boundary layer at a concentration of 0.1~5ppt. For more reactive iodocarbons, CH2ClI, CH2I2 and CH2BrI have been observed in the coastal environment at sub-ppt levels (Carpenter et al., 1999; 2003) or ppt levels (Peters et al., 2005). At remote areas, C2H5I and CH2ClI have been detected at sub-ppt level (Yokouchi et al., 1997; Chuck et al., 2005; Varner et al. ,2008), but no measurements have been reported for CH2I2 and CH2BrI. In this study, we report hourly measurements of CH2ClI CH2I2 and C2H5I as well as CH3I in the atmosphere at Hateruma Island in the East China Sea (24.05° N, 123.8° E) and at Cape Ochiishi at the north-eastern edge of Hokkaido (43.15° N, 145.5° E) throughout the whole year. Controlling factors of their concentrations in the atmosphere as well as their emission rates from the ocean will be discussed.

P-Transformation.2 ID:4380 **10:30**

Conceptual Study of Water Vapor and Bry Transport Across the Tropical Transition Layer

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The process of water vapor transport across the tropical transition layer (TTL) combines slow advection with both microphysical and convective processes. The bromine budget of the stratosphere can not be reconciled with ground-based, balloon or satellite bromine monoxide (BrO) measurements using long-lived bromine containing substances alone. Much recent modeling work has invested the contribution of the very short-lived bromine substances bromoform (CHBr3) and combined with dibromomethane (CH2Br2) upon the stratospheric Bry budget. In this study we examine these very short lived (VSL) as well as long-lived bromine species including their interaction of the water vapor transport processes through the TTL. Lagrangian back trajectories initiated in the stratosphere are generated using ECMWF Interim reanalysis data. A convective parameterization using the ERA-Interim water vapor data is presented. A simplified microphysical and bromine chemical box model representation along the trajectories is used and the effect of convective processes explored. We present the transport of the VSL bromine in a conceptualized form and investigate sensitivities to the general chemical, microphysical and convective processes that result in VSL bromine contributing to the total stratospheric bromine budget.

P-Transformation.3 ID:4472 10:30

Dependance of the stratospheric ozone depletion potentials of very short lived substances on emission location

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We present Lagrangian estimates of Ozone Depletion Potentials for very short-lived species that include dependence on source location. The estimates are based on the calculation of ensembles of trajectories driven by winds from large scale meteorological datasets. The trajectory calculations can be divided into two parts to reflect the different timescales and physico-chemical processes in the troposphere and the stratosphere with significant computational saving. The inclusion of a convective parametrization shows a variability in the geographical pattern of surface emission influence on the stratosphere and stratospheric trajectory calculations yield estimates of the expected stratospheric residence time, a concept related to the stratospheric age of air in the adjoint transport model. Stored trajectory information can be used to generate ODPs for arbitrary arbitrary linearized chemistry. We have characterized the strong dependency of ODPs for VSLSs on the emission location: it presents surface pattern variability and seasonality influenced by tropospheric transport systems such as convective regions and the Asian Monsoon but also by stratospheric features. Estimates of the ODPs of bromoform, 1-bromopropane and methyl iodide are discussed with respect to previous values found in the literature.

P-Transformation.4 ID:4364 10:30

Estimations of free tropospheric mixing rates in the Arctic using Lagrangian tracer reconstructions

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The role of mixing between pollution plumes and background air masses over the Arctic has been studied

using a multiple parcel model. Data was analysed from aircraft campaigns carried out in July 2008 as part of POLARCAT-France/GRACE in western Greenland. An event of long-range transport of pollution from Siberian forest fires to Greenland was observed with mixing playing an important role in the dilution of pollutant concentrations. These processes are poorly represented in global models. Reconstructions of measured CO concentrations were performed using a modified version of FLEXPART, including stochastic perturbations to the wind field, and initialised with CO concentrations field from the LMDz-INCA global model. This tool was used to estimate vertical and horizontal diffusion coefficients in the Arctic free troposphere. The DLR-Falcon also carried an experiment where an inert petrofluorocarbon tracer was emitted into the free troposphere and re-sampled 20 hours later. The model was adapted to simulate the injection of artificial tracer with initial concentration fields calculated using a diffusive Gaussian plume-growth approach. Results were used to reconstruct the PFC measurements and provide important additional constraints on mixing rates since the tracer was only influenced by mixing. The use of inert tracer and CO reconstructions allows further refinement of these estimates as well as information about the vertical/horizontal aspect ratio of layers/plumes during long-range transport across the Arctic.

P-Transformation.5 ID:4479 10:30

Forecasting intercontinental transport of pollution into Canadian High Arctic using Langrangian particle dispersion model.

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A forecast system has been developed to interpret measurements at the Polar Environment Atmospheric Research Laboratory (PEARL). PEARL is located in the Canadian Arctic at 610 m above sea level on Ellesmere Island (80oN, 86oW). We have used trajectory and particle dispersion models FLEXTRA and FLEXPART to construct 10-day backward trajectories starting from PEARL and 5-day emission tracer transport forecasts. FLEXPART is a Lagrangian particle dispersion model that simulates the long-range transport, diffusion, dry and wet deposition, and radioactive decay of tracers released from point, line, area or volume sources. FLEXPART was originally developed to simulate the dispersion of dangerous substances from point sources (Stohl et al., 1998), however, it has been used for many other purposes, including studies of intercontinental transport (Damoah et al, 2004), pyro-convection (Damoah et al., 2006) and as a forecast and analysis tool for flight planning (Forster et al., 2004).

For the emission tracer forecasts we have used as emission basis the EDGAR version 3.2 inventory for the year 2000 with a resolution of 1x1 degree, except for most of North America where the inventory of Forst and McKeen (2004) was used. Tracer masses are carried by particles following trajectories calculated using GFS winds from NCEP and stochastic components for turbulence and convection. Tracer forecasts are run separately for anthropogenic emissions from Europe, Asia and North America and their transport to PEARL monitored. Currently, we produce total column of carbon monoxide (CO) tracer; nitrogen oxides (expressed as NO_2), and sulfur dioxide (including direct emissions of sulfate) can be included. The species are run as passive tracers for 20 days, after which tracer particles are dropped from the simulation. Using this system, back trajectories from PEARL will be generated to explore possible origins for pollutants that arrive during selected short episodes.

P-Transformation.6 ID:4426 10:30

Evaluating the ability of a chemical transport model to simulate Arctic tropospheric pollution.

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Previous comparisons of global chemical transport models have shown that they exhibit large variability in their Arctic chemical budgets. This indicates that the processes controlling Arctic tropospheric composition are not well understood or represented within models. New in-situ gas-phase datasets collected during the POLARCAT campaign are now available. This provides an opportunity to evaluate a 3-D chemical transport model in the challenging Arctic environment and understand the current weaknesses in such models. To further compliment the analysis, retrievals from the TES instrument aboard the AURA satellite and Arctic surface station data has also been utilised. This allows the model to be tested at a variety of locations and on a range of time-scales looking at the model's representation of the seasonal cycle and inter-annual variability of CO and O3.

Results show that the standard model is unable to simulate observed concentrations of CO and also overestimates the stratospheric flux of O3 to the Arctic troposphere. This has led to a re-evaluation of CO sources and sinks in the region. Different emissions datasets for both biomass burning and anthropogenic sources have been tested within the model at different temporal resolutions. A particular focus of the work has been on the treatment of boreal biomass burning within models and quantifying its contribution to the Arctic troposphere compared to anthropogenic sources.

P-Transformation.7 ID:4200 10:30

Global evolution of chemical tracers

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Global mass conservation in advection schemes is an important requirement when using a chemical transport model. This is because when analysing the change in chemical tracers over time it has to be possible to be able to differentiate between changes in mass that arise due to the numerics and those that are due to the chemistry. Most models include a fix in order to maintain mass conservation and counter the numerical changes to tracer mass. A model that is conservative over time without needing a 'mass fix' term would be beneficial as they are not always accurate. A mass fix term will also not counter spurious features in the tracer field that may arise from the numerics. This work focuses on conservation within a global tracer advection model. The model uses a finite volume scheme and so is inherently mass conservative. The model uses an upwind advection scheme based on the NIRVANA scheme and all tracers are passive. The main change made to improve the conservation of the model involved a grid alteration over the Polar Regions. The resulting experiments revealed that in the case of winds specified analytically (such as solid body rotation) the model produced mass change on the order of a fraction of a percent over the period of a month. Global tracer mass is not accurately conserved when driven by analyses, even though the continuity equation is used to obtained vertical velocity from horizontal divergence and surface pressure. The non-conservation arises through time variation in the winds. The error is much smaller with ERA-Interim (2.5% change over 6 months) than with earlier analyses (15%) indicating that ERA-Interim is more consistent in time. The implementations for stratospheric ozone hole simulations are discussed.

P-Transformation.8 ID:4391 10:30

Improving process understanding of transpacific transport with high-resolution models and observations

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Global chemical transport models are essential tools for estimating intercontinental transport of air pollution, but their ability to adequately resolve the relevant processes requires further study. Fine-scale processes affect both the export of pollution from Asia (e.g. deep convection, rapid frontal lifting, and topographic venting) and the import of Asian pollution to surface air over the North American west coast (e.g. mixing of free troposphere air into the boundary layer, orographic flow, and land-sea breezes). We conduct high-resolution simulations using the Weather Research and Forecasting model with fully coupled chemistry (WRF-Chem) for a hemispheric domain covering East Asia and North America (60x60km^2), and a nested domain (20x20km^2) over the North American west coast. The hemispheric WRF-Chem simulation is driven with boundary conditions from the global MOZART-4 model (2.8x2.8 degree). We evaluate the regional and global simulations with observations from EANET (Asia), AQS and CASTNet (United States) surface networks as well as TRACE-P (western Pacific, spring 2001) and INTEX-B (eastern Pacific, spring 2006) aircraft field campaigns. Analysis of WRF-Chem simulations and TRACE-P measurements indicate the venting of Asian pollution by deep convection that develops rapidly along the leading edge of frontal system convergence bands. This rapid venting is not adequately resolved in either of the two global models compared with the observations, suggesting a potential underestimate of Asian outflow. Further analysis for the INTEX-B period consistently suggests that the continental outflow of SO_2 and O_3 at a regionally representative EANET site (26.8N, 128.2E) is simulated more accurately in the regional WRF-Chem model than in the global MOZART model. We focus on a plume that was lifted from the coast of China, transported rapidly across the Pacific as indicated by daily CO maps from the AIRS instrument aboard the NASA Aqua satellite, and arrived ~3 days later above Seattle with sulfate > 5ug/m^3, CO > 300 ppbv, and O_3 > 100 ppbv as observed by the NCAR-C130 flight. This plume is absent in the global model simulation, and we intend to explore further with the hemispheric WRF-Chem simulation. The nested WRF-Chem analysis is designed to diagnose Asian O_3 influence in U.S. surface air with a focus on the high-altitude sites in spring where chances are best for detecting an influence.

P-Transformation.9 ID:4480 10:30

How much air-mass mixing occurs during long-range transport in the free troposphere?

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Lidar sounding of aerosol, ozone and water vapour as well as in-situ measurements at the Zugspitze summit (Germany, 2962 m a.s.l.) have been used for the characterization of atmospheric layers that have travelled over many thousand kilometres. In some cases contributions from rather different sources co-exist in such air masses. This is frequently the case if the air stream gets close to frontal systems. However, without convergence or stirring the composition of air masses may remain more or less unchanged even after travelling over very long distances. One example is stratospheric air intrusions. Measurements of water vapour in recent years with our differential-absorption lidar (DIAL) have repeatedly demonstrated relative

humidities between 0 and 2 % even in very thin intrusion layers. The corresponding mixing ratios are so low that we must assume that any mixing with surrounding air masses must have occurred in the upper troposphere, i.e., in the remote arctic source regions. Ozone in these air tongues, as observed at the Zugspitze, rarely reaches stratospheric values and also CO frequently does not decrease at all, a fact which is not easy to understand. Another example is air from the subtropical Atlantic that, e.g., reaches Central Europe in prefrontal air streams. These layers exhibit sharp edges in the ozone distribution and carry along ozone, nitric oxides and CO with low concentrations as found over the remote oceanic regions. All these observations induce a challenge for the model development since models have a tendency of overestimating atmospheric mixing due to the limited spatial resolution.

P-Transformation.10 ID:4546 10:30

Studying the effect of meteorology on calculations with the EMEP Unified model

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The EMEP model is used operationally for assessment of the regional levels and the transboundary fluxes of acidifying compounds, ozone and particulate matter (PM) in Europe, and also pre-operationally for chemical weather forecasting. Accurate model calculations are essential for reliable assessment of the compliance with regulatory target values. Furthermore, correct description of the PM chemical composition is important for trustworthy modelling of PM source allocation and the impact of emission reductions. During the past decade, the PARLAM-PS (based on HIRLAM-v.3) weather prediction model was used to prepare meteorological input to the EMEP model. Using the same meteorological model is essential for consistent calculations of pollution trends. Recently, updating the meteorological driver has caused changes in EMEP model results. The most prominent is a considerable decrease of the surface concentrations of most pollutants, leading in several cases to model underestimation of observations. Here, we look at the effect of using different meteorological drivers, namely the HIRLAM-v.7 and ECMWF models, on EMEP model results, with a special focus on its performance for PM. We analyse the reasons for the significant discrepancy between model calculated PM concentrations when two versions of the HIRLAM meteorological model were employed. In particular, the seasonal variation and spatial distribution of precipitation, as well as the atmospheric stability and vertical mixing were found to be responsible for the decrease of surface concentrations, accompanied by their increase aloft. Also, the effect of meteorology on source-receptor calculations has been investigated, which may have implications for designing emission reduction strategies. Finally, we compare the effects due to using different meteorological drivers with those that are due to inter-annual meteorological variability.

P-Transformation.11 ID:4437 10:30

Modeling study of thunderstorm effects on the composition and chemistry of the upper troposphere during the 2006 North America Monsoon

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Recently the existence of an enhanced ozone region in the upper troposphere (UT) over the southern U.S. during July and August has been documented (Zhang et al., 2003; Li et al., 2005; Cooper et al., 2006, 2007,

2009). These studies have hypothesized that the ozone is a result of thunderstorms and chemistry occurring during the North American monsoon (NAM) when an UT anti-cyclone traps the air. To examine the role of thunderstorms on UT composition and chemistry, we have conducted a high-resolution simulation using the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) for the NAM region so as to explicitly simulate convective systems, their convective transport and production of nitrogen oxides from lightning, and subsequent chemistry.

This presentation focuses on the mid July to early August 2006 period when the anti-cyclone sets up over the southern U.S. Modeled meteorology, UT ozone, and carbon monoxide are evaluated with meteorological analysis data, ozonesondes, and TES satellite observations. We examine the time variation of the mass of different species in the 5-15 km region and at the 10 km altitude. We further analyze these species in the context of boundary layer, stratospheric, and lightning-NO_x tracers. Finally we make estimates of ozone production from various chemical and transport sources.

P-Transformation.12 ID:4255 10:30

The impact of deep convection on the radical budget over Canada in summertime

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The mixing ratios of numerous organic trace gases and ozone were measured in the upper troposphere over Canada during the ARCTAS aircraft campaign in July 2008. A comprehensive suite of inorganic species and organic species, including NMHCs, halogenated organics and oxygenated volatile organic compounds were measured using a variety of instruments on- board the NASA DC-8. The research objectives of ARCTAS included the study of biomass burning plumes and their effect on the atmosphere. A natural extension of the objectives included the chemical impact of deep convection on the upper troposphere over Canada. A significant number of convective outflow events were identified by the elevated mixing ratios of organic species in the upper troposphere relative to background conditions. Several dramatic convective events were observed in which isoprene was detected at hundreds of pptv at altitudes higher than 10 km. First generation oxidation products of isoprene were detected as well allowing a calculation of the photochemical lifetime and hence the timescale of the convective events. The impact of deep convection on the local upper tropospheric formaldehyde and HOx budgets is assessed and a global model, MOZART, is used to extrapolate our findings into a global context.

P-Transformation.13 ID:4421 10:30

Assessment of the lightning nitrogen species emissions in the Upper Troposphere and of their contributions on tropospheric chemistry

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NO_y species play a major role in the tropospheric chemistry by controlling ozone. In the upper troposphere, NO_y is principally emitted on the form of NO_x through aircraft, surface emissions, stratospheric intrusions and lightning activity. This study aims to discriminate lightning emissions and to evaluate their influence on the NO_y and O_3 distributions over the northern midlatitudes. We use 5 years (2001-2005) of aircraft observations (NO_y, CO, O_3, Relative Humidity) from the MOZAIC program, ground based and satellite observations of clouds (GOES, METEOSAT) and lightning (LIS, NLDN, ATD), combined with systematic simulations of the FLEXPART model, in order to assess the location of NO_x sources and to quantify lightning activity and the main transport agents driving NO_y and O_3 distributions. During fall and winter lightning emissions are generally characterised by low NO_y/CO correlation, with NO_y values higher than 1 ppbv and low CO. In one case sampled over the Atlantic, mean NO_y (0.70 ppbv) and CO (83 ppbv) concentrations were associated with lightning activity over the Gulf of Mexico, 72 hours prior to the measurements, with more than 80.000 CG flashes. During summer, most of the large-scale plumes with high NO_y are associated with lightning NO_x emissions and pollution. Nevertheless, during July 2003, a 1000 km zonal plume influenced by clean convection was observed over the Atlantic (87 ppbv of CO, 90 ppbv. of O_3, 2.1 ppbv of NO_y), associated with lightning activity over the Gulf of Mexico with more than 100.000 flashes one to four days before the observation. Based on the observations, transport and lightning activity properties, we estimate the NO_y and O_3 produced for each episode. It is finally expected that the outcome of such an exploitation of the MOZAIC records will serve as a basis to build a comprehensive climatology of NO_y in the upper troposphere.

P-Transformation.14 ID:4168 10:30

Record of Concentration, $\delta^{13}C$ and $\delta^{18}O$ of Atmospheric CO over the last millennium from Antarctic Ice Cores

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A record of preindustrial biomass burning over the past ~650 years in the extratropical Southern Hemisphere is derived from the carbon monoxide (CO) isotope record based on new measurements of CO concentration, $\delta^{13}C$, and $\delta^{18}O$ from 40 Antarctic ice core samples covering the time period from 1360 to 1900. CO concentration decreases by ~25% (14 ppbv) from 1360 to around 1600, then recovers completely from 1700 to the late 1800s, followed by a very slight decrease by 2000. $\delta^{13}C$ and $\delta^{18}O$ of CO decrease by about 2‰ and 4‰ respectively from 1360 to 1600, then increase by about 2.5‰ and 4‰ respectively from preindustrial times to the late 1800s. Present day values of both isotopic ratios are close to those observed during the 1600s. Based on these observations, we conclude that biomass burning strongly modulated the CO budget during preindustrial times and decreased significantly from the early 20th century to the present day. This decrease was nearly compensated by the concomitant methane increase, which is an isotopically depleted source of atmospheric CO, bringing present day CO concentrations close to its maximum level observed over the last 650 years.

P-Transformation.15 ID:4289 10:30

In Situ Spectral Study Of Ozone Hydrates Formation And Their Reaction With Chlorine-containing Compounds Under Stratospheric Conditions

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Ozone is one of the unique chemical substances: at the same time it demonstrates favorable and harmful activity on the people’s life, along with easiness of decomposition its amount in the Earth’s atmosphere is reasonable. Nevertheless chemistry of ozone up to date is not investigated totally. Only few literature data concern ozone hydrates: they are supposed to belong to type II clathrates with (H2O/O3) ratio equal to 5.75. The experiments on the laboratory simulation of heterogeneous chemical processes were carried out with the use of vacuum flowing gas-discharged device under stratospheric conditions. Ozone was synthesized from the purified oxygen in gas-discharge tube and trapped at liquid nitrogen temperature. Thin reactant films were produced on the thermostated gold reaction mirrors of the specially designed grazing angles reflection cuvette under 10-4 torr and 77K. Reactions were studied at 77K-290K by FTIR spectroscopy at 1 cm-1 resolution. Outgoing gases were fixed by time-of-flight mass spectrometer. Spectrum of thin ice film covered by ozone at 77K consisted of vibration bands of H2O, O3 and H-bonds in adsorbed state. Growth of temperature to 85K lead to increasing of ozone bands (660, 678, 1036, 1264, 1300 cm-1) and H-bonds, especially 2107 cm-1. New absorption lines at 704, 1001, 1106, 2045 and strong doublet 2350-2380, 2784 см-1 were also observed. The substance formed did not react with HCl untill 220K, while normally reaction of HCl, adsorbed on the ice film, with ozone takes please at 77K. This fact point out on formation of gas clathrate hydrate which decompose only at temperatures higher than 220K. Its reactions with CHCl3 and CHBr3 were studied also. Thus, the present work show the possibility of ozone hydrates formation under stratospheric conditions.

P-Transformation.16 ID:4485 10:30

Production and Transport of Ozone in the Amazon: Comparison of WRF-Chem and CCATT-BRAMS Simulations with In-Situ Observations from the BARCA Campaign

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In the Amazon Basin, biogenic, biomass burning, and urban emissions react photochemically to produce ozone, which is then transported by convection and regional circulations. As the Amazon basin becomes increasingly populated and developed for agriculture, the ability to predict regional ozone distributions will be critical to mitigating pollution that harms human health, agriculture, and natural ecosystems. However, major uncertainties remain in characterizing emissions, photochemical processes and pollutant transport in the Amazon basin. In this study, basin-wide CO and O3 aircraft measurements taken during the LBA/BARCA campaign during the dry-to-wet transition of 2008 are used to evaluate the WRF-Chem and CCATT-BRAMS coupled chemistry and meteorology models, constrain emissions and analyze ozone budgets in the Amazon. The WRF-Chem (Weather Research and Forecasting with Chemistry) model

developed at NOAA/ESRL was adapted in order to more realistically represent tropical meteorological and chemical processes. New WRF-Chem implementations include updated land cover and surface properties and soil moisture initialization from an off-line hydrological model, EDGAR4 urban emissions updated with vehicle emissions inventories for South American cities, and chemistry initial and boundary conditions from the MOCAGE global model. CCATT-BRAMS (Coupled Chemistry-Aerosol-Tracer Transport model coupled to the Brazilian developments on the Regional Atmospheric Modeling System) is currently in operational use for regional air quality forecasting at Brazil's National Institute for Space Research (INPE). Simulations were conducted with both models using RACM (Regional Atmospheric Chemistry Mechanism) and parameterizations for shallow and deep convection and 1D plume rise, with a 30/10km 2-way nested-grid configuration covering the Amazon Basin. The performance of the meteorological models was evaluated using LBA flux tower and surface meteorological station data and the TRMM precipitation product. Simulated CO and O3 mixing ratios were compared with in-situ observations from the BARCA campaign, and the model performance in reproducing both individual flight tracks and regional distributions was evaluated.

P-Transformation.17 ID:4529 10:30

Ozone budget in the lower troposphere over Amazonia

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During LBA/BARCA (Balanço Atmosférico Regional de Carbono na Amazônia) field experiment, measurements of several trace gases and aerosols were made onboard INPE´s “Bandeirante” aircraft. Flights transected the Amazon basin from the Tropical Atlantic Ocean shore to the northwestern part of the Amazon basin, passing over a variety of ecosystems from the surface to an altitude of about 5 km in two different periods. Phase A was in November 2008, during transition from dry to wet season, with rainfall to the west and south of Manaus, but much drier conditions with widespread vegetation fires in the eastern and northern Amazonia. Phase B occurred during the opposite transition, from wet to dry season, when the ITCZ was located over the equator, abnormally south of its normal position in May, favoring events of northern hemisphere inflow to the Amazon basin. Precipitation was above average and fires only occurred far to the south. Here, we examine the observations of ozone through comparison with mixing ratios simulated by CCATT-BRAMS, a non-hydrostatic and fully compressible mesoscale chemistry-transport model, aiming to use the model results to assess the ozone budget in the Amazonia during these two different periods. The observed ozone zonal gradient across the basin below 700 mb was analyzed through the relative contribution of precursors emissions from urban, biomass burning and biogenic sources, as well as chemical production and loss, dry deposition and convective transport mechanisms.

P-Transformation.18 ID:4577 10:30

Cross-Border Transport of Ozone and its Precursors between Canada and the US

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Transport of pollution across geopolitical borders may complicate effective implementation of regional or nationwide air pollution control strategies. The problem is further confounded for secondary pollutants that can be transported across a border as the pollutant or precursors. In such situations, source attribution from various sources becomes an important and yet challenging task. Contributions from various local, regional, or global sources to air pollution at certain locations are usually evaluated using receptor- or source-based models. However, various approaches can be taken for evaluation of source impacts. In this work we will explore various approaches for source attribution that differ in terms of the impacted air quality metrics, as well as attribution methodologies. In particular, we will look at three methods of source attribution in zero-out simulations, adjoint contribution evaluation, and cross-border flux estimation. Zero-out and flux simulations are performed using the community multiscale air quality (CMAQ) model of the USEPA. For adjoint calculations, we will employ the adjoint version of CMAQ. All simulations will be carried out for the summer of 2007 and covering the main portion of the North American border between Canada and the US (excluding Alaska). We will explore deficiencies and strengths of each method while looking at the impact on concentrations and ozone exposure as two air quality metrics of concern.

P-Transformation.19 ID:4231 10:30

Spatial and Temporal Variability of Trace Gases over the United States Derived from Regional Model Output

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The determination of the spatial and temporal resolution needed by a geostationary instrument to adequately resolve trace gas species and aerosols associated with the emission and transport of air pollution will be explored for several regions of the United States. WRF/Chem and CMAQ were used to generate 4-km and 1.5 km horizontal resolution trace gas and aerosol fields, respectively, over the Eastern US for a high ozone episode that occurred on July 9, 2007. In addition, CMAQ was used, at a 4-km horizontal resolution, to model a pollution event over Los Angeles, CA on July 4, 2007. For each model run, O3, NO2, CO, HCHO, and SO2, and PM2.5 were analyzed. Using a spatial autocorrelation analysis, horizontal variability was calculated as a function of separation distance in an attempt to quantify the resolution needed to capture various amounts of explained variance. This process was repeated for each species, all showing the decay of the autocorrelation with greater separation distance. For the Northeast US, autocorrelations of NO2 and SO2 displayed the most rapid decay, whereas over Los Angeles, no particular species exhibited significantly faster decay than the others. In addition, a temporal correlation analysis was performed to determine the optimal temporal resolution needed from a geostationary instrument. Results from spatial and temporal variogram analyses will also be shown for several trace gas species. These variograms show how trace gas gradients in both space and time change with instrument resolution.

P-Transformation.20 ID:4296 10:30

What happens at night matters for this morning: the effect of nighttime chlorine activation on oxidant budgets in urban air

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Three field campaigns have now established that nighttime heterogeneous chemistry of reactive nitrogen leads to substantial levels of the chlorine atom precursor nitryl chloride (ClNO2). The implications of this chemistry on daytime oxidant and nitrogen oxide budgets is expected to be significant, yet detailed treatments are lacking. I use a combination of conceptual and detailed chemical box models of urban photochemistry and nighttime processes to assess the effects of intense nitryl chloride formation under wintertime conditions. Direct effects include enhanced HOx formation and consequent ozone production rates. The effects on NOx abundance and lifetime are more subtle due in part to feedbacks involving HOx. A poor understanding of the vertical extent of this chemistry continues to plague quantitative estimates of its regional importance.

P-Transformation.21 ID:4465 10:30

Investigation of strong ozone plumes downwind of New York during ICARTT including enhancement of background ozone due to Alaskan wildfires

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Well defined urban plumes originating from New York City (NYC) were studied in the aircraft measurements during the ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) field campaign in 2004. They extended vertically to over 2 km near NYC and were transported above the stable marine atmospheric boundary layer. Maximum ozone mixing ratios of about 160 ppbv were observed 600 km downwind of NYC. In this study, the WRF-Chem (Weather Research and Forecasting-Chemistry) model was used to simulate the transport and chemical transformation of the urban plumes (‘New York plumes’) during the 2 day period in July 2004. For the simulation, anthropogenic and biogenic emissions were generated based on the EPA National Emission Inventory 2005 and the Biogenic Emissions Inventory System Version 3.13. The simulation results were compared to in-situ observations from surface stations, the NOAA P3 aircraft, and the Ronald H. Brown research vessel and airborne lidar measurements, focusing on ozone mixing ratios and meteorology. Chemical and meteorological features of the episodic New York plumes were well captured in the simulations. However, the model systematically underestimated the observed ozone by about 20 ppbv in the layer above 1 km. In order to explain the ozone discrepancies between the model and the observations, the contribution of remote biomass burning to background ozone was investigated using the ‘online’ plume rise module in the model. From these simulations, it appears that large wildfires in Alaskan and Canadian forest areas in summer 2004 affected the New York plumes. In addition, we examined the impacts of chemical initial and boundary conditions on the simulated ozone using the output of the global chemical transport model MOZART4.

P-Transformation.22 ID:4207 10:30

Physical and chemical processing of tropospheric ozone during long range transport

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Understanding the processes influencing air masses during transport is important for both air quality and climate issues. For example, the lifetime of trace gases in the troposphere can be affected by long range transport, and descent following high altitude transport can lead to ozone (O3) production downstream of pollution sources.

The Lagrangian photochemical model “ensemble CiTTyCAT” (Cambridge Tropospheric Trajectory model of Chemistry and Transport) is used to investigate the interactions between physical and chemical processes influencing tropospheric O3 during long range transport.

Observed air mass composition and photochemical activity is reproduced by the model in North Atlantic transport cases with robust Lagrangian links (from the International Transport and Chemical Transformation Lagrangian 2004 experiment). The integrated change in O3 following air masses, ΔO3, is reproduced to within the model and observational uncertainties.

The model is used to investigate ΔO3 partitioned by process in different regimes: low altitude anthropogenic pollution transport over the N. Atlantic, high altitude boreal fire emissions and West African Monsoon (WAM) flow. In low altitude N. Atlantic transport, there is chemical and physical O3 loss in the lowest layers. In the high altitude fire emissions case, chemical O3 production dominates.

Net chemical production of O3 in the WAM is 4-5 times higher than in the boreal fire emissions case, as photolysis rates, temperature and humidity are higher in the tropics and the air mass is continually passing over fresh emissions. The large photochemical O3 production in the WAM is counteracted by physical losses, particularly dry deposition over vegetation. The physical and chemical processes are inextricably linked and the processing responds non-linearly to perturbations.

P-Transformation.23 ID:4196 10:30

Three-dimensional smoke plume dynamics and aerosol properties and their transformation measured by a ground-based lidar

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Biomass burning is a major contributor of particulate matter and other pollutants to the atmosphere. The largest uncertainty in modeling the long-range transport of smoke plumes are the plume injection height, dispersion, and the diurnal cycle of the plumes. We have developed a ground-based, mobile elastic scanning lidar (light detection and ranging) instrument to study the three-dimensional plume dynamics and the transformation of optical properties of smoke particles over large wildfires. The two-wavelength lidar (355 nm and 1064 nm) has an operational range up to 20 km with a spatial resolution of 2.4–9.6 m. The scanning angles are 0°–180° (azimuth) and 0°–90° (vertical). The lidar is lightweight, can be easily assembled in an hour in the field, and has been deployed in rugged mountainous terrain near 20 fires since 2003.

We will present the lidar measurement results for two large fires in northwestern United States: the Kootenai Creek Fire in western Montana on August 27–28, 2009, and the Tripod Complex Fire in central Washington on August 8–21, 2006. We will show the profiles of multiple layers of smoke plumes, the upper and lower boundaries of the plumes, and their spatial and temporal evolution. The selected profiles of aerosol backscattering, extinction coefficients, and optical depth will also be presented. The profiles are derived from the lidar data using the lidar measurement methodology adapted to the specifics of smoke-polluted atmospheres. In addition, we will show a time series of the progression of smoke plume dynamics, its interaction with clouds, and its diurnal cycle from early morning to late afternoon for the I-90 Fire in western Montana on August 9–16, 2005. The lidar observations are being used to validate the plume rise sub-models in our air quality-forecasting model, Weather Research and Forecasting – Smoke Dispersion (WRF-SD).

P-Transformation.24 ID:4187 10:30

NO_x and PAN emissions from biomass burning: Implications for tropospheric ozone

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Aircraft campaigns in the Arctic (*e.g.*, ARCTAS-B) and in the tropics (*e.g.*, TRACE-A) have shown that the NO_x emitted by biomass burning is rapidly converted to PAN within the concentrated smoke plumes. However, most emission inventories used in global models do not account for this subgrid chemistry, thus overestimating NO_x emissions and underestimating PAN emissions from biomass burning. This can have a large impact on the simulation of tropospheric chemistry, especially ozone, in regions impacted by biomass burning. Here we examine the impact of these emissions of PAN on global tropospheric chemistry using the GEOS-Chem model. We use observations from TES and OMI as well as aircraft data to evaluate the model simulations. In the Arctic, aircraft and TES observations during ARCTAS-B both show little evidence for ozone formation within boreal smoke plumes. Adjusting model emissions to match aircraft observations of CO and the observed enhancement ratios of NO_x and PAN in boreal smoke plumes reduces model ozone by 5% throughout large regions of the Arctic. We will discuss the impact of PAN emissions from tropical biomass burning on ozone levels, and the sensitivity of model ozone to the injection height of emissions.

P-Transformation.25 ID:4491 10:30

FOREST FIRE IMPACT ON AIR QUALITY: THE LANCON-DE-PROVENCE 2005 CASE

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In Mediterranean regions, forest fires are a risk to the environment and to communities. Wildfires also represent a significant source of gas and aerosols. Depending on the meteorological conditions, these emissions can efficiently perturb air quality and visibility far away from the sources. The aim of this work is to simulate the interactions of a mediterranean fire with its environment both in terms of dynamics and air quality. A new coupled fire-atmosphere system Forefire / Meso-NH was applied for the first time to a typical mediterranean fire to study the impact of fire on the dynamics and air quality. The coupled atmosphere-wildfire modeling was able to simulate well documented perturbations induced by forest fires on the atmosphere, in particular fire-induced increase and convergence of surface winds. The impact of fire on atmospheric pollutants was simulated several hundreds of kilometers downwind of the burnt area. Ozone destruction was obtained near the fire in the high-NOx regime whereas ozone production was simulated

downwind of the fire.

P-Transformation.26 ID:4377 10:30

Global distribution of the effective aerosol hygroscopicity parameter for CCN activation

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The effective hygroscopicity parameter (K) offers a simple way of describing the influence of chemical composition on the CCN activity of aerosol particles, this facilitates calculation of the climate effects of aerosols. Andreae and Rosenfeld (2008) reviewed observed values of K and suggested that continental and marine aerosols on average tend to cluster into relatively narrow ranges of effective hygroscopicity (continental K = 0.3 ± 0.1; marine K = 0.7 ± 0.2). However, currently, the availability of observational data is not sufficient to obtain a clear picture of the global distribution and characteristic regional differences of K . In this study we use a general circulation model to calculate the global distribution of K and analyse the variation in K both between and within regions.

We find that the simulated global mean value of K at the surface is 0.27 ± 0.21 for continental and 0.72 ± 0.24 for marine regions. Over the continents K appears to be quite uniform, but Africa and South America have a lower mean K (0.15 – 0.17) than other continental regions (0.21 -0.36). At the top of the planetary boundary layer, K values can deviate significantly form surface values (by up to 30 percent). Marine values of K are seen to be influenced by continental outflow, with reduced values in marine regions where there natural and anthropogenic aerosol pass from the continents over the ocean (e.g. the Gulf of Mexico and the Saharan outflow region) .

P-Transformation.27 ID:4191 10:30

Aerosol formation and growth in anthropogenic sulfur plumes

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Within the past ten years, global and regional chemical-transport models with online aerosol microphysics have become powerful tools for understanding how humans may be changing aerosols, clouds and climate. However, large uncertainties in processes such as new-particle formation and emissions limit the predictive ability of these models. Related to both of these uncertainties is the question of how to represent sub-grid aerosol processes in large-scale models with grid-box lengths of 10s of km or larger. Sub-grid SO2 oxidation in power-plant plumes with condensation of H2SO4 onto newly-formed and existing particles is an important example of these difficult sub-grid aerosol processes. We have developed a modeling framework with aerosol microphysics in the System for Atmospheric Modelling (SAM)[1], a Large-Eddy Simulation/Cloud-Resolving Model (LES/CRM), to explore these plumes in detail and develop parameterizations of plume microphysics for global and regional models. We evaluate the model with

airborne data obtained in the plume of the W. A. Parish power generation facility near Houston, TX during the TexAQS 2006 field campaign[2], as well airborne data obtained in the plume of the Conesville power generation facility near Conesville, OH during the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) campaign[3]. Finally, we show how the effective downwind plume aerosol emissions can be greatly modified by both meteorological and background aerosol conditions. The ultimate goal of this project is to develop improved emissions schemes for sub-grid nucleation in global and regional models. [1]Khairoutdinov, M. F., and D.A. Randall. (2003) J. Atmos. Sci., 60, 607-625 [2]Parrish, D. D., et al. (2009) J. Geophys. Res., 114, D00F13 [3]Fountoukis, C. et al. (2007) J. Geophys. Res., 112, D10S30

P-Transformation.28 ID:4115 10:30

Assessment of particulate matter present in the Atmospheric aerosol and its adverse health effects

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Particulate matter (PM) has been widely studied in recent years due to its potential health impact and need for its control. The particulate matter can pass through the natural protective mechanism of human respiratory system and plays an important role in genesis and augmentation of allergic disorders. Sources of air pollution in the area and the unique problem arising out of the emission from the vehicles, industries, etc. have been described. A study of the atmospheric particulate matter (APM) and associated anions present in aerosol has been carried out for the city of Raipur India using principal component analysis (PCA) and positive matrix factorization (PMF) models. The data are analysed to evaluate the critical situation arising out of the emission of air pollutants and the impact on human health due to respirable diseases (RDs) to middle class sub-population in the area are assessed. A strategic air quality management plan has been proposed. The particulate matters (PM10 and PM2.5) in the ambient air at residential site of Raipur city were collected using sequential speciation air equipped with temperature and humidity sensor. The annual mean mass concentration distribution of PM2.5 and PM10 was ranged from 9.0 – 207 and 28 – 464 µg m-3 with mean, median and STD values of 93, 196; 82, 158 and ± 57 and 116 µg m-3, respectively. The seasonal evolution of the daily contributions confirmed the interpretations of these sources. Ambient air quality was monitored along with micrometeorological data and the results are discussed. The status of air pollution in the area has been evaluated and a questionnaire survey was conducted to estimate the allergic symptoms and exposure to assess the respiratory disorders.

Allen, A.G., Nemitz, E., Shi, J.P., Harrison, R.M., Greenwood, J.C. Atmos. Environ. 35, 4581–4591, 2001. Mouli, P.C., Mohan, V.S., Reddy, J.S. Environ. Monit. Assess. 117, 291–305, 2006. Mishra, U.C.J. Aerosol Sci. 19, 1165–1169, 1988.

P-Transformation.29 ID:4217 10:30

Towards high pollution episode forecasting of PM10/PM2.5 under very stable nocturnal conditions and steep terrain using the WRF-Chem model.

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This ongoing study presents a system to predict high pollution events developed under special meteorological conditions during the winter over the City of Santiago de Chile. An accurate forecast of these

episodes is of extreme importance since the government has to take actions in advance to prevent excessive air quality deterioration. The forecasting system is based on the fact that during these episodes a high correlation (over 0.95) is observed between measurements of elevated levels of PM10/PM2.5 and carbon monoxide (CO). Thus, by accurately forecasting CO, which behaves closely to a tracer on this scale, a PM estimate can be made without involving an aerosol-chemistry model. This simplification is convenient to the authorities that responsible for operational air quality forecast since it reduces computing resources. However the very stable nocturnal conditions over steep topography that creates the maximum in concentrations are hard to represent by models. The new system is based on the WRF-Chem model with optimum settings, determined through extensive testing, that best describe both meteorological and air quality measurements. Some of the important configurations choices relate to the boundary layer (PBL) and cloud schemes, which can have an effect on the radiative feedbacks that modify the nocturnal PBL. Finally, a forecast for the whole winter period for year 2008 is performed, evaluating the forecasting performance and comparing it to previous prediction models.

P-Transformation.30 ID:4330 10:30

Aircraft Measurement of Polar Organic Compounds in PM10 Aerosols over Central China: Contributions of Primary Emissions and Secondary Photooxidation

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Atmospheric aerosol samples (PM10) were collected by aircraft at low-middle altitudes (0.5-3.0 km) over Central East to West China during summer 2003 and spring 2004. They were analyzed for different types of polar organic compounds using GC/MS. More than 80 species were detected in the aerosols and grouped into different compound classes, including fatty acids, n-alcohols, sugar compounds, sterols, hydroxy-/polyacids, organic nitrogen compounds (oxamic acid and urea), and biogenic secondary organic aerosol (SOA) tracers for the photooxidation of isoprene (e.g., 2-methyltetrols), α/β-pinene (e.g., pinic acid), and β-caryophyllene (β-caryophyllinic acid). Total concentrations of the identified organics were 746-3537 ng m-3 (average 1986 ng m-3) in summer versus 223-1339 ng m-3 (519 ng m-3) in spring. Fatty acids are the most abundant compound class, followed by n-alcohols, sugar compounds and biogenic SOA tracers. The total concentrations of biogenic SOA tracers are higher in summer (5.80-153 ng m-3, average 50.8 ng m-3) than in spring (5.76-66.8 ng m-3, 22.2 ng m-3). This difference is especially significant for isoprene SOA tracers (41.6±38.0 ng m-3 in summer versus 14.3±10.5 ng m-3 in spring). On the contrary, concentrations of α/β-pinene and β-caryophyllene SOA tracers are similar between spring and summer. Interestingly, a strong correlation (R2=0.83) was found between levoglucosan and β-caryophyllinic acid for the spring samples whereas no correlation (R2=0.17) for the summer samples, indicating that biomass burning could be a main source of β-caryophyllinic acid over Central China during springtime. Total concentrations of organic nitrogen compounds are comparable to those of biogenic SOA tracers without any differences between spring and summer. The urea concentrations decreased with an increase in the altitude, suggesting that it may be emitted from the anthropogenic sources on the ground surface. This study demonstrates that both biogenic SOA and organic nitrogen are significant components of organic aerosols over Central China.

P-Transformation.31 ID:4600 10:30

Development and Application of a Geographical Tagged-Species Source Apportionment Method to Characterize Transport of Primary PM Components in a Regional Air Quality Model

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Atmospheric transport of particulate matter (PM) and its precursors is a serious issue for human health problems as well as air quality management. Regional Eulerian PM air quality models are designed to help understand how PM is formed and transported between regions or countries. However, most PM air quality models do not have built-in source apportionment capabilities to isolate and quantify the contribution of various source categories or regions to the PM burden at a given receptor. The work presented here focuses on the development, evaluation, and application of a geographical, emissions-based source apportionment method within the regional air quality modelling system AURAMS (A Unified Regional Air quality Modelling System).

AURAMS was developed by Environment Canada to describe the formation of ozone, particulate matter (PM), and acid deposition in the troposphere. The off-line AURAMS chemical transport model is driven with meteorological fields from the Canadian operational weather forecast model GEM and solves chemical and micro-physical processes related to size-segregated, internally-mixed aerosol particles made up of 9 chemical components (sulphate, nitrate, ammonium, elemental carbon, primary organic matter, secondary organic matter, crustal material, sea-salt, and particle-bound water) and their interaction with gaseous co-pollutants in multiple phases.

The tagged-species source apportionment method that was implemented in AURAMS can track primary organic matter emissions from selected regions throughout a North American continental domain (Canada, U.S., and northern Mexico). The method has been checked in terms of mass conservation and the hypotheses related to primary processes. As a sample application, the relative contributions of Canada and the U.S. to North American ambient organic carbon concentrations for a one-month simulation will be discussed. These results will also be compared to results from two brute-force “zero-out“ sensitivity simulations.

P-Transformation.32 ID:3559 10:30

Particle phase products from the reaction of nopinone with OH radicals

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β-Pinene is one of the most abundant monoterpenes after α-pinene in the atmosphere. The oxidation of these compounds leads to semi-volatile compounds that form secondary organic aerosol (SOA). In addition, these oxidation products can be oxidized further to form a wide range of secondary oxidation products that also play an important role in the formation of SOA. The knowledge about the particle phase products is an essential step towards a better understanding of the atmospheric degradation of the monoterpenes. We present the results from a series of chamber experiments of the known β-pinene oxidation product, nopinone, with OH radicals. The produced particulate products were sampled using a denuder/PTFE filter device. After extraction, the filter samples were analysed using HPLC/(-)ESI-TOFMS. The influence of the reaction conditions were investigated by using different OH sources and changing the particle acidity. The following seed particles were used for the experiments: sodium sulfate ($pH \approx 7$), ammonium bisulfate ($pH \approx 0.1$) and two mixtures of ammonium sulphate/sulphuric acid (pH ≈ 0.6, pH ≈ 2). OH radicals were produced using two different methods: ozonolysis of tetramethylethylene, photolysis of methylnitrate and photolysis of ozone in the presence of nitrogen oxide. The particle phase analysis revealed the compounds with the mass to charge ratios (*m/z*) 171, 185, 203 and 231 corresponding to terpenylic acid, pinic acid, 3-methyl-1,2,3-butanetricarboxylic acid and diaterpenylic acid acetate, respectively. The highest percentages of these compounds in SOA are observed in the presence of neutral particles, a dramatic decrease in mass percentage is observed with increasing particle acidity, suggesting the consumption of monomeric compounds through acid catalysed reactions. Higher SOA yields were also observed in the presence of NO_x than in the absence of NO_x. This is consistent with other monoterpene oxidation studies that the low HO_x/NO ratio leads to lower SOA yield.

P-Transformation.33 ID:4264 10:30

Secondary aerosol formation over urban atmosphere in the Indo-Gangetic Plain, northern India

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The chemical composition of ambient aerosols (PM2.5 and PM10) sampled during day-time (n=20) and night-time (n=14), spread over two weeks, provides an unequivocal evidence for the formation of secondary aerosols over an urban location (Kanpur) in the Indo-Gangetic Plain (IGP). The aerosol mass dominated by fine-mode particles (PM2.5/PM10 mass ratio = 0.46 – 0.86) and their chemical composition suggest that total carbonaceous aerosols (TCA = 1.6*OC + EC) account for ~50% of the PM2.5 mass; whereas mass fraction of water-soluble inorganic species (WSIS) is no more than 20%. The PM2.5 mass and concentration of carbonaceous species (EC, OC) show significant increase during night-time, indicating their efficient trapping due to decrease in boundary layer height and/or increase in the source strength. But the average WSOC/OC ratio is significantly higher in the day-time samples (0.66 ± 0.11) compared to that in the night-time (0.47 ± 0.07); suggesting increased contribution of secondary organic aerosol (SOA) during day-time. The formation of particulate NO3- increases by a factor of four during night-time; whereas fractional contribution of SO42- shows enhancement (~20%) during day-time. The particulate NH4+ neutralizes ~85% of the acidic species (NO3- and SO42-) in PM2.5. The contribution of TCA and WSIS was maximum (~75% of PM2.5) on 30th Oct 2008, marked by a thick haze formation (also supported by MODIS data). The formation of secondary aerosols over an urban polluted region has, thus, attracted much attention in recent years in view of their importance in degrading the air-quality, visibility, haze formation and enhancing the aerosol absorption and scattering properties.

P-Transformation.34 ID:4136 10:30

Identification of higher molecular weight carboxylic acids as important components of atmospheric organic aerosols

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Water-soluble organic compounds (WSOC) are found to be an important fraction of the total organic mass in atmospheric aerosols. It was suggested that WSOC are a mixture of polar oxygenated multifunctional compounds with different functional groups, like carboxylic acids as very important constituents. Studying the chemical diversity of the WSOC is essential for understanding the physico-chemical properties of atmospheric organic aerosols and their role in atmospheric processes. The present study was focused mainly

on the identification of higher molecular weight carboxylic acids (e.g. succinic, glutaric, adipic, pimelic, suberic, azelaic, maleic, fumaric, cis-pinonic, o-phthalic, etc.) in urban aerosols ($PM_{2.5}$, PM_{10}) using a novel Hydrophilic Interaction Chromatography – Negative Electrospray Ionization Tandem Mass Spectrometry (HILIC-(-)ESI-MS/MS) method. The separation of the acids was achieved on XBridge Amide BEH column (Waters) using gradient elution with mobile phase composed of 90/10 (v/v) acetonitrile/water mixture containing 10mM ammonium acetate (pH = 5) and 10 mM aqueous ammonium acetate (pH = 5). Using a Multiple Reaction Monitoring (MRM) and Neutral Loss (NL44, loss of CO_2) scanning MS/MS, several carboxylic acids were identified in the urban $PM_{2.5}$ and PM_{10} aerosols. Among them the most intense peaks were found for succinic, o-phthalic, maleic, azelaic and glutaric acid. Other detected carboxylic acids were pimelic, adipic, suberic and cis-pinonic acid. Using NL44 other peaks were also identified. Some of the detected acids also gave the same MRM transitions with their studied analogues, but had different retention times. For example, the isomers of o-phthalic acid (like m- or p-phthalic acids) gave the same MRM transition (m/z 165.0 → m/z 121.1).

P-Transformation.35 ID:4252 10:30

Slow aging in secondary organic aerosol observed by liquid chromatography coupled with high-resolution mass spectrometry

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Model biogenic secondary organic aerosol (SOA) prepared via ozonolysis of limonene has been observed to turn brown when exposed to small amounts of gaseous or aqueous NH_3. Our hypothesis is that the chromophoric compounds responsible for this color change are conjugated imines. These compounds are only present in relatively small amounts, hence standard mass spectrometry is insufficient to unambiguously detect these compounds. However, a combination of HPLC, UV-Vis detection and high resolution electrospray ionization mass spectrometry (HR-ESI-MS) allows assignments of chemical formulae to hundreds of components in aged SOA, including likely chromophores. Peaks corresponding to nitrogen-containing compounds elute simultaneously with compounds exhibiting peaks in the visible spectrum. LC-MS analyses, in combination with MS/MS analyses, provide extra dimensions of information that can resolve ambiguities in ESI-MS spectra.

P-Transformation.36 ID:4565 10:30

Secondary organic aerosol using the volatility basis set approach: Comparisons of 3-D model results with observations over the U.S during the summer of 2006

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Regional scale air quality forecast models generally under-predict the organic aerosol (OA) component of PM2.5 aerosol observed in urban and rural areas of North America using traditional, single-product approaches to organic vapor/aerosol phase partitioning from the oxidation of a given non-methane organic carbon (NMHC) compound. More recent parameterizations of secondary OA (SOA) combine laboratory measurements of SOA formation with a multi-product representation of NMHC oxidation and semi-ideal solution partitioning theory. The products of a given oxidized NMHC span a spectrum of volatilities, which are represented by a base set of vapor/aerosol equilibrium volatility concentrations. Applying this volatility basis set (VBS) approach within a 3-D Eulerian model framework requires some physical and chemical assumptions regarding the unknown SOA and semi-volatile vapors. This work examines the sensitivity of SOA for a well-documented VBS algorithm recently incorporated within the WRF/CHEM air quality model. Several key assumptions are examined through rigorous comparison of model SOA predictions to available surface network and aircraft observations collected during the summer of 2006. The surface comparisons include daily average measurements from the IMPROVE, STN and SEARCH networks located throughout the U.S. Aircraft observations of aerosol composition collected during the TexAQS/GoMACCS-2006 intensive field campaign test the model’s ability to capture the underlying mechanism responsible for observed rapid SOA formation from the Houston and Dallas urban centers. Predicted SOA, and the relative importance of biogenic versus anthropogenic SOA sources, are found to be very sensitive to assumptions regarding the dry deposition of SOA and semi-volatile vapors, the density of condensed SOA, and the assumed rates of photochemical aging of organic vapors from higher volatility to lower volatility species. Model simulations with grid sizes ranging from 4km to 60km show that predicted SOA is also sensitive to model horizontal resolution due to inherent nonlinearities within the semi-ideal solution assumption. The larger goal of improving PM2.5 forecasts is also discussed in terms the VBS sensitivity analysis and comparisons with the North American PM2.5 AIRNow network administered by the U.S. EPA.

P-Transformation.37 ID:4180 10:30

Laboratory studies on optical properties of secondary organic aerosols generated during the photooxidation of toluene and the ozonolysis of alpha-pinene

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Recently researches suggested that some of organic aerosols can absorb solar radiation especially at the shorter visible and UV wavelength. Although quantitative characterizations of optical properties of secondary organic aerosols (SOAs) are required in order to confirm the effect of SOAs on atmospheric radiation balance, the light absorption of SOAs has not been well studied so far. In this study, controlled laboratory experiments to measure optical properties of SOAs generated during the photooxidation of toluene in the presence of NOx and the reactions of alpha-pinene with O3 have been conducted. Extinction and scattering coefficients of the SOAs were measured by a two wavelength custom-built cavity ring-down aerosol extinction spectrometer and a three wavelength integrated nephelometer, respectively. Refractive indices of the SOAs were determined so that the measured particle size dependence of the extinction and scattering efficiencies were reproduced by calculations using Mie scattering theory. As a result, significant light absorption was found for the SOAs generated during the photooxidations of toluene in the presence of NOx, and light absorption at 355 nm was larger than that at 532 nm. On the other hand, no significant absorption was found both at 355 and 532 nm for the SOAs generated during the reactions of alpha-pinene with O3. Using the obtained refractive indices, mass absorption cross section values of the toluene SOAs with diameters of 100-600 nm are calculated to be 1.0-1.8 m2g-1 at 355 nm and 0.1-0.2 m2g-1 at 532 nm. The results indicate that light absorption by the SOAs formed from the photooxidation of aromatic hydrocarbons have a potential contributing to the radiation balance especially at UV wavelength.

P-Transformation.38 ID:4133 10:30

How efficient is secondary organic aerosol formation from glyoxal in aqueous particles and clouds?

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Traditional models describe secondary organic aerosol (SOA) formation by condensation of low-volatility or semivolatile gas species. Over the past decade, the formation of SOA mass in cloud droplets has been identified as a potentially significant contribution to the total SOA budget. Even more recently, it has been shown that the aqueous phase of particles might act as a reactor for efficient reactions of glyoxal leading to considerable SOA amounts. However, to date none of these aqueous phase SOA sources is included in most models due to the lack of suitable parameterizations. An air parcel model is used in order to compare the efficiency of SOA formation in these two aqueous regimes. It will be shown that under a wide range of conditions - in terms of cloud contact time, cloud liquid water content, and ambient relative humidity at subsaturated conditions - approximately equal amounts of SOA from glyoxal are formed in cloud droplets and aqueous particles despite the much smaller water content of the latter. The different chemical mechanisms in concentrated particles and dilute cloud droplets will be discussed in order to explain the reasons for the similarity of the SOA yields.

Based on the parcel model results, SOA yields from glyoxal in clouds can be expressed as a function of cloud liquid water content and contact time (cloud fraction). In a similar way, SOA yields in aqueous particles can be parameterized using the ambient relative humidity and aerosol loading as a proxy for the total water amount.

P-Transformation.39 ID:4164 10:30

Ozone production potential of volatile organic compounds: a new approach

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The role of VOC (Volatile Organic Compounds) in the photochemical formation of of ozone has been recognised since the 1950s. The need to improve this understanding of the chemistry involved in the production of photochemical smog has been a major driving force in the development of numerical models of atmospheric chemistry for the last several decades. Such models are now used routinely to provide operational forecasts of air quality, and have helped in the regulation of the emissions of ozone precursor substances, including VOC. The current state-of-the- art for determining the different abilities of different kinds of VOC to form ozone is the use of so-called incremental reactivity techniques. These techniques involve performing several model runs in which the concentrations of the VOC of interest are slightly perturbed, and relating these perturbations to the change in the modelled ozone production. In this poster, modelled ozone production potentials of VOC calculated using new analysis techniques which do not require a different perturbation run for each individual VOC, but rather calculate all ozone production potentials from a single model run are presented. The technique also provides more information about the mechanistic pathways of ozone formation than is available using traditional incremental reactivity techniques. A particular strength of this new technique is the ability to compare different chemical oxidation mechanisms.

P-Transformation.40 ID:4161 10:30

VOCs and SVOCs Reduction by Heterogeneous Nanomaterials

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Anthropogenic volatile organic compounds (VOCs) and semi volatile organic compounds (SVOCs) are emitted from various sources including vehicles (as unburnt hydrocarbons), industrial applications, solvent and detergent use and wood combustion. They contribute to outdoor air pollution by photo-oxidation in the presence of NOx to form tropospheric ozone, a smog precursor. VOCs and SVOCs affect the climate as they are precursors to secondary organic particles affecting cloud-particle interactions. VOCs also contribute to indoor air pollution, which is a nuisance to health. Elimination of those pollutants can be achieved by using demanding techniques (high temperatures, expensive catalysts and/or high energy requirements). We develop an easily manufactured, inexpensive and adaptable (no extra energy required, operating temperature <100 C, green adsorbents) technique involving nanomaterials for the recovery by adsorption of VOCs and SVOCs from gas streams. We will discuss our results regarding the efficiency of our constructed fibers in air circulation in comparison to existing filters for the removal of major VOCs components.

P-Transformation.41 ID:4159 10:30

Application of an alternative method for measuring carbohydrates in routine filter samples collected by the FRM network

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Biomass burning is one of the major sources of organic carbon aerosols. The most common method used in order to try to determine the contribution of biomass burning to the total organic carbon aerosol is through the use of smoke marker measurements. However, the traditional methods used to measure the smoke marker levoglucosan, an anhydrosugar produced during the combustion of cellulose, are quite labor intensive and expensive due to the solvents, reagents, and instrumentation needed. Therefore, these analyses are generally conducted only for selected filter samples from special studies or on composited filter samples. This prevents one from understanding the impact of biomass burning both spatially and temporally. Therefore, an alternative method to measure levoglucosan in aerosol samples has been developed using high-performance anion-exchange chromatography with pulsed amperometric detection (HPAEC-PAD). Along with levoglucosan, this method is able to simultaneously measure other carbohydrates including sugar alcohols/polyols (e.g., mannitol) and sugars (e.g., glucose).

As part of the Federal Reference Monitor (FRM) network a daily 47 mm Teflon filter is collected routinely to obtain a measurement of the total PM2.5 mass. Once this measurement is completed then the entire filter is archived, generally refrigerated and in a Petri dish, providing a filter sample that can then be analyzed by additional methods.

Here we will present results from the application of HPAEC-PAD analysis on one year of archived FRM filters from 10 sites in the Upper Midwest. Data for levoglucosan and various other carbohydrates along with WSOC (water-soluble organic carbon) will be shown. All of this data will be used to investigate the spatial and temporal trends as well as the sources of these various species. A determination for the impact of biomass burning to the total organic carbon concentration in this region will also be provided.

P-Transformation.42 ID:4329 **10:30**

Molecular compositions and transformations of dicarboxylic acids, ketoacids, α-dicarbonyls in marine aerosols over the western North Pacific during long- range transport

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A rapid industrial development in China and East Asian countries for last two decades may have seriously changed the air quality of the North Pacific. To better understand a long-term atmospheric changes of organic aerosols in the western North Pacific, we collected marine aerosols on weekly basis at a remote island, Chichijima (27°04'E; 142°13'N) in 2001-2009. The island is located in the boundary of westerly and easterly wind regimes. The aerosol samples were analyzed for dicarboxylic acids, ketoacids and α-dicarbonyls employing butyl ester derivatization followed by GC determination, together with total carbon (TC) and water-soluble organic carbon (WSOC). Homologous series of saturated diacids (C2-C11) were detected with a predominance of oxalic (C2) acid followed by malonic (C3) and succinic (C4) acids. Unsaturated diacids, including maleic (M), fumaric (F), phthalic acids, were also detected together with ketoacids and α-dicarbonyls. Concentrations of total diacids fluctuated significantly in a range of 10-600 ngm-3 with winter/spring maximum and summer minimum. The maximum was explained by a combination of enhanced emissions of polluted aerosols and their precursors in Asia and enhanced atmospheric transport to the North Pacific due to the intensified westerly winds. Concentration ratios (range 0.2-28, av. 2.8) of C3 to C4 acid showed a maximum during summer, indicating more oxidation of longer-chain diacids to shorter ones. Azelaic acid (C9) that is a specific photo-oxidation product of unsaturated fatty acid such as oleic acid showed a sharp increase relative to other diacids in summer, suggesting enhanced sea-to-air emission of unsaturated fatty acids followed by photochemical oxidation during summer. On the other hand, M/F ratios (range 0-8.7, av. 1.1) significantly decreased from winter to summer due to photochemical cis-to-trans isomerization. We also discuss long-term trends in the concentrations of diacids and related compounds as well as TC and WSOC.

P-Transformation.43 ID:4483 **10:30**

A secondary organic aerosol scheme in 10 reactions: the no frills approach to global aerosol modelling.

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Box modellers produce large chemical mechanisms to describe the atmosphere explicitly. Whilst large schemes are useful for representing the detail of the chemistry their computational expense means they cannot be used to simulate the global scale.

We have designed a secondary organic aerosol (SOA) scheme which describes the production of 1 generic lumped anthropogenic, and 1 generic lumped biogenic condensable product in just 10 reactions. It is derived from simulations of the highly explicit Master Chemical Mechanism (~13500 reactions) and its smaller relative, the CRIv2-R5 (~550 reactions) which fill chemical composition space. A parameterisation of the key processes producing our chemical species is optimized within this chemical composition space. Under the range of conditions simulated (from clean to very polluted), we find our parameterized SOA scheme sits within 20% of the two larger scheme's results.

We have incorporated our SOA scheme into TOMCAT/GLOMAP, our global aerosol microphysics and chemical transport model. We present global fields of anthropogenic and biogenic SOA from this study,

which compare favourably with current global estimates for SOA burdens.

P-Transformation.44 ID:4218 10:30

Identification of biogenic SOA based on NMR spectral signatures observed in chamber experiments and at pristine forested environments

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During the last years major efforts have been made to understand secondary organic aerosol (SOA) formation by the oxidation of biogenic volatile organic compounds. Nevertheless current estimates of global SOA production remain extremely uncertain due to the lack of observations capable to discern the biogenic SOA from other oxidized organic aerosol types. In the present study new results are shown on the chemical characterization of biogenic SOA produced during photo-oxidation and ozonolysis of terpene mixtures representative for VOCs emitted by conifer tree species. Reaction chamber experiments were conducted in the frame of the EUCAARI and PolySOA projects in the SAPHIR and PSI simulation chambers. Two subsequent samplings were performed in different time intervals of each experiment to compare fresh and aged SOA. The water extracts of the collected SOA samples were analyzed by proton nuclear magnetic resonance spectroscopy (1H-NMR) and the resulting data were further elaborated with multivariate statistical methods. Positive matrix factorization (PMF) has been applied to the 1H-NMR spectra of biogenic SOA generated in laboratory, providing basically two factors associated to fresh and aged SOA. These laboratory experiments provided spectral signatures that were helpful for the interpretation of field data. In fact, a series of NMR spectra recorded for submicron aerosol particles collected in pristine forested environments dominated mainly by conifers was also subjected to PMF analysis and the resulting factors were compared to those obtained for laboratory biogenic SOA. Among the NMR factors extracted by applying PMF to a series of samples collected at the Finnish forest station of Hyytiälä, one factor was found to fit unambiguously with the factor associated to aged biogenic SOA obtained in reaction chambers. These findings suggest that 1H-NMR spectroscopy combined to statistical multivariate analysis can be profitably employed to apportion the bulk organic composition of atmospheric aerosol into its source contributions.

P-Transformation.45 ID:4163 10:30

Impact of convection on air quality during the HOOVER/COPS/TRACKS campaign

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We present a detailed analysis of the impact of convection on air quality and the redistribution of trace gases within the vicinity of a severe storm by using the WRF-chem model. Deep convective clouds can have a major impact on atmospheric chemistry as they can rapidly transport trace gases from the lower to the upper troposphere. At the same time, highly soluble species are efficiently scavenged due to uptake on

hydrometeors and subsequent removal by precipitation. Furthermore, deep convection can influence atmospheric chemistry by changing UV fluxes and thus photochemical reaction rates as well as by the formation of NO molecules via lightning. All of these processes change chemical reaction pathways in the troposphere. As part of the HOOVER/COPS/TRACKS campaign aircraft measurements of trace gases were made in the outflow region of a severe convective cell which developed ahead of a frontal system over Germany on 19 July 2007. We focus first on understanding the main meteorological characteristics of the storm system, in particular its initiation and the rapid development of the precipitating cell. This case study will then be employed to investigate the removal of pollutants from the atmosphere by scavenging processes. Furthermore, we focus our case study on modifications of chemical reaction pathways in the upper troposphere due to upward transport of boundary layer tracers as well as the release of trace gases such as H2O2 from ice particles.

P-Transformation.46 ID:4393 10:30

Analysis of air quality trace gas spatio-temporal variability over the USA using the WRF-chem regional model

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Ozone and carbon monoxide play key roles in the photo-chemical processes occurring in the atmosphere, with strong consequences for tropospheric chemistry, air quality and climate. Both molecules are highly variable, particularly in the troposphere, and need to be accurately monitored in order to provide a better insight into, for example, pollution episode development and pollution transport on regional to global scales.

In this work, we analyse ozone and carbon monoxide concentrations simulated by the WRF-chem regional model over the USA in order to characterize their geographical and temporal variations at the surface, in the lowermost troposphere and in the free troposphere along with their spatio-temporal correlations. Model sensitivity studies are used to differentiate the variability due to dynamics, photochemistry and emissions. We also present comparisons with surface measurements available from several ground-based stations over the USA.

P-Transformation.47 ID:4271 10:30

Improving Santiago's air quality network design for better capturing dispersion patterns of a passive tracer

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Addressing the question of how to best sample airborne pollutants in order to evaluate the fulfillment of air quality monitoring objectives is non trivial. Santiago is the main urban center of Chile, allocating ca. 40% of its population, which is oftentimes subject to poor air quality conditions. Santiago's air quality network was primarily conceived to address population's exposure to acute air pollution in the late 1980's. Today it is still largely devoted to evaluate the compliance or not of air quality standards, particularly those referred to inhalable particulate matter. But as acute episodes have become less frequent over the last 20 years, more

attention is being paid to address more subtle and typically elusive objectives. In this work, we use a least square approach, combined with simulated annealing and other optimization methods to improve Santiago's network for addressing the meso-scale dispersion of carbon monoxide. This methodology allows comparing different observing configurations. In particular, we evaluate the effects of adding and removing stations according to the history of Santiago's network.

P-Transformation.48 ID:4320 10:30

Air Quality during the 2008 Beijing Olympics: Secondary Pollutants and Regional Impact

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We present the first results of the atmospheric measurements of trace gases and aerosols at three surface sites in and around Beijing before and during the 2008 Olympics. We focus on secondary pollutants including ozone, fine sulfate and nitrate and the contribution of regional sources in summer 2008. The results reveal different responses of the secondary pollutants to the control measures from primary pollutants. Ambient concentrations of vehicle-related nitrogen oxides (NOx) and volatile organic compounds (VOCs) at an urban site dropped sharply in the first two weeks after full control was put in place, but the levels of ozone, sulfate and nitrate in PM2.5 increased significantly, compared to the period prior to the full control; wind data and back trajectories indicated the contribution of regional pollution from the North China Plains. Air qualities (for both primary and secondary) improved significantly during the Games, which was also associated with the changes in weather conditions (prolonged rainfall, decreases in temperature, and more frequent air masses from clean regions). A comparison of the ozone data at three sites in eight episodes, when the air masses were from the southeast-south-southwest sector, showed that regional pollution sources contributed 34%-88% to the peak ozone concentrations in urban Beijing. Ozone production efficiencies at two sites were low (~3 ppbv/ppbv), indicating that ozone formation was being controlled by VOCs. Compared with data from 2005 at a downwind site, the concentrations of ozone (O3), sulfur dioxide (SO2), total sulfur (SO2+PM2.5 sulfate), carbon monoxide (CO), reactive aromatics (toluene and xylenes) sharply decreased (by 15-62%) in 2008, but there were no significant changes in the concentrations of PM2.5, fine sulfate, total odd reactive nitrogen (NOy), and longer lived alkanes and benzene. We suggest that these results indicate the success of the government's efforts in reducing emissions of SO2, CO, and VOCs in Beijing. However, further control of regional emissions is needed for significant reductions of ozone and fine particulate pollution in Beijing.

P-Transformation.49 ID:4506 10:30

EUROCHAMP-2 – A European Infrastructure Project for Atmospheric Simulation Chambers – Current Progress and Future Directions

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Since June 2004 the European Commission in Brussels is funding the Integrated Infrastructure Initiative EUROCHAMP (http://www.eurochamp.org), whose overall goals are: - the integration of existing environmental reaction chambers into a Europe-wide infrastructure - the initiation of an effective interdisciplinary collaboration between the community of atmospheric scientist and colleagues from other disciplines (e.g. cultural heritage protection and human health). In addition to the development of e.g. standardized data protocols, a central database for environmental chamber studies and instrument

intercomparisons, the project includes two so-called joint research activities (JRA). The JRAs are dealing with the development and refinement of analytical equipment (e.g. characterization of oxygenated VOC‘s, radical measurements, nitric acid measurements, characterization of aerosols) and the improvement of chemical modeling techniques (techniques for the evaluation and development of oxidant mechanisms and models, techniques for the evaluation and development of aerosol models, development and deployment of statistical tests and sensitivity analysis methods). The presentation will review the current progress of the project in particular with respect to aforementioned joint research activities.

P-Transformation.50 ID:4563 10:30

Diurnal and seasonal variation of the thermal inversion layer for its application to the study of the atmospheric pollution on the city of Metropolitan Lima

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In the present work is characterized the diurnal and seasonal variation of the thermal inversion layer on Lima city. This characterization was made in two periods of radiosonde data, the first period was of 14 years (1992-2005) at 12 UTC, and the second period was of two months (February and August 2009), where we made 3 releases per day (12, 18 and 00 UTC) only for 15 days each month. Also, is reported the concentration of pollutans SO2 and Nox.

The results indicate that the thermal inversion layer on the city of Lima has a large seasonal variability, since during the summer months the height of this layer is below of the 500m, while in the winter months the values are about 800m, being this one of the main factors that influence the ascent or descent in the concentration of pollutants emitted into the atmosphere. This seasonal varation is mainly due to the influence of the South Pacific Anticyclone and other climatic factors.

P-Transformation.51 ID:4582 10:30

Scavenging of nitrates by frozen precipitation: Impacts on the sensitivity of upper tropospheric O3 to variations in convection and lightning NOx

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Precipitation scavenging of nitrates provides the primary removal mechanism for reactive nitrogen (NOx) in the lower troposphere. In the upper troposphere, removal of NOx via scavenging of HNO3 and HNO4 by frozen precipitation has the potential to significantly impact O3 production. Simulations with the UC Irvine global chemistry transport model (CTM) using observed ice uptake rates for HNO3 show that scavenging of nitric acid by frozen precipitation decreases global mean O3 by ~3 DU relative to a simulation with no ice uptake. Here, we present simulations that examine how ice scavenging of nitrates impacts the sensitivity of tropospheric ozone to variations in convection and lightning NOx. We show, for example, that ice scavenging of HNO3 reduces the sensitivity of O3 to a 25% perturbation in NOx production per lightning flash by more than 35% over large regions. We compare the results from the UCI CTM to results from the CAM-Chem coupled chemistry-climate model, and discuss implications for how future changes in lightning and convection might impact tropospheric O3.

P-Transformation.52 ID:4267 10:30

Influences of In-Cloud Aerosol Scavenging and Cloud Processing on Aerosol Concentrations and Wet Deposition in ECHAM5-HAM

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In-cloud scavenging and cloud processing strongly influence global aerosol three-dimensional distributions. This study uses the ECHAM5-HAM GCM to compare three in-cloud scavenging approaches that are commonly used in global models: prescribed scavenging fractions, scavenging diagnosed from the cloud microphysical parameters, and a prognostic aerosol cloud processing approach. We show that predicted aerosol concentrations in the middle and upper troposphere differ by up to one order of magnitude between the various in-cloud scavenging schemes, with the greatest changes for black carbon. The predicted global and annual mean aerosol burdens change by up to 50% between the scavenging parameterizations. The relative contributions of cloud nucleation and impaction scavenging processes are quantified. Impaction scavenging is shown to account for near to 50% of the annual and global mean deposition attributed to in-cloud scavenging for dust. We examine the influence of aerosol cloud processing in both stratiform and convective clouds on the predicted aerosol three-dimensional distributions.

P-Transformation.53 ID:4321 10:30

Spatio-temporal variability of atmospheric NH_4^+, NO_3^-, and SO_4^{2-} concentrations over Indian region

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In the present-day scenario of growing anthropogenic emissions, the aerosol abundances of NH_4^+, NO_3^-, and SO_4^{2-} (formed from their gas-phase precursors) have potential to introduce non-linearity in several of the atmospheric chemical processes. The spatio-temporal variability in the concentration of these anthropogenic constituents has been studied during wintertime from an urban (Hisar; HSR) and remote high-altitude sites (Mt. Abu; MTA and Manora Peak; MP) of north-western India as well as over MABL of Bay of Bengal (BoB). On average, abundances of NO_3^-, SO_4^{2-} and NH_4^+ (13.3, 12.7 and 6.3 µg m^{-3} respectively) within the urban source region (HSR) are 2 to 5 fold higher compared to those at high-altitude sites (MTA, MP) and over MABL. The NO_3^-/nss- SO_4^{2-} weight ratio of 1.07 at HSR is significantly higher than that at remote sites (MTA = 0.29, MP = 0.23) and BoB (0.10). The decrease in NO_3^-/nss- SO_4^{2-} ratio, away from the source region, could be attributed to preferential removal of NO_3^- (associated with coarser dust particle) during the long-range transport. Likewise, equivalence ratio of NH_4^+/ nss- SO_4^{2-} are also higher over urban region (HSR =1.22) in contrast to relatively lower ratios at MTA (0.76), MP (0.39) and BoB (0.56); suggesting the efficient neutralization of SO_4^{2-} by NH_4^+ over urban region. A lack of correlation among NH_4^+ and NO_3^- (over remote sites and MABL) suggests association of the latter with mineral dust and sea-salt. The nature and influence of atmospheric transport processes, at temporal and spatial scales in the tropical regions, needs systematic evaluation in order to integrate observations and modelling capabilities.

P-Transformation.54 ID:4198 10:30

Total OH reactivity of vehicular exhaust with a chassis dynamometer

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Total OH reactivity for exhaust gas of gasoline vehicles was measured under nine different driving conditions with a chassis dynamometer at the National Institute for Environmental Studies. Analyses of trace species including 50 kinds of VOCs and NOx as well as OH reactivity were carried out. The chemical compositions of vehicular exhaust were found to depend on the temperature of the engine or catalysts. For all driving cycles, the calculated OH reactivity was confirmed to be underestimated, implying the existence of unknown species in the exhaust gas. The percentage contribution of OH reactivity to the unknown species during "Cold" start was about 17.5%, which was almost the same as that for "Hot" start at 17.0%. However, the absolute value of OH reactivity for "Cold" start was about ten times higher than that for "Hot" start.

P-Transformation.55 ID:4332 10:30

Modelling Atmospheric OH-reactivity

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As the focus on climate, air quality, visibility and public health has increased, so has the research on secondary organic aerosol (SOA) formation. SOAs are formed from atmospheric oxidation of volatile organic compounds (VOCs). The hydroxyl radical (OH) is the most important oxidant of the atmosphere, and knowledge of the atmospheric concentration and lifetime of OH is necessary to further the understanding of the SOA formation. However, the overall OH sink term is currently poorly constrained in models in the absence of direct measurements of the OH radical and its total OH-reactivity. A better understanding of the OH-reactivity is needed. We have modelled the atmospheric OH-reactivity in a boreal forest and investigated the contributions from atmospheric inorganic species, methane, isoprene, monoterpenes and other important VOCs. We have used SOSA; a one dimensional vertical chemistry-transport model together with measured data from Hyytiälä, SMEAR II station, Southern Finland, August 2008. In order to ascertain how well we understand the OH initiated photochemical processes, we have compared our calculated OH-reactivity with measured ambient OH-reactivity from the BFORM (Boreal Forest OH Reactivity Measurements) campaign, August 2008. Here the total atmospheric OH-reactivity was measured using the the Comparative Reactivity Method (Sinha et al., 2008, ACP). We found that the simulated OH-reactivity underestimates the measurements by about 30%. This was expected due to the thousands of unknown organic compounds not included in our model simulations. In this work we will show how much different individual compounds contribute to the total measured OH-reactivity. Reference: Sinha, V., Williams, J., Crowley, J. N. & Lelievold, J. (2008). Atmos. Chem. Phys, 8, 2213-2227.

P-Transformation.56 ID:4305 10:30

Hydroxyl radical in the troposphere: lessons learned from methyl chloroform simulations

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We have used an AGCM-based Chemistry Transport Model (ACTM) for the simulation of methyl chloroform (MCF; CH_3CCl_3) and methane (CH_4) in the height range from the earth's surface to about 90 km. Tropospheric loss rates are calculated based on two sets of hydroxyl (OH) fields: (1) Spivakovsky et al. (2000), manually adjusted for MCF simulation by the transport model (TM5) for the period 2000-2006, (2) ACTM, manually adjusted for MCF decadal trends. Loss rates due to photolysis and chemical reactions (with Cl and O^1D) were parameterized in the ACTM based on JPL Publication 06-2 (Sander et al., 2006) for appropriate absorption cross-sections and reaction rates. Surface emissions for the period of simulations (1988-2008) are constructed using the EDGAR3.2 emission distribution and global totals from McCulloch and Midgley (2001). The model results are compared with more than two decades of surface observations from the NOAA cooperative network and the multi-institutional AGAGE network, and campaign based balloon-borne vertical profiles.

The Pearson's moment correlations for model and observed time series of separated synoptic and seasonal variations are found to be generally greater than 0.3 (N~350) and 0.6, respectively, for individual years in the period of 1990-2007 at 5 AGAGE sites (Mace Head, Trinidad Head, Ragged Point, Cape Matatula, Cape Grim), where continuous measurements are available. This comparison suggests that the MCF emission inventory and both the OH distributions in the troposphere are valid. The balloon-borne measurements of vertical profiles in the stratosphere from Hyderabad and southern France, on an event basis, are also well captured by the ACTM simulations, suggesting a realistic representation of MCF photochemistry and troposphere-stratosphere transport. Note that the global total MCF emissions decreased from 715 Gg in 1990, rapidly to 102 Gg in 1996, and then slowly to 5 Gg in 2008. The ACTM simulations show that MCF is transported from the northern (NH) to southern (SH) hemispheres through the upper troposphere in the first half of the 1990s when the NH emissions were large compared to SH emissions. The equilibration in MCF concentrations between the two hemispheres in the 2000s helps to explain the temporal evolution in MCF concentrations, e.g., the difference between Mace Head and Cape Grim, decreased from about 50 ppt in 1991 to none in 2006.

The observed 50% decrease in the MCF concentration differences between the Barrow and Mauna Loa flask observation sites in 1996 successfully tracks the largest MCF emission decrease of from 241 Gg in 1995 to 102 Gg in 1996. Further, the concentration differences simulated using Spivakovsky/TM5 OH appear to be in much closer agreement with the observed differences while the simulation using the ACTM OH tends to underestimate by a fraction of a ppt. This underestimation of MCF concentration difference for the simulation using ACTM OH is demonstrated clearly in the differences between the Alert and South Pole sites, which are the furthest apart in terms of latitude. The NH/SH ratio of OH concentration for Spivakovsky/TM5 OH is 0.98 and it is 1.25 for the ACTM OH. The higher OH loss of MCF in the NH for the simulation using ACTM OH results in the weaker inter-hemispheric gradient in MCF concentrations, even though both the OH fields are adjusted for simulating the MCF decadal growth rates successfully.

Our results suggest that the NH/SH ratio as well as the absolute concentration of OH can be estimated using MCF simulations, and this impacts on the simulations of other tropospheric species, such as methane and carbon monoxide.

P-Transformation.57 ID:4146 10:30

The role of hydrocarbons on free tropospheric HO_x.

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The hydroxyl (OH) radical is the major oxidizing agent in the troposphere and is responsible for the destruction of most hydrogen-containing gases. Over the past decade it has emerged that measured HO_x in the upper troposphere exceeds that predicted by models and that a range of HO_x precursors may contribute to this discrepancy. Such precursors include formaldehyde, alkyl peroxides, acetone and isoprene derivatives which are thought to be transported from lower altitudes. A global chemistry transport model (STOCHEM) is employed with a reduced form of the common representative intermediates mechanism (CRIv2 – R5). This common representative intermediates mechanism was created using the MCM v3.1 as a reference benchmark. The reduced mechanism contains 196 species competing it 555 photochemical reactions.

The contribution of formaldehyde to free tropospheric HO_x is investigated. New laboratory photolysis parameters for formaldehyde have shown that the radical channel of photolysis is a more important pathway for the oxidation of formaldehyde. Photolysis via the radical channel leads to two hydroperoxy (HO_2) radicals being produced. The effect of faster radical channel photolysis for formaldehyde is investigated as a potential source of free tropospheric HO_x. An increase of up to 5 % in OH concentration is modelled for the upper troposphere.

High levels of OH have been measured in forested regions co-located with high emissions of isoprene. Current modelling studies predict low OH levels in the presence of high emissions of isoprene. The possibility of HO_x recycling during isoprene oxidation has recently been proposed. A parameterized OH recycling mechanism has been included in STOCHEM leading to an increase of up to 30 % in OH concentration.

P-Transformation.58 ID:4345 10:30

HOx regeneration in isoprene oxidation: global model evaluation of unexpected reaction sequences

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There is now overwhelming evidence that the traditional isoprene chemical oxidation mechanism used in CTMs requires substantial revision. Lelieveld et al. (Nature, 2008) showed that strong OH regeneration must be invoked in order to explain observed HOx levels in isoprene-rich, pristine conditions, and suggested that it occurs through the reactions of isoprene peroxy radicals with HO2. Paulot et al. (Science, 2009) showed in a laboratory study that the OH-reaction of isoprene hydroperoxides generates OH along with dihydroxy epoxides possibly contributing to secondary organic aerosol formation. However, measurement campaigns at mid-latitudes indicate that HOx regeneration occurs even at mid-latitudes (Hofzumahaus et al., Science, 2009; Ren et al., J.Geophys.Res., 2008) where isoprene peroxy radicals are expected to react mostly with NO. A likely explanation is provided by the theoretical study of Peeters et al. (Phys.Chem.Chem.Phys., 2009), which showed that isomerization reactions of specific isomer/conformer peroxy radicals from isoprene lead to the formation of HOx radicals and photolabile hydroperoxy aldehydes (denoted HPALDs), at rates such that the traditional reactions of the isoprene peroxy radicals (with NO and HO2) are outrun in most non-urban atmospheric conditions. We present a detailed global modelling study aiming at quantifying the impact of the new reactions proposed by Paulot et al. and Peeters et al. on the global tropospheric composition. The calculated concentrations of key compounds will be evaluated against measurement campaigns at Tropical (e.g. GABRIEL) and mid-latitudes (INTEX-A, PROPHET), and sensitivity simulations will be performed to estimate the remaining uncertainties.

P-Transformation.59 ID:4522 10:30

To what Extent is Isoprene Oxidation a Sink for Hydroxyl Radicals?

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The large terrestrial emissions of isoprene have been assumed to result in a substantial depression of hydroxyl radical (OH) levels, in particular over the tropical forests. The GABRIEL field campaign in 2005 revealed that the forest sustains high hydroxyl radical concentrations, being about 5e6 molec/cm3 on average. Atmospheric chemistry models (box and 3D) strongly underestimated the OH concentrations, showing a lack of knowledge about the oxidation mechanism of isoprene. These results sparked experimental and theoretical work providing valuable new insights. The new results have been implemented in our comprehensive isoprene oxidation mechanism, reconciling the model calculations with OH observations during GABRIEL. Our model predicts that isoprene oxidation could have an OH recycling efficiency close to 100% under low-NOx conditions. A budget analysis of the oxidation mechanism supporting this intriguing result will be shown. Under moderate NOx conditions the model results significantly differ from the ones of traditional oxidation mechanisms. The impact of these new results on the atmospheric composition will be assessed using a global model, with a special focus on the production and recycling of hydroxyl radicals.

P-Transformation.60 ID:4295 10:30

Investigating the impacts of HOx recycling in the oxidation of isoprene: Sensitivity studies of past, present and future atmospheres using the UKCA model.

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For several decades now the importance of isoprene to global atmospheric chemistry has been highlighted. However, our ability to understand many of the fundamental processes involving emissions and oxidation of isoprene remain questionable. In particular recent evidence suggests that the current understanding of low NOx oxidation of isoprene is poorly represented in numerical models. In this work we present results from a range of sensitivity experiments that focus on exploring the effects of inclusion of recent mechanistic changes, based on laboratory and theoretical studies, concerning the oxidation mechanism of isoprene. We start with a mini survey of some of the most commonly used isoprene mechanisms and note that with a range in complexity from 25 to over 500 reactions, all mechanism perform poorly under conditions of low NOx. We show that based on our current knowledge, intermolecular reactions of the isoprene hydroxy-peroxy

radicals are the most favourable route to novel chemistries for recycling HOx radicals in isoprene oxidation. We focus our main simulations on using the UKCA chemistry climate model to which a modified isoprene mechanism has been developed. The UKCA model is a state of the art coupled chemistry, aerosol and climate model which uses the UK Met Office Unified Model at its core. The results of sensitivity studies concerning climate scenarios relevant to the pre industrial, present day and future are presented. The results suggest that inclusion of a HOx recycling mechanism from the oxidation of isoprene has significant effects to the modelled levels of HOx, more than doubling the levels of HOx in regions which are currently much lower than observations suggest. The effects of increased HOx are presented in terms of their impact on more long lived and climatically important gases such as O3, CO and CH4 and discussed in terms of local and global scales.

P-Transformation.61 ID:4405 10:30

The phase dependency of atmospheric sulphate production

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Sulphur is a major component of the atmospheric aerosol. However, the natural and anthropogenic emissions are usually in the form of SO2 (S(IV)) or SO2 precursors that are quickly converted to sulphur dioxide by multiphase chemical processes. The oxidation step from S(IV) to S(VI), required to form sulphate aerosols, is relatively slow in the gas phase (at least in the lower troposphere), such that the chemical conversion in the other phases is more important. In this study we analyse the importance of gas, aerosol and cloud phase oxidation with the help of a global atmospheric chemistry general circulation model with new and relatively detailed descriptions of the atmospheric aerosol and cloud chemical processes.

P-Transformation.62 ID:4588 10:30

Laboratory Study of Photochemical Properties of Atmospheric Pollutants.

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One of the most important parameters in estimating the environmental impact due to emission of a compound is its residence time in the atmosphere, which is driven by the reaction of a compound with hydroxyl radicals (OH) for many atmospheric trace gases. The atmospheric lifetime is important for estimating ozone depletion potential (ODP) and global warming potential (GWP) of industrial compounds which are needed for evaluation of their environmental impact and regulatory purposes. The sources of critically evaluated photochemical data for atmospheric modeling, NASA/JPL Publications and IUPAC Publications, recommend uncertainties within 10%-60% for the majority of OH reaction rate constants with only a few cases where uncertainties lie at the low end of this range. These uncertainties can be somewhat conservative because evaluations are based on the data from various laboratories obtained during the last few decades. Nevertheless, even the authors of the original experimental works rarely estimate the total combined uncertainties of the published OH reaction rate constants to be less than ca. 10%. Thus, uncertainties in the photochemical properties of potential and current atmospheric trace gases obtained under controlled laboratory conditions still constitute a major source of uncertainty in estimating the compound’s environmental impact. The purposes of the present work was to illustrate the potential for obtaining accurate

laboratory measurements of the OH reaction rate constant over the temperature range of atmospheric interest. We will provide results of such higher-accuracy measurements obtained for various groups of halogenated compounds. Accurate IR absorption measurements of potential greenhouse gases will be discussed.

P-Transformation.63 ID:4335 **10:30**

Some Limitations of MM5 Mesoscale Meteorological Model for Air Quality Applications

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The performance of MM5 mesoscale model using different planetary boundary layer (PBL) and land surface model (LSM) parameterizations is evaluated and compared using high temporal and spatial resolution GOTE2001 campaign data at local scale over the Greater G€oteborg area along the Swedish west coast during 7–20 May 2001. The focus is on impact of PBL and LSM parameterizations on simulated meteorological variables important for air quality applications such as global radiation, diurnal cycle of near-surface air temperature and wind, diurnal cycle intensity, near-surface vertical temperature gradient, nocturnal temperature inversion,boundary layer height, and low-level jet. The model performance for daytime and nighttime and under different weather conditions is also discussed. The purpose is to examine the performance of the model using different PBL and LSM parameterizations at local scale in this area for its potential applications in air quality modeling.

The results indicate that the influence of PBL and LSM parameterizations on simulated global radiation, diurnal cycle of near-surface air temperature and wind speed, diurnal cycle intensity, vertical temperature gradient, nocturnal temperature inversion and PBL heights, which are critical parameters for air quality applications, is evident. Moreover, the intensity and location of LLJ are simulated well by all schemes, but there also exist some differences between simulated results by using different PBL and LSM schemes. Therefore, the choice of PBL and LSM parameterizations is important for MM5 applications to air quality studies.

Also, the sensitivity of sea breeze simulations to combinations of PBL and LSM parameterizations in the model for an observed SB case over this area is investigated. The results show that the different combinations of PBL and LSM parameterization schemes result in different SB timing and vertical circulation characteristics. The results have significant implications for convective initiation, air quality studies and other environmental problems in coastal areas.

P-Transformation.64 ID:4352 **10:30**

Integrating model chemical mechanisms with databases of reaction kinetics

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The role of chemistry in atmospheric modeling is central to research into climate change and air quality. The description of chemistry in these models, known as the 'chemical mechanism', is becoming increasingly complex. The validity of such chemical mechanisms relies largely on fundamental laboratory measurements of the kinetics of individual chemical reactions. For many years, expert scientists have evaluated published kinetic data and made it available in peer-reviewed literature and, for the last 10 yrs, via the World- Wide-Web (see the IUPAC database at http://www.iupac- kinetic.ch.cam.ac.uk/). Keeping up to date with the latest laboratory data is a time-consuming and error prone task. Changes to the mechanism need to be carefully checked and assessed to understand any differences in the model results. Consequently, it is not unusual for the kinetic data in complex models to be out of date. The work presented here seeks to develop a facility that would make significant improvements to the way in which the atmospheric chemical modeling community (automatically) update their chemical mechanisms. To achieve this, an important first step is to be able to describe a model's chemical mechanism in a concise but accurate way that a kinetic reaction database can understand. We define a syntax based on XML (eXtensible Markup Language). XML is now used in many areas of informatics, including atmospheric modeling. This has been done through a combination of CML (chemical markup language) and MathML (mathematical markup language). The ASAD chemical modeling software, used in the UKCA community chemistry aerosol model and p-TOMCAT chemical transport model, will provide a “test case” for the new XML structure. We have developed our current schema to a more concise and acceptable form for describing model chemical mechanisms. This simplified structure will be presented and its benefits in linking directly with kinetic databases will be described.

P-Transformation.65 ID:4443 10:30

The Sensitivity of Chemistry and Composition of the Troposphere on the Incident Solar Flux prescribed at the Top of the Atmosphere in a global CTM.

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Atmospheric composition, the oxidative capacity of the troposphere and resident chemical lifetimes of important trace gas species are essentially determined by the absolute rate of formation of free-radicals (e.g. OH) via fast photochemical reactions. Therefore an accurate estimate of the actinic flux with respect to altitude, location and season is a crucial step towards being able to capture the short- to medium-term fluctuations in chemical species typically seen by atmospheric observations. These variations are determined by solving the radiative transfer equation which is dependent on (e.g.) total overhead ozone, clouds, aerosols, etc as well as the incident solar flux at the top of the atmosphere (TOA). Here we use a version of the ‘8-band’ approach for calculating photolysis frequencies, which has been optimized for the troposphere in the global CTM TM5. This flexible parameterization allows the investigation of the influence of different estimates for the incoming solar irradiance at TOA on photochemistry. For this purpose we utilize the recently compiled high resolution solar reference spectrum of Dobber et al (2008) that is currently used for retrievals from the Ozone Monitoring Instrument (OMI). We then compare against simulations which use a number of different solar spectra commonly used by the atmospheric science community. We subsequently quantify the sensitivity of global large-scale CTM calculations towards the choice of the TOA solar spectrum via comparisons against measurements and a detailed chemical budget analysis.

P-Transformation.66 ID:4152 10:30

Ozone oxidation of secondary organic aerosol material formed by glyoxal and methylglyoxal

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Glyoxal and methylglyoxal may form oligomeric secondary organic products upon uptake to aqueous tropospheric aerosol particles. The reaction products identified by us and others include unsaturated species such as aldol condensation products and imidazoles which may be vulnerable to O3 oxidation in the atmosphere. Depending on the lifetimes of these species in an oxidizing atmosphere, they may affect aerosol optical properties and could potentially be used as tracer species for glyoxal or methylglyoxal SOA formation.

Here we report the results of kinetic studies of the O3 oxidation of methylglyoxal and glyoxal SOA products. Experiments were performed using an aerosol flow tube reactor coupled with an aerosol chemical ionization mass spectrometer (Aerosol-CIMS) for monitoring aerosol composition. Aerosols were generated by atomizing dilute aqueous aerosol mimics containing glyoxal or methylglyoxal and ammonium sulfate, and relative humidity in the reactor was maintained at ~65%. A scanning mobility particle sizer was used for determining aerosol size distributions and surface area. Oxidation mechanisms, products, and atmospheric lifetimes of 1H-imidazole-2-carboxaldehyde (formed by glyoxal) and aldol condensation products formed by methylglyoxal will be discussed.

P-Transformation.67 ID:4343 10:30

Uncertainty in modelled tropospheric composition due to rate constant uncertainty.

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Our understanding of the global distribution of species in the atmosphere is driven by field measurements and global models. Both suffer from uncertainties. Quantifying the uncertainties in the global models is difficult due to a plethora of input and boundary parameters. In this study we focus on the range in the concentrations of tropospheric species produced by a global model due to uncertainties in the chemical gas-phase rate constants. We use the GEOS-Chem composition transport model and perform multiple perturbation simulations where we change selected rate constants within the 1-sigma uncertainty determined from laboratory studies. Our choice of rates to perturb is determined by a recent box model full Monte Carlo study. We determine the impact of these uncertainties on the O3 budget and the global OH distribution. Finally we make recommendations to the gas kinetics community as to which reactions should be studied in more depth to improve our understanding of global tropospheric composition.

P-Transformation.68 ID:4338 10:30

Atmospheric Chemistry of i-Butanol: Kinetics, Mechanisms, and Products of Chlorine Atom and OH Radical Initiated Oxidation

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Alcohols are of interest as potential (e.g. butanol) or existing (e.g. ethanol) biofuels which can be blended with gasoline for use in automotive engines. To facilitate assessments of the environmental impacts of the use of alcohol biofuels we have embarked on a systematic study of the atmospheric chemistry of short chain alcohols. As part of this work the atmospheric chemistry of i-butanol was investigated using the smog chamber at Ford Motor Company, Dearborn, Michigan. Relative rate techniques were used to measure k(Cl

+ i-butanol) = (2.06 ± 0.40) x 10-10 and k(OH + i-butanol) = (1.13 ± 0.14) x 10-11 cm3 molecule-1 s-1 in 700 Torr of N2/O2 diluent at 296 ± 2K. Product studies of the Cl atom initiated oxidation of i-butanol were performed at 296 ± 2K in 700 Torr total pressure of N2/O2 diluent in the presensce and absence of NO. Iso-butyraldehyde, acetone and formaldehyde were observed as products. The results are discussed with respect to the the previous litereature data and the atmospheric oxidation mechanism of i-butanol.

P-Transformation.69 ID:4182 10:30

Kinetics and Mechanism of CF3CF2CF2CF2CF2CF2CF2CF2H Reaction with OH Radicals

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The rate constants for CF3CF2CF2CF2CF2CF2CF2CF2H reaction with OH radicals were obtained to be (2.56 ± 0.08) × 10-13 exp[-(1630 ± 130)/T] cm3 molecule-1 s-1 at 253–328 K by using relative rate method. Using FTIR smog chamber technique, COF2 was identified to be a unique product for CF3CF2CF2CF2CF2CF2CF2CF2H initiated by OH radicals at 298K.

P-Transformation.70 ID:4484 10:30

The latest updates to the GRAHM mercury model and their effect on the long-range transport of atmospheric mercury.

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Two major changes have been implemented in Environment Canada’s Global Regional Atmospheric Heavy Metals modeling system (GRAHM): 1) Vertical tracer transport by sub-grid scale convection 2) Wet deposition scheme.

It is widely accepted that deep convective storms have a major impact on the chemical composition of the troposphere and the long range transport of long lived tracers. The long lifetime (6 months to 2 years) of atmospheric gaseous elemental mercury, the dominant form of mercury in the troposphere, makes it prone to long range transport. We discuss the impact in GRAHM of the modified Kain-Fritsch (KF) scheme previously presented in association with A Unified Regional Air-quality Modeling System (AURAMS). The scheme uses the calculated KF mass fluxes to modify the chemical composition of the atmosphere.

Chemical transport models that simulate the long-range transport of atmospheric mercury are generally used in policy support in order to determine the deposition of atmospheric mercury, hence our effort to improve the wet deposition scheme in GRAHM. A new wet deposition scheme has been implemented to better account for below cloud scavenging as well as particle to particle collection. This scheme is a hybrid between the AURAMS and the Harvard wet deposition schemes. The model is tested against data from the Mercury Deposition Network (MDN).

Global simulations with these modifications to GRAHM will be presented. The preliminary analysis of these simulations shows that convective transport has an effect on the surface distribution of gaseous elemental mercury, as well as on the deposition of atmospheric mercury, especially in Africa and South America.

Simulations with emissions exclusively from East-Asia will also be presented to show the effects of these changes on the trans-Pacific transport of mercury.

P-Transformation.71 ID:4336 10:30

Mercury Oxidation by OH and Ozone: combined measurement and modelling studies in a high ozone environment suggest they play a very minor role

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In light of the recent interest in mercury as a global scale pollutant, and the ongoing debate as to its major oxidation pathways in the atmosphere, numerous kinetic, modelling and field studies have been carried out over the last ten to fifteen years. Mercury's lifetime is long enough for it to be found in the atmosphere and environment in remote areas such as the Arctic and Antarctica. The major removal process for mercury from the atmosphere is oxidation to Hg(II), which is more soluble than Hg(0), and subsequent scavenging via wet deposition processes. Which oxidants are responsible has been the subject of not a little debate, although it is now clear from observations, and modelling, of Arctic atmospheric mercury depletion events that Br-containing compounds do oxidise Hg in the atmosphere. The role of ozone and the OH radical is less certain; laboratory measurements suggest they both oxidise gas phase Hg(0), but the published rates vary and the gas phase products, if any, have not been unequivocally identified. Using data from a number of cruise measurements in the Mediterranean MBL between 2000 and 2009, the possible formation of Hg(II) from both ozone and OH has been studied. The Mediterranean MBL, particularly in the summertime provides an ideal environment to study these reactions due to the consistently high ozone concentrations, and high solar radiation intensity – during some cruises the average concentration of ozone was over 60 ppb. The diurnal variation seen in gas phase Hg(II) concentration in the MBL is completely irreproducible if OH or ozone are assumed to give gas phase products on reaction with Hg, however high concentrations of Hg associated with particulate matter were not observed, suggesting that the role of these oxidants in the atmospheric cycling of Hg is grossly overestimated.

P-Transformation.72 ID:4392 10:30

Estimating the contribution of shipping emissions to high ozone concentrations during a summer anticyclone in the Mediterranean MBL

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During the summer of 2005 the Italian Research Council R.V. Urania a performed series of atmospheric measurements along a route from Naples, south through the Strait of Messina, around the southern tip of Italy and then north through the Adriatic to the Gulf of Trieste and back again. The average ozone concentration during the entire measurement campaign was over 60 ppb, with some daytime peaks exceeding 100ppb. As part of a ongoing systematic investigation of the sources contributing to these high ozone concentrations the WRF-Chem model has been used to investigate the influence of different emission source categories on the regional ozone concentration. The model has been used with an outer domain covering most of northern Europe and all of the Mediterranean (81 by 81 km) and a nested domain (27 by 27

km) covering the whole of Italy, the Adriatic and part of the Tyrrhenian Sea. The EMEP emission database combined with the RADM2 chemical mechanism gave a reasonable comparison to observations along the whole ship's path, with the exception of a few occasions when the measured ozone values were particularly low, probably because the research vessel was close to a significant anthropogenic source. When the ship emissions are removed from the emissions inventory the modelled ozone concentrations change markedly. Along the route taken by the R.V. Urania the maximum daytime concentrations are generally around 10 ppb lower than when ship emissions are included, and sometimes as much as 15 ppb lower. Where maritime traffic is particularly high, (the Strait of Gibraltar and the Strait of Sicily) the difference in modelled ozone concentrations reaches 20 ppb. The amount of maritime traffic combined with the strong anticyclonic conditions encountered clearly mean that shipping is a major factor affecting summertime air quality in the Mediterranean.

P-Transformation.73 ID:4122 10:30

Towards a better representation of ship plume chemistry in a Global Chemistry-Transport Model

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Carrying about 90% of the world trade, International shipping is a cornerstone of the global economy and as such, a growing sector. In order to transport their load from one part of the globe to another one, ships burn bunker fuel resulting in the release of trace gases and particulate matter in the marine boundary layer.

In particular, NOx emissions are emitted in large amounts as a result of currently available propulsion technologies in regions where background concentrations of this species are usually low. Several modeling studies and field observations have already shown the potential from shipping emissions to perturb the tropospheric composition and the radiative forcing and to affect human health and ecosystems. However, large-scale models tend to overestimate the impacts of these emissions because they are not able to reproduce the non-linear character of the NOx-Ox-HOx-VOC chemical system occurring at the ship plume scale.

In this study, we present results from the implementation of the Cariolle et al., 2009 parameterization in the global chemistry-transport model: MOZART-4. We show that this parameterization reduces, as expected, concentrations of O3 along the ship corridors and we compare these results to previous studies. Finally, we estimate the radiative forcing resulting from shipping NOx emissions by using the offline Edwards&Slingo's radiative transfer model.

P-Transformation.74 ID:4319 10:30

Source identification and budget of elevated levels of formaldehyde within the ship plumes: A ship-plume photochemical/dynamic model analysis

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Elevated levels of formaldehyde along the ship corridors have been observed by satellite sensors, and were also predicted by global 3D chemistry-transport models. In this study, three likely sources of the elevated HCHO levels were investigated to identify the detailed sources and examine the contributions of the sources of the elevated levels of HCHO in the ship corridors using a ship-plume photochemical/dynamic model: (1) primary HCHO emission; (2) secondary HCHO production via the atmospheric oxidation of NMVOCs emitted from ships; and (3) atmospheric oxidation of CH4 within the ship plumes. From multiple simulations, CH4 oxidation by elevated levels of in-plume OH radicals was found to be the main factor responsible for the elevated levels of HCHO in the ship corridors. More than ~90% of the HCHO for the base case is produced by this atmospheric chemical process, except in the areas close to the ship stacks where the main source of the elevated HCHO levels would be primary HCHO. Because of active CH4 oxidation by OH radicals, the instantaneous chemical lifetime of CH4 decreased to ~0.45 yr inside the ship plume, which is in contrast to chemical CH4 lifetime of ~1.1 yr in the background. A variety of likely ship-plume situations at three latitudinal locations in the global ship corridors was also studied to determine the extent of the enhancements in the HCHO levels in the ship-going MBL. It was found that the ship-plume HCHO levels could be 20.5-434.9 pptv higher than the background levels depending on the latitudinal locations of the ship plumes, MBL stability and NOx emission rates. On the other hand, NMVOC emissions from ships were not found to be a primary source of photochemical HCHO production inside ship plumes due to their rapid and individual dilution.

P-Transformation.75 ID:4492 10:30

Atmospheric Organic Material and the Nutrients it Carries to the Ocean

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Organic material is present in the atmosphere both in the particulate and in the gas-phase as result of emissions, atmospheric transport and physico-chemical transformations and deposition to the surfaces. Due to their impact on air quality, the oxidizing capacity of the atmosphere, visibility, climate, human health and the ecosystems, atmospheric organics have attracted extensive attention from the scientific community. C/N/P cycles are mainly coupled through photosynthetic fixation of these elements by biological activity. C, N and P are main constituent of proteins and living organisms. Photosynthetic organisms utilize P, C, N and other essential nutrients to build their tissues. Biological productivity relies on the availability of these nutrients. There is increasing evidence that a significant fraction of N and P deposition occurs as ON and OP. However it is an open question the environmental impact of these amounts of nutrients. This paper is revisiting the budget of non methane organics in the global troposphere and provide updated picture of the role of organics in transporting nutrients particularly nitrogen and phosphorus to the oceans. This information is of particular interest in view of the involvement of atmospheric organic material in several climate/biosphere (terrestrial and marine)/chemistry feedbacks. It builds upon earlier works and makes use of the recently acquired knowledge on atmospheric organic material, emissions, transformation and deposition to the surfaces.

P-Observations 2.1 ID:4399 10:30

The effect of tropical islands on the chemical mixing of biogenics from the surface layer to the lower stratosphere

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In recent years much interest has been shown in the composition of the tropical atmosphere from the surface to the tropopause. During June 2008 an extensive measurement campaign (OP3) was conducted from the ground level in the canopy of the tropical rainforest of Borneo concurrently with aircraft flights over coastal areas, a mixture of virgin rainforest and palm plantations on the island. The aim of this campaign was to gain a better understanding of the chemical processes governing the tropical boundary layer and emissions from changing vegetation types. Tropical islands present a set of meteorological conditions that have the potential to loft surface emissions into the upper troposphere/lower stratosphere. Presented here is a discussion of the impact of introducing surface emissions to this region of the atmosphere on the chemical processing throughout the entire air column, with reference to high resolution model data and measurements from the OP3 campaign. The data show the lifting lifetimes and the effect these tropical chimneys have on the distribution of a range of biogenics and NOx, which in light of changing land usage have implications for the oxidative impact of the tropical region on global oxidative capacity.

P-Observations 2.2 ID:4579 10:30

Observations of Stratosphere-Troposphere Exchange Using Radar Windprofilers and Balloon-Borne Ozonesondes

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Twice-daily ozonesondes have been launched during a number of short campaigns, incorporating nearby radar windprofiler measurements, in Ontario and Quebec between 2005 and 2009. This novel combination of observations has demonstrated the existence of a strong relationship between apparent rapid changes in tropopause height and stratosphere-troposphere exchange (STE). In general, stratospheric intrusions appear to follow the passage of upper level cyclones which temporarily force the mid-latitude jet stream south of the observing locations. Since total ozone variations at extratropical latitudes are well-correlated with synoptic scale meteorological disturbances, particularly tropopause height, this suggests that it may be possible to relate rapid changes in total ozone to the occurrence of ozone intrusions. Further examination shows that where the radar- determined tropopause differs from the WMO thermal tropopause, the radar is typically responding to the sharp gradients of potential temperature and humidity at the lower edge of a stratospheric intrusion. Radar appears to be a particularly good intrusion detector. In addition to explaining the radar's success at finding STE events, this fact can potentially be used to study the descent of layers of stratospheric origin in the troposphere.

P-Observations 2.3 ID:4575 10:30

The Asian UTLS – a characterization by a combined model-observation approach

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In-situ observations of atmospheric compounds are often by nature local, and limited in time and by the number of measured species. From this fact arises the motivation to complement such observations with three-dimensional information based on models, which is routinely done in the form of back-trajectory calculations. We extended this Lagrangian approach considerably through a unique combination of atmospheric-chemistry models, which allows to separate and quantify transport, mixing, and chemistry processes along these trajectories.

We employed a model hierarchy consisting of the global Eulerian atmospheric-chemistry general-circulation model EMAC and the corresponding trajectory-box model CAABA to study the characteristics of air masses in the upper troposphere / lower stratosphere (UTLS) above Asia. Model output has been compared to monthly in-situ airborne measurements of long- and short-lived tracers from the CARIBIC project during the years 2006 and 2007. For this purpose, chemical mixing ratios along the flight tracks have been recorded on-line during a model simulation, which included a full atmospheric-chemistry mechanism, transient biomass burning emissions, and nudging towards ECMWF meteorology. Besides the direct comparison of tracers in the UTLS, trajectory clusters based on ECMWF operational analysis data and on the global model have been evaluated for further interpretation of observation-model comparisons.

New and unique Lagrangian model diagnostics within the model hierarchy enable the separation and quantification of transport, mixing, and chemistry contributions of various chemical tracers on their way into the Asian UTLS. Thus, important areas of air mass convergence and mixing are identified as well as dominant chemical reactions during the transport, and corresponding chemical lifetimes. Especially the latter analysis reveals important variation in lifetimes - and thus in transport budgets towards the UTLS – with respect to the trajectory source regions and emissions. In this way, the airborne observations above Asia are complemented with a 3-D dynamical and chemical history of the observed air masses.

P-Observations 2.4 ID:4583 10:30

Solar remote sensing FTIR measurements of trace gases in the marine boundary layer

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Measurements of Carbonyl Sulfide and other trace gases in the marine boundary layer have been made by Solar remote sensing Fourier transform spectroscopy. Spectra were recorded from the University of Wollongong, Australia (34.5°S, 150°E, 20m above sea level) in September and October 2009 and March 2010 at very high solar zenith angles at dawn. The use of spectra recorded at dawn with the sun rising over the Tasman Sea greatly increases the pathlength and thereby increases the sensitivity of the measurements of many marine boundary layer species. Here we present some initial results from these measurements for a

number of gases including Carbonyl Sulfide.

P-Observations 2.5 ID:4461 10:30

Pilot Study Preliminary Results for a Marine Boundary Layer Site near Ucluelet on Vancouver Island

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A one year pilot study is taking place from April 2010 to April 2011 on the west coast of Vancouver Island to determine its feasibility as a Marine Boundary Layer (MBL) site. The MBL site is located at Amphitrite Point on Vancouver Island (48.9217, -125.5411) at an elevation of about 20 meters above sea level. Due to the Beaufort Mountain range bisecting Vancouver Island from NW to SE, the winds in the region are predominantly parallel to the island or WNW in the summer and ESE in the winter. Back trajectories show that transpacific air should impact the site on the majority of days with typical meteorological patterns. Occasionally air reaching the site may be impacted by the Seattle area or the Vancouver area. Measurements at the site include particulate as well as gas samples. Preliminary data from the spring 2010 will be presented.

P-Observations 2.6 ID:4292 10:30

Characterisation of the Cape Verde Observatory's air mass origin and chemistry using the NAME dispersion model

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The UK Met office's NAME atmospheric dispersion model has been used to develop station footprints for the Humberto Duarte Fonseca WMO Global Observatory on the Cape Verde islands in the Atlantic Ocean. The model output has been run every 3 hours over a period of three years, tracking air mass origin over the previous 10 days. The most common air mass trajectories types have been identified and each back run was assigned to one trajectory type.

The long term trace gas measurements at Cape Verde from the past three and a half years were used to calculate an average composition (of O3, NO2, CO, CN, VOCs etc.) for each type of trajectory over the whole period. Saharan dust episodes are particularly common at the station during the winter months but air from the coastal areas just off the African continent are the main route to the islands all year round. Many air masses originate from North America or from Europe a few days before and tracking the pollutant loadings of these gives us an idea of the extent of the oxidation that occurs during transport. This seasonal and geographical characterisation of the chemistry of the air masses arriving at the Cape Verde islands can elucidate the long term pathways and transformations of pollution across the Atlantic Ocean.

P-Observations 2.7 ID:4406 10:30

Chemical composition of aerosols in a tropical region considering the influence of local and long distance transport of pollutants

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Aerosols can affect the climate by changing the way radiation is transmitted through the atmosphere. Direct observations of aerosols are quite limited, making any attempt to estimate their global effect difficult. Due to the uncertainties of aerosol radiative forcing measurements around the world, especially in Southern Hemisphere tropical regions, to a better estimate of their effects is necessary. The objective of this study is to determine the chemical composition of aerosol samples collected in the Rio de Janeiro metropolitan area, under the influences of different local (natural, industrial and traffic) and long-range transport. The aerosol samples were collected for one year (2008-2009). Concentration levels of aerosols, metals, water- soluble ions, and soluble organic and nitrogen fractions were measured. Preliminary results showed that the mean aerosol concentrations were 36 µg/m3; 50 µg/m3 and 74 µg/m3 at the natural, downtown and industrial sites, respectively. An increase in the average particle concentrations was observed in the period from May to September probable due to air stagnation increasing pollutant concentrations. Average concentrations of anions were 1.7 µg/m3 for Cl-, 2.7 µg/m3 for NO3-, 2.5 µg/m3 for SO4= and for cations were 2.9 µg/m3 for Na+, 1.0 µg/m3 for Ca++, 0.3 µg/m3 for K+ and Mg++. Low levels of NH4+ and organic acids (0.04-0.10 µg/m3) were measured. Average water-soluble organic compounds (WSOC) were detected in similar concentrations to the main ions species (2.1 µg/m3). The studied species concentrations are influenced by the site locations. At the natural site, lower concentrations of NO3- and SO4= were detected as well as when air masses came from the ocean. According to satellite images, Saharan dust reached the sampling sites, increasing from two to five times aerosols, Ca2+and K+ concentrations. This is the first study reporting the increase of some aerosol species in Rio de Janeiro due to African outbreaks. Other results of nitrogen compounds and metals will be presented at the meeting.

P-Observations 2.8 ID:4306 10:30

Dry deposition of trace elements in central India

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The vast industrialization and urbanization result in substantial increase of the aerosol concentration in urban sites of India. The aerosol concentration is reached maximum in the winter season (December/January) of a year with serious environmental and health hazards. The nature of atmospheric deposition reflects the quality of water and precipitates. Therefore, thirteen resuspended dust deposited in the roof of building of Raipur city (capital of Chhattisgarh state, India) from the residential(Re), commercial(Co) and industrial(In) sites were collected in January, 2009. The dust of mesh size ≥ 0.1 mm was sieved out. The concentration of trace elements and ions in dust were determined. The metals i.e. Al, Fe and Mn showed the highest concentration, ranged from 2.6 – 4.0, 5.2 – 12.4 and 0.3 – 9.5% with mean value of 3.0±0.2, 9.2±1.3 and 1.9±1.6%, respectively at 95% probability. Similarly, the concentration of Cr, Ni, Cu, Zn, Cd and Pb is ranged from 54 – 527, 25 – 172, 37 – 157, 72 – 408, 63 – 251 and 0.10 – 0.91 mg kg-1 with mean value of 127±68, 49±21, 74±19, 275±48, 115±30 and 0.29±11 mg kg-1, respectively. Four metals i.e. Fe, Mn, Zn and Cd exhibited the highest value in the industrial sites. Whereas, other metals i.e. Cr, Ni, Cu and Pb showed the highest value the in commercial sites. The toxic metals i.e. As, Hg and Se species are quantified. The correlation of the metals with ions i.e. Cl-, NO3-, SO42-, Na+, K+, Mg2+ and Ca2+ are described. The sources of the metals and ions are apportioned by using various models.

P-Observations 2.9 ID:4370 10:30

Atmospheric composition over Namibia (austral Africa) from the MOZAIC programme.

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The general aim is to document the climatology of vertical profiles over Windhoek (Namibia) from 2006-2008 MOZAIC data. This dataset is very dense, i.e. two profiles per day every two day for ozone, carbon monoxide and meteorological parameters. Windhoek is pretty far away from the African regions where ozone precursors are emitted, either from biomass fires, or from deep continental convection (LiNOx), or from anthropogenic sources. It is a place where the impact of long-range transport of these emissions can be documented. We will focus on the two seasons during which intra-seasonal variabilities are the largest, i.e. spring (SON) and summer (DJF). During SON, intra-seasonal variabilities come from the long-range transport of lower- and upper-tropospheric CO plumes emitted by biomass fires over both southern Africa and southern America. SON is also the season during which stratospheric intrusions may have an impact on the upper tropospheric ozone distribution down to the latitude of Windhoek (22S). During DJF, intra-seasonal variabilities come from the long-range transport of mid- and upper-tropospheric O3 plumes that we intend to attribute to ozone production processes activated by LiNOx emissions over convective regions both over southern Africa and southern America.

The results show (i) the climatology in SON and DJF, (ii) representative case studies of each class (i.e., low-tropospheric and upper-tropospheric CO plumes, stratospheric intrusions, LiNOx plumes), (iii) the history and origin of case studies using the Lagrangian dispersion model Flexpart and satellite data for the detection of biomass fires and lightning flashes, and (iv) MOZAIC composites of every class of plumes and differences between the composite and the climatology. General characteristics of the plumes, as well as components of intra-seasonal variabilities coming either from the long-range transport (meteorological situation) or from the emissions are discussed.

P-Observations 2.10 ID:4374 10:30

Lulin Atmospheric Background Station (LABS, 2,862m MSL): A newly established baseline site in East Asia

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The Lulin Atmospheric Background Station (LABS) in Taiwan held its grand opening for operation on 13 April 2006. It is located at the Mt. Lulin (2,862 m MSL; 230 28'07"N, 120052’25"E) in central Taiwan. The

LABS is unique because its location and altitude can enhance the global network of GAW (Global Atmosphere Watch) in the Southeast Asian region where no high-elevation baseline station is available. Our site is located between the GAW Waliguan station (3,810 m) in Tibetian plateau and Mauna Lao Observatory (3,397m) in Hawaii. Trajectory study indicates that this site provides us a great of opportunity to observe a variety of air masses originated from source regions, giving a distinctive contrast of atmospheric changes. Present continuous operations include precipitation chemistry, aerosol chemistry, trace gases (CO, CO2, O3, CFCs, NOy), mercury, atmospheric radiation, and meteorological variables. Mission oriented measurements of HCFCs, HFCs, and VOCs were also performed on a needed basis, mainly to help deconvolute long range transport phenomena from local circulation. Till present time, the average concentrations of CO, O3 and PM10 are about 121 ppb, 34 ppb and 10 microgram m-3, respectively. The average pH value of precipitation is 5.73 with the major ions of SO42-, NO3- and NH4+. The average concentrations of mercury such as GEM, RGM and PHg are about 1.78 ng m-3, 41.7 pg m-3 and 9.0 pg m-3, respectively. The average aerosol optical depth is 0.101. The background concentrations of CO, O3 and PM10 are estimated to be about µ82 ppb, 28 ppb and 6 microgram m-3, respectively. About 32 % of the days in a year can be categorized as polluted, which mainly occurred in March, and the concentrations of above three pollutants could elevate to as high as twice their background values. To summarize the results, the maximum concentration of pollutants generally occurred during spring time, especially in March, corresponding to the biomass burning from SE Asia. The LABS is also one of US/NOAA CCGG network and NASA AeroNet. Relevant data will be presented.

P-Observations 2.11 ID:4360 10:30

Trace Species over the Central Himalayan and Indo-Gangetic Plain Regions: Contribution of Regional Pollution and Long-range Transport

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Satellite based observations and model results show that the northern India is the most polluted region in India. In view of this, for the first time, observations of various trace gases (O3, NO-NOy, CO, SO2, CO2 and Hydrocarbons), aerosols (AOD, black carbon, aerosol number concentration and chemical composition) and meteorological parameters are initiated at a high altitude site (Nainital, 29.4N, 79.5E, 1958m) in the central Himalayas. Surface ozone and black carbon observations are also made at two other low altitude sites (Pantnagar, 29.0N, 79.5E, 231m; Dehradun, 30.3N, 78.1E, 640m) in the Indo-Gangetic regions. Observations at Nainital are representative of regional environment in the Northern Indian subcontinent and are also useful to study influences of long-range transport. Observations at Pantnagar and Dehradun are used for studying sources of regional pollution.

Observations of ozone, NO-NOy and CO suggest that daytime net ozone production is not dominating over Nainital. While other two sites show daytime higher ozone typical of urban/rural sites. In contrast to the diurnal variations, seasonal variations are more-or-less similar at these three sites with a spring maxima and summer/monsoon minima. Seasonal variations in trace species over this region are different than those over the western India. Based on residence time of air masses, net ozone production over Northern Indian Subcontinent in regionally-polluted air masses is estimated to be ~3 ppbv/day in spring but no clear build-up is seen in other months. Background ozone levels are estimated to be 30-35 ppbv and contribution of long-

range transport is estimated to be 8-11 ppbv during January-March. Role of springtime biomass burning is studied and enhancements in ozone and BC levels at Nainital due to fires are estimated to be ~18 ppbv and ~1725 ng/m3 respectively. Significant influences of biomass burning on tropospheric column NO2, aerosol properties and lower tropospheric CO over Northern India are seen. The clear sky shortwave aerosol radiative forcing at the surface and TOA during fire-impacted periods is seen to increase by about 26 and 8 W/m2 with respect to low fire activity periods. More detail on results, including preliminary results of ozone vertical distributions will be presented.

P-Observations 2.12 ID:4567 10:30

Modelling the multi-isotope composition of CO and related trace gases with the comprehensive AC-GCM EMAC.

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The unique information on sources, chemical modification and sinks of atmospheric trace gases can be inferred from their isotope composition. An appropriate modelling approach is vital to the interpretation and use of an increasing number of isotope analyses. However, the exact implementation of isotopic information is a challenge, and many studies use simplifications which limit their applicability. We present a modelling study with an elaborate isotopic chemistry mechanism employed in the atmospheric chemistry general circulation model EMAC. The Modular Earth Submodel System (MESSy) framework incorporated in EMAC allows an explicit, more realistic treatment of the isotope effects (i.e. fractionation in chemical and physical processes) per species basis. Conformably, we present the results of the global modelling of the stable carbon and oxygen isotope composition of the bulk of tracers, with a focus on carbon monoxide. The sensitivity of the CO isotopic composition to the key atmospheric processes is estimated; in particular, we investigate the yield of CO from methane, the only distinctively 13C-depleted source of carbon in CO. The isotope mass balance imposes yield values significantly lower than unity, pointing to efficient removal processes acting upon the intermediates of the CH4 to CO conversion chain. In contrast to prevalent simplified approaches, in EMAC the yield is a diagnosed variable, rather than an assumed one. Concurrently, the first-time a global synthesis of the mass-independently fractionated (MIF) oxygen isotope composition of CO is presented. The intrinsic MIF enrichment in CO induced by its oxidation by OH versus the ozone-impelled MIF source effect (due to reactions with unsaturated hydrocarbons) is assessed. The latter is found to be as large as +1‰ and +0.7‰ in Δ17O(CO) for NH and SH, respectively, a relatively minor enrichment compared to the dominating OH effect of +4‰ to +6‰ in background air. This renders a considerable importance of CO MIF for studying the OH budget.

P-Observations 2.13 ID:4471 10:30

Temperature, relative humidity, mass divergence, and ozone response to strong rainfall events in the western tropical Pacific: Observations and models

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Strong precipitation events are identified at several locations in the western tropical Pacific using the TRMM 3B42 gridded rainfall product. Radiosonde (IGRA) and Ozonesonde (SHADOZ) soundings are then used to construct composite anomaly patterns of temperature, relative humidity, mass divergence, and ozone, from 24 hours prior to the rainfall events to 24 hours after the rainfall events. The observed composite anomaly patterns are compared with those derived using data output from four general circulation models (AGCM3, AGCM4, GFDL2.1, ECHAM5) and two reanalysis products (ERA-40 and ERA-Interim). For the most part, the models do not realistically represent the mid-level warming, moistening, and divergence associated with congestus clouds that precedes the rainfall maximum. The surface cold pool that develops about the strong precipitation events is either not present, or is weaker than observations. In addition, the models do not fully represent the mid-level stratiform convergence that follows the precipitation. The inability of models to reproduce these features suggests that the convective parameterizations used by the models and reanalysis do not realistically simulate the short timescale interactions between moist convection and the background atmosphere.

P-Observations 2.14 ID:4184 **10:30**

Vertical ozone measurements in the troposphere over the Eastern Mediterranean basin

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Vertical ozone profiles measured in the period 1994–2008 in the framework of the MOZAIC project (Measurement of Ozone and Water Vapor by Airbus in Service Aircraft) for flights connecting Central Europe to the Eastern Mediterranean basin (Cairo, Tel-Aviv, Heraklion and Rhodes, Antalya) were analyzed in order to evaluate the observed high rural ozone levels in the Mediterranean area during summertime. In total 237 summer (JJA) profiles were analyzed with the following distribution: Cairo, Egypt (94 profiles), Tel-Aviv, Israel (85 profiles), Heraklion-Rhodes, Greece (36 profiles) and Antalya, Turkey (22 profiles). This work is an extension of a previous study including 77 MOZAIC profiles collected in Heraklion, Rhodes and Antalya during the period 1996-2002 (June – September) so that to include the Middle-East region.

In the lower troposphere and in the 1000-3000m layer the average profiles over Cairo and Tel-Aviv show comparable values in the 60-72 ppb range, while the corresponding averages for Heraklion, Rhodes and Antalya are also quite comparable, though in the 55 – 62 ppb range. At all stations a rapid decrease of the average ozone mixing ratios is observed in the lowest 1000m, the sharpest decrease being observed at Tel-Aviv followed by Cairo, whereas the decrease is less significant at Heraklion, Rhodes and Antalya. These three airports show the higher average values (50–55 ppb) in the surface layer below 500m than the Middle-East profiles. An examination of the average profiles for the 7% highest and the 7% lowest ozone mixing ratios within the vertical interval 1000-1500m asl shows the following: a) profiles over Cairo, Heraklion, and Rhodes show similar characteristics with large differences (40–50 ppb) between the minimum and maximum values, b) profiles over Antalya and Tel-Aviv exhibit smaller differences (20-30 ppb). An analysis of composite weather maps for the high and low ozone cases as well as back-trajectories suggests that the main factor leading to high tropospheric ozone values in the area is the presence of anticyclonic conditions especially over the European continent transporting ozone and precursors, in association with subsidence over Eastern Mediterranean. On the other hand, the lowest ozone levels are associated with low-pressure anomalies over the European continent leading to westerly circulation over Mediterranean towards the eastern region diffusing the air pollutants and also with extension of the Middle East low over the Eastern Mediterranean

P-Observations 2.15 ID:4216 10:30

Twenty years of tropospheric ozone variability measured at the Observatoire de Haute Provence from 1990-2009

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Understanding of the tropospheric mid-latitude ozone trends in the free troposphere becomes again a critical issue, as long range transport of continental emissions varying in opposite directions appears to control the ozone variability and its role in the climate. The tropospheric ozone variability can be studied from vertical profiles obtained using two types of instruments during the time period 1990-2009 at Observatoire de Haute Provence in Southern France (44N, 6E), balloon borne ozonesondes and a UV DIAL lidar providing data in the altitude range 3-14 km.

In our study, we have been doing calculations of the seasonal variation for 2 km layers in the troposphere (2-8 km) for four five years periods. Since it is done separetely for the lidar and sonde records, differences can be discussed both in terms of instrument accuracy and of sampling bias due to different air mass histories. Sondes are launched every week on the same day, while lidar profiles are taken 1 or 2 times per week and for low cloud coverage. The comparison is based on the shape of the seasonal variability and the absolute values of the seasonal averages, especially the spring or summer maximum. The shape of the seasonal variability is similar but differences of the order of 3-5 ppb occurs in the seasonal averages with a systematic negative bias for the lidar in 1996-1999 for all season.

Differences related to sampling conditions are analyzed with 3-days three dimensional backward trajectories calculated for each vertical profile during twenty years. Four source regions are identified: [Europe (lat>45N)], [Eastern Mediterranean], [Spain/Africa/North America (lat<40N)] and [North America (lat>40N)]. Thus, two criteria are discussed in this analysis : the fraction of profiles being above a given region during the last three days and the starting altitude of the trajectories. We did not find large differences in the starting altitude, but more situations from the South-West region, sampled by the sounding balloons after 2000. Another way to perform this comparison is to calculate the frequency of the ozone layering in a given profile. For the time period 1996-1999, it was found that more layering occurs for the lidar with an unusual seasonal variation (no summer peak). This points toward a peculiar lidar sampling in 1996-1999.

The 20-years trend will be presented using all the lidar and sonde data. Considering that, we cannot identify a clear bias in a given direction due to instrumental effect

P-Observations 2.16 ID:4225 10:30

Is Europe getting cleaner? Analysis of European ozone trends 1995-2006

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The GEOMON (Global Earth Observation and MONitoring) project has produced a harmonised dataset of trace gases from ~400 ground-based measurement stations from a variety of regional, national and European air quality networks (e.g. EMEP, GAW). Forty-one of these sites typically categorised as rural and/or background, with good ozone data coverage form 1995-2006, have been selected for analysis.

Customised tools have been developed in R to assess European trace gas trends. Long-term and seasonal trends have been characterised at all sites using loess regression. Additionally, ozone was deseasonalised and linear trends were quantified for monthly means, 5th and 95th percentiles (to illustrate changes in mean, background and peak concentrations respectively). Linear trends for seasonal ozone means were quantified in a similar manner.

With the introduction of national and European legislation over the past 20 years, and the modernisation or removal of industrial sources, European ozone precursor emissions have resulted in significant reductions in ozone since the 1980's. However, between 1995-2006 analysis of ozone 5th and 95th percentiles (representing background and peak levels, respectively) show that the majority of sites in central and northwestern Europe have increased with a range of 0.11-0.67 ppbv/yr and 0.19-0.64 ppbv/yr, respectively (significance $p<0.01$). Increasing ozone 5th and 95th percentiles are consistent with observations of 5th percentile trends across the Northern Hemisphere, but inconsistent with observations of decreasing 95th percentiles. Isolated sites in peripheral Europe display negative trends in ozone 5th percentiles, with a range of -0.26- -0.70 ppbv/yr (significance $p<0.01$), whilst seven sites located in the U.K., and central or eastern Europe exhibit decreasing trends in ozone 95th percentiles (range -0.30 - -1.62 ppbv/yr, $p<0.01$).

A comparison to EMEP NOx emissions inventories or each site has been carried out, in order to investigate links between the control of anthropogenic precursors emissions and European ozone trends.

P-Observations 2.17 ID:4238 10:30

Long-term changes in ozone over Europe since the early 1990s – comparison of ozone measurements from sondes, regular aircraft (MOZAIC) and alpine surface sites.

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Intercontinental transport of ozone and its precursors has been the subject of several recent studies because precursor emissions from the northern continents have changed substantially over the past 20 years. There is a need for a credible set of time series data for ozone with which to evaluate models of the effects of emissions changes and of long range transport. In our study we compare the following ozone measurements: (i) ozone sondes, available since the late 1960s from Uccle (Belgium), Hohenpeissenberg (Germany) and Payerne (Switzerland); (ii) continuous surface ozone measurements from high alpine sites including Zugspitze (Germany, available since 1978), Jungfraujoch (Switzerland) and Sonnblick (Austria) (both available since around 1990); (iii) measurements from regular aircraft measurements (MOZAIC, available since 1994). In order to analyze the consistency of the different archives, monthly anomalies of the individual data series were determined and compared, including comparisons at the altitudes of the mountain sites (3000-3700 m). After about 1998, the time series of monthly anomalies are fairly consistent, whereas in the earlier period there are some substantial differences. The MOZAIC data are generally more consistent with the mountain sites than the ozone sonde records (measured with Brewer Mast sensors) during 1994-1998. Prior to 1990, large deviations among the three ozone sonde series were found, suggesting some data

quality problems. The high alpine surface ozone series show an increase in ozone during the 1990s, as does the MOZAIC data for 1994-2000. However, after about 2000, all data series (sonde, aircraft, and mountain sites) indicate stabilization of ozone. This is in contrast to recent results for western North America.

P-Observations 2.18 ID:4263 10:30

Temporal trends in near-surface ozone and N,°tropospheric column ozone at a tropical coastal site Trivandrum (8.5 E): effects of precursors and meteorology°77

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Ozone plays a vital role in tropospheric chemistry through its reactions with water vapour producing the highly reactive OH radicals. Tropospheric ozone has two major sources namely (1) transport from stratosphere and (2) photochemical production from gases like CH4, non-methane hydrocarbons (NMHC), other volatile organic compounds (VOCs), CO, and NOx (NO and NO2. The mixing ratio of ozone at any location is controlled by availability of precursors, photochemistry, meteorological conditions and vertical transport. Continuous measurements of ozone and NOx (NO+NO2) are carried out at Trivandrum (8.55ºN, 77ºE) by using an on-line UV Photometric Ozone analyzer (Model 49C of Thermo Electron Corporation) and a Chemiluminescence NOx 9841B of ME). This paper presents the seasonal®analyzer (Model ML trends in near-surface ozone and nitrogen oxides at the tropical coastal site Trivandrum. In addition, the seasonal changes in the satellite-retrieved (TOMS/OMI) tropospheric column ozone data during 2005-2009 also have been investigated in detail and compared with the surface data. The monthly variation is found to be the same in both tropospheric and surface ozone over a year. Meteorological data available from an Automatic weather station, airflow patterns obtained from NCEP/NCAR reanalysis and airmass backtrajectories obtained from HySplit model are used to understand the meteorological effects in the temporal behaviour of ozone at this location. High mixing ratios are observed in the winter months in both tropospheric and surface ozone. The prevailing northerly/north-easterly airflow which favours advection of precursors from inland locations causes the high mixing ratios during winter. During monsoon when airflow is from ocean, these gases exhibit low mixing ratios. It is also observed that, the mesoscale circulations - seabreeze and land breeze- modifies the diurnal patterns. The contribution of surface ozone to total tropospheric column ozone varies with season. The long-term trends in tropospheric ozone are also investigated in this paper.

P-Observations 2.19 ID:4368 10:30

Links between tropospheric O3 and weather conditions over India: evidence from spaceborne observations

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Between November and May, South Asia is affected by an 'Atmospheric Brown Cloud' (ABC), characterized by high levels of pollutants (aerosols, O3 precursors) in the lower troposphere. In the tropical Upper Troposphere (UT), O3 is dynamically influenced by interactions between midlatitude and tropical systems. Our study aims at characterizing the impact of severe weather conditions upon both lower and upper tropospheric O3 over the Indian sub-continent based upon O3 data from the Infrared Atmospheric Sounding Interferometer (IASI). The high spatio-temporal coverage and high sensitivity of the IASI data will enable such a study on a continental scale for the first time. We will present our results as follows: 1/-

IASI O3 data: O3 atmospheric profiles are retrieved from the IASI radiances with the SOftware for a Fast Retrieval of IASI Data (SOFRID) developed at Laboratoire d'Aérologie and based on the RTTOV radiative transfer code. We will show that SOFRID-IASI O3 data in the tropics are sensitive both to the free and upper troposphere. 2/- Validation of IASI O3 : We will validate our O3 profiles focusing on the tropical troposphere using O3 in-situ measurements from the MOZAIC program (unique data in Central India) and the SHADOZ network in 2008. 3/- Application to the Indian sub-continent: (a). pollution in the lower troposphere: We will first establish the relationship between the South Asian ABC (based on MODIS Aerosol Optical Depth observations) and high concentrations of O3 observed by IASI during the 2008 post monsoon season. We will then focus on quantifying the 'cleaning' effect of tropical storms upon lower tropospheric O3 over India based on a couple of cases in 2008. (b). Upper tropospheric dynamics: Episodes of interactions between a tropical cyclone and the Sub-tropical Westerly Jet will be documented using meteorological analyses and IASI data.

P-Observations 2.20 ID:4382 10:30

Quantifying the influence of stratospheric import on ozone concentrations at the Mt. Bachelor Observatory using 7- beryllium observations

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There has been much debate about the degree to which lower tropospheric O3 concentrations are influenced by transport of ozone from the upper troposphere/lower stratosphere [UT/LS, see review by Stohl, 2003]. Previously published estimates of Stratosphere to Troposphere O3 transport rely heavily on European high-altitude observational records of 7-Beryllium (7Be), a radionuclide tracer of stratospheric air [e.g. Schuepbach et al., 1999; Gerasopoulos et al., 2001; Zanis et al., 2003; etc.].

Here we present the first observational record of 7Be from a high altitude observatory in the western United States. Spanning the time period of April 22nd through May 28th, 2009, our research group analyzed the 7Be activity of 42 bulk aerosol samples collected at the Mt. Bachelor Observatory (MBO) near Bend, Oregon (43.979 N, 121.687 W, 2.7 km amsl). Comparing this new timeline of 7Be concentration to MBO observations of O3, CO, relative humidity and temperature, our results indicate that, during the study period, at least three separate tropopause folding events led to significant transport of O3 rich air from the UT/LS air down to at least the 730 hPa surface over the cascade mountains of central Oregon. For each of these three events, we confirmed stratospheric origin by comparing the 7Be concentration with concurrent observations of CO, aerosols and water vapor, computing back trajectories from the Hybrid Single Particle Lagrangian Trajectory Model (HYSPLIT), and by analyzing available satellite observations of tropopause fold events. Maximum observed O3 concentrations during these three tropopause fold events ranged from 64 ppbv to 94 ppbv, compared to a full period average O3 concentration of just 46 ppbv. These results indicate that 730 hPa springtime O3 concentrations over the western US are heavily influenced by import of stratospheric O3 during tropopause fold events.

P-Observations 2.21 ID:4514 **10:30**

The Use of Satellite based GOME and SCIAMACHY data to Study the Tropospheric Ozone amount over the Tropics

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The Global Ozone Monitoring Experiment (GOME) launched in April 1995 is measuring the sunlight back scattered by the surface in nadir viewing mode (240-790 nm) to detect O3, NO2, BrO, OClO, HCHO and SO2. SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric ChartographY) launched in March 2002 is measuring sunlight, transmitted, reflected and scattered by the earth atmosphere or surface (240 nm - 2380 nm). SCIAMACHY measurements yield the amounts and distribution of O3, BrO, OClO, ClO, SO2, H2CO, NO2, CO, CO2, CH4, H2O, N2O, p, T, aerosol, radiation, cloud cover and cloud top height in limb and nadir mode. Over the tropics e.g. biomass burning is extensive. During these burning events large amounts of aerosols and trace gases like nitrogen oxide NOx, hydrocarbons, formaldehyde (HCHO) and carbon monoxide (CO) are emitted into the troposphere. In photochemical reactions tropospheric O3 is produced. GOME- and SCIAMACHY-data were analysed to observe an increasing of this trace gas during the fire event and to compare then these results with the data of a “non-burning-season” to calculate the additional impact.

P-Observations 2.22 ID:4213 **10:30**

The Bihar Pollution Pool as observed from MOPITT (version 4 data), CALIPSO and tropospheric ozone residual data

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The Bihar pollution pool covers an extensive region in the eastern parts of the Indo-Gangetic basin and is most intense during winter. We use recently improved (version 4) retrievals of carbon monoxide (CO) from the MOPITT observations along with the aerosol data from the latest version of the CALIPSO lidar instrument to provide new insight into the vertical structure of this pollution. In addition, the tropospheric ozone residual products from the TOMS/SBUV and OMI/MLS database are examined to characterize this pollution pool. The feature is seen primarily in the lower troposphere from about November to February with strong concomitant increase in the CO, aerosol and ozone tropospheric columns. The height resolved aerosol data from CALIPSO confirm the trapping of the pollution pool at the lowest altitudes. The CALIPSO subtype data indicate polluted dust to be the dominant aerosol type. In winter the dust is probably of local origin. The observations suggest that MOPITT can capture this low altitude phenomenon even in winter conditions as indicated by the averaging kernels. The simultaneous observations of the feature in both the tropospheric ozone column products suggest photochemical production of ozone from the enhanced precursor abundances at low altitudes.

P-Observations 2.23 ID:4259 **10:30**

Effects of regional pollution and long range transport on the lower tropospheric ozone over an urban site in western India

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The balloon borne measurements of vertical distribution of ozone and meteorological parameters were made fortnightly from April 2003 to July 2007 at Physical Research Laboratory, Ahmedabad (23.03º N, 72.54º E, 49m amsl). The seasonal distributions of ozone are investigated for the boundary layer (1-2 km) and lower troposphere (2.5 to 4 km) using back-trajectory analysis. Three geographical sectors are defined for the regional pollution, marine region and long range transport, and residence time of trajectories for balloon flight days are estimated. These residence times are tagged with the corresponding ozone mixing ratios. The ozone levels increase with increasing residence time till 4-6 days for all seasons except winter in the regional sector. The significant ozone build up is absent during winter probably due to high levels of local pollutants. The slope of ozone vs residence time is extrapolated to zeroth day to get the "background ozone mixing ratio". The average background ozone mixing ratios are found to be about 28 ppbv for boundary layer and 42 ppbv for the free troposphere. The background ozone mixing ratio is comparable to average ozone mixing ratio during summer-monsoon season in the boundary layer. The seasonal variation of ozone shows higher ozone levels during late autumn, winter and early spring (> 50 ppbv) and minimum during summer-monsoon (~30 ppbv) in the boundary layer. The higher ozone levels during late autumn, winter and spring seasons are dominated by regionally polluted airmass whereas poor ozone levels during summer-monsoon are influenced by marine airmass. In the lower troposphere, seasonal variation of ozone is found to be affected by regional pollution and long range transport. Detailed results will be presented.

P-Observations 2.24 ID:4578 **10:30**

High-resolution tropospheric ozone fields for INTEX and ARCTAS from IONS ozonesondes

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The IONS-04, IONS-06 and ARC-IONS ozone sounding campaigns over North America in 2004, 2006 and 2008 obtained approximately 1400 profiles, in five series of coordinated and closely-spaced (typically daily) launches. Although this coverage is unprecedented, it is still somewhat sparse in its geographical spacing. Here we use forward and back- trajectory calculations for each sounding to map ozone measurements to a number of other locations, and so to fill in the spatial domain. This is possible because the lifetime of ozone in the troposphere is of the order of weeks. The trajectory-mapped ozone values show reasonable correlation with estimates from OMI and, where they overlap, to each other. Further validation with MOZAIC profiles and surface station data will be presented. A variable-length smoothing algorithm is used to fill data gaps. The total tropospheric ozone column maps calculated by integrating the smoothed fields agree well with similar maps derived from TOMS and OMI/MLS measurements. The resulting three-dimensional picture of

the tropospheric ozone field for the INTEX and ARCTAS periods facilitates visualization and comparison of different years and seasons, and will be useful to other researchers.

P-Observations 2.25 ID:4261 10:30

Characteristic variations of O3, CO, NOx, and NMHCs at two urban sites of Indo-Gangetic plain.

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Variations in surface O3, CO, NOx and NMHCs were investigated during December 2004 at two urban sites of Hissar (25°5′N, 74°46′E) and Kanpur (26°2′N, 80°2′E) in the Indo-Gangetic Plain (IGP) as a part of a major land campaign. Measurements of surface O3, CO, and NOx were made using online analysers, while methane and NMHCs (C2-C5) were analysed from the air samples collected twice in a day. The IGP is a most densely populated as well as polluted region of India along the foothills of Himalaya. Measurements at both the sites can be influenced by the diverse activities of emissions and local meteorology. Generally, the sky conditions are clear as well as foggy in the month of December. The mixing ratio of O3 shows strong diurnal variations with maxima of 80-85 ppbv in the after noon hour at both sites for clear sky days, but on foggy days the diurnal patterns were not clear and the levels were very low (10-15 ppbv). The time series variations of all species show almost similar trends at Hissar and Kanpur. There were events of higher levels of CO and NOx coinciding with lower values of ozone during night hours. The average mixing ratios of ozone, CO and NOx were 32, 550 and 16 ppbv for Hisar and 28, 300 and 12 ppbv for Kanpur respectively. The abundances of ethane (25%), ethene (23%), and acetylene (14%) were dominant in light (C2-C5) NMHCs at both the sites. The detailed results will be presented for these two sites along with a comparison with other observations and results from the 3D model MOZART.

P-Observations 2.26 ID:4280 10:30

Inter-annual variability of satellite derived ozone Column, carbon monoxide and aerosol optical depth over major cities in Canada during 2000 – 2009

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Detailed analysis of multi satellite sensor data (MODIS, OMI AURA, MOPITT, AIRS) of atmospheric properties (aerosol optical depth, angstrom coefficient, carbon monoxide, ozone) have been carried out over major Canadian cities (Vancouver, Calgary, Montreal, Toronto, Ottawa, Windsor, St. Johns, Fort McMurray, Yellowknife) for the period 2000-2009. A low optical depth (less than 0.3) is observed over most of the cities, higher aerosols optical depth more than 0.3 is found over Windsor and Toronto, that may be influenced by the nearby US cities. In general, the aerosol optical depth is found to be higher during summer season compared to winter season, an elevated carbon monoxide is observed over cities close to forests in British Columbia. The elevated carbon monoxide, ozone and aerosol optical depth is found to be associated with the forest fire events. Cities located in the eastern and western parts show contrast difference in the ozone column and aerosol optical depth during summer and winter season. The low and high variability of atmospheric properties during 2000 – 2009 will be discussed in view of the dust transport over Canada, anthropogenic activities and cold air mass from the northern sides.

P-Observations 2.27 ID:4268 **10:30**

PRELIMINARY OBSERVATIONS OF ATMOSPHERIC SURFACE OZONE, NITROGEN OXIDES, CARBON MONO OXIDE OVER INDIAN TROPICAL STAION, UDAIPUR

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Atmospheric surface ozone is the one of the leading positive radiative active forcing agent, trace gases which also acts as secondary pollutant. Its contribution towards as positive radiative forcing agent is the order of 17 to 20% of total earth radiative forcing effect and it is second rank candidate after the Atmospheric Carbon Dioxide trace gases. The concentration of ground level ozone primarily depends upon its precursors (CO, NOx, CH4, Nonmethane Hydro Carbon) as well as meteorological conditions and transport of air mass from far away polluted regions. Ozone is also a oxidizing agent, increasing concentration of which can alter the oxidizing capacity of the atmosphere and in turn can effect earth atmospheric chemistry and solar radiation energy budget or the earth climate. It also heath hazard for human beings, animals and plants if its exceeds certain limits. Recent reports on the basis of modeling studies have clearly shown that surface ozone is enhancing at rate of above 2% per year due to anthropogenic activities like increased of vehicular, energy consumption activities and industrial activities etc., over Urban and agriculture areas. Therefore, the regular information of ozone and its precursors on the ground levels is needed for setting the ambient air quality threshed levels and understanding photochemical air pollution in Urban and semi Urban area. In view of the above, the Surface Ozone Analyser (Thermo Scientific Make ,Model 49-i), Nitrogen Oxides Analyser(Thermo Scientific Model 42-i TL) and Carbon Monooxide (Thermo Model 48- TLE) analyzer have been installed at Department of Physics, University College of Science campus, Udaipur (Geo. Lat 24.6'N, Geo. Log. 73.6' E, Allti.,598) under the Research Project funded by Department of Science & Technology, New Delhi in November, 2009.The Ozone analyzer is calibrated using internal calibration unit and external Charcoal and Exhaust Unit ,the calibration procedure of Nitrogen Oxide analyzer is completed using permeation tube and oven and CO analyzer is calibrated using external Zero Air gas Cylinder and standard CO gas cylinder of 2ppm level. The real time and continuous round the clock observations of Surface Ozone, NO, NO2, NOx and CO have been carried out at each one minute interval using the above analysers. From the analysis of recorded data from Nov., 2009, the preliminary results of diurnal variations of O3 and its precursors in winter season over Udaipur will be presented in the paper.

P-Observations 2.28 ID:4431 **10:30**

Correlations between O3 and HNO3: Analysis for the ARCTAS field campaign using aircraft, satellite observations and Mozart model simulations

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Reactive nitrogen compounds play an essential role in the processes that control the ozone abundance in the lower atmosphere, in particular HNO3, which is one of the principal reservoir species for the nitrogen oxides. However, there remains a significant lack of data for simultaneous observations of O3 and HNO3, despite the fact that the correlations between these species are particularly important for characterizing air

masses and evaluating how ozone depends on nitrogen compounds. As a consequence, the chemical link between O3 and HNO3 remains poorly known in the lower layers.

In this study, we use aircraft observations of O3 and HNO3 from the NASA ARCTAS and NOAA ARCPAC campaigns during spring and summer of 2008 together with HNO3 and O3 satellite data from the IASI instrument and a global chemical transport model (MOZART) to better understand the sources, transport and variability of these compounds in the Arctic. The results are discussed in terms of O3-NOy chemistry, and the role of HNO3 as a reservoir of NOx is also investigated. These analyses also help us to quantify the contribution of the stratosphere to the tropospheric ozone budget in the Arctic.

P-Observations 2.29 ID:4326 10:30

Role of drop distortion in enhancing the lightning activity in clouds formed over cities

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Atmospheric pollutants are believed to modify the electrification and the lightning activity in thunderclouds. Results of a laboratory simulation experiment in which distilled/polluted water drops are suspended in a vertical wind tunnel to study their distortion characteristics in absence and presence of a horizontal electric field, are discussed in this context. Distortion of water drops is more when drops are formed from the water polluted with Ammonium Sulphate or Potassium Nitrate than that from distilled water and the difference in distortions is more in higher electric field. Further, the polluted water drops falling in horizontal electric field can trigger a discharge and the discharge can propagate as a streamer in lower electric fields as compared to that from distilled water drops. The difference in electrical conductivity of polluted and unpolluted water drops is most likely the key factor for manifestation of these differences. It is proposed that the enhanced distortion of polluted drops coupled with the change in their characteristics to trigger and propagate a discharge in lower electric field may significantly contribute to the enhancement of lightning activity observed in the clouds formed over big cities.

P-Observations 2.30 ID:4407 10:30

Measurements of NO2-profiles during the CINDI campaign

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Satellite measurements of tropospheric constituents have advanced significantly over the last decade, now providing near global measurements of a number of species relevant in tropospheric chemistry (e.g. CO, NO2, HCHO, SO2, CHO.CHO). With improving spatial resolution, these data sets move towards air pollution monitoring and even air quality assessment. However, the integrating nature of this type of remote sensing measurements makes the link to in-situ data as they are usually provided by monitoring networks difficult. This is a problem for both validation and interpretation of the satellite data. Multi-axis differential absorption spectroscopy (MAXDOAS) measurements can improve on this situation in several ways. Firstly, they can be used to provide directly the integrated tropospheric columns which are the quantity needed for validation. Secondly, they offer some vertical resolution, information which is needed in the retrieval of satellite data. Furthermore, their relatively high temporal resolution enables MAXDOAS to sample the diurnal cycle of trace gases which is not covered by satellite instruments, yielding insight in the photochemistry of atmospheric pollutants. However, a good estimate on the accuracy of NO2 tropospheric columns and profiles for different atmospheric conditions (clouds/aerosols) and viewing geometries using the MAXDOAS technique is still missing. Here the CINDI campaign held in June to July 2009 in Cabauw provides the unique opportunity to compare and validate the MAXDOAS NO2-profiles with in-situ, NO2-lidar and NO2-sonde measurements. Furthermore it is possible to intercompare the different MAXDOAS results and finally come to recommendations for harmonization / standardization of instruments settings and retrieval algorithms. In addition to the real data a MAXDOAS model study has been initiated to identify more clearly the pros and cons for each method and to elaborate more clearly the general limitations of this technique. Results from both studies will be shown and discussed.

P-Observations 2.31 ID:4203 10:30

Nitrogen oxides in the UTLS: Five years of observations from CARIBIC

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Since December 2004 a unique set of nitrogen oxides data has been obtained in the upper troposphere and lower stratosphere. These data have been acquired within the CARIBIC project (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container, www.caribic-atmospheric.com) on a monthly base using a Lufthansa Airbus A340-600. During nearly 200 flights between Frankfurt in Germany and airports in South America, Asia, North America and South Africa the sum of all reactive nitrogen species (NOy) and NO has been observed in the tropopause region.

Nitrogen oxides play a key role in atmospheric photochemistry, particularly in controlling the cycling of OH and the production of ozone in the upper troposphere and lower stratosphere (UTLS) and therefore have a strong impact on the radiative forcing of the atmosphere. The budget of nitrogen oxides in the UTLS is controlled by a variety of different sources and processes, chiefly: long-range transport, lofting from the boundary layer, lightning, and air traffic emissions.

The acquired data set is suitable for a reliable base assessing the atmospheric relevance of nitrogen oxides. The large scale seasonal and regional distribution of nitrogen oxides at the UTLS is presented and compared to other measurements and the results of model simulations. The data are analysed along with species as CO, O3, and aerosol particles. Tracer correlations are used to investigate the contribution of different sources on the nitrogen budget. Case studies show the influence of biomass burning events and air traffic emissions.

P-Observations 2.32 ID:4209 10:30

Measurement of atmospheric NH3, NO and NO2 at different environments over India

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Mixing ratios of atmospheric NH3, NO and NO2 were measured during 2008-10 over three different meteorological environments i.e. New Delhi (Delhi), Dibrugarh (Assam) and Thiruvanathapuran (Kerala) to study their diurnal, day-to-day variations, source strength and role in acid-base chemistry of atmosphere. NH3, NO and NO2 were measured continuously and precisely using NH3- and NOx-Analyzer, which operates on chemiluminescence method having estimation efficiency (>90%) is much higher than the chemical trap method (reproducibility 4.7%). Significant diurnal, day-to-day variations of mixing ratio of atmospheric NH3, NO, and NO2 were observed during the study period at all the locations. At Delhi, NH3 mixing ratio ranges from 1.02 – 46.17 ppb with average mixing ratio of 20.23 ± 9.42 ppb during winter whereas the mixing ratio ranges from 1.05 – 29.26 ppb with an average value of 17.47 ± 3.25 ppb during autumn. The mixing ratio of all measured trace gases varying significantly at all the locations might be due to source strength and meteorological conditions over the seasons. Result reveals that the mixing ratio of atmospheric NH3 is positively correlated (during winter) with the atmospheric mixing ratio of NO (r^2 = 0.79), NO2 (r^2 = 0.91) and negatively correlated with ambient temperature (r^2 = -0.32). Early morning increase in NO2 mixing ratio may be attributed to conversion of NO to NO2 with the interaction of O3. The surface wind direction and wind speed analysis indicates that major possible local sources of atmospheric NH3 are contributed from agricultural activities, road side traffic and industries.

P-Observations 2.33 ID:4476 10:30

NO2 variations over the ocean observed by MAX-DOAS

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Atmospheric aerosol and gas measurements by Multi-Axis-Differential Optical Absorption Spectroscopy (MAX-DOAS) on a Japanese research vessel, Kaiyo, were conducted during an ocean cruise over Japan-Guam-Bali-Indian Ocean from February to May 2009. Clear latitudinal variations of NO2 and aerosol extinction were observed by MAX-DOAS with maxima over Japan and Indonesia, as well as by satellite observations during the cruise. Relatively higher NO2 concentrations for 0-1 km were sometimes observed over the ocean (~0.25-0.4 ppbv) as compared to the background level (< 0.1 ppbv) partly due to advection of polluted air masses. The estimated error in NO2 mixing ratio (errors from the residual in the fitting and additional change in AOD) during the cruise was as small as ~20% (~0.02 ppbv), even in the low background value case over the ocean. We will also present the other trace gases variations, such as H2O, SO2, HCHO, and CHOCHO during the cruise.

P-Observations 2.34 ID:4544 10:30

The concentration profile of NO2, SO2 and O3 in Dar es Salaam, Tanzania and its link to atmospheric corrosion

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While many investigations on atmospheric pollution have been conducted in Europe, North America, South America and various parts of Asia, only a handful of such studies have been reported for Africa, especially Sub-Saharan Africa. Atmospheric pollution, which is associated with atmospheric pollutants, viz particulate matter and gaseous pollutants, cause a wide spectrum of effects, such as atmospheric corrosion of various materials. It is now an increasingly important environmental problem due to the increase of industrial activities, traffic, biomass burning, and the ever increasing demand for power, mostly supplied by coal power plants introducing air pollutants into the atmosphere. Atmospheric corrosion of local building materials, e.g. galvanized corrugated iron sheets widely used as roofing materials, along the coast of Dar es Salaam and neighbouring regions, is a major problem. As part of solving this problem, the ambient air quality to which the local building materials are exposed to, was determined by measuring concentration levels of NO2, SO2 and O3 gaseous pollutants at exposure sites, S1, S2 and S3 - aligned at intervals 0 km, 15 km and 40 km from the coastline over a period of 3 years to observe the influence of marine atmosphere. The average concentrations of NO2, SO2 and O3 from S1 (19.7, 3.5 and 25.3 µg/m3) during 2005 – 2007, in comparison to completed studies, are indicative of a polluted area. This is related to the meteorological conditions as well as the air mass movements, as shown by the backward air masses trajectories models. Possible sources for the concentration variations were identified for S1, S2 and S3. Atmospheric corrosion of construction materials confirmed the higher corrosivity of the coastal site compared to the inland site. Furthermore, an analysis of the corrosion products formed on the surfaces of galvanized roofing samples indicate a variety of corrosion products that can be formed, depending on the site atmospheric conditions.

P-Observations 2.35 ID:4323 10:30

Remote Sensing Measurement of 2-Dimensional NO2 and SO2 Distributions from Power Plant Stacks Using GIST Imaging-DOAS System

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Anthropogenic emissions of nitrogen and sulfur compounds from power plants account for significant portion of the total emissions to the atmosphere and are of great concern due to their impact on the atmospheric environment. This study includes remote measurements of these compounds to understand their subsequent chemical reactions in the atmosphere, which are important for quantitative assessment of their contributions to local and regional air quality. Slant column densities (SCDs) of power-plant-generated NO2 and SO2 were remotely measured using imaging differential optical absorption spectroscopy (Imaging-DOAS) with scattered sunlight as a light source. The vertical profiles of NO2 and SO2 SCD above the power plant stacks were simultaneously probed using a pushbroom sensing method. Measured SCDs were converted to mixing ratios to determine the rates of NO2 and SO2 increases at the center of the plume. Knowledge on the rates of NO2 and SO2 increases is important since SO2 and NOx compete for the same oxidizing radicals, and the amount of NOx is related to SO2 oxidation rate and sulfate formation. Here, we introduce a retrieval scheme to determine the rate of NO2 and SO2 increases in the power plant plumes using the I-DOAS technique and present the results from Taean thermal power plant. The derived mixing ratios measured on October 17, 2007 reached up to 12.6 ppm for NO2 and 30.1 ppm for SO2 with SCD values of 4.38 x 10e17 (NO2) and 4.44 x 10e17 (SO2) molecules/cm2, respectively. For validation of imaging-DOAS measurement results those derived NO2 and SO2 mixing ratios were compared to those measured by in-situ analyzers and continuous stack monitoring system.

P-Observations 2.36 ID:4310 10:30

MAX-DOAS Monitoring Network Observations of NO2 and Aerosols over Japan, China, Korea, and Russia

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Air quality change in Asia and surrounding regions is paid attention because of the rapid and substantial increase in the human activity in this region. We have established a long-term MAX-DOAS (Multi-Axis Differential Optical Absorption Spectroscopy) monitoring network over the region to provide vertically-resolved (or integrated) NO2 densities suitable for the validation of satellite data and chemical transport models. Standardized instruments were deployed at 6 stations: Cape Hedo (26.87N, 128.25E, starting in March 2007), Yokosuka, Japan (35.32N, 139.65E, April 2007), Gwangju, Korea (35.23N, 126.84E, February 2008), Hefei, China (31.91N, 117.16E, March 2008), Zvenigorod (55.70N, 36.78E, October 2008), and Tomsk, Russia (56.48N, 85.05E, January 2009). Scattered sunlight at 6 elevation angles (3, 5, 10, 20, 30, and 90 (sometimes 70) degrees) is received sequentially by a rotating mirror and then delivered to a temperature-regulated spectrometer by a bundle of optical fibers. A set of measurements at the 6 angles took 30 minutes. A single algorithm comprising of DOAS spectral fitting and inversion to yield vertical column densities (VCD) from slant columns was applied for all of the spectra and thus provided a systematic data set. Observations of an O4 band at 476 nm provided optical path information (and thus aerosol extinction) together with NO2 (460-490nm). A wide range of the observed tropospheric NO2 VCD ((0.5-60)x10**15 molecules cm-2) benefited the satellite data validation. Clear summertime minimum was found for the 3-year observations at Cape Hedo and Yokosuka. Generally-found daytime decreases were attributable to the photolysis, oxidation by OH, and strong emissions in the morning. However, daytime increases were found in winter in Yokosuka, suggesting transport from other polluted regions. The levels on Sundays were lowered for Yokosuka and Gwangju where transportation sector emission is dominant, while they were not for Hefei, suggesting that dominant sources and/or weekly emission patterns are different.

P-Observations 2.37 ID:4156 10:30

Optical properties of aerosols during dry and wet season over Amazonia

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The biogeochemical cycles, among many critical processes in land-atmosphere interactions, are of fundamental importance in the global radiative balance. In particular, aerosol particles change the solar radiation balance and atmospheric temperature profile, affects cloud microphysics, and has other important

effects. The Amazon Basin, as a huge ecosystem, is of particular interest because of its influence in the local and global climate. Aerosol particles have a strong connection with the carbon uptake in this area, through change in diffuse to direct radiation. As part of the LBA/BARCA experiment, an airborne campaign was performed for large scale measurements of the physical properties of the aerosol particles over the Amazon Basin. The campaign occurred during November/December-2008 (Phase-A) and May-2009 (Phase-B), covering a large part of the Amazon region. A non-pressurized turbo-prop Brazilian EMBRAER Bandeirante airplane was used, and more than 150 vertical profiles were obtained, up to 4000m, including over the tropical Atlantic Ocean. The aircraft was equipped for optical scattering measurements (TSI-Nephelometer), absorption (Thermo-MAAP5012), particle size distribution (TSI-SMPS), optical particle size distribution (OPC Lasair II), total particle concentration (TSI-CPC3010), and for trace gases measurements, including O3, CO, CO2, CH4, and N2O. Measurements of the aerosol vertical profile show a very well mixed atmosphere, with a weak signal from the top of the boundary layer, in particular during the wet season (Phase-B). Light scattering was about 2-6 Mm-1, at the boundary layer, going down to 1 Mm-1 at 3500m, and absorption coefficients was bellow 1 Mm-1, which is close to the detection limit of the MAAP instrument. The obtained data are being analysed in conjunction to greenhouse gases, and in terms of optical properties.

P-Observations 2.38 ID:4167 10:30

Spatio-temporal variability of light-absorbing carbon concentration in a residential area impacted by woodsmoke

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Residential wood combustion (RWC) is responsible for 33% of the total carbon mass emitted in Europe. With the new European targets to increase the use of renewable energy, there is a growing concern that the population exposure to woodsmoke will also increase. This study investigates observed and simulated light-absorbing carbon mass (MLAC) concentrations in a residential neighborhood (Lycksele, Sweden) where RWC is a major air pollution source during winter. The measurements analysis included descriptive statistics, correlation coefficient, coefficient of divergence, linear regression, concentration roses, diurnal pattern, and weekend versus weekday concentration ratios. Hourly RWC and road traffic contributions to MLAC were simulated with a Gaussian dispersion model to assess whether the model was able to mimic the observations. Hourly mean and standard deviation concentrations measured at six sites ranged from 0.58 to 0.74 µg m-3 and from 0.59 to 0.79 µg m-3, respectively. Both the temporal and spatial variability decreased with increasing averaging time. Low-wind periods with relatively high MLAC concentrations correlated more strongly than high-wind periods with low concentrations. On average, the model overestimated the observations by 3-5 fold and explained less than 10% of the measured hourly variability at all sites. Large residual concentrations were associated with weak winds and relatively high MLAC loadings. The explanation of the observed variability increased to 31-45% when daily mean concentrations were compared. When the contribution from the boilers within the neighborhood was excluded from the simulations, the model overestimation decreased to 16-71%. When assessing the exposure to light-absorbing carbon particles using this type of model, we suggest employing a longer averaging period (i.e., daily concentrations) in a larger area with an updated and very detailed emission inventory.

P-Observations 2.39 ID:4199 10:30

Highly sensitive, rapid in-situ optical NO2 detection for airborne peroxy radical measurements

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The first measurement campaign OMO (Oxidation Mechanisms Observations) of the new high-altitude long-range (HALO) aircraft aims at the study of oxidative processes of the upper troposphere / lower stratosphere (UT/LS region). Here, peroxy radicals (HO2 and RO2, R being an organic chain) are of major importance, and information about their concentration levels is essential to improve our understanding of the mechanisms of chemical processing in the troposphere. One of the techniques to detect the total sum RO2* of peroxy radicals ([RO2*] = [HO2] + [RO2]) is to quickly convert them to NO2 in a chain reaction (PeRCA Peroxy Radical Chemical Amplification). An amplification factor (called chain length CL) of 50-200 can be achieved depending on the ambient pressure; thus typical RO2* upper tropospheric concentrations of 10-50 pptv results in the formation of ~0,5-2,5 ppbv NO2. To overcome the limitations of a chemiluminescent detection scheme using luminol which usually prevents measurements below ambient pressures of less than 250 mbar, a novel optical detector based on cavity enhanced absorption spectroscopy (CEAS) and using optical feedback has been developed. Resonator calibration with ring-down time acquisition permits direct absorption coefficient measurements. A high finesse V-cavity with 40 cm mirror distance is employed; the mirror reflectivity of 99,993% results in an optical path length of 5,7 km. The detector operates with a continuous gas flow of 1 std.l./min. at a pressure of 100 mbar, which allows the resonator to be flushed with a t90/10 time of three seconds. A NO2 1σ-detection limit of 0,23 ppbv / (Hz)^0,5 (data rate of the detector: 9 Hz) is achieved which already allows for peroxy radical detection. However, the use of cavity mirrors with an even higher reflection is possible to boost this limit. A comparison with the previously used chemiluminescence luminol detector will be presented and both detectors' performance discussed.

P-Observations 2.40 ID:4170 10:30

CH4 and N2O emissions from a sphagnum peatland and a fertilized sandy soil using a new portable infrared laser spectrometer

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A new type of portable infra red spectrometer (SPIRIT : SPectromètre Infra-Rouge In situ Troposphérique) using a quantum cascade laser and a patented new long multipass optical cell has been set up for the simultaneous flux measurements of two Greenhouse Gases (GHG): nitrous oxide (N2O) and methane (CH4), at the air land interface. The basics of the instrument, the data derivation for trace gas concentration determination in the atmosphere, and the chamber method to derive emission fluxes of these GHG from lands are described. The analytical performances of SPIRIT are tested in two types of lands in Region Centre (France): (i) an anthropogenized sphagnum peatland (Laguette; Neuvy sur Barangeon) characterized by vascular plants invasion (ii) a sandy soil in the site of INRA-Orléans. The ability of SPIRIT to assess with precision spatial and temporal dependence emissions of these GHG in the field is demonstrated. In addition emission modes (diffusive episodes and bubbling events) can be observed and quantified due to the high frequency (1 Hz) of the concentration measured. SPIRIT adaptation for detailed process-oriented studies of GHG flux emissions is also demonstrated by the investigation of emission dependence as a function of biotic

and abiotic parameters (including diurnal cycle sensibility and emission modes); processes of C exchanges between different compartments of the biota can be studied. Such investigations are required for a better understanding of the lands to atmosphere exchange mechanisms of GHG and for the prediction of feedbacks on GHG emissions in response to anthropogenic or climate change perturbations of terrestrial ecosystems. Implications of SPIRIT in other air lands studies are also presented.

P-Observations 2.41 ID:4181 **10:30**

Microphysical Properties of Low Altitude clouds in East Asia

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In-situ measurements of cloud properties, namely, cloud droplet size distribution, liquid water content (LWC), and droplet shape information, were made using the Cloud Aerosol and Precipitation Spectrometer (CAPS) instrument (DMT, Boulder, CO) onboard the chartered King Air aircraft during the Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign, which was conducted over the East China Sea and Yellow Sea in March-April 2009. In total, 21 flights were conducted and cloud properties were measured during 9 maneuvers (7 flights) within stratus and stratocumulus clouds. Mean cloud droplet number concentrations (CDNC) were between 300 and 1900 cm-3, and they were greater by factors of 2 to 5 than those observed during previous stratus/stratocumulus measurements, such as those observed off the coast of California. In accordance with these high CDNC, mean effective diameters are systematically smaller than previous observations when data with the same LWC are compared. These results indicate that microphysical properties of stratus/stratocumulus clouds over the East China Sea and Yellow Sea in spring are quite unique. These high CDNC are consistent with high aerosol number concentrations in the accumulation mode observed below clouds during A-FORCE and high cloud condensation nuclei (CCN) concentrations previously observed at Gosan station on Cheju Island, Korea (33.28°N, 126.17°E) in spring.

P-Observations 2.42 ID:4561 **10:30**

Space-based observations of HCHO/NO2 from Aura's Ozone Monitoring Instrument during the 2008 Beijing Summer Olympics and Paralympics

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In preparation of the Beijing summer Olympic and Paralympic games, strict emission controls were imposed between July and September 2008 on motor vehicle traffic and industrial emissions to improve air quality standards for the competitors. We assess the effectiveness of these controls using the Ozone Monitoring Instrument (OMI) measurements of HCHO and NO2 column amounts, where the ratio serves as a proxy for ozone production sensitivity. The 2005-2007 OMI record of HCHO/NO2 show that Beijing is generally in a VOC-limited regime through-out most of the year. However, during the summer of 2008, when emission reductions were imposed, the HCHO/NO2 ratio was much greater than unity indicating a strong NOx-sensitive regime. This was due to dramatic reductions in tropospheric NO2 largely in response to vehicular restrictions within the province. This sustained enhancement of the HCHO/NO2 ratio over Beijing and transition to a strong NOx-limited regime is a unique feature in the OMI data record over eastern China.

OMI also measured summertime increases in HCHO concentrations of around 18% in 2008 compared to 2005 due to enhanced vegetation cover as part of Beijing's greening initiative for the Olympics. After the traffic controls were lifted NO2 concentrations returned to average values and the HCHO/NO2 ratio returned to a VOC-limited regime typical of the autumn-winter-spring seasons.

P-Observations 2.43 ID:4275 **10:30**

Airborne measurement of peroxy radicals in the African upper troposphere

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The radical chemistry reaches special significance in the free troposphere. In the presence of enough NO from lightning and aircraft emissions, peroxy radicals can be responsible for rapid and effective production of O3 in upper tropospheric layers and thus have a global impact. While radical field observations are still limited, model underestimation of HO2 radicals has been reported in different free tropospheric air masses. On top of this, unexpected high radical concentrations have been recently observed in various remote and polluted environments indicating the existence of unknown propagating radical reactions. In that context, uplifting of peroxy radical precursors from the boundary layer by convection can play a critical role in the processing of long lived trace gases and in determining the global O3 budget.

Airborne measurements of the total sum of peroxy radicals (RO2*=HO2+RO2) made within the European project AMMA in the upper tropical troposphere in Africa will be presented. The observed RO2* vertical distribution and mixing ratios are indicative of enhanced in situ production of peroxy radicals in the upper troposphere during convective episodes. Examples of air masses having different photochemical histories will be shown. RO2* mixing ratios of high variability and up to 50-60 pptv are observed in the outflow of mesoscalic convective sytems where O3 production rates up to 1.3 ppbv h-1 are calculated. In air masses affected by biomass burning plumes photochemical active layers with RO2* up to 50 pptv are detected.

P-Observations 2.44 ID:4327 **10:30**

Trends in total column ethane measured over south eastern Australia from 1996-2010

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Ethane is the second most abundant atmospheric hydrocarbon (after methane). Its major sources are biomass burning, vegetation and fossil fuels and its major sink is by reaction with OH· in the atmosphere. Ethane (C2H6) has an atmospheric lifetime of 2-3 months and a seasonal cycle that reflects the concentrations of atmospheric OH·, with C2H6 concentrations peaking in the winter (when OH· is low). Global surface measurements have shown a rapid decline in C2H6 concentrations from the mid 1980's to the current day that have been attributed to reduced leaks from natural gas & oil fields. These trends have been echoed in northern hemisphere total column measurements of C2H6 made from USA and Japan using Fourier transform infrared (FTIR) solar absorption spectra. However, previously published FTIR measurements of total column C2H6 from Lauder, New Zealand from 1993 -2000 failed to show a statistically significant trend in southern hemisphere C2H6.

Here we report the results of a 14 year time series of C2H6 total column measurements from solar absorption FTIR spectra acquired at Wollongong, Australia (34'S, 151'E). The downward trend in our data is in agreement with the global surface measurements confirming a decrease in southern hemisphere C2H6 over the last two decades.

P-Observations 2.45 ID:4183 **10:30**

Year long measurements and analysis of OVOCs at Mace Head, Ireland

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We present results from year long measurements (2008-2009) of a range of Oxygenated Volatile Organic Compounds (OVOCs) made at the background air station, Mace Head, Ireland. The OVOC measurements were made using a high frequency GC-MS pre-concentration system. In this work specific attention is applied to understanding how the daily, weekly, and seasonal trends in OVOCs manifest themselves at Mace Head, with a specific focus on acetone (CH3COCH3). The location of this site allowed for clean-air background measurements of acetone to be made as well as measuring elevated mixing ratios of acetone during times when the air masses had travelled from Europe. An estimated lifetime of 22.5 days (due to both reaction with OH and photolysis, based on an OH concentration of 9.1x10-5 molec cm-3, rates of reaction taken from MCM) means that long range transport of acetone is possible. Overall, a median mixing ratio of 297 ppt was seen. Acetone was shown to peak during the summer months, with a median mixing ratio of 691 ppt being measured, compared with a low median value during the winter months of 209 ppt. Acetone was shown to increase with increasing temperature, with high mixing ratios of greater than 10 ppb correlating with temperatures at Mace Head of greater than 20 °C. The periods during which the air masses had travelled over Europe were isolated and a photochemical trajectory model was used to identify the key emission sources and reaction processes that contributed to the mixing ratios seen during these times. It was shown that during several case studies, direct emissions of acetone contributed the most to the mixing ratios seen at Mace Head, averaging 75 % during most events, compared with a 25 % contribution from secondary production.

P-Observations 2.46 ID:4250 **10:30**

Seasonal and diurnal variations of volatile organic compounds from tall tower measurements in the Upper Midwest

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Diurnal and seasonal cycles in concentration provide constraints on sources and sinks of volatile organic compounds (VOCs), which play a key role in atmospheric chemistry as precursors of ozone and secondary organic aerosol. Here we present VOC measurements from our ongoing field campaign at the University of Minnesota Trace Gas Observatory, a 244 m Ameriflux tall tower 28 km south of the Twin Cities, Minnesota, USA. VOC concentrations are measured by proton-transfer mass spectrometry (PTR-MS) at a sampling height of 200 m above ground level, providing a high-resolution signal with a regional-scale footprint to examine the importance of both biogenic and anthropogenic sources. We use wintertime tracer-tracer correlations to estimate direct anthropogenic VOC emission rates, and apply the observed diurnal and

seasonal variations to better understand the processes and seasonality driving biogenic and anthropogenic VOC emissions.

P-Observations 2.47 ID:4127 10:30

Vertical variations of polycyclic aromatic hydrocarbons in PM10 samples in Bangkok atmosphere

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Vertical variations of polycyclic aromatic hydrocarbon (PAH) concentrations in PM10 were investigated in order to assess the factors controlling their behaviors in urban atmosphere of Bangkok city, Thailand. Air samples were collected every three hours for three days at three different levels (i.e. site-1:38 m; site-2: 158 m; site-3: 328 m above the ground level) at Bai-Yok Suit Hotel (site-1 and site-2) and Bai-Yok Sky Hotel (site-3) in February 18th-21st 2008. PAH extraction was conducted by Soxhlet in dichloromethane and quantification by GC-MS. The total PAH concentrations measured at site-1, site-2 and site-3 were 12.158 ± 11.504 ng m-3, 5.951 ± 7.346 ng m-3and 2.776 ± 2.182 ng m-3 respectively. The B[a]P concentration showed, at ground level, value 3.11 fold higher than the United Kingdom Expert Panel on Air Quality Standard (UK-EPAQS; i.e. 0.25 ng m-3). In contrast, the B[a]P concentrations exhibited, at middle and top level, values 1.41 and 0.54 fold of the UK-EPAQS. PAHs displayed a diurnal variation with maximums in day time (06.00 am – 09.00 am) because of the traffic rush hour. Pearson correlation analysis, t-test and binary ratio analysis of PAHs revealed following results; 1. Traffic emissions appear to be responsible for diurnal variations of PAHs at all levels, 2. Meteorological conditions such as wind speed and solar radiation may responsible for the diurnal variations of PAHs at the middle and top levels.

P-Observations 2.48 ID:4242 10:30

Relationships between the surface layer dynamics and ambient VOCs concentrations in an urban area influenced by industrial emissions

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The transport and dispersion of pollutants depends on the temporal and spatial variety of sources, photochemical reactions and meteorological phenomena, particularly on a local scale. The specific gas marker measurement associated with the characterization of the surface layer behaviours may constitute a tool to study industrial emission dynamics. Our objective is to develop a methodology to examine the interdependence of the behaviours of specific type of industrial sources and the influences of meteorological conditions. Meanwhile, this paper will focus on the evolution of the concentration of Volatile Organic Compounds (VOCs) as a function of dynamic parameters. A measurement campaign of 90 VOCs, among which 20 are oxygenated volatile organic compounds (OVOC), was performed by using gas chromatography (used in an on-line mode) during July and September 2009 at two urban sites exposed to industrial emissions in the Urban Community of Dunkerque, in northern France. The VOC concentrations were examined by taking into account several meteorological parameters (wind directions, atmospheric stability…). The ambient VOC pollution is not only predominated by wind direction, but also by turbulence intensity and the highest levels were recorded for slightly unstable conditions. Different behaviours of some VOCs in the same wind direction allow the nature of different sources to be identified. And so, from this analysis, it's

possible to distinguish between pollutants emitted from chimneys and those due to surface emissions. These specific markers will make it possible to use a positive matrix factorization (PMF) modeling to calculate the contribution of an industrial source and to study the evolution of this contribution in regard of meteorological conditions.

P-Observations 2.49 ID:4537 10:30

Observations of BVOC (Biogenic Volatile Organic Compound) Fluxes and Vertical Gradients in a Ponderosa Pine Forest during BEARPEX 2009

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During summer 2009 an intensive field campaign (Biosphere Effects on AeRosols and Photochemistry EXperiment - BEARPEX) took place in Blodgett Forest, a Ponderosa pine forest in the Sierra Nevada Mountains of California. The campaign was aimed to understand the biosphere-atmosphere interactions during a period of intense photochemical activity, to elucidate the fate BVOC (Biogenic Volatile Organic Compounds) in the atmosphere, and explore the processes of secondary organic aerosol formation. In this study, a PTR-MS (Proton Transfer Reaction – Mass Spectrometry) was used to measure 19 compounds (masses) including methanol, isoprene + MBO (2-Methyl-3-butene-2-ol), monoterpenes, sesquiterpenes, and some oxygenated BVOCs at 5 heights of a vertical gradient from the forest floor to above the canopy. Fluxes of the 4 dominant BVOCs were measured above the canopy with the Eddy covariance technique. In parallel with BVOC measurements, ozone fluxes and gradients, and meteorological parameters (PAR, temperature, relative humidity, wind speed, and wind direction) were recorded in order to test the dependence of BVOC from meteorological conditions and to test the hypothesis that BVOC remove atmospheric ozone through gas-phase reactions. Data will be compared to previous from the same site collected at different periods of the year to explore the dynamics of BVOC concentration and oxidation products as a function of varying environmental conditions.

P-Observations 2.50 ID:4366 10:30

Size-resolved Aerosol Measurement and Its Light Attenuation Characteristics in Seoul, Korea

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Aerosol can scatter and absorb the light, which cause visibility impairment in the atmosphere. Light attenuation efficiency depends on aerosol size distribution, chemical composition, and relative humidity. In order to investigate the amounts of light attenuation efficiencies for PM1.0, PM2.5, and PM10, several intensive visibility monitoring conducted from May 2007 to Nov. 2009 in the urban area of Seoul, Korea. Semi-continuous 2-hour aerosol sampling produced time-resolved chemical composition for visibility impairing aerosol and real-time optical measurements made a light extinction coefficient, a light scattering coefficient, and a light absorption coefficient using a transmissometer, a nephelometer, and an aethalometer, respectively. 455 data sets were used to determine the light extinction efficiencies for size-resolved aerosol. From the results, visibility varied with chemical compositions of PM1.0, PM2.5, and PM10.And under consideration of water growth function f(RH), the light extinction efficiencies of PM1.0, PM2.5, and PM10 were estimated to be 8.6, 4.5, and 2.7 m2/g respectively.

P-Observations 2.51 ID:4497 10:30

Multi-site Characterization Of Aerosols: Implications To Radiative Forcing

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Extensive land-campaign measurements have been performed using a suit of major instruments involving AERONET Sun/Sky sun-photometer, MICROTOPS-II, shortwave pyranometer from December 1 2006 to April 30 2007 as a part of the Indian Space Research Organization-Geosphere Biosphere Program (ISRO-GBP). The observation sites spread over a wide geographical area from central to southern tip of peninsular India, and include Ahmedabad (23.02N, 72.52E), Pune (18.53N, 73.8E), Sinhgad (18.36N, 73.75E), Gadanki (13.45N, 79.17E) and Trivandrum (8.53N, 76.86E), representing arid, urban, high-altitude, rural and marine environments respectively. At all locations, the aerosol optical depth (AOD) showed a significant day-to-day variability. The Angstrom exponent exhibited values ranging between around 0.4 (over arid region) and 1.8 (over rural region). The retrieved volume size spectra over all the stations during the period of study indicated bi-modal distribution with varying concentration for each mode. Interestingly, very low concentration of coarse-mode particles was noticed over the marine site (Trivandrum) even up to a distance of about 200 meter from the coast. The maximum (0.84) and minimum (0.73) single scattering albedo (SSA) values at 440nm are observed at Trivandrum and Ahmedabad, respectively. The difference among SSA values at 440 and 1020nm is found to be higher for Trivandrum and less for Ahmedabad. The synergy of results from these complimentary measurements is reflected in the computed regional aerosol radiative forcing and heating rates using the radiative transfer model (SBDART), which can serve as indicators for understanding the regional climate change scenarios.

P-Observations 2.52 ID:4441 10:30

ANALYSIS SEASONS OF 60 YEARS OF SOLAR RADIATION DATA FROM BUENOS AIRES ARGENTINA, 1949-2009.

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Measurements of global radiation at the surface have a great importance for the evaluation of climate change and global warming, because they are potentially sensitive indicator of anthropogenic disturbances. Tenders in Buenos Aires Argentina Global Radiation and clearness index for the period 1949 to 2009 was evaluated on data for daily surface solar radiation. Season means for two variables were calculated for the Buenos Aires observatory. Linear regression analysis was used to characterizer long-term season tender in these variables. The preliminary results show significant decrease in global radiation and clearness index during all season, the largest slope is present at summer and lowest at winter. Although there is some evidence that conditions have improved in the last decades, the consistent spatial and temporal variation of these variables support of theory that increased of aerosols loading were at least partially responsible for the observed decreases in the global radiation and clearness index over Buenos Aires. The radioactive forcing caused by clouds, greenhouse gases and aerosols has been major topic of concern for the intergovernmental Pane of Climate Change.

P-Observations 2.53 ID:4340 10:30

Measurement of chemical composition and optical properties of aerosol particles at Fukue Island at Japan in spring 2009

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An intensive field campaign was carried out in Fukue Island (32°5'N, 128°4'E) in spring 2009. Fukue Island is located at western end of Japan. To investigate chemical and optical properties of aerosol particles, PM2.5 samples were collected on the quartz filters using a high-volume (500L/min) air sampler. Using these filters, polycyclic aromatic hydrocarbons (PAHs), water-soluble ions and metals were measured by high performance liquid chromatography (HPLC), ion-chromatography, and Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES), respectively. Also, EC (elemental carbon) and OC (organic carbon), total mass concentration and scattering coefficient for PM2.5 were measured by semi-continuous EC/OC instrument, synchronized hybrid ambient real-time particulate monitor (SHARP monitor),and nephelometer, respectively . During the campaign, the total PM2.5 mass concentration ranged from 3 to 35 ug/m3. By comparing those results, we will discuss the variation of composition, mass closure, and relationship between scattering coefficients and chemical species for PM2.5.

P-Observations 2.54 ID:4466 10:30

Long-term measurements of aerosol optical properties in Amazonia

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For the first time in Amazonia, a ground based long-term aerosol experiment was conducted, in the framework of the European Integrated project on Aerosol Cloud Climate and Air Quality Interactions (EUCAARI). Continuous measurements of aerosol properties were performed for two years at the Cuieiras forest reserve, 60 km NNW of Manaus in Central Amazonia. Measurements of aerosol scattering, absorption, size distribution, mass and number concentration, among others, were taken under dry conditions (< 40% RH). Aerosol concentration and optical properties showed strong seasonal variability, with higher loadings observed during the dry season (Jul-Oct), at the onset of the biomass burning period, in comparison to the wet season (Nov-Jun). Although the forest site is preserved, measurements have shown that the location was occasionally affected by regional transport of polluted air masses, either from biomass burning or urban plumes. From wet to dry season, aerosol scattering coefficients increased from 10 Mm-1 to 100 Mm-1, while PM10 aerosol mass concentrations increased from 10 ug/m³ to 40 ug/m³, in average. Coarse mode particles dominate the volume size spectra, either at dry or wet season. Based on Mie theory, closure calculations between aerosol scattering, absorption and size distribution observations indicate that coarse mode aerosols, possibly from biogenic origin, have a significant impact on aerosol extinction and single scattering albedo in Amazonia. Occasional long range transport of Sahara soil dust also affects the observed aerosol optical properties. In situ observations are being compared to atmospheric column observations (LIDAR, sun-photometers). These analyses provide a detailed description of the optical properties of Amazonian aerosol, as well as a better comprehension of its potential influence over regional and global

climate.

P-Observations 2.55 ID:4272 **10:30**

Measurements on atmospheric aerosols and of the Aerosol Optical Depth during 2006 at Uccle, Belgium

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From January through April and from July through November 2006 continuous measurements of the AOD were performed at Uccle with a Brewer ozone spectrophotometer and PM2.5 and PM10 filter samples were collected for mass measurement and chemical aerosol characterisation. Samples were collected during the daytime only and on days with no or few clouds. There were a total of 109 sampling days. The boundary layer mixing height was estimated from ECMWF data and the AOD data and mixing heights were averaged over each filter sampling. The range of the daily mean AOD values (at 320 nm) was 0.05–1.08 and the AOD values were clearly higher in spring and summer than in winter and fall. The median for the sample-averaged mixing height was 870 m and it was substantially higher in spring and summer than in fall and lowest in winter. The median PM2.5 mass level was 14 $\mu g/m^3$ (range: 3–70 $\mu g/m^3$) and the median was much larger in winter (i.e., 35 $\mu g/m^3$) than in the other three seasons (10–14 $\mu g/m^3$). The relation between the daily mean AOD values and the PM2.5 and PM10 mass data was poor. It was estimated to which extent our daily mean AOD data could be explained by the boundary layer aerosol. To this end, the PM2.5 aerosol mass data were multiplied by the sample-averaged boundary layer height and a mass extinction efficiency of 5 m^2/g. The calculation indicated that the boundary layer aerosols contributed, on average, for 23% to the vertical column-integrated AOD and that the contribution for the individual days ranged from 6% to 63%. There was a slight tendency for a larger contribution of boundary layer aerosols during winter than during the other three seasons. Secondary inorganic aerosols was the major component in each season for both PM2.5 and PM10.

P-Observations 2.56 ID:4451 **10:30**

Submicron aerosol over the central Pacific: observations of non-refractory composition under clean marine conditions

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Understanding aerosol concentrations that constitute a clean versus polluted marine atmosphere is essential for understanding how human perturbations are affecting current aerosol loadings. As a result, diverse studies have attempted to quantify background atmospheric conditions from land and ship-based sites. However, many of these sites are subject to potential influences from local and/or long range transport. Here we show preliminary results from three studies conducted in the remote central Pacific suggesting lower organic and ionic concentrations than results obtained from other sites. Submicron aerosol properties were measured aboard the NCAR C-130 aircraft over the southeast Pacific during the Pacific Atmospheric Sulfur Experiment (PASE) in September/October 2007 out of Christmas Island (10°30'S, 105°40'E), over the

southeast Pacific out of Arica, Chile during the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx) in October/November 2008, and over the central Pacific aboard the R/V *Ka'imimoana* during August and September 2009. All experiments used a High Resolution – Particle Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS) along with other aerosol measurements. During VOCALS the lowest median concentrations of chemical species such as aerosol sulfate (0.13 µg m^{-3}), organics (0.06 µg m^{-3}), and black carbon mass (4.9 ng m^{-3}), as well as gas-phase CO (60 ppb), were encountered in the marine boundary layer along 20°S and west of 78°E. We believe these low values can be attributed, at least in part, to collection taking place 1) in the southern hemisphere, where there is less population and landmass, 2) over a remote area of the ocean, far removed from continental influence, and 3) over the oligotrophic ocean, therefore less primary and secondary organic aerosol. Using Black Carbon and CO to establish background conditions, the data from the three campaigns are shown and will be discussed in terms of describing the meteorology and aerosol chemistry that constitute these background conditions.

P-Observations 2.57 ID:4519 10:30

Characterization of water soluble ionic and carbonaceous aerosol over Bay of Bengal during ICARB-W

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To characterize the physico-chemical properties and radiative effects of atmospheric aerosols and trace gases over Bay of Bengal, an extensive experiment "Integrated Campaign for Aerosols, Gases and Radiative Budget "(ICARB) was carried out under the Geosphere Biosphere Program of the Indian Space Research Organization (ISRO-GBP) during winter season 2009. In the present study, concentration of water soluble ions such as NH4+, SO4--, NO3-, Cl-, F-, Na+, K+, Ca2+ and Mg2+ were estimated. The average concentration of NH4+, SO4--, and NO3- over BOB found to be 1.96, 8.68, 1.92, µg/m3 respectively. The percentage of non sea-salt fraction (nss) and sea salt fraction (ssf) was estimated to understand the contribution of major ions other than the marine sources. Dominance of non sea salt SO4-- was observed over this region. Concentration of SO4-- was found to be positively correlated with NH4+ (r2=.676) similarly concentration of NH4+ found to be positively correlated with NO3- (r2=.867) which may indicate the presence of ammonium sulfate and ammonium nitrate as major water soluble particle. Higher concentration of organic carbon (OC) is found in the Indian costal area which ranges from 10 -16 µg/m3, this may be due to strong influence from the Indian continental air masses. In the mid of Bay of Bengal concentration of OC ranges from 4-6 µg/m3 whereas ambient concentration of OC over eastern part of BOB ranges from 4-12 µg/m3. Similarly, the trend of Elemental carbon was found same like OC but with lower values.

P-Observations 2.58 ID:4532 10:30

A study of optical properties of Amazon Basin aerosols using remote sensing and in situ measurements

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The Amazon tropical rainforest constitutes a unique and complex ecosystem, and an important source of aerosol particles for the global atmosphere for both dry and wet season. Remote sensing techniques are extremely useful because of the high spatial coverage and recent instruments are dedicated to the study of aerosols and clouds, and their influence on climate. Satellite measurements usually present very large uncertainties when compared to in situ measurements. We have analyzed data from 2000 to 2007 acquired by the radiometers CERES and MODIS onboard the Terra satellite, which together provide shortwave and longwave radiative fluxes on the top of the atmosphere, aerosol optical depth and surface reflectance among other critical data. For the year 2000, a combined analysis of CERES and MODIS provides a value of -14 + or - 5 W/m2 for the shortwave direct radiative forcing of the whole Amazon region during the dry season. For the in situ validation, we have used the AERONET sun photometer network data, which provides very detailed information on aerosol distribution and optical properties. We have used local solar flux measurements from pyranometers from the SolRad-Net network, which are collocated to each sun-photometer. In an attempt to reduce the uncertainty of the calculated radiative forcing we have used the radiative transfer code SBDART and the above mentioned in situ measurements. We have also compared CALIOP and AERONET AOD retrievals for four Amazon sites within 50km and 100km radius for three years, and have found that CALIOP consistently underestimates the AOD. The observed bias is a high -47%. This bias seems to have occurred due to the proximity of clouds near the area where measurements were taken and also due to the misidentification of biomass burning aerosols with clouds.

P-Observations 2.59 ID:4562 **10:30**

The Organic Fraction of Aerosols in the Caribbean

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As part of the Rain in Cumulus over the Ocean Experiment, we investigated the organic fraction of aerosol particles collected simultaneously at two tropical marine sites in the Caribbean region: Dian Point, Antigua and Cape San Juan, Puerto Rico. Two size-resolved low-pressure impactors (Dp from ~0.054 to ~18 μm), stacked-filter units (Dp < 1.7 μm) and a high-volume sampler were used to collect the samples. Thermal-optical analysis was used to determine the concentrations of aerosol total carbon, organic carbon, and elemental carbon. Neutral compounds, mono-, di-, and poly-acids were analyzed using an HPLC-TOC and acetate, formate, malonate, and oxalate using ion chromatography. The water-soluble organic carbon (WSOC) was determined with a total organic carbon (TOC) analyzer. The EQSAM3 (EQuilibrium Simplified Aerosol Model) was used to determine the water uptake of the collected aerosol samples. Air masses influenced by marine (M) aerosols, African dust (AD), and anthropogenic pollution (AP) from North America were studied. Particulate organic matter (POM) was up to 12% of the total aerosol mass and up to 27% of the non-sea salt aerosol mass, and its concentrations were ~80 ng/m^3 on average during the M and AP periods. WSOC as well as oxalate tend to be higher for the fine fraction particularly during the presence of AD. POM previous results generated also in Puerto Rico showed up to 4 times higher concentrations of POM. These discrepancies might be due to seasonal variations or a long trend. Results showed that the chemical composition of the organic fraction of aerosols, its contribution to the total aerosol mass, and the effect they have on cloud properties are still not well understood. Answer to questions such as the organic aerosols' temporal/spatial variation, dependence on the seasonal biological activity, mixing state, and sources are needed to fully understand their role in the Caribbean climate and their true extent and origin.

P-Observations 2.60 ID:4450 **10:30**

PARASOL Satellite Observations of Aerosol Distributions over Mexico: Methodology for the Evaluation of Regional Modeling (CHIMERE)

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Aerosols have significant radiative and environmental impacts, affecting human health, visibility and climate. Therefore, they are regulated by air quality standards worldwide, and monitored by regional observation networks. Even though remarkable progresses in aerosol modeling by chemistry-transport models (CTM) and measurement experiments have been made in recent years, there is still a significant divergence between the modeled and observed results. In this context, satellite observations offer very interesting perspectives, with good horizontal and temporal coverage. The observed distributions are generally characterized using aerosol optical depth (AOD) retrievals. However, these AOD retrievals remain a highly challenging task, mostly because it depends on a variety of different parameters such as cloud contamination, surface reflectance contributions and a priori assumptions on aerosol types, each one of them incorporating its own difficulties. Therefore, comparisons between CTM and observations are often difficult to interpret. In this presentation, we will discuss comparisons between regional modeling (CHIMERE CTM) over Mexico and satellite observations obtained by the POLDER instrument on board the PARASOL micro-satellite. After a comparison of the model AOD with the retrieved AOD, we will present an alternative methodology for direct comparison to the measured reflectance. Preliminary evaluation of the CHIMERE simulation over Mexico will be presented.

P-Observations 2.61 ID:4525 **10:30**

Determination of carbonyl compounds in airborne particulate matter (PM10) from different regions of the São Paulo state.

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Most of the research to date has focused on urban areas,where organic emissions are large and have dramatic effects on air quality and particulate mass concentrations. The atmospheric chemistry of non-urban areas represents an important area of study, since very little is known about the influence of mixed environments. The n-alkan-2-ones and n-alkanals are a group of oxygenated compounds that is present in the particulate matter. Some published papers have shown that they are produced from direct emission from the vegetation wax or from oxidative processes, cooking smoke, wood burning smoke, and automobile exhausts. There are few studies about these compounds worldwide, and never were studied before in Brazil, where the burning of different fuels (gasohol, biodiesel) has produced them. Samples of particulate matter (PM10) were collected in the state of São Paulo (Brazil): (i) University City - SPA, urban site with heavy traffic of light and heavy vehicles, (ii) ESALQ - PRB, region impacted by agricultural activity and the burning of sugar cane, (iii) Core Forest Santa Virginia - MAT, a region with low human impact. The PM10 concentration and air mass back trajectories showed that the urban site SPA and the forest site MAT receives influences of the biomass burning region. These average concentrations exceeded the daily levels recommended by the World

Health Organization (MP10 = 50 µg m-3), although they are within the limits of the Brazilian legislation CONAMA Nº 3 28/06/1990 (PM10 = 150 g m-3), both for daily patterns. The correlation between the n-alkanes (precursors; previously determined by the group) and nalkanals (product) through the process of oxidation was observed in SPA 2008 and PRB 2007, confirming the importance of the secondary aerosol formation. The same behavior is not observed for n-alkan-2-ones and probably, the n-alkan-2-ones were enriched by direct anthropogenic emissions.

P-Observations 2.62 ID:4237 10:30

WATER SOLUBLE AND INSOLUBLE COMPONENTS IN FINE AND COARSE AEROSOLS COLLECTED IN THE METROPOLITAN AREA OF BUENOS AIRES, ARGENTINA

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The content of metals and metalloids in urban particulates is receiving increasing attention due to their impact on human health and the environment. Total content of trace elements has become insufficient to evaluate overall pollution levels because the effect of trace elements in the environment strongly depends on the association form in the solid phase to which the element is bound. To this end, a two step chemical sequential leaching procedure was used to establish the distribution of Al, Ba, Ca, Cr, Fe, Mn, Pb, Se, Ti and Zn between water soluble and residual fractions. Concentrations of ions such as Cl-, SO42-, Na+ and NH4+ were determined in the soluble fraction. A total of 113 coarse (PM2.5-10) and fine (PM2.5) samples were collected during one year in Buenos Aires, Argentina using a GENT SFU, in a sampling site influenced mainly by traffic and residential sources. A data analysis and receptor model has been carried out to elucidate the potential origin of the elements of interest. In general, for both particles sizes, element concentrations in the soluble fraction were below 50%, while Se (PM2.5-10) and Mn (PM10) exhibited the highest levels (~ 40%). Concentrations of toxic or potentially toxic elements namely, Cr, Mn and Pb were below levels of concern. As expected, geological elements such as Al (0.02 – 0.084 µg m-3 for PM2.5-10 and PM10 respectively), Ca (0.968 – 1.35 µg m-3) and Fe (0.076 – 0.229 µg m-3) were mainly detected in the residual fraction which would be indicative of being environmentally immobile elements. The presence of Cl-, SO42- and Na+ made an important contribution to the water soluble fraction. The significant correlation between Na+ and Cl- suggests that there are mainly originated from sea salt.

P-Observations 2.63 ID:4520 10:30

Impact of megacity aerosol emissions on air quality and climate in the Met Office climate model HadGEM2

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Megacities are a substantial source of anthropogenic pollution. This affects the air quality of hundreds of millions of people both within the cities themselves and in the wider world. The megacity pollution can also have an impact on global climate, either directly or via complex interactions within the Earth system.

The impact of megacities on particulate burden, surface concentration, deposition and resultant radiative forcing are determined using the Met Office Hadley Centre climate model (HadGEM2). The study perturbs aerosol and aerosol precursor emissions from a selection of megacities as defined by the EU MEGAPOLI project. In addition to large cities this definition includes, conurbations (e.g. Boston-New York-Washington)

and highly industrialized regions (e.g. the Po valley). The specific megacities in this study are chosen to provide a global sample. From this, the differences due to the location of the megacities can be investigated. The remote responses away from megacity perturbations are considered in addition to local effects. Aerosol types investigated include sulphate, nitrate, black carbon and organic carbon.

P-Observations 2.64 ID:4334 10:30

Characterization of solvent-extractable organics in urban aerosols using a high resolution time-of-flight aerosol mass spectrometer and a hygroscopicity tandem differential mobility analyzer

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To investigate the characteristics of atmospheric particulate organics with different polarities, aerosol samples collected in the urban area of Nagoya, Japan, in spring and summer 2009 were extracted with water and organic solvents and subjected to the analyses using a high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) and a hygroscopicity tandem differential mobility analyzer. The aerosol samples were collected on quartz fiber filters (50% cut-off diameter: 0.95 μm) and extracted with water, methanol and ethyl acetate. Particles introduced to the instruments were generated by atomization of the extracts. The absolute concentrations of the solvent-extractable organics were determined from additional HR-ToF-AMS analyses for particles generated from the extracts with known amounts of phthalic acid. The concentrations of the organics extractable with water, methanol and ethyl acetate were in the ranges of 1.9–5.4, 2.6–7.1, and 1.4–5.0 $\mu g\ m^{-3}$, respectively. Further, the oxygen-to-carbon ratios were determined to be in the ranges of 0.51–0.71 (water), 0.31–0.48 (methanol), and 0.27–0.32 (ethyl acetate) from the millimass spectra. The quantification of water soluble fractions in ethyl-acetate extractable organics using the HR-ToF-AMS and the comparison with the organic carbon concentrations measured using a carbon analyzer suggest that a major part of the organics is extractable with water and/or ethyl acetate. From the hygroscopic growth measurements of water and ethyl acetate extracts, the κ values of the extracted organics were calculated, with the consideration of the water uptake by inorganic salts using the AIM model (http://www.aim.env.uea.ac.uk/aim/aim.php). By excluding the samples with large nitrate fractions to minimize a possible artifact caused by the evaporation, we determined κ of extracted organics to be from 0.00 to 0.16 at 85% relative humidity (RH). The aerosol water retained by these organics was estimated to comprise up to 70% of the total aerosol water at 85% RH.

P-Observations 2.65 ID:4262 10:30

Chemical Composition of aerosols over the marine environments of Bay of Bengal and Arabian Sea

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Aerosols in the marine atmosphere primarily compose of in situ generated sea-salt particles and non-sea-salt particles, formed from oceanic biogenic activities or transported from continental regions. This paper presents the results of the ship-based measurements conducted as part of the Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB) under the Geosphere Biosphere Programme of Indian Space Research Organization (ISRO-GBP). Aerosol sampling has been carried out during the premonsoon months of March to May 2006 over Bay of Bengal(BoB) and Arabian Sea(AS) and during winter months of

December 2008-January 2009 over BoB to understand the mass concentration and chemical composition of aerosols. Aerosol mass loading as well as that of various chemical constituents showed distinct spatial variation over BoB and AS. Aerosol mass concentration is found to be high during winter period. While the effect of advection from nearby landmass is more pronounced over BoB, the wind-induced sea salt production is found to be more significant over Arabian Sea during premonsoon. The total aerosol mass concentration as well as the concentration of individual chemical components showed enhancement over the northern BoB due to advection of continental aerosols from Indo Gangetic Plains as revealed by the chemical composition of aerosols and the airmass back trajectories. Based on these measurements mean chemical models are evolved for both the oceanic regions. While the sea-salt aerosols contributed ~31% of the aerosols mass over AS, it was ~11% over BoB. Abundance of nss-SO4 has been observed over both the oceanic environments, with higher concentration over BoB, revealing significant anthropogenic influence. Mineral dust contribution is fairly uniform in both these oceanic environments. These models have been compared with those obtained from previous campaigns over Indian land mass and oceanic regions.

P-Observations 2.66 ID:4418 **10:30**

Atmospheric pollution in Dakar: focus on aerosols in the frame of the POLCA program

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Black carbon (BC) were measured from April 2008 to July 2009 at 2-mn intervals using a seven-wavelength Aethalometer at an urban site in Dakar (Senegal), within the framework of the POLCA program (French acronym for "POLlution des Capitales Africaines"). Daily mean [BC] values varied from about 1000 to 20000 ng m-3. However, short-term spikes exceeding 50000 ng m-3 were commonly observed, mainly occurring during the morning and evening rush-hour periods, suggesting the paramount role of traffic. BC values were highest from November to April (13000 ng m-3) and lowest from May to August (6000 ng m-3). Diurnal, seasonal, and mondays to Thursdays/Fridays/saturdays and sundays trends are discussed. Differences in BC diurnal variations in Dakar and Bamako (two POLCA sites) reveal that BC sources strongly vary at regional scale. It appears that BC levels in Dakar were lower than at other West African capitals (e.g. Bamako), but comparable to reported values for European and Asian mega-cities. BC concentrations, as measured by Aethalometer, is comparable with measured BC on filters using a DRI analyzer (thermo-optical method), though consistently about 1.8 times higher. Using Al content of aerosols, Bowen ratio (Bowen, 1966) and other aerosol chemical contents (Galy et al., 2007), we have reconstructed PM2.5 mass concentrations in Dakar. Average PM2.5 mass concentrations for the study period from 16 November 2007 to 11 February 2008 were 29 µg m-3, well above the WHO threshold of 10 µg m-3. This paper clearly highlights high pollution levels in African large cities with ensuing important impacts on population health. More detailed results will be issued from the intensive POLCA campaigns.

P-Observations 2.67 ID:4498 **10:30**

Distributions and diurnal variations of dicarboxylic acids and related compounds in suburban aerosols collected at Mangshan, northern China

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Total suspended particles (n=58) were collected at Mangshan, a suburban area located in the north of Beijing, China in autumn 2007. They were analyzed for low molecular weight α,ω-dicarboxylic acids (C2-C12), ketoacids (ωC2-ωC4, pyruvic acid), α-dicarbonyls (C2-C3) as well as aromatic (phthalic, iso and tere-phthalic) acids, using a capillary gas chromatography. Molecular distributions of diacids demonstrated that oxalic (C2) acid was the most abundant species followed by succinic acid (C4) and malonic acid (C3). Higher carbon number diacids were less abundant. C2 diacid constituted 38–77% of total diacids. For most compounds, the concentrations were higher in daytime than nighttime. This result indicates that diacids are produced by photochemical oxidation of organic compounds, which are emitted from anthropogenic sources in Beijing, and then transported to Mangshan area by the northward wind in daytime. However, higher concentrations of C2 were observed in nighttime rather than in daytime. Positive correlations of C2 with glyoxylic acid (ωC2) and pyruvic acid (Pyr), and high relative humidity at night suggests that secondary production of C2 occurs probably via aqueous phase oxidation of both ωC2 and Pyr in nighttime. High C3/C4 and Ph/C4 ratios suggest that automobiles exhaust and fossil-fuel combustion products from industries in Beijing are important anthropogenic sources. This study also demonstrates that secondary organic aerosols are significantly produced in the vicinity of Beijing.

P-Observations 2.68 ID:3548 10:30

Source provenance of size- and time-resolved fine ambient particle fractions from the southeast of Beijing

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The Beijing area suffers heavy air pollution and associated particle emissions caused by the rapid economical growth during the past decades. The aerosol affects the regional air quality and visibility as well as cloud formation processes and the radiative balance of the atmosphere by its chemical, optical and microphysical properties. The project targets to study microphysical and chemical parameters of fine aerosol in order to associate the chemical composition of particles with their ability as cloud condensation nuclei. For this purpose, two measurement campaigns were performed at a background site near Beijing. The winter campaign was carried out in March 2009 and the summer campaign took place from mid July 2009 to mid August 2009. PM_1 samples were continuously sampled every 24 hours using a DIGITEL high volume sampler and size- and time-resolved aerosol samples were collected using 11-stage Berner impactor. This study presents the results of the chemical characterization of the PM_1 and impactor samples, which were analyzed for the mass concentration, inorganic ions and carbon sum parameters such as elemental, organic and water soluble organic carbon (WSOC). The WSOC fraction was characterized for hygroscopic substances such as dicarboxylic acids and saccharides. Additionally, fatty acids were analyzed to investigate surface active substances. Polycyclic aromatic hydrocarbons, alkanes and hopanes were analyzed for anthropogenic emissions and metals for crust material. During the winter time the highest concentrations were observed during periods with prevailing wind directions from southern areas, while northern wind directions led to lower concentrations. The main components of the fine particles are inorganic ions and carbonaceous material. The organic carbon fraction is mostly dominated by WSOC. High concentrations of tracers such as levoglucosan indicate an influence of biomass burning as one source of organic particles in the area.

P-Observations 2.69 ID:4145 10:30

Long-term observation of aerosols in Beijing, China, from 2001 to 2009: The impact of the Olympic Games 2008 on the air quality of Beijing city

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This research has been achieved in order to quantify the long-term trend of the air quality in Beijing, China where the outstanding and historic development has undergone in the past 10 years at the beginning of the 21st century. We have measured the concentrations of PM10, PM2.5, and chemical species in aerosols such as ionic species and polycyclic aromatic hydrocarbons at Tsinghua University, Beijing. PM10 concentration in Beijing from 2001 to 2009 was 147.0±89.6 µg/m3 (n=3113). It was approximately five times higher than those observed in several other cities in the world. PM2.5 concentration in Beijing from 2005 to 2009 was 77.4±65.9 µg/m3 (n=1164). The air quality in Beijing has not been improved during the past 10 years in terms of the aerosol pollution. Many regulations that have been applied to prevent the air quality in Beijing from the air pollution during the Olympic Games 2008 made the larger aerosols that have the particle size larger than 2.5µm reduced 25%, however it didn't work well for the reduction of much smaller aerosols that have the particle size smaller than 2.5µm. Calcium and sulfate have significantly decreased during the Olympic period.

P-Observations 2.70 ID:4298 10:30

Layered structures of the physical,chemical and optical properties of the particulate matter in the boundary layer over Beijing, China

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Profiles of particulate matter were measured several times over Beijing extending from the surface to 1000m or so, by using a tethered balloon observing system. The tethered-balloon-borne instruments included an optical particle counter to measure number concentration of particles in several channels and a 2-stage low-volume aerosol impactor (LAI) to directly collect particles for electron microscopic analysis afterwards, and together with meteorological radio sonde for getting temperature and relative humidity profiles. Meanwhile the corresponding lidar signals were used to make comparison with the balloon-borne observations. The results are reported separately for clear and hazy days by categorizing the days as clear or hazy on the basis of visibility data. Obvious inversions at the altitude about 500-600m high appeared under hazy weather, below which particulate matter seemed to be confined, meanwhile clear weather was identified by relatively uniform vertical structure of aerosol profiles in the low atmosphere. Lidar signals evidenced the fact well, in that the aerosol layer of relatively large extinction value extended to higher altitude than hazy days. Different particle type's profiles could also be seen between the cases in hazy and clear weather. Relatively large depolarization ratios in the area in some cases were not only caused by mineral dust but also sea-salt particles, especially in dry condition, probably the crystallized sea-salt particles.

P-Observations 2.71 ID:4555 10:30

Carbonaceous aerosol measurements at the "Demokritos" urban background site in Athens, Greece – Concentration levels and potential sources

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The aim of this work was to examine the EC / OC and BC concentration levels at an urban background station in the Athens city, as well as their potential local and/or regional sources. All measurements were conducted at the "Demokritos" site, in the periphery of the Athens Metropolitan area, during October 2009 – February 2010. PM2.5 elemental and organic carbon concentrations were measured in a 3-hr basis by the use of a Semi-Continuous OCEC Field Instrument (Sunset Laboratory Inc.). The NIOSH Method 5040 thermal protocol was applied during samples analysis. Black carbon was measured continuously by an aethalometer (Model AE31, Magee Scientific). 24-hr mean OC and EC concentrations were measured equal to 2.1 ± 0.6 μg m-3 and 0.6 ± 0.3 μg m-3, respectively. Regression of EC and OC 3-hr mean values yielded a relatively low correlation coefficient (r = 0.63), indicative of the presence of different sources affecting each carbon fraction. This was also evident by the differences in the EC and OC diurnal cycles pattern. Elemental carbon concentrations presented a clear peak during morning and evening rush hours, linking EC with vehicular traffic. OC peak concentrations were observed during afternoon and evening hours, which could be attributed to formation of secondary organic aerosol. 3-hr mean EC concentrations determined thermally and optically were highly correlated (r = 0.93), with a slope value equal to 0.87 and negligible intercept (-0.01). A high correlation was also observed between optically determined EC and BC concentrations (r = 0.89). The measurement period was characterized by stagnant conditions (1-hr mean wind speed values below 2 m s-1 for approximately 85 % of the data). The results of Conditional Probablity Funtion (CPF) analysis of wind direction and concentration data suggest that the measured EC / OC concentration levels were mainly of local origin.

P-Observations 2.72 ID:4590 10:30

Morphology and Chemical Analysis of Atmospheric particles at a low-income high population density location of the City of Gaborone, Botswana

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Airborne aerosol particles were collected in a low-income high population density location of the city of Gaborone Botswana using a High Volume air-sampler. Samples were collected in the summer and winter months at a location bordering a busy high way and the low income locality. Morphologies and elemental compositions of aerosol particles of different sizes were obtained by Scanning electron microscopy coupled with energy-dispersive spectrometry. Preliminary results on diurnal variations on both morphology and elemental composition will be discussed including air-quality health related correlation factors.

P-Observations 2.73 ID:4134 10:30

The effect of anthropogenic pollution on the fractionation of gaseous mercury in coastal urban areas

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The origin of one half of primary emitted mercury is estimated to be anthropogenic. This results in emission hotspots of mercury in urban/industrial areas. Also other anthropogenic emitted species like SOx, NOx and VOC can be expected to be elevated in urban plumes. There is evidence that bromine is the main oxidant of gaseous elemental mercury (GEM) in the atmosphere. One source of bromine is release from sea spray above the ocean. The difference in meteorological conditions and chemical composition in the marine boundary layer compared to the terrestrial boundary layer combined with mercury emissions from coastal urban areas could cause a different pattern in fractionation and deposition of mercury at the coast than is seen at inland urban sites. Investigations are thus needed to better understand emission loads, fractionation, and impact of mercury on air, soil, and water in coastal urban areas.

We investigate this issue using measurements of GEM and reactive gaseous mercury (RGM) in 15 coastal cities and their marine boundary layer. A closer examination of 3-4 days of continuous harbor measurements is carried out for three urban areas in the Southern Hemisphere (Sydney (Australia), Christchurch (New Zealand) and Valparaiso (Chile)). The results will be discussed.

P-Observations 2.74 ID:4270 10:30

Ground-Based Measurements of Tropospheric Species Over Toronto and Comparisons with the GEOS-Chem Model

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We present tropospheric trace gas columns retrieved using a ground-based Fourier Transform Infrared (FTIR) spectrometer at the University of Toronto Atmospheric Observatory (TAO), which has been recording high-resolution solar absorption spectra on a regular basis since May 2002. The trace gas retrievals are performed using the optimal estimation method implemented semi-empirically with the SFIT2 algorithm.

The time series of the total columns from May 2002 to October 2009 are presented here for tropospheric species CH4, C2H6, N2O, CO, HCN and O3. The retrievals were done using the newly recommended spectral microwindows from the Network for Detection of Atmospheric Composition Change (NDACC). Three of the time series are compared to the GEOS-Chem model, which is a global 3-D chemical transport model of atmospheric composition driven by assimilated meteorological fields from the Goddard Earth Observing System (GEOS). The CH4 time series is also compared to recent GOSAT observations. We assess both the measurement retrieval parameters and the model emissions inventories, and interpret the measurements in terms of atmospheric processes.

P-Observations 2.75 ID:4375 10:30

Air quality in South Africa – past trends and future challenges. A case study at a regional background site.

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Certain aspects of air quality in developed countries have shown vast improvements during the last couple of decades. In contrast, developing countries have historically rather placed emphasis on economic growth, with the acceptance of certain environmental and health risks.

On the African continent South Africa (SA) plays a leading role in many aspects and is regarded as the industrial and economic hub of southern Africa. Within a global air quality perspective SA is also relatively important, due to the fact that i) biomass burning plumes from southern Africa affects Australia and South America, ii) an anthropogenic NO2 hotspot, over the South African Highveld area, is clearly visible form satellite observations, iii) NO2 transport from this region has been tracked as far as Australia and iv) SA is regarded as one of the top ten sulfur emitting countries in the world.

Considering the importance of SA within the continent and indeed within the global perspective, the question should therefore be asked: Where is air quality in South African heading?

In this paper a 16 year data series from a regional background station is discussed in order to make some conclusions with regard to past air quality trends and the possible future direction that air quality in SA might be heading.

P-Observations 2.76 ID:4193 10:30

Chemical composition of PM2.5 and PM10 collected in urban and biomass burning sites of São Paulo State, Brazil.

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Aerosol particles affect the climate both directly by scattering and absorbing incoming solar radiation and indirectly by acting as cloud condensation nuclei. On the global scale, biomass burning represents an important source of atmospheric aerosols and greenhouse gases, and has demonstrated positive and negative effects on climate. The chemical composition of aerosol is variable, thus it is important the knowledge and determination of molecular markers to identification emission of specific sources. In order to characterize the aerosol from urban (August 2008 and March-April 2009) and biomass burning (November 2008) in São Paulo State, Brazil, particulate matter samples were collected (PM2.5 and PM10). In this work were determined PM2.5 and PM10 concentrations, inorganic ions, organic acids and monosaccharide anhydrides as levoglucosan, galactosan e manosan. PM2.5 mass concentrations in 2008 were from 17 to 83 µg m-3 at SPA (av. 47 µg m-3) and 6 to 26 µg m-3 in PRB (av. 18 µg m-3), and from 4 to 42 µg m-3 in 2009 at SPA (av. 13 µg m-3). The PM10 mass concentration range in 2008 is 35 to 98 µg m-3 at SPA (av. 64 µg m-3) and 27 to 43 µg m-3 (av. 35.2 µg m-3) at PRB. The differences can be explained considering seasonal influences, meteorological condition, and local emission sources of the aerosol. Sulphate, nitrate, ammonium, elementar carbon (EC) and particulate organic material (POM) are major components in mass balance of the total PM2.5 and PM10. On average 68% of PM10 was made up of fine particles at SPA08 site and 46% at PRB08 site. At SPA08 sulfate, potassium, ammonium, nitrate, fosfate concentrations were higher in PM2.5 than in PM2.5-10. Converselly at PRB08 site sodium and magnesium concentrations were higher in PM2.5-10 than in PM2.5.

P-Observations 2.77 ID:4194 10:30

Seasonal variation of water-soluble ions in atmospheric particulate matter PM10 collected in Sao Paulo city, Brazil

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Human activities can change the composition and concentration of chemical species in the atmosphere. Atmospheric composition changes affect the photosynthesis processes, the aquatic life, the vegetation, the biogeochemistry cycle and the human health. Sao Paulo is the largest city of the Latin America and presents intense vehicular traffic and high pollutants emission. The objective of this study is to investigate the seasonal variation of the water-soluble ions determined on the atmospheric particulate matter collected in Sao Paulo, Brazil, in 2003 (n=28). Samples were collected twice a month using a high volume sampler and the ionic fractions were determined by ion chromatography. The average concentration of PM10 was the highest in the winter (70 ng m-3) and it was the lowest in the spring season (31 ng m-3). Vehicular emissions, biomass burning and photochemical reactions are the main sources of pollution in the winter (dry season). In the wet season the influence of long range transport was observed by air mass back trajectories. The constant burning of solid waste and biomass on small properties and at landfills is probably responsible for the chlorine concentrations (221-387 ng m-3). Formic/acetic acid ratios suggest photochemical and biogenic emissions of formic acid in the winter. Sulfate and oxalic acid showed a good correlation only in summer (r2=0.7). Previous studies have suggested that in-cloud and heterogeneous formations can yield a good correlation between these species. On the other side potassium and oxalate in agreement can suggest biomass burning origin.

Conclusion Local sources have a strong impact on the concentrations of particulate matter and pollutants. Remote sources also contribute to the concentrations of pollutants produced by biomass burning and soil resuspension.

P-Observations 2.78 ID:4202 10:30

Continuous measurements of PM10 and PM1 chemical composition at the high elevated Nepal Climate Observatory – Pyramid NCO-P (5079 m a.s.l.)

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The so-called Atmospheric Brown Cloud (ABC), is a wide polluted layer extending from the Indian Ocean to the Himalayas during the winter and pre-monsoon seasons (November to April). This phenomenon strongly impacts air quality, visibility and the energy budget of the atmosphere over the entire Indian sub-continental area. Here we present 2-years of measurements of the chemical composition of PM10 and PM1 from the Nepal Climate Observatory – Pyramid (NCO-P), a permanent aerosol monitoring station located in the Khumbu valley at 5079m a.s.l. beneath Mt. Everest. Despite its high altitude, the average PM10 mass apportioned by the chemical analyses is of the order of 10 µg/scm, with almost a half of this mass accounted for by organic matter, elemental carbon (EC) and inorganic ions, the rest being mineral dust. The concentrations of carbonaceous and inorganic ionic aerosols follow a common time trend with a maximum in the premonsoon season, a minimum during the monsoon and a slow “ramp-up” period in the postmonsoon

and dry seasons, which is the same phenomenology observed for other Nepalese Himalayan sites in previous studies suggesting a common seasonal pattern along the southern side of the upper Nepalese Himalayas. Such seasonal cycle can be explained by the seasonal variations of dry and moist convection and of wet scavenging processes characterizing the climate of north Indian subcontinent. Valley breeze circulation provides an efficient mechanism for bringing polluted boundary layer air rich of carbonaceous material, sulphate and nitrate salts in the Asian free troposphere. In contrast, mineral dust, which is the major component of PM10 at NCO-P are impacted to a smaller extent by valley breezes and follow a unique seasonal cycle which suggest multiple source areas from central Asia and long-range transport from far western arid regions.

P-Observations 2.79 ID:4266 10:30

Evaluation of simulated PM concentrations over Greece using MODIS satellite measurements

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The main objective of the present study is the evaluation of photochemical simulations over Greece that are performed with the air quality model CAMx off-line coupled with the mesoscale meteorological model MM5. The photochemical model was implemented with grid spacing over the Southern Balkans in 10 km spatial resolution for the year 2007. An anthropogenic emission inventory for NOx, NMVOCs, CO, NH3, SO2 and PM10 was compiled to support the model implementation. A natural emission model driven by MM5 was also used to calculate the biogenic NMVOCs, sea salt and wind-blown dust emissions. Particulate and gaseous chemical boundary conditions for CAMx were derived from the global chemistry transport model TM4-ECPL. The evaluation of model results is performed through the comparison of simulated tropospheric PM10 column amounts with collocated satellite measurements. MODIS/Terra and MODIS/Aqua daily level 2 data at the spatial resolution of a 10kmx10km (at nadir) pixel array are analysed for the year 2007 and the adopted domain of the CAMx model. The columnar aerosol mass concentration, defined as the total aerosol mass in a vertical column of atmosphere, is used for the comparison with the PM10 output of the CAMx model. The consistency between the spatial patterns and temporal variations of PM columnar data from model results and from satellite retrievals is examined. Emphasis is given both on the heavily polluted areas of Greece like the major urban agglomerations and on rural sites. The similarities and discepancies between model results and remote sensing data are discussed. The atmospheric processes or model input data (for e.g. sources of emissions) critical for the improvement of the performance of the photochemical model are investigated and identified. Future work to provide a robust modelling tool for the determination of atmospheric aerosols levels in Greece is outlined.

P-Observations 2.80 ID:4523 10:30

Examination of PM10 and CO trends in Southern Chile at Temuco (38º 46´S, 72º 38´W, 128 m.a.s.l): 2003-2009

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Temuco is a fast growing city in Southern Chile that suffers from heavy air pollution every year in autumn and winter. The main source of inhalable particulate matter (PM10) and other pollutants is residential wood burning (RWB), used for heating the houses and cooking. The government has set up an air quality monitoring network. The monitoring network has 2 stations: Las Encinas (LET) and Padre Las Casas (PLC). Here we analyze measurements of PM10, carbon monoxide (CO) and meteorological parameters for the period 2003- 2009. Preliminary results show that annual concentrations of PM10 increased in 30% since 2003 reaching 92 ug/m3 in 2009 (March-August) at LET. Station PLC does not show a significant trend over these six years with a annual PM10 average concentration of 55 ug/m3. During 2008 and 2009, the Chilean air quality standard was exceeded 35 days, with 24 hours running average maximum concentrations of 414 and 436 ug/m3, respectively. CO mixing ratios are available since 2005 at LET showing a diurnal cycle during the cold months (Mar-Aug). The average concentrations CO at daytime were 0.7 ppmv. Nighttime CO averages was of 2.5 ppmv. The average CO concentrations from September to February were less than 0.8 ppmv. CO and PM10 correlate well during the cold months.

P-Observations 2.81 ID:4214 10:30

Measurements of Particulate Matter in Seoul and its Upwind Deokjeok Island in the Yellow Sea in Spring 2003

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Measurements for particulate matter (TSP, PM10 and PM2.5) were made at Seoul and at Deokjeok Island, which is about 70 km off the west coast of Korean Peninsula, from March to May in 2003. Deokjeok Island is considered a background site, upwind of Seoul along with prevailing westerlies in the region. Ionic composition in PM2.5, and organic and elemental carbons in PM10 were also measured at the same sites about two weeks around one or two days of Asian dust (AD) in March and April, respectively, and throughout May. The effects of AD and long-range transport of air pollutants were investigated by comparing the variations in pollutants between Seoul and Deokjeok Island. In general, the effects of AD were not significant during the measurement periods. Measurements indicate that the concentration of coarse particles (TSP portions larger than PM10) at both sites was higher than that of PM10 and the concentration of particulate matter in Seoul was higher than that at Deokjeok Island during the AD period. These concentrations are attributed in large part to local emissions of fugitive dust over that of long-range transport of AD. Most pollutants, including sulfate and nitrate, were higher in Seoul than in Deokjeok Island. Unlike nitrate, variations in sulfate concentrations between Seoul and Deokjeok Island showed a high correlation. We understood that sulfate was more affected by long-range transport while local emissions were more important in the determination of nitrate. Many researchers report that the Northern Hemisphere, including the Korean Peninsula, was affected by intense Siberian forest fires in May 2003. We also found an increase in K+, particularly during a smog episode in late May. However, continuous accumulation of sulfate for around two weeks and the high concentration of nitrate, along with low wind speed in the smog episode, are associated with a contribution of long-range transport from China and local emissions as well as that of Siberian forest fires.

P-Observations 2.82 ID:4494 10:30

VERTICAL PROFILES OF PM10 CONCENTRATIONS DERIVED FROM MOBILE LIDAR MEASUREMENTS IN THE FRAMEWORK OF THE MEGAPOLI EXPERIMENT

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A compact and mobile lidar has been deployed around Paris onboard a van during the MEGAPOLI (Megacities: Emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation) summer experiment in July 2009. The measurements performed with this Rayleigh-Mie lidar have been converted into PM10 concentrations profiles using optical to mass relationships determined during previous campaign around Paris. This method is here described and an example of application on the 1st July 2009 is presented and compared with ground-based stations from Airparif network. Such an approach is a powerful way for the validation of air quality mesoscale models.

P-Observations 2.83 ID:4144 **10:30**

Seasonal variation of black carbon aerosol at Happo, a remote mountain site in Japan

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Black carbon (BC) aerosols efficiently absorb solar visible radiation and contribute to the radiative forcing of the atmosphere on regional and global scales. The spatial distributions of BC are mainly determined by the transport and emission rates. Asia was estimated to be one of the largest source regions of BC in 2000. Therefore, it is important to study the distribution of BC in the outflow of Asia. For this purpose, we made continuous measurements of mass concentrations of BC (MBC) with a filter-based absorption photometer (continuous soot monitoring system; COSMOS) at a remote mountain site, Happo, Japan (137°48′E longitude and 36°41′N latitude, 1800 m ASL) for 2 years from August 2007 to August 2009. The MBC did not show significant diurnal variations, suggesting little influence from nearby sources. The average and median values of MBC were 0.27±0.18 μg m-3 and 0.23±0.03 μg m-3, respectively. The MBC showed well-defined seasonal variations with spring maximum and summer minimum. We also calculated MBC using a CMAQ model, which was driven by a WRF model and compared them with the observed MBC values. The contribution of BC emitted from East Asia to the observed MBC was also estimated from the model calculations.

P-Observations 2.84 ID:4512 **10:30**

Black carbon air pollution and their environmental impacts in Central India

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Black carbon has recently emerged as a major contributor to global change, probably second only to CO2 as the main driver of change. Black carbon (BC) particles strongly absorb the sunlight and generate heat in the atmosphere, which warm the air and can affect regional cloud formation and precipitation pattern. Black carbon plays significant role in the atmospheric particulate and thereby the research of carbonaceous particulate has recently become one of the most favorite topics in field of atmospheric science. The main region for its popularity is that carbonaceous particulate represents an extreme diversity within the atmospheric particulates. The winter particulate pollution is severe in several urban parts of India. In the context of Central India, the sampling campaigns of particulate matters (PM) in the winter season of year 2006 - 2007 were carried out in the city of Raipur to characterize the ambient mass distribution of coarse particulate matters (PM10) and their sources. The coarse particulate matters (PM10) from different locations of Raipur city were collected using a low volume sampler. The mass distribution of PM10 in the ambient air was found to be ranged from 221 - 760 µg m-3. The mass concentrations of species i.e. organic carbon, elemental carbon and trace elements in the particulates are discussed. The sources and environmental impacts of the carbonaceous particulates are described.

P-Observations 2.85 ID:4526 10:30

Four-years of black carbon measurements at 5000 m asl in the Himalayas.

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The Asian continent hosts the two world's most populated countries, India and China, which are also the most fast growing ones and the largest emitters, very close to the highest world mountain ridge: the Himalayas. Since pollutants are released from different sources and interact, the observations of the seasonal and diurnal variations of pollutants at an high Himalayan remote site gives an insight in the extent of the anthropogenic influence on the larger regional and global atmospheric scale. The NCO-P (5079 m) is operated in the Himalayas since March 2006 in the framework of Ev-K2-CNR SHARE and UNEP-ABC projects; besides other important information on the atmospheric background conditions it monitors the intensity and frequency of pollution transport at such altitude, due to both valley winds and large scale circulation.

Surprisingly high pollution levels with black carbon concentrations up to 5 µg m-3 (PM1 60 µg m-3, ozone > 90 ppbv) were observed during pre-monsoon hot-spots for 4 continuous year. This suggests that Himalayan valleys could represent a "natural chimney" by which BC and other pollutants can be directly vented to high Himalayas and middle/upper troposphere where their reach a longer life time. Moreover, high levels of BC in long range polluted air mass transports affect the atmospheric composition at the station. These high level of pollutants at such altitude could be open to significant climatic effects.

BC is an important component of ABCs and directly warms the atmosphere in the Indian-monsoon region. This absorbing aerosol remains one of the largest sources of uncertainty in analyses of climate change.

Here we will provide an overlook of the frequency and intensity of pollution transports up to NCO-P ABC-GAW station observed in first 4 years of continuous monitoring, also providing a preliminary estimate of radia-tive effects on radiation budget and snow-ice melting.

P-Observations 2.86 ID:4286 10:30

Microphysical Properties of Black Carbon and Light Scattering Particles over California during CARB 2008

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Black carbon (BC), light scattering particles (LSP), and trace gases were measured on board the NASA DC-8 aircraft over California during the ARCTAS-CARB campaign conducted in June 2008. A single particle soot photometer (SP2) was used for measurements of BC and LSP. The aerosol concentrations showed large variations related to various plumes from different regions of California. We used SO2 and CH3CN as tracers to identify the fossil fuel (FF) and biomass-burning (BB) plumes, respectively. FF plumes were often observed near the surface, while the influence of BB plumes was observed up to 4 km. The influence of BB at higher altitudes suggests the role of the convective up-lift of air parcels due to thermal energy at fire bases. The average BC mass concentrations in the FF and BB plumes were about 90-500 ng m-3 and 300-700 ng m-3, respectively. The mass concentrations of BC showed tight correlations with the mixing ratios of CO and CO2 in the two types of plumes. The slopes of the BC-CO2 correlations were 17.9±8.7 ng m-3/ppmv and 271.5±87 ng m-3/ppmv for FF and BB plumes, respectively. On the other hand, the slopes of the BC-CO correlations were similar in these plumes. The mass concentration of BC also showed good correlation with the volume concentration (Vsc) of LSP. The Vsc/BC ratios were much higher in BB plumes than those in FF plumes. The number and mass-size distributions of BC and LSP showed single mode lognormal fits for the two types of the plumes. BC particles in fresh BB plumes were more thickly coated than in the fresh FF plumes indicating that BC was already thickly coated upon emissions by BB. These data are useful in characterizing microphysical properties of primary aerosols from different sources, and for improved understanding of the evolution of the properties associated with aging.

P-Observations 2.87 ID:4436 **10:30**

Lidar observations of the Kasatochi volcano aerosol plume

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The eruption of Kasatochi volcano on 7-8 August 2008 injected about 1.7 Tg of SO2 into the troposphere and lower stratosphere. A week later the emissions were observed by the Dalhousie Raman Lidar as an aerosol plume in both the troposphere and stratosphere above Halifax, Nova Scotia. The stratospheric plume was observed for four months as it gradually dispersed. Descent and vertical eddy diffusion brought the plume down to the tropopause, and so may partially explain aircraft observations of Kasatochi aerosol in the upper troposphere over those same four months. Aerosols from volcanic injections into the troposphere were also observed above Halifax, but only in the few weeks after the eruption. Source attribution for the aerosols is made using FLEXPART, a Lagrangian particle transport model. The aerosols appear to have reached the surface near Halifax, although the net contribution to PM2.5 could have been no more than 5 micrograms per cubic meter. Identification of Kasatochi aerosols beyond the initial detections were not possible due to the complex mixing processes in the troposphere combined with other natural and anthropogenic aerosol sources.

P-Observations 2.88 ID:4545 **10:30**

A comparison of DOAS observations by the CARIBIC aircraft and the GOME-2 satellite of the 2008 Kasatochi volcanic SO_2 plume

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The 2008 Kasatochi volcanic eruption emitted ≈1.5 - 2.5 Tg SO_2 into the upper troposphere and lower stratosphere. Parts of the main volcanic plume (gases and particles) reached central Europe a week after the eruption and were detected there by the CARIBIC (Civil Aircraft for Regular investigation of the Atmosphere based on an Instrument Container) flying observatory. A detailed study on the CARIBIC observation of the plume will be given, focusing in the aerosol concentration and composition but mainly on the DOAS observation of the SO_2 column densities. The Kasatochi eruption influenced the aerosol composition for at least three month.

The plume was also observed by the GOME-2 satellite instrument, only a few hours after the CARIBIC aircraft had crossed the plume, thus giving a unique opportunity to compare results. Trajectories and local wind speeds are investigated in detail using the GOME-2 and CARIBIC observations for better comparison of the results from these two observational systems. A comparison of the spatial pattern with the local observations of the wind speed and the trajectory models TRAJKS and HYSPLIT showed a slight discrepancy, which has to be considered for satellite validation. The online measured wind speed and wind direction agreed quite well with the model data, and therefore also led to the same discrepancy as the models. Hence, it appears that detailed analyses of wind speeds are required.

Emitted and secondary particles, partly measured and sampled by the CARIBIC in situ instruments, affected the DOAS SO_2 measurements, of both CARIBIC and GOME-2. Moreover the plume was situated above a cloud by which the observed radiation and the column densities were strongly affected. Overall GOME-2 and the CARIBIC SO_2 measurements agree very well. The major uncertainties remain the actual wind speed needed to properly correct the advection of the plume between the different overpass times, and to smaller degree the effect of clouds and aerosol.

P-Observations 2.89 ID:4416 **10:30**

Multi-component vertical profile retrievals for ground-based MAX-DOAS

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We attempt to retrieve lower-tropospheric vertical profile information for 8 components from ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements. The components retrieved include aerosol extinction coefficients (AEC) at two wavelengths 357 and 476 nm, NO2, HCHO, CHOCHO, H2O, SO2, and O3 volume mixing ratios (VMRs). This method was applied to MAX-DOAS observations performed at Cabauw, the Netherlands (52.0ºN, 4.9ºE) in June- July 2009 during the Cabauw

Intercomparison campaign of Nitrogen Dioxide measuring Instruments (CINDI) campaign. For the lowest layer of retrieved profiles at 0-1 km, two channels of AEC values reveal consistent variations. NO2 showed typical diurnal variations with maximum in early morning and minimum in the afternoon. Positive correlations between HCHO and CHOCHO were seen, as expected. H2O VMR agreed well with that derived from NCEP surface data, and was used to judge cloudy cases after conversion to relative humidity. All these results support the capability of MAX-DOAS observations applicable to various air quality studies.

P-Observations 2.90 ID:4260 10:30

Retrieval and analysis of vertical columns of sulphur dioxide from OMI over central Chile

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Over the central region of Chile there are several sources of sulphur dioxide (SO_2), such as copper smelters, that oxidize to sulphate which affects regional air quality and global climate. Satellite observations provide valuable information on these emission sources however care is needed to fully account for atmospheric scattering in the heterogeneous environment. In this work we use a local air mass factor to convert the OMI slant columns into vertical columns using a high spatial resolution (4km horizontal) regional chemical transport model (*Polyphemus*) to represent the vertical profile of the SO_2. We also use a radiative transfer model (LIDORT) to compute scattering weights. These observations will provide constraints on emissions of sulphur dioxide trough inverse modeling.

P-Observations 2.91 ID:4274 10:30

Atmospheric SO_2 concentrations over high-dust region in western India: Temporal variability and transformation to SO_4^{2-}

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In the present-day scenario of increasing atmospheric pollution, an understanding of the factors influencing the formation of sulphate (SO_4^{2-}) from its precursor gases has special significance due to its dominant role as cloud condensation nuclei (CCN). With this objective, surface level SO_2 and fine-mode SO_4^{2-} concentrations have been measured from an urban site, Ahmedabad (23.0^{0}N, 72.6^{0}E, 49m asl), and a high-altitude remote site, Mt. Abu (24.6^{0}N 72.7^{0}E, 1680 m asl), located in the high-dust semi-arid region of western India. The selected sites have distinct differences with respect to emission sources, wind regimes, atmospheric transport and abundance of mineral dust. As a specific case, SO_2 concentration at urban Ahmedabad, during Jan '09, varied between 5 to 30µg/m^3; whereas that at Mt. Abu (remote site) varied as 1 to 8µg/m^3 in Feb'09. The S-ratio, defined as the ratio of particulate S (SO_4^{2-}) to total S (SO_2 and SO_4^{2-}), has been used as a parameter to study the chemical conversion efficiency of SO_2 to SO_4^{2-} via both homogeneous and heterogeneous phase oxidation pathways. The observations, spanned over winter and summer seasons, suggest that atmospheric oxidation efficiency of SO_2 to SO_4^{2-} enhances with increasing dust content in the atmosphere. For example, over Ahmedabad, the average S-ratio is 0.37 and 0.55 during Aug-Sept'08 and Jan '09, respectively; and that at Mt. Abu (lower abundance of dust), the S-ratio is 0.28 during Feb '09 and 0.18 during Sept-Oct '09. It is conceivable that heterogeneous phase chemistry (in presence of mineral dust) is of major significance, in addition to the homogeneous gas phase chemistry of SO_2. Validation of our experimental data with a global 3D chemical transport model, GEOS-Chem, is being investigated.

Keywords: Gas-to-particle conversion, S-ratio, Sulphate, SO_2, homogeneous and heterogeneous oxidation of SO_2

P-Observations 2.92 ID:4536 10:30

Column measurements of atmospheric carbonyl sulfide at Wollongong, Australia

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Carbonyl sulfide is the most abundant atmospheric sulfur-containing species, and responsible for a considerable amount of the stratospheric aerosol layer during non-volcanic periods. COS has an oceanic source and is taken up be soils and vegetation. Recently, considerable interest has centred on the possibility of the vegetative uptake of COS provided a valuable constraint on the photosynthetic fraction of NPP.

Here we present a 13 year time series of carbonyl sulfide column measurements obtained from solar absorption Fourier transform infrared measurements at Wollongong, Australia. Wollongong is located on the eastern coast of Australia, ideally situated to observe the effects of both ocean and vegetation on atmospheric carbonyl sulfide amounts.

P-Observations 2.93 ID:4412 10:30

High frequency continuous observations of Carbonyl Sulfide

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Carbonyl sulfide (COS) is the most abundant sulfur containing gas in the atmosphere. COS is a surrogate for CO_2 uptake by plants during photosynthesis, but, unlike CO_2, is not emitted by plants during respiration. Recent studies have suggested that COS may provide unique constraints on carbon assimilation by the biosphere that are independent of the influence of respiration. While the loss of COS is dominated by vegetative uptake, COS is also subject to loss through uptake by oceans and soils, the magnitudes of which are not known. Globally, COS mixing ratios have been found to vary from 350 to 550 pptv (parts per trillion).

Present measurement methods for measuring COS rely on canister sampling followed by laboratory analysis using gas chromatography. While accurate and precise, this method is highly labour intensive, with limited sampling sites and global coverage. We have developed a method for the direct, continuous, measurement of COS using an infrared laser absorption technique with a measurement precision (3 std deviation) of less than 10 ppt in 30 s. The instrument uses a continuous wave quantum cascade laser operated at near room temperature, allowing for long term unattended operation. We present continuous data observed at a 1 second sampling rate obtained from a suburban Boston sampling site during spring 2010.

P-Observations 2.94 ID:4496 10:30

Characterizing mineral dust aerosol in the upper troposphere

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Mineral dust is an efficient ice nucleus with a strong potential to alter the properties of upper tropospheric cirrus clouds and, consequently, the Earth's radiation budget. While mineral dust in the atmosphere is primarily of natural origin (emitted by the world's deserts), changing land use patterns and increased desertification can lead to a significant anthropogenic fraction (~20%) in the future. Given that it is the most abundant aerosol in the atmosphere (by mass) it is important to improve our ability to detect and predict mineral dust concentrations, particularly in the vulnerable upper tropospheric regions where small amounts of dust can lead to large changes in cirrus clouds. To this effect, we performed an extensive trajectory modeling study to explore the availability of mineral dust particles as ice nuclei for cirrus clouds, also tracking the particles' history of processing by water and mixed-phase clouds. We found that dust emissions from Asian deserts have a higher potential for interactions with high clouds, despite being climatologically much smaller than African emissions. Moreover, for "classical" cirrus-forming temperatures, our results show that only mineral dust IN that previously underwent mixed-phase cloud-processing are likely to be relevant. We discuss the limitations and implications of our modeling study in detail.

At present, observations of upper tropospheric mineral dust are sparse, coming primarily from aircraft campaigns. While nadir-viewing remote sounders provide global coverage of optically thicker lower tropospheric dust plumes, the limb- viewing satellite geometry is ideally suited to detecting low-concentration mineral dust in the upper troposphere due to its long observation path length (~50-100 km), high vertical resolution (~1-2 km) and good geographic coverage. Since 2001, the OSIRIS instrument on board the Odin satellite has been recording height-resolved spectra of limb-scattered sunlight between 280 – 810 nm with a grating spectrometer and 1351 detector pixels (~1 nm spectral resolution). OSIRIS also comprises an infrared imaging system that is not a part of our study. Using the fully three-dimensional radiative transfer code Sasktran, we simulated the sensitivity of limb-viewing OSIRIS measurements to mineral dust in the upper troposphere. We will show how it is influenced by a number of factors, including the spatial distribution of the dust aerosol, the real and imaginary index of refraction of mineral dust, satellite viewing geometry, scene brightness, and measured wavelength. Finally, we will outline when mineral dust can be discriminated from ice crystals.

P-Observations 2.95 ID:4486 10:30

Multielemental composition of size fractionated road dust deposited in a megacity.

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An intensive campaign was carried out to collect road dust samples in 67 points of a grid of 1.5 x 1.5 km, covering an area of ~150 km2 in the city of Buenos Aires, Argentina. The content of 15 elements (Al, As,

Ca, Cd, Cu, Fe, Mg, Mn, Mo, Ni, Pb, S, Sb, Sn, and Zn) was determined in the different sizes of the samples (A < 37 µm, 37< B < 55, 55< C < 75 and 75< D < 100). X-ray powder diffraction was selected to detect major components: minerals, silicate glass and sulphates. Individual mineral dust particles were analyzed by SEM- EDAX showing that sulphur tended to reside on the particles surface, while Al, Ca, Mg, K, Fe and Si were all an integral part of the particles. Elements content was quantified by ICP OES in the four particle size ranges. Special attention was paid to the determination of key traffic related elements (TRE´s): Cu, Mo, Pb, Ni, S, Sb and Zn. Concentrations of elements were averaged over the monitoring sites. Geological elements exhibited the higher concentrations: Fe (28-43 mg g-1) > Al (13-40 mg g-1) >> Ca (1.7-6.9 mg g-1), followed by Mg (2.5-5.5 mg g-1) > S (0.2-1.7 mg g-1). Concentrations pattern showed high variability in the four fractions but in all cases, higher concentrations were detected in the smaller size fractions (A and B). The TRE´s concentrations (mg g-1) varied from < 0.3 (for Ni) to 1.53 (for Zn). The smallest size fraction were particularly enriched in Sb > Cd > Cu which are of particular interest from the environmental and human health standpoint. To the best of our knowledge this is the first study devoted to the multielemental characterization of the Buenos Aires road dust.

P-Observations 2.96 ID:4604 10:30

WOODSMOKE SOURCE APPORTIONMENT AND HOME INFILTRATION STUDY IN THE RURAL ANNAPOLIS VALLEY, NOVA SCOTIA, CANADA

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Woodsmoke has recently received increasing attention as an important source of particulate matter, particularly in rural areas. Indoor woodstoves and outdoor wood boilers are widely used in the Annapolis Valley, in rural Nova Scotia, and the area's topography is conducive to trapping emissions especially during thermal inversions. In an effort to quantify the impact of these factors on local air quality, the source contribution of residential wood burning to ambient PM2.5 at one fixed site in the Annapolis Valley, was analyzed during the winter 2008/2009 (phase 1). 4 x 24-hr PM2.5 filters, continuous PM2.5 using TSI Inc. Dust Traks, and associated gases (VOC, NO2, O3) were measured over 50 days between 24 February 2009 and 7 May 2009 near the town of Middleton, located in the longitudinal centre of the Annapolis Valley. The PM2.5 chemical species analyzed included levoglucosan (woodsmoke marker), elements, ions, C14 (confirmatory marker for woodsmoke), organic and elemental carbon. g m-3 observed fromµThere were numerous short-term excursions over 100 PM2.5 Dust Trak measurements. There was no significant difference (P = 0.524) between the Dust Trak PM2.5 and Dust Trak PM10 concentrations suggesting that the PM10 size fraction was dominated by PM2.5. Chemical Mass Balance (USEPA CMB v8.2/SPECIATE v4.2) conducted on 16 of the 50 PM2.5 samples thus far yielded an average source contribution of residential woodsmoke to the total PM2.5 concentration of 56.2% (range 32.6% – 73%), with 12.4 % NH4NO3 (range 5.0% - 20%), 24.1% SO4 (range 11.0% - 47%), 9.2% (NH4)2SO4 (range 2% - 19.8%), 3.9% unknown mass (range 0.6% - 14%) with no CMB statistically significant input from vehicle emissions. In phase 2 of the study, an investigation of woodsmoke home infiltration in 30 homes was conducted during winter 2009/2010, together with an evaluation of the effectiveness of electrostatic air cleaners at reducing indoor PM2.5 concentrations. Significant (P<0.05) reductions in indoor PM2.5 were observed between days where air cleaners were in use to days where they were not in use.

P-Observations 2.97 ID:4358 10:30

Trends in wet deposition in South Africa

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Potential adverse environmental impacts due to increased anthropogenic emissions into the atmosphere necessitate the execution of long-term atmospheric deposition programmes. Atmospheric deposition of emitted chemical species back to the earth's surface through wet and dry processes plays an important role in controlling the concentrations of pollutants present in the troposphere. Deposited species can provide essential nutrients to ecosystems or can cause disturbances. Deposition of chemical species was measured at selected sites in southern Africa during the DEBITS (Deposition of Biogeochemical Important Trace Species) programme. The study of the chemical composition of rain (wet deposition) is essential to trace the temporal and spatial evolution of atmospheric chemistry.

Earlier collection of rainwater utilising wet-only samplers was conducted at Louis Trichardt and Amersfoort from 1986 until 1999 and at Skukuza during the period of 1999-2002. Collection of rain water at these sites recommenced in 2008 and is still ongoing. The study area is classified as semi-arid savannah. Skukuza, which is situated in the Kruger National Park and Louis Trichardt situated in the Limpopo Province are considered to be background and rural sites, while Amersfoort represent industrial activity in the Mpumalanga Highveld. Back trajectory analysis of the region further indicate that air mass originating from the Mpumalanga Highveld Industrial region are circulated over the sites at Skukuza and Louis Trichardt, impacting these sites with no significant local emission sources.

The most important inorganic and organic ions present in the collected rainwater samples were determined with an ion chromatograph. The chemical composition of the rainwater samples collected at each of the sites for the various sampling periods was compared. The variation in concentrations for each of the measured chemical species over the sampling periods and the observed changes in trends are reported.

P-Observations 2.98 ID:4348 10:30

Atmospheric nucleation in the eastern Mediterranean marine boundary layer

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Atmospheric nucleation has been reported to take place in varying locations and environments worldwide. Most of the observations were made during daytime, suggesting that photochemical processes play a key role in new particle formation mechanisms. Night-time observations have so far been scarce in literature and none of them refers to the marine boundary layer (MBL). At the environmental research station of the University of Crete at Finokalia, Crete (35°20´N, 25°40´E, 250 a.s.l) measurements of atmospheric aerosol properties were performed during the period April 2008 – April 2009. The site is considered representative for the MBL conditions of Eastern Mediterranean. Size distributions of atmospheric ions and charged aerosol particles with diameters 0.8-42 nm were measured with an Air Ion Spectrometer (AIS). Concurrent measurements of aerosol particles number size distributions with diameters 8 – 900 nm were performed with Scanning Mobility Particle Sizer (SMPS). At the Finokalia station atmospheric nucleation has been found to be a reoccurring phenomenon both during day and night. However the characteristics governing such processes were found to be quite different. There were 53 nucleation events recorded during day-time and the event to non-event days ratio was 0.21. Common observation for all the events was that the initial nucleation evident in AIS data was then followed by condensational growth of the new particles to larger diameters and the subsequent recording of the event by the SMPS system. During the measuring period 42 events of night-time nucleation were recorded as well. The event to non-event days ratio was found to be 0.14. A common feature for all of the events is that no particle growth was observed probably due to the lack of significant amounts of condensable vapours during the night. Atmospheric nucleation at Finokalia, both for day and night, was found to be more frequent from February to April.

P-Observations 2.99 ID:4516 10:30

Atmospheric versus Riverine Nutrient Inputs to the Northeastern Levantine Basin of the Eastern Mediterranean

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Aerosol and rain samples were collected at a rural site located on the coastline of the Eastern Mediterranean, Erdemli, Turkey between January 1999 and December 2007. Riverine sampling was carried out at five Rivers (Ceyhan, Seyhan, Göksu, Berdan and Lamas) draining into the Northeastern Levantine Basin (NLB) between March 2002 and July 2007. Samples were analyzed for macronutrients of phosphate, silicate, nitrate and ammonium (PO43-, Sidiss, NO3- and NH4+). Phosphate and silicate in aerosol and rainwater showed higher and larger variation during the transitional period (March-May, September) when air flows predominantly originate from North Africa and Middle East/Arabian Peninsula. Deficiency of alkaline material were found to be the main reason of the acidic rain events whilst high pH values (> 7) were associated with high Sidiss concentrations due to sporadic dust events. In general, lowest nitrate and ammonium concentrations in aerosol and rainwater were associated with air flow from the Mediterranean Sea. Unlike NO3- and NH4+ (Dissolved Inorganic Nitrogen, DIN), there were statistical differences for PO43- and Sidiss solubilities in sea-water and pure-water. Solubilities of PO43- and Sidiss were found to be related with air mass back trajectories and pH. Atmospheric and riverine fluxes demonstrated that DIN and PO43- fluxes to NLB were dominated by atmosphere (~ 90 % and ~ 60 % respectively) whereas the input of Si was mainly derived from riverine runoff (~ 90 %). N/P ratios (atmosphere~233; riverine~ 28) revealed that NLB receives excessive amounts of DIN and this unbalanced P and N inputs may provoke even more phosphorus deficiency. Molar Si/N ratios (atmosphere + riverine) suggested Si limitation which might cause a switch from diatom dominated phytoplankton communities to non-siliceous populations in NLB.

P-Observations 2.100 ID:4449 10:30

Impact of Fires on Air Quality in the Mediterranean during the Summer 2007: Constraint from Satellite Observations (IASI/METOP, A-Train)

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Fires burn large areas of forest every year in Southern Europe, emitting huge amounts of trace gases and aerosols, sometimes resulting in severe degradation of air quality. Due to the difficulty of evaluating the associated emissions, this strong perturbation is most often not accounted for in air quality forecasting. In this presentation, we will discuss the constraint provided by satellite observations on pollution emissions and transport from fires, in particular by the IASI/METOP and A-Train missions (MODIS, PARASOL, CALIPSO). IASI/METOP allows the global monitoring of several key species for atmospheric chemistry analysis, with unprecedented spatial sampling and coverage. Its ability to detect a large series of species within fire plumes has recently been demonstrated, and could significantly improve current evaluation of the impact of such extreme pollution events on air quality. Here, we will discuss the information provided by the carbon monoxide (CO) retrievals, one of the main species measured by IASI, on fire emissions transport mechanisms and pathways. Therefore, IASI retrievals will be compared to simulations from the CHIMERE regional chemistry and transport model for the case study of the large fires which burned in Greece during August 2007. We will then present an analysis of the information provided by IASI on chemistry within the transported plumes using the retrievals for the shorter lived species and for ozone. Finally, the IASI observations will be coupled to the aerosol observations from PARASOL and CALIPSO in order to assess the impact of this specific fire event on air quality in the Euro-Mediterranean region (both PM2.5 and ozone).

P-Observations 2.101 ID:4220 10:30

Air pollution over the Western Mediterranean: Observations from a cruise ship during the years 2006-2009.

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Observations from monitoring stations as well as from field campaigns have shown high levels of air pollution both by ozone and by particulate matter in the Mediterranean Basin. However much of the area is not adequately covered by air pollution monitoring stations and there is a particular lack of observations over the sea. In order to help to fill in the gap of observations and to learn more about sources of air pollutants and transformation processes, the JRC has initiated a long-term monitoring programme in collaboration with the Italian cruise line Costa Crociere. Measurements have been performed regularly during spring-summer-autumn since 2006 from a monitoring station placed on Costa Crociere cruise ships following a fixed weekly route in the Western Mediterranean. Ozone and Black Carbon (using an Aethalometer) are measured, along with SO2, NOx and particle size distributions. Four week-long campaigns with filter-sampling and subsequent chemical analysis have been carried out and also time-resolved analysis of the elementary composition of submicron aerosols was performed by streaker-sampling followed by PIXE analysis. The present study provides an overview of the observations done during the period 2006-2009, showing concentration levels and air pollution 'hot spots' in the Western Mediterranean. The data have been analysed with the aim of gaining knowledge about the contributions of different sources to air pollution with ozone and particulate matter in the area. This analysis has been carried out by a combination of back-trajectory calculations and observations of correlations among trace gases as well as chemical and elemental components of aerosol. Among the findings are that SO2 concentrations generally show a geographical distribution similar to that of tracers of heavy fuel combustion, typical of ship ship emissions. High ozone concentrations are found to be strongly related to breeze circulation and subsiding air masses.

P-Observations 2.102 ID:4547 10:30

A comparison of wind tunnel measurements and CFD model simulations for pollutant dispersion from a tunnel portal for different wind directions

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The exhausts from tunnel heads in urban area may be considered as important local air pollutant emission source due to the concentrated release of polluted air, which can affect air quality in its vicinity. Hence, it is important to have suitable tools to assess the dispersion from roadway tunnels for planning purposes. Mercure_Saturne, a three-dimensional computational fluid dynamics (CFD) model developed at CEREA and now part of the open source software (www.code-saturne.org), was applied to simulate pollutants dispersion of an experimental study which had been performed on a model tunnel in the atmospheric boundary layer wind tunnel of the Laboratoire de Mécanique des Fluides et d’Acoustique (LMFA), France. The k-eps turbulence closure was applied. A total of 28 configurations for 7 wind directions relative to the tunnel axis (0, 30, 60, 90, 120, 150, 180°) and 4 different velocity ratio between the tunnel exit velocity and the wind velocity (0.25, 1, 2 and 4) were simulated. Only one grid with the tunnel geometry was generated and the wind direction is varied on the boundaries. Measurements of wind, turbulence and concentration are used for the comparison. First we have performed detailed comparison of the profiles, especially for concentrations but also for wind and turbulence which help in the interpretation of the results. Sensitivity analyses are performed for the turbulence conditions at the tunnel head due to incomplete measurements (no epsilon). In addition to these point wise comparisons we have also computed statistical parameters to characterize the differences between measurements and simulations by following the Model validation kit procedures. We discuss these different statistics for all the cases simulated and for the different tunnel exit conditions used for the sensitivity analysis.

P-Observations 2.103 ID:4344 10:30

Observed and Modeled Variations of Tropospheric Molecular Hydrogen over Japan

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To elucidate temporal and spatial variations of tropospheric molecular hydrogen (H2), air samples were systematically collected once per month using a chartered light aircraft (0~4km) and commercial jet airliners (4~11km). Their H2 concentrations were determined against our gravimetrically prepared standard gases

using gas chromatographs with a HgO reduction detector. The observed H2 concentration showed clear vertical gradient and height-dependent seasonal cycle. The H2 concentration showed small seasonal variation in upper altitude and mean concentration decreased with decreasing height. At lower altitudes, peak-to-peak amplitude of seasonal cycles were large and maximum and minimum concentrations appeared in July/August and November, respectively. We compared observational results from our observation and from NOAA/ESRL observation network with those from model simulation using a global chemical transport model, CHASER. The model was coupled with land process model (MATSIRO) and H2 uptake by enzymes in soil, which determined by a function of both soil moisture and soil temperature, was calculated. The model reproduced spatial distributions and seasonal variations of surface stations and airborne observations well. Simulated H2 concentrations show large seasonal amplitudes on the continent surface of northern high latitudes, with the maximum and minimum in boreal spring and autumn, respectively. The global burden of H2 in the troposphere is 163 Tg and its overall lifetime in the troposphere is 1.86 years. Soil uptake is 63.9 Tg, with the contribution of 73% of total H2 sink. The results show that not only soil moisture but also soil temperature plays an important role in the seasonal variation of soil uptake and H2 concentration at northern high latitudes.

P-Observations 2.104 ID:4121 **10:30**

Constraining the budgets of hydrogen and carbon monoxide through in-situ measurements and modelling.

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Hydrogen (H2) is the second most abundant atmospheric trace gas, it acts as an indirect greenhouse gas by removing the hydroxyl (OH) radical and increasing the lifetimes of other greenhouse gases. Recent increased attention in H2 is due to its possible introduction as a clean energy fuel alternative. However, its current atmospheric budget is poorly constrained, particularly its source from primary and secondary emissions. Carbon monoxide (CO) is central in controlling the abundance and distribution of the OH radical. The secondary production of CO from the oxidation of volatile organic compounds (VOCs) remains the most poorly constrained sector of its global budget. This study investigates the primary and secondary production of H2 and CO using a combination of in-situ measurements and modelling. High-frequency measurements of H2 and CO at an urban location provide detailed information on urban time-series, diurnal cycles as well as sources and sinks of both H2 and CO. These urban measurements help to improve assessments of primary anthropogenic emissions of H2, largely from transport. Modelling of the secondary production of H2 and CO carried out using a box model simulating tropospheric conditions tracked the oxidation of a range of VOCs to produce individual conversion factors for VOCs oxidation to H2 and CO. Global budget estimates of the secondary production of H2 and CO were made based on these conversion factors. To maintain the oxidation balance within global chemical transport models which do not include a detailed suite of non-methane VOCs (NMVOC) it may be necessary to include the H2 and CO source from NMVOC oxidation.

Transformation 1.1 ID:4223 **INVITED 14:00**

Enhancing the prognostic capability of global aerosol models: Atmospheric aqueous chemistry and its role in secondary organic aerosol (SOA) formation

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A growing body of laboratory and field evidence suggests that SOA forms through aqueous-phase reactions in clouds/fogs and wet aerosols. Specifically, water soluble gases undergo photochemistry, acid catalysis, and reactions with inorganic constituents in atmospheric waters forming lower volatility species (e.g., oxalic acid, oligomers, and organosulfates). Lower volatility products are retained, at least in part, after water evaporation. Retention of some aqueous species (e.g., glyoxal and methylglyoxal) is enhanced by oligomerization via hemiacetal formation and aldol condensation during water evaporation. Field evidence is provided by the atmospheric abundance of oxalate - which is not explained by gas phase chemistry, by the association of oxalate with sulfate – known to be formed through aqueous reactions, by measurements of organic acids below and above clouds, and by stronger associations between SOA surrogates and aerosol water than between SOA surrogates and aerosol organic matter in some locations. Importantly, aqueous SOA is formed from more volatile precursors (e.g., C2, C3) than "traditional" smog-chamber like SOA (>C7) and forms products with higher O/C ratios. SOA formation via aqueous reactions could explain the observation that aged atmospheric aerosols have higher O/C ratios than observed in smog chamber SOA and could help explain gaps between measured and modeled organic aerosol. Several publications now predict that the amount of regional (northeastern USA) and global SOA formed through aqueous pathways is comparable in magnitude to that of traditional SOA formed through partitioning of semivolatile products of gas phase reactions, although uncertainties are large. This presentation reviews the current understanding of aqueous SOA formation. Detailed chemical theory and experiments conducted at a range of concentrations are used to provide insights into differences between chemistry occurring in aerosol water and cloud water. Prospects for incorporating this pathway into global models will be discussed.

Transformation 1.2 ID:4276 14:30

Organosulfates of C_9-C_{11} hydroxy carboxylic acids: novel tracers for a marine secondary organic aerosol formation process

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Organosulfates are a novel group of secondary organic aerosol (SOA) constituents that have only been recently reported and are formed by sulfation of hydroxyl-containing SOA. We demonstrate that organosulfates are also formed in a marine environment during summer conditions that are characterized by algal blooms. Aerosol samples were collected at Amsterdam Island (37.52 °S, 77.32 °E), a pristine marine site in the southern Indian Ocean, during an austral summer campaign. Using liquid chromatography/mass spectrometry with electrospray ionization detection in the negative ion mode, organosulfates of isomeric C_9 – C_{11} hydroxy acids were detected and structurally characterized. The molecular weights of the organosulfates of the C_9 – C_{11} hydroxy acids were 254, 268, and 282, and the $[M – H]^-$ isotopic pattern indicated the presence of sulfur and multiple oxygen atoms. Ion trap fragmentation of the deprotonated molecules resulted in the formation of the bisulfate ion $[HSO_4]^-$ (m/z 97), indicating a sulfate group, and the neutral loss of SO_3 (80 u), consistent with the presence of a carboxyl group. The organosulfates showed a different time course than methane sulfonic acid (MSA), suggesting a different biogenic source process, and were most abundant in the beginning of the campaign. The hydroxy acids are explained by oxidation of the double bond in hydroxylated unsaturated fatty acid residues present in algal biomass at the sea-air interface. To our knowledge, the formation of organosulfates from oxidation of marine biomass is a novel aerosol formation process. MSA and the organosulfates only explain a fraction of the water-soluble organic carbon (on average, 20 ± 8 % and between 1 and 5%, respectively). However, they both could contribute to the cloud condensation nuclei effects that have been observed above algal bloom regions and have been attributed to SOA from biogenic emissions. No evidence was found for SOA from the photooxidation of isoprene.

Transformation 1.3 ID:4240 **14:45**

Organic Aerosol: from oxidation to optical depth

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Organic aerosol in the atmosphere consists of a multitude of organic species which are the products of a variety of chemical reactions. Recent years have shown that the complexity of formation processes and atmospheric aging is not well captured by simplified model descriptions and thus uncertainties on the global budgets of OA remain large. In an effort to reduce some of the uncertainties on these budgets from observational constraints, we will present here two investigations: (1) Exploring observed bulk organic aerosol composition and how changes associated with physical and chemical aging processes can be simply described using a Van Krevelen diagram. (2) Investigating remote observations of aerosol optical depth and whether these measurements provide a constraint on the organic aerosol budget simulated in global models.

Transformation 1.4 ID:4128 **15:00**

Evolution of organics in the atmosphere: Dependence on technology of diesel vehicles and wood burning facility

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Recently it was shown that organics are an important fraction of PM1 in the whole Northern hemisphere (Jimenez et al., 2009). Most of the organics are found nearly everywhere as oxygenated organic aerosol and is interpreted mostly as secondary organic aerosol. Robinson et al. (2007) showed recently that semi-volatile or intermediate volatile organics are crucial for the formation of secondary organic aerosols. We show now in smogchamber experiments that the amount of primary and! secondary organic aerosol strongly depend on technology of the diesel vehicle or also on the wood burning technology when wood is used for heating. We show that a diesel oxidation catalyst strongly reduces the formation of secondary organic aerosols. Pellet burners are shown to be much cleaner than a log-wood stove not only concerning their primary carbonaceous emissions but concerning their secondary organic aerosol formation potential.

Jimenez, J.L. et al. (2009) Evolution of Organic Aerosols in the Atmosphere, Science, 326, 1525-1529.

Robinson, A.L. et al. (2007) Rethinking Organic Aerosols: Semivolatile Emissions and Photochemical Aging, Science, 315, 1259-1262.

Transformation 1.5 ID:4571 15:15

Are Aromatic Hydrocarbons Generated from the Atmospheric Oxidation of Biogenic Hydrocarbons Such as α-Pinene?

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Anthropogenic sources release into the troposphere a wide range of volatile organic compounds (VOC), including aromatic hydrocarbons. Biogenic volatile organic compounds (BVOCs) are also widely emitted, and are estimated to account for approximately 90% of total hydrocarbon emissions. When released into the troposphere, these BVOC and VOC undergo chemical oxidation, e.g. by hydroxyl radicals (OH) and ozone (O_3) during daytime, and by nitrate radicals (NO_3) and ozone at night. Care must be taken to distinguish direct anthropogenic emissions and products from BVOC and its oxidation products. For example, an aromatic hydrocarbon, p-cymene, was recently reported in the air above a forest canopy that had significant emissions of α-pinene, and its concentration increased as a function of altitude, suggesting its possible formation in the atmosphere. Consequently, the aim of the present study was to see if p-cymene is generated or not by the oxidation of α-pinene. Oxidation of α-pinene by O_3, NO_3 and OH has been studied at 1 atm air under dry conditions, at high relative humidity (70% RH), and with deliquesced sodium bisulfate (acid source) on the walls of a Teflon reaction chamber (100 L). Reactants and products were measured using GC-MS. The results of the work will be presented and the atmospheric implications discussed.

Observations 4.1 ID:4601 INVITED 08:30

Atmospheric Organic Particulate Matter: Revisiting its Sources, Properties and Impacts

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An overview of the development of our understanding of the sources, formation mechanisms, physical and chemical transformations of atmospheric organic aerosol (OA) is presented. Until recently, organic particulate material was simply classified as either primary or secondary with the primary component being treated in models as nonvolatile and inert. However, this oversimplified view fails to explain the highly oxygenated nature of ambient OA, the relatively small OA concentration gradients between urban areas and their surroundings, and the concentrations of OA during periods of high photochemical activity. A unifying framework for the description of all components based on their volatility distribution (the volatility basis set) can be used for the treatment of a wide range of processes affecting organic aerosol loadings and composition in the atmosphere. These processes include direct organic particle and vapor emissions, chemical production of organic PM from volatile precursors, chemical reactions (aging) in all phases, as well as deposition of both particles and vapors and chemical losses to volatile products. The combination of this new framework with the recent results of laboratory studies can resolve some of the discrepancies between OA observations and laboratory results.

To illustrate the advances in the experimental techniques and theoretical tools in the study of atmospheric organic aerosol we will combine ambient measurements from recent field campaigns in the US, Europe, and Mexico City with chemical transport models. We focus on the ability of these models to reproduce observations but also on their response to changes in emissions and climate.

Observations 4.2 ID:4143 09:00

Using ambient refractory particle mass to calibrate black carbon measurements made by laser-induced incandescence, thermal-optical transmittance, and filter-based photo-absorption techniques

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The total mass concentrations of fine-mode particles that were refractory at 400 °C (Mref) agreed with the mass concentrations of black carbon (BC) measured by the thermal-optical transmittance method (Mtot) in Tokyo, where the dominant sources of BC were emissions from diesel vehicles. The masses of the same refractory particles were also used to calibrate BC mass measured by a single particle soot photometer (SP2), which uses laser-induced incandescence. This calibration agreed well with that of fullerene soot, which indicates the consistency of the standards. Total BC mass concentrations measured with the filter-based absorption photometer continuous soot monitoring system referred to COSMOS (Mcosmos) operated at a wavelength of 565 nm with a heated inlet were calibrated by Mtot at six different sites in Asia with a variability of about 7% in 2004–2007. Mcosmos values were also measured simultaneously with total BC mass concentrations measured with an SP2 (Msp2) in Tokyo for 9 days in 2009. Mcosmos and Msp2 were highly correlated ($r2 = 0.97$) and agreed to within about 10% on average. These results demonstrate that Msp2, Mcosmos, and Mtot are nearly identical to Mref. The use of mass concentrations of refractory particles for calibration of BC mass concentrations determined by means of different techniques gave consistent results, which allowed for the unambiguous definition of BC.

Observations 4.3 ID:4333 09:15

Observations of large HNO3-containing particles and redistribution of reactive nitrogen in the 2010 winter Arctic stratosphere during RECONCILE

Hans Schlager [1], *Stephan Borrmann* [2], *C. Michael Volk* [3], *Jens-Uwe Grooß* [4], *Andreas Dörnbrack* [1], *Fred Stroh* [4], *Marc Von Hobe* [4]
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The sedimentation of HNO3-containing particles removes reactive nitrogen from air parcels in the polar vortex. Due to this denitrification chlorine remains longer in its active form and causes enhanced ozone destruction. It is assumed that large low-density HNO3-containing particles (so called NAT rocks) are mainly responsible for the denitrification in the winter Arctic stratosphere. However, observations of these large HNO3-containing particles are very sparse and details of the denitrification process are not well understood. This makes it difficult to predict denitrification and ozone loss accurately in models. Here we present the in situ detection of large HNO3-containing particles and redistribution of reactive nitrogen in the 2010 winter Arctic stratosphere during the RECONCILE (Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions) campaign using the NOy, particle, and tracer instruments on board the Geophysica high altitude aircraft. We discuss the spatial distribution of these large particles and the ambient conditions under which they were detected. For the first time large HNO3-containing particles were measured embedded in nitric acid trihydrate and supercooled ternary solution polar stratospheric clouds. We also observed for the first time these large particles during evaporation in renitrified air masses. The measured denitrification and renitrification layers during RECONCILE will be compared to model simulations of the redistribution of reactive nitrogen in the Arctic polar vortex considering the nucleation and sedimentation of large NAT particles using the model CLaMS (Chemical Lagrangian Model of the Stratosphere).

Observations 4.4 ID:4502 09:30

Transport effects on the vertical distribution of ozone over marine regions surrounding India

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As a part of the Integrated Campaign for Aerosol, gases and radiation Budget (ICARB) conducted during 18th March to 10th May, 2006, balloon borne ozone-sondes were launched to study vertical distribution of ozone over the Bay of Bengal (BoB) and the Arabian Sea (AS). A total of 29 ozone soundings were made on alternate days during the cruise. Higher ozone levels were observed above Marine Boundary Layer (MBL) up to 3 km over the BoB as compared to over the AS due to transport from the Indo-Gangetic plain, which is known to be a polluted region. This has been confirmed using back trajectories and Potential Source Contribution Function (PSCF). These measurements also show higher levels of ozone up to 3 km in the northern parts of the two marine regions especially over the BoB as compared to southern regions. These layers are characterized by higher temperature and lower humidity which indicate advection of continental air. On the contrary, in the free troposphere, ozone levels are lower over the BoB as compared to AS. This is due to strong convective activities over the BoB, which lifts poor ozone air in the MBL to the free troposphere. On the contrary, ozone rich air gets transported downwards over the AS especially over the northern AS. Thus, the two marine regions across the Indian sub-continent have opposite vertical transport,

which results in different ozone distributions. Detailed results will be presented.

Observations 4.5 ID:4543 09:45

Decadal trends in tropospheric ozone over East Asian Pacific rim during 1998-2007: Comparison to European and North American records, and implications for emerging Asian emissions impacts

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We examine springtime ozone trends at nine remote locations in East Asian Pacific rim during the last decade (1998-2007). The observed decadal ozone trends are relatively small at surface sites but are substantially larger at a mountainous site. The level and increasing rate of ozone at the mountainous site are both higher than those observed at background sites in Europe and North America. We use a regional chemistry-transport model to explore the observed changes and how changes in Asian anthropogenic emissions have contributed to the observed increasing trends. The model with yearly-dependent regional emissions successfully reproduces the levels, variability, and interannual variations of ozone at all the surface sites. It predicts increasing trends at the mountainous site, suggesting that increasing Asian anthropogenic emissions account for about half the observed increase. However, the discrepancy between the observation and model results after 2003 (the time of largest emission increase) suggests significant underestimation of the actual growth of the Asian anthropogenic emissions and/or incompleteness in the modeling of pollution export from continental Asia. These findings imply that improving emissions inventory and transport scheme is needed to better understand rapidly evolving tropospheric ozone in East Asia and its potential climatic and environmental impacts.

Observations 4.6 ID:4287 10:00

Aerosol characterization in the Amazon Basin during AMAZE-08: Fine particle composition and source apportionment

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As one of the major global sources of biogenic natural aerosol particles, the Amazon Basin plays an important role on the links between biological processes and climate. The sources, transformations, and properties of Amazonian aerosol particles are currently not well understood or quantified. In Feb to Mar 2008, as a part of the AMAZE-08 field campaign, submicron Amazonian particles were analyzed for the first time using an Aerodyne High-resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS). Mass concentrations, diel cycles, and size distributions of inorganic and organic species as well as the organic elemental composition were obtained. Results were compared with GEOS-Chem model predictions. Mass spectral characterization, including positive matrix factorization analysis, was used to test for and show the

minor contribution of primary biological particles in the submicron mode, the importance of long-range transport, and insights into the nature of the organic material. Moreover, in-cloud processing of isoprene and terpene oxidation products was investigated. The results show that biogenic secondary organic aerosol (SOA) production can dominate the submicron organic loading during times of diminished long-range transport influence. The campaign-average elemental O:C is similar to that measured in environmental chambers for SOA particles and to that expected for two oxygen-atom addition to each C5H8-mer of a biogenic volatile organic compound. Long-range transport can increase the organic loading by 50% and alter the oxidation state of organic material. The results presented provide mechanistic insight and quantitative parameters that can serve to increase the accuracy of models of the formation, transformations, and cloud-forming properties of biogenic natural aerosol particles.

Observations 4.7 ID:4219 **10:15**

North Atlantic marine boundary layer organic aerosol: sources and fluxes

Darius Ceburnis [1], *Andrius Garbaras* [2], *Soenke Szidat* [3], *Andre Prevot* [4], *Cristina Facchini* [5], *Colin O'Dowd* [1]
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It has become well documented that biological activity in the Earth’s oceans can contribute very significantly to primary as well as secondary organic aerosol, especially in the biologically productive North Atlantic Ocean. While the evidence linking coincident organic aerosol enrichment to biological activity has been compelling a direct proof of the link has been missing. As much important issue has been a magnitude of pollution in anthropogenically perturbed Northern Hemisphere. Using stable carbon and radiocarbon isotope analysis of organic aerosol we were capable not only proving a direct link between organic aerosol and biological activity, but provided a first quantitative estimate of marine organic carbon source at 80% and the magnitude of anthropogenic signal at about 20% in clean marine boundary layer over the North Atlantic. Dual isotope analysis is able to separate unambiguously and quantitatively marine, terrestrial non-fossil and anthropogenic fossil sources in all air masses. An intense debate is on-going on the importance of primary versus secondary organic aerosol in marine boundary layer. Gradient flux method has been demonstrated to be a promising tool by separating organic carbon into primary and secondary components and providing a first estimate of chemical fluxes. In general water insoluble organic carbon has been demonstrated to be of primary origin, which has been supported by laboratory experiments, while water soluble organic carbon was mainly secondary. While gradient method depends on a number of assumptions it nevertheless presents important insights into aerosol chemical species. An alternative novel method developed a combined organic-inorganic sea-spray source function using wind speed and ocean chlorophyll concentration. When implemented in the REMOTE regional climate model and/or global chemical transport model TM5 it compared well with available observations.

Transformation 2.1 ID:4469 INVITED 14:00

Heterogeneous chemistry in the dark: New insights into atmospheric budgets for reactive nitrogen and halogens

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The oxidative capacity of the atmosphere depends critically on the sources, sinks and lifetimes of trace species such as reactive nitrogen and halogen compounds. A key uncertainty in nitrogen oxide and chlorine budgets is the efficiency with which they are consumed and / or produced by heterogeneous or multiphase reactions (i.e., those where a gas phase molecule reacts on or in a solid or liquid substrate suspended in the atmosphere) that occur in the nighttime atmosphere. Recent advances in optical instrumentation and mass spectrometry have enabled detailed atmospheric measurements of these nighttime nitrogen oxide and halogen compounds in the lower atmosphere. A key finding from recent field campaigns is that release of ClNO2 (nitryl chloride) formed from heterogeneous/multiphase uptake of N2O5 is surprisingly efficient and remarkably widespread. This halogen species is unreactive in the dark but photolyzes in sunlight to provide atomic chlorine. Production of ClNO2 spans a wide variety of environments and may represent a large fraction of the global tropospheric halogen budget. If so, tropospheric halogens may derive in large part from anthropogenic emissions of NOx rather than from natural sources. This presentation will describe results from recent measurements of N2O5 and ClNO2 and the evidence for ClNO2 as a major source of active chlorine in the lower atmosphere.

Transformation 2.2 ID:4301 14:20

OH variability in the period 1988-2008 inferred from the global methyl chloroform budget

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Inter-annual variability in the oxidizing capacity (the OH climate) is driven by variability in the sources and the sinks of the OH radical. Short-wave solar radiation, ozone, and water vapor variations may induce variations in OH production, but also the abundance of NO is known to stimulate OH recycling. Important sinks for OH are CO and CH4, but regionally non-methane hydrocarbons can play an important role as well. Thus, many natural processes influence the OH abundance, ranging from biomass burning (CO emissions), El Nino Southern Oscillation (water vapor, wetland CH4 emissions), and the ozone layer (radiation). On top of the natural factors, anthropogenic emissions may perturb global OH chemistry, possibly leading to long-term trends. One important question is how large the inter-annual variations in OH can be, as they provide indications about the sensitivity of the atmospheric oxidation capacity to perturbations. Previous analyses of the methyl chloroform budget seem to indicate that the inter-annual variability in OH is in the order of 7-8%. This, however, is at odds with the CH4 budget, that allows maximum variations in the order of maximum 5%. In this contribution, a simple analysis of the methyl chloroform budget will be presented in which realistic assumptions about the methyl chloroform emission uncertainty are used to estimate OH variability. Preliminary results show that variability in the derived OH is smaller than 2%, with variations that can be

linked to know variations in the OH climate.

Transformation 2.3 ID:4224 14:35

Are megacity's oxidizing environments changing?

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Elevated levels of ozone, which are harmful to human health, have been observed in many urban areas and megacities. The main precursors to ozone formation are volatile organic compounds (VOCs) and nitrogen oxides (NOx). Until now the reduction of ozone through the regulation of ozone precursors has been focused on control of VOC emissions from road transport and petrol evaporation as these are the largest sources of VOCs in urban areas. As air quality regulations further reduce anthropogenic emissions, one would expect that the role that biogenic emissions play in the formation of ozone will become increasingly important. Isoprene, a tracer for biogenic emissions, has one of the highest photochemical ozone creation potentials of a large number of volatile organic compounds (VOCs). Additionally, biogenic VOCs have been shown to have a significant impact on summer-time ozone formation not only in rural areas where the most vegetation is present, but also in urban areas. However, while their importance is increasing, their overall contribution to reactivity with respect to OH remains relatively small. Nitrogen dioxide emissions however, have been increasing in the past 10 years and their relative contribution to reactivity with respect to OH has also been increasing. That the oxidizing environment of a megacity (London) and the surrounding region has been changing is shown from the examination of the long-term trends in VOCs, carbon monoxide, methane, and NOx at a number of sites in and around London. The relative loss of these compounds with respect to the OH radical shows that the oxidizing balance of megacities and the surrounding regions have been and are changing over time. Data from monitoring sites in and around London are used to compare the oxidative environment in the city centre to urban background and semi-rural locations.

Transformation 2.4 ID:4397 14:50

A study of halogen chemistry in the tropical Atlantic Ocean boundary layer

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The role of halogenated species (chlorine, bromine, iodine) in the oxidative chemical processes occurring in the marine boundary layer (MBL) has been investigated for several years, but many uncertainties remain in our knowledge of the sources, sinks and reactivities of these species. Measurements taken at the Cape Verde Atmospheric Observatory (16°51'N, 24°52'W) from 2007 to 2009 and on the RRS Discovery during the RHaMBLe (Reactive Halogens in the Marine Boundary Layer) campaign in May-June 2007 have shown that significant concentrations of bromine and iodine species are present in the tropical MBL.

In this work, the one-dimensional model MISTRA (v7.4.1), which includes a detailed description of meteorological and microphysical processes and up-to-date gas and aqueous phase chemical mechanisms, was used to simulate the observations of bromine and iodine species in the region surrounding the Cape Verde islands under different scenarios. Sea-air fluxes derived from measurements of organic iodine compounds during the RHaMBLe cruise were included in the model.

Using the model results and the field observations we have investigated the sources of inorganic iodine in

the tropical MBL and tested our understanding of gas-phase iodine chemistry. We found that the measured fluxes of organic iodine are not able to explain the observations of inorganic iodine in Cape Verde, suggesting that other sources exist in the open ocean. The uncertainties in the chemical mechanism and potential source mechanisms will be discussed. The model results were then used to analyze the impacts of the observed levels of halogens on the concentrations of ozone and of dimethyl sulphide (DMS) in the tropical MBL and the consequences for the nitrogen and sulphur budgets.

Transformation 2.5 ID:4439 15:05

An evaluation of upper tropospheric NOx/ozone chemistry during INTEX-A using CMAQ with a modified CB05 chemical mechanism and lightning NO emissions

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Aircraft measurements during the summer 2004 INTEX-A field campaign over North America and the western Atlantic revealed unexpectedly large amounts of NOx in the upper troposphere. Modelers have found that doubling the lightning-NO production per flash brings model-calculated and measured NOx during the INTEX-A campaign into better agreement; however low-biases in upper tropospheric NOx still remain. These biases could indicate continued problems with the parameterization of deep convection and its associated lightning; however, the too rapid conversion of NOx to HNO3 and PAN in the model simulations may also indicate a fundamental problem with upper tropospheric NOy chemistry. In this study, upper tropospheric photochemistry during the INTEX-A period will be evaluated using CMAQ with the standard CB05 chemical mechanism and with a CB05 based mechanism that includes adjusted radical amplification and cycling efficiency. Results will be compared to a variety of data sets including INTEX-A measurements, SCIAMACHY NO2 retrievals, and IONS ozonesondes.

Transformation 3.1 ID:4493 15:50

Pollutant Plume Processing during Long-range Transport to the Arctic

Kathy Law [1], *Antoine Auby* [1], *Francois Ravetta* [1], *John Burkhart* [2], *Anke Roiger* [3], *Hans Schlager* [3], *Steve Arnold* [4], *Arctas Science Team* [5], *DLR-Falcon Science Team* [6], *ATR-42 Science Team* [7]
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As part of IPY POLARCAT, several aircraft campaigns investigated the long-range transport of pollutants to the Arctic. In summer 2008, anthropogenic plumes from North America, and forest fire plumes from Canada and Siberia, were sampled by the French ATR-42 and German DLR-Falcon during flights from Kangerlussuaq (Greenland). The origins of the various plumes were identified in terms of their chemical composition and results from a Lagrangian multi-parcel model, FLEXPART. Post-campaign analysis

identified pseudo-Lagrangian links between upwind flights made by the NASA DC8 over Canada and the downwind flights over Greenland. Changes in trace gas and aerosol plume concentrations allow quantification of processes influencing pollution during long-range transport into and out of the Arctic. Simulations using a photochemical model, CiTTyCAT, have been used to investigate the photochemical and aerosol evolution of plumes transported from North America to Greenland. For example, we quantify processes influencing O3 concentrations during long-range transport including photochemical processing, wet deposition and the role of PAN as a NOx reservoir in Arctic. Aerosol processing may also be quantified as well as impacts of aerosols on photochemistry (e.g. via impacts on photolysis rates). The role of mixing between polluted plumes and other air masses (notably of upper tropospheric origin) is also investigated. Estimates of in-situ photochemical O3 production and mixing rates from Lagrangian studies can be used to validate results from global chemistry-transport models and to provide constraints on the different contributions to the Arctic ozone budget.

Transformation 3.2 ID:4235 16:05

Assessment report from the Task Force on Hemispheric Transport of Air Pollution

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The Task Force on Hemispheric Transport of Air Pollution (TF HTAP) under the Convention on Long Range Transboundary Air Pollution (CLRTAP) was established to improve our understanding of the intercontinental transport of air pollution in the Northern Hemisphere. The Task Force, led by the United States and the European Commission, is releasing an assessment of hemispheric transport in summer of 2010. This assessment focuses on atmospheric transport of ozone, fine particles, mercury, and persistent organic pollutants. For each of these pollutants, measurements, emissions, modeling, and impacts are considered. The report draws from peer-reviewed scientific literature, other traceable reports, and original analyses, including a set of multi-model experiments to estimate source-receptor relationships. This presentation will outline the process by which the assessment was created and summarize the main findings and recommendations of the assessment.

Transformation 3.3 ID:4317 16:20

Investigation of ship-plume chemistry using a newly-developed photochemical/dynamic ship-plume model

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A photochemical/dynamic ship-plume model, which can consider the ship-plume dynamics and ship-plume chemistry, simultaneously, was developed to gain a better understanding of atmospheric impact of ship emissions. The model performance was then evaluated by a comparison with the observation data measured on a NOAA WP-3D flight during the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) airborne field campaign. The simulation conditions and parameters, such as meteorological conditions, emission rates, and background gas and particulate species concentrations, were obtained directly and/or inferred indirectly from the ITCT 2K2 observation data. The model-predicted concentrations showed good agreement with the observed concentrations of five ambient species (NOx, NOy, ozone, HNO3, and H2SO4)

at the eight plume transects by the WP-3D flight with strong correlations around the 1:1 line. In addition, a set of tests were carried out to approximate the magnitude of the reaction probability of HNO3 onto sea-salt particles in the model-observation comparison framework. These results suggest that the reaction probability of HNO3 onto sea-salt particles may be in the order of 0.05-0.1. The equivalent NOx lifetime throughout the “entire plume” was also estimated from photochemical/dynamic ship-plume modeling. The NOx lifetimes estimated throughout the entire ship plume ranged from 2.64 hrs to 3.76 hrs under stable to neutral stability conditions. The short NOx lifetime over the entire ship plume clearly shows that the ship-plume chemistry shortens the NOx lifetime considerably. Therefore, the ship-plume chemistry model should be used to model the changes in ship-plume chemical compositions and better evaluate the atmospheric impact of ocean-going ship emissions.

Wake-up 1.1 ID:4247 INVITED 09:00

Chemistry-Climate Interactions and the Role of Short-lived Climate Forcers

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Short-lived aerosols, tropospheric ozone and methane influence climate in numerous ways. I will discuss current understanding of the chemistry-climate interactions that govern these species effect on the Earth's radiation budget, and the manifold climate responses to these various forcing agents, paying particular attention to assessing the impacts of individual activities rather than individual pollutants. I will contrast the impacts of the short-lived species with those of carbon dioxide as a function of space and time, and briefly discuss how this affects the choice of metrics used to compare pollutants or to evaluate the response to emissions changes.

Wake-up 1.2 ID:4283 09:30

Bounding the Role of Black Carbon in Climate

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Black carbon is the most strongly absorbing aerosol in the atmosphere. It is a large contributor to present-day climate forcing, along with other short-lived pollutants. Emission changes, either due to technological shifts or to targeted reductions, will also affect total forcing in the near future. Understanding the magnitude of these potential changes is important, despite the substantial scientific uncertainties. We present a study sponsored by the Atmospheric Chemistry & Climate initiative, which relied on about thirty co-authors. Its goal has been determining a best estimate and uncertainties for the magnitude of radiative forcing and climate response to a unit change in emission. While black carbon aerosol is the focus of the study, it is never emitted in isolation. Thus, we examine sources that are rich in black carbon, but we estimate the impact of all short-lived, co-emitted pollutants from those sources. We review and update estimates of direct atmospheric forcing by black carbon, and account for aerosol mixing state and vertical distribution. Without changing atmospheric burden, forcing estimates are somewhat higher than those of the Intergovernmental Panel on Climate Change (IPCC). We account for the probable bias and uncertainties in atmospheric concentrations to yield new forcing estimates. We also account for changes in clouds and snow albedo (presented elsewhere), and for the efficacy of atmospheric, cloud and snow forcing. Finally, we present net impacts for several sources rich in black carbon: diesel engines, wood cooking, wood heating, industrial coal, and agricultural burning.

Wake-up 1.3 ID:4499 INVITED 09:45

The Impacts of Megacities on Air Quality and Climate Change: An IGAC Perspective

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With increasing global population and continuous urbanization, in 2008, for the first time in human history over half the world's population resides in urban areas. In the mean time, the number of megacities (cities with population over 10 million) has reached to 19 and expected to reach 27 in 2025. Megacities are the areas with the most intensive human activities; they impact on terrestrial and aquatic ecosystems, and are major sources of air pollutants and greenhouse gases, due to intensive economical and social activities as well as energy consumption in megacities and their surrounding regions. It's long been recognized that air pollution has negative impacts on human and ecosystem health. More recently, the role of anthropogenic emissions in altering climate has come to the fore. While the former has typically been considered a local issue and the latter global, they are linked through atmospheric concentrations of short-lived species such as aerosols and ozone. Experience of developed counties show that the pronounced air quality degradation accompanied past development can be avoided. The scientific and engineering knowledge accumulated as earlier developing megacities dealt with their air quality problems is a significant resource for current and future developing megacities. With the growing trend towards urbanization, understanding the role of megacities in local to global atmospheric chemistry is critical to effectively addressing both of these issues. An assessment on the impacts of megacities on air quality and climate change will be presented. The importance of mega-cities as an emission source of global greenhouse gases and the impacts of megacities on climate through emission and photochemical formation of ozone and aerosols will be discussed.

Wake-up 1.4 ID:4239 10:15

Atmospheric chemistry in volcanic plumes

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Recent field observations have shown that the atmospheric plumes of quiescently degassing volcanoes are chemically very active, pointing to the role of chemical cycles involving halogen species and heterogeneous reactions on aerosol particles that have previously been unexplored for this type of volcanic plumes. Key features of these measurements can be reproduced by numerical models such as the one employed in this study. The model shows sustained high levels of reactive bromine in the plume, leading to extensive ozone destruction, that, depending on plume dispersal, can be maintained for several days. The very high concentrations of sulfur dioxide in the volcanic plume reduces the lifetime of the OH radical drastically, so that it is virtually absent in the volcanic plume. This would imply an increased lifetime of methane in volcanic plumes, unless reactive chlorine chemistry in the plume is strong enough to offset the lack of OH chemistry. A further effect of bromine chemistry in addition to ozone destruction shown by the model studies presented here, is the oxidation of mercury. This relates to mercury that has been coemitted with bromine from the volcano but also to background atmospheric mercury. The rapid oxidation of mercury implies a drastically reduced atmospheric lifetime of mercury so that the contribution of volcanic mercury to the atmospheric background might be less than previously thought. However, the implications, especially health and environmental effects due to deposition, might be substantial and warrant further studies, especially field measurements to test this hypothesis.

Wake-up 2.1 ID:4414 INVITED 11:00

Observing the troposphere with IASI: emission, chemistry and transport

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Since about one decade thermal infrared (TIR) optical sounders are providing measurements of several reactive species, such as carbon monoxide or ozone, with good sensitivity to the troposphere. They add to the products available from UV-visible satellite sounders and these altogether contribute in drawing a more complete picture of the tropospheric composition, its changes over space and time, and its impact on the global environment. There is currently a suite of TIR instruments in operation, with very different instrumental designs, driven by specific science objectives. Among these, IASI, the Infrared Atmospheric Sounding Interferometer onboard MetOp satellite, has the advantage as an operational mission supporting Numerical Weather Predictions, of a relatively high spatial resolution (12 km footprint at nadir) and high spatial and temporal sampling. It covers an extended portion of the thermal infrared (645-2760 cm-1) without gaps, and therefore offers unprecedented possibilities for the sounding of the troposphere in near-real time and for capturing sudden changes in the atmosphere.

This presentation gives an overview of the IASI achievements after 2.5 years of operation and discusses the long-term plans for IASI-like missions in the framework of the GMES EU program (after 2020). The chemistry products available from the mission (CO, O3, HNO3, NH3, HCOOH and CH3OH) are reviewed, with detailed characterizations of accuracy and vertical sensitivity. Based on these, the capabilities of IASI to contribute to several aspects of atmospheric chemistry, from the identification of local sources to the long-range transport of pollution, are discussed with concrete examples. The presentation also highlights the recent exploitation of IASI data for operational applications (monitoring of fires, volcanoes, dust events and pollution episodes).

Wake-up 2.2 ID:4401 INVITED 11:30

Inferring surface fluxes of trace gases from space-borne data: current and future science

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Only over the past decade has the necessary technology begun to match our desire to observe surface air pollutants and climate-relevant trace gases in the lower troposphere where we live and breathe. A new generation of Earth-observing satellites, capable of probing the lower troposphere, are already orbiting the Earth with more ready for launch or in the planning stages. Consequently, this is one of the most exciting times for Earth system scientists who study the countless current-day physical, chemical and biological interactions between the Earth's land, ocean and atmosphere. I will present some recent results that exemplify the Earth system approach using space-borne data, and also discuss how we can address the new challenges associated with observing projected changes during this century.

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