

IGAC *tivities*

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A Note From the IGAC Chair: *Guy Brasseur*

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Integration and Synthesis: The "Aspen Meeting"

Over the last year, a number of members of the atmospheric chemistry community have been busy preparing chapters of the IGAC integration and synthesis report. This document, which will be published as part of the IGBP Book Series, will address two general, but important questions. First, what has the scientific community learned over the last 10-15 years regarding the chemistry of the atmosphere and its relations with the biosphere? And second, what are the remaining important questions that must be addressed in the future?

Three main topics will be reviewed in relation to the first question: (1) interactions between the atmosphere and the terrestrial and marine biosphere, (2) the photochemistry of the troposphere, and (3) atmospheric aerosols. Advances in instrumentation and models will also be reported. Answering the second question will provide an opportunity for the community to express what should be the major scientific priorities for the decade ahead. Preparation of this part of the document is being led by the Commission on Atmospheric Chemistry and Global Pollution of IAMAS, one of the parent organizations of IGAC.

Preliminary chapter texts are currently under "friendly" review by a few experts in the respective fields. Extensive discussion of revised texts will occur during a gathering of 100-120 scientists on 27 April through 2 May 2000 at the Aspen Institute in Aspen, Colorado (USA). During this meeting an executive summary will be drafted and the basis for the report chapter on future scientific priorities will be developed. Several program managers specializing in atmosphere chemistry and related disciplines are being invited to participate.

This integration and synthesis is an important milestone for IGAC because it will summarize 10-15 years of very active research. But more importantly, it will endeavor to relate this research to the broader context of global change science, thus contributing to the synthesis effort being undertaken by IGBP, IGAC's other parent organization. Finally, it will help to establish the basis for new and challenging scientific directions for our community.

The Aspen meeting is open to the entire community. If you wish to attend, please let us know by contacting Ms. Harriet Barker (Fax: (+1-303) 497-8770; Email: harriet@atd.ucar.edu) who is coordinating its planning.

In closing, I would like to thank the many scientists who have contributed to the integration and synthesis report thus far, and especially the lead authors of all the chapters. Hopefully, this very demanding exercise will be useful to the entire community as well as to decision makers in government and industry who rely very often on scientific expertise to make important societal choices and political decisions. More and more, issues related to air quality, global and regional pollution, and climate and global change will be on the political agenda.

The East Asian/North Pacific Regional Experiment (APARE)

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The East Asian continental rim region is characterized by anthropogenic emissions that are already high in many locales and that are rapidly growing throughout much of the region. For example, recent projections indicate that anthropogenic NO_x emissions from Asia, primarily from the combustion of hard coal and liquid fuels for power, transportation and industry will continue to rise for the foreseeable future. An increase in NO_x emissions of 350% from 1990 to 2020 is predicted if no further pollution controls are implemented [*van Aardenne et al.*, 1999]. Furthermore, emissions from eastern Asia are increasing more rapidly than in most other parts of the world. Within two decades, emissions from eastern Asia could account for roughly half of the S and N and a third of the C emitted from all anthropogenic sources worldwide [*Galloway et al.*, 1998]. The Asia/western Pacific region has a unique mixture of aerosols and trace gases because of these distinctive patterns of emissions in combination with the particular meteorological conditions affecting the region.

Aeolian dust and gaseous and particulate pollutants from the Asian continent are transported eastward over the Pacific, especially in the spring, and the effects of these materials are widespread [*Berntsen et al.*, 1999; *Jaffe et al.*, 1999]. For example, growing levels of nitrogen oxides from the expanding transportation sector in Asia will change the oxidizing capacity of the atmosphere beyond the regional scale [*Elliot et al.*, 1997]. Similarly, recent modeling studies indicate that increasing emission from fossil fuel combustion in Asia will affect surface ozone concentrations in the United States [*Jacob et al.*, 1999]. These authors suggest that the long-range transport of Asian O₃ could even offset some expected benefits of reduced emissions of NO_x and hydrocarbons in the western US.

Anthropogenic materials in the atmosphere can also influence climate by altering the Earth's radiative balance. Concerns over climate forcing have led to the development of the ACE-Asia experiment, which focuses on the Asia/Pacific region. ACE-Asia is the third in a series of Aerosol Characterization Experiments (ACE) planned under IGAC auspices (for further information see <http://saga.pmel.noaa.gov/aceasia/>). The TRACE-P (TRansport and Chemical Evolution over the Pacific) Pro-

gram, part of NASA's Global Tropospheric Experiment, will focus on outflow from Asia with the goals of (1) understanding and quantifying the export of chemically and radiatively important gases and aerosols, and their precursors and (2) determining and understanding the evolution of these air masses (for further information see <http://www-as.harvard.edu/chemistry/trop/tracep/>). Continental outflow also affects biogeochemical cycles of the North Pacific [*Duce et al.*, 1991], and the associated effects on marine biology and chemistry could also have climate implications.

Despite the recent interest and obvious urgent need for systematic studies of atmospheric chemistry and the impacts of natural and anthropogenic substances in this region, little research had been done on these topics until the early 1990s. The East Asian/North Pacific Regional Experiment (APARE) was established as an IGAC Activity in March 1990 largely to address this need. Thus, the APARE program has been concerned with the oxidizing capacity of the regional atmosphere, the tropospheric ozone budget, the deposition of acidic substances, and the effects of continental emissions on marine atmospheric chemistry and biogeochemistry. Marine activities carried out in conjunction with APARE include cooperative endeavors with the Joint Global Ocean Flux Study (JGOFS), another IGBP Core Project. The following goals and tasks are the focus of APARE:

Goals

- To assess transport and chemical transformations of air pollutants over the East Asian continent and the northwestern Pacific Ocean.
- To determine the deposition of primary and secondary pollutants (sulfate, nitrate, organics) in the East Asian region.

Tasks

- Emission inventory and air chemistry database
- Surface measurements of important trace gases
- Intensive field programs
- Ground-surface monitoring network

Scientists from Australia, China, Japan, Korea, Taiwan and the United States took part in the first APARE planning meeting, which was held in Tsukuba, Japan. Dr. Hajime Akimoto, then at the National Institute of Environmental Sciences, was elected as convener of APARE. Under his leadership, the APARE community has successfully undertaken several sub-programs, including the

Pacific Exploratory Mission-Western Pacific (PEM-West), Perturbation of the East Asian Continental Air Mass to the Pacific Oceanic Troposphere (PEACAMPOT) and CATS (Chemistry of the Atmosphere, Taiwan Station), which were proposed and accepted as international projects by the APARE Coordinating Committee.

Review of APARE/IGAC Activities

- 1st Planning Workshop, March 1990, Tsukuba
- 2nd Planning Workshop, March 1991, Tsukuba
- 1st Intensive Field Campaign, PEM-West PEACAMPOT/CATS: Phase A, Sept.-Oct., 1991
- 3rd Planning Workshop, Nov. 1992, San Francisco
- International Conference on Regional Environment and Climate Change in East Asia, 1993
- 4th Steering Committee Meeting, Dec. 1993, Taipei
- 2nd Intensive Field Campaign, PEM-West/PEACAMPOT/CATS/H.K. Station: Phase B, Feb./March, 1994
- Regional Environmental Changes in East Asia, APARE/IGAC special issue of *Terrestrial Atmospheric and Oceanic Sciences*, Vol. 6, Number 3, Sept. 1995
- 5th Planning Workshop, January 1996, Tokyo
- PEM-West A Special issue, *J. Geophys. Res.*, 101, D1, Jan. 1996
- 6th Planning Workshop, March 1997, Hong Kong
- PEM-West B Special issue, *J. Geophys. Res.*, 102, D23, Dec. 1997
- 7th Planning Workshop/Tour, March 1998, Seoul/Cheju
- APARE/TRACE-P Planning Meeting, Oct. 1998, Hong Kong
- 8th Planning Workshop/Tour, June 1999, Honolulu/MLO

At the 8th Planning Workshop in 1999, the APARE Coordinating Committee endorsed both the TRACE-P campaign and the ACE-Asia experiment. Multi-platform field campaigns for both of these programs are planned for spring 2001 with particular emphasis on continental outflow, including studies of trace gas and aerosol composition and reactions, aerosol physical and optical properties, radiative effects of the aerosols, and chemical evolution. At present, scientists from China (including Hong Kong), Japan, Korea, and Taiwan are operating a network of ground surface monitoring sites in East Asia in conjunction with APARE. These sites will form the backbone of an ACE-Asia ground station network, providing data on aerosol chemical, physical and optical properties that will be used to assess the spatial and temporal (seasonal and inter-annual) variability of these properties and the factors controlling this variability.

The current members of the APARE Coordinating Committee are:

M. Uematsu	(Univ. of Tokyo, Convener)
M. Anson*	(Hong Kong Polytechnic University)
R. Arimoto	(New Mexico State University)
Y. Kondo	(Nagoya University)
C.M. Liu*	(National Taiwan University)
R. Newell	(Massachusetts Inst. of Technology)
S.G. Shim	(Korea Inst. Science & Technology)
J. Tang	(Chinese Meteorol. Administration)
Y. H. Zhang	(Peking University)

*will rotate to new members during 2000.

The APARE community is rather unique because it is a non-governmental organization composed of scientists who are doing atmospheric chemistry research in the Asia/Pacific Rim region. This Activity presents valuable opportunities for those interested in exchanging information concerning ongoing projects, for discussing the scientific results from their experiments, and for coordinating future projects with scientists from multiple countries through APARE workshops, especially those in the Asian countries. The APARE Coordinating Committee exists to communicate, create, and conduct atmospheric research with scientists interested in the East Asia region. At the most recent meeting, the Committee agreed to begin contacting scientists from other Asian countries to broaden the geographical coverage and possibly enlarge the scope of APARE. The 9th workshop will be held at Xi Ning, China in June 2000.

The following articles in this issue of *IGAC Activities* summarize several recent scientific projects undertaken in cooperation with APARE.

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Biomass burning and lightning experiment (BIBLE)

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Introduction

The Earth Observation Research Center of the National Space Development Agency of Japan (NASDA) is implementing its scientific activities in association with the NASDA Earth observing satellites. The program for atmospheric chemistry—Global Atmospheric Chemistry Experiment (GLACE)—covers tropospheric and stratospheric chemistry, and is designed to be carried out in close collaboration with international projects such as IGAC and SPARC. One of the core activities of GLACE is the Biomass Burning and Lightning Experiment (BIBLE). Started in 1997, BIBLE is a tropospheric chemistry research project which uses aircraft as a sampling platform to study important natural and anthropogenic processes in the tropical Asia/Pacific region.

Objectives of BIBLE

Tropical Asia, in particular the region stretching from Indonesia to northern Australia, is a place where trace gas and aerosol distributions in the troposphere are strongly influenced by deep convection, frequent lightning and biomass burning. Quantitative studies of these processes in the region have been limited because of the lack of simultaneous measurements of key species. The BIBLE activity is comprised of *in situ* observations aboard an aircraft supplemented with simultaneous satellite observations and numerical modeling efforts. BIBLE uses a Gulfstream II (G-II) jet plane chartered by NASDA. Coordinated ground-based measurements of trace gases and aerosols, including balloon-borne ozonesonde measurements, are also carried out at strategic times. Instruments used for the BIBLE aircraft measurements conducted in 1998 are listed in Table 1.

The NO concentration in the tropical maritime troposphere is usually quite low, and as a result, ozone tends to be destroyed there rather than produced (ozone transported into the region is photochemically removed) [Crawford *et al.*, 1997]. However, very different conditions occur in the presence of biomass burning products, when lightning occurs, and during periods of strong convective transport. The BIBLE campaign was planned to evaluate these three influences.

During the dry season, biomass burning is an important source of ozone precursors and aerosols for the tropical troposphere, and ozone formation can occur in biomass burning plumes originating in Indonesia and tropical Australia. Late in the dry season (September-October) in the El Niño years of 1994 and 1997, large scale biomass burning occurred in Indonesia, leading to significant increases in tropospheric ozone [Fujiwara *et al.*, 1999; Kita *et al.*, 2000]. Biomass burning has been reported to occur regularly in northern Australia in the dry season. Major scientific objectives involving biomass burning for BIBLE include the following:

- Estimate the amounts of trace gases and aerosols emitted by biomass burning
- Study the vertical transport of biomass burning plumes
- Study the long-range transport and chemical transformation of the biomass burning plumes
- Evaluate the impact of production of ozone precursors on ozone photochemistry

During the rainy season, lightning and convective transport play important roles in the distributions of trace gases in tropical Asia [Kawakami *et al.*, 1997; Koike *et al.*, 1997]. In particular, it has been shown that convective activity associated with the Walker circulation influences trace gas distributions in subtropical regions of the western Pacific [Koike *et al.*, 1997]. Major scientific objectives involving lightning and convective transport to be studied during the 2000 BIBLE mission are to:

Table 1. Instruments aboard the GII Aircraft during the BIBLE Experiments in 1998

Species/items	Technique(s)	Sampling interval
O ₃	UV absorption	1 s
NO and NO _Y	Chemiluminescence	1 s
CO	G C/HgO-reduction	20 s
PAN	Cold trap/GC-ECD	15 min
CO ₂	IR absorption	1 s
H ₂ O	Dew point hygrometer	10 s
NMHCs (C2-C10)	Whole air sampling/GC	5 min
Halocarbons (C2-C10)	Whole air sampling/GC	5 min
Alkyl nitrates (C1-C4)	Whole air sampling/GC	5 min
Aerosol size distribution	MASP*	10 s
CN	CN counter	1 s
JNO ₂ , JO(1D)	Radiometers	1 s

*Multiangle Aerosol Spectrometer Probe

- Statistically evaluate the NO_x production rates by lightning
- Make a detailed study of NO production by single thunderclouds
- Characterize the impact of convection (ITCZ, SPCZ) on the transport of trace gases
- Evaluate the efficiency of transport across the ITCZ and SPCZ

Thus far the BIBLE science team has conducted a test flight campaign (BIBLE-T) and two major deployments (BIBLE-A and B) as listed in Table 2. During BIBLE-A, the G-II flew from Nagoya (36°N) to Bandung (7°S), Indonesia via Saipan (15°N), Biak (1°S), and Darwin (12°S). The return route was Bandung-Biak-Saipan-Nagoya. The data for BIBLE-T and BIBLE-A will be available to the public around July 2000. A third deployment (BIBLE-C) is to be conducted in late 2000.

Table 2. Major Deployments for BIBLE

Phase	Period	Major base
T	April, 1998	Nagoya, Japan
A	September 24–October 10, 1998 (Dry season)	Bandung, Indonesia
B	August 30–September 14, 1999 (Dry season)	Darwin, Australia
C	November–December 2000 (Rainy season)	Darwin, Australia

After BIBLE-C, the program scientists are planning a new aircraft campaign mostly in and around Japan in May 2001, in close coordination with US NASA's TRACE-P program, which is planned for April 2001. One of the important objectives of TRACE-P is to determine the chemical composition of the Asian outflow over the western Pacific. The BIBLE campaign, in combination with TRACE-P, will significantly enhance the understanding of the seasonal progression of the outflow from continental Asia along with the associated photochemistry.

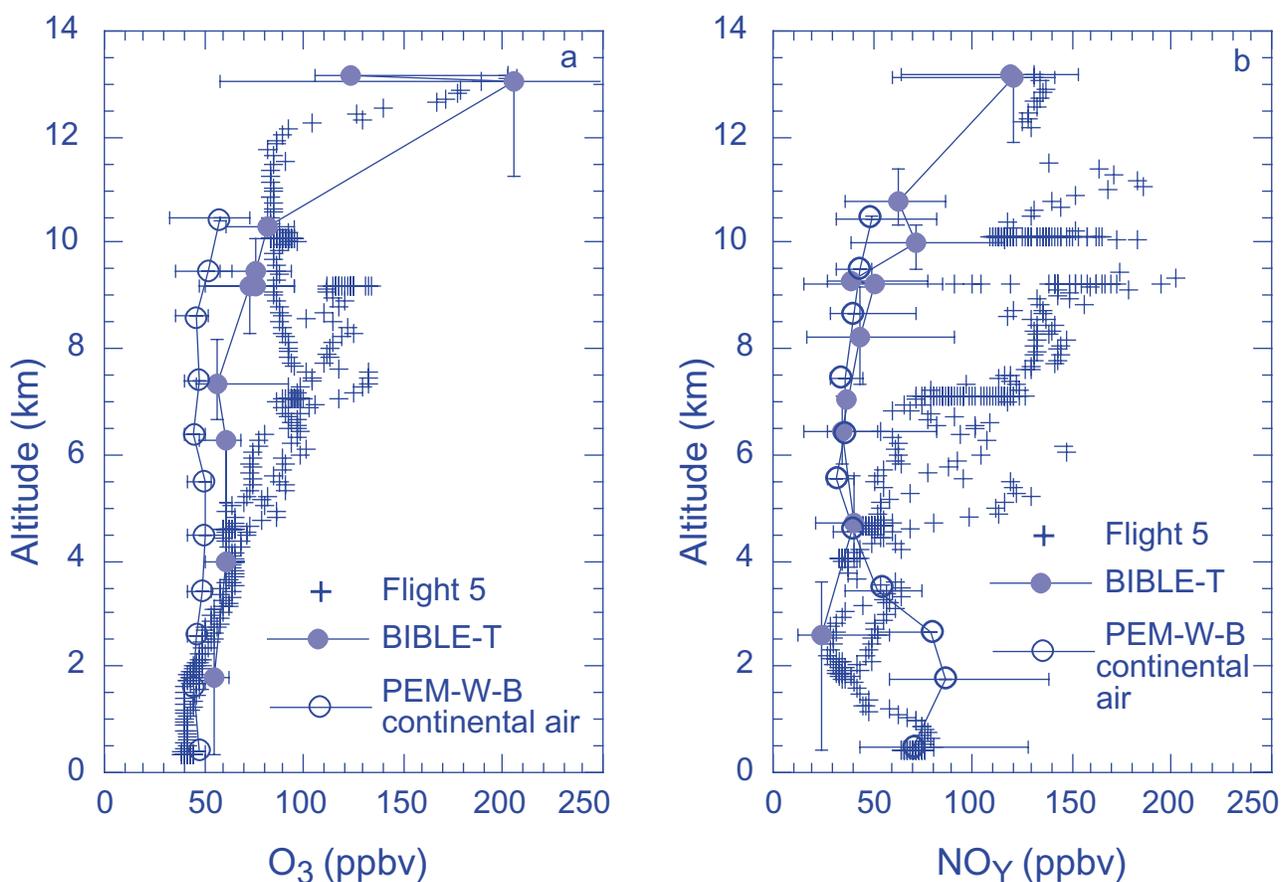


Figure 1: Ozone (a) and NO_y (b) profiles obtained in April 1998 during BIBLE-T. The data for flight 5 are shown as crosses. The median values for all BIBLE-T observations (closed circles) and PEM-West B continental air (open circles) are shown for comparison.

Measurements over the western Pacific Ocean

The data obtained over the mid-latitude and subtropical Pacific Ocean are most relevant to APARE. These data were obtained during BIBLE-T and transit flights during BIBLE-A and B. Some of the interesting features obtained by these measurements are described below.

BIBLE-T: Influence from surface sources in Northeastern China

Five flights were made from Nagoya between April 17 and 24, 1998. The region covered by these flights was 26 to 44°N and 136 to 144°E. Profiles of O_3 and NO_y are shown in Figure 1. For reference, the median values obtained in the continental air masses during PEM-West B conducted in March 1994 [Kondo *et al.*, 1997] are also shown. For further comparison, median O_3 profiles from the Japanese ozonesonde stations at Naha (26°N), Kagoshima (32°N), Tateno (36°N), and Sapporo (43°N) were derived. The median O_3 values at 2-12 km in April of 1993-97 were 60-80 ppbv, which were close to the BIBLE-T values. The median ozonesonde values in February-March were about 10 ppbv lower than April. The median BIBLE-T NO and NO_y values were 20-80 and 400 ± 100 pptv, respectively, similar to those for PEM-West B at 4-11 km. However, the NO , NO_y , and NO/NO_y values during flight 5 made on April 24 were much higher than the average BIBLE-T values in the upper troposphere.

A detailed meteorological analysis indicates that the air mass sampled during this flight was strongly influenced by convection over northeastern China 1 to 2 days prior to the sampling. The convection, which reached up to

12 km, was associated with cyclonic activity. The NMHC and CO concentrations suggest that high O_3 and NO_y values were anthropogenic in origin. Strong convection, such as that encountered during BIBLE-T, can transport pollutants far over the Pacific Ocean in conjunction with the strong westerly flow. In April, solar UV intensity is higher than February-March. If the mixing ratios of key precursors of ozone, such as NO , CO , and NMHC, remain similar or increase from March to April, the net photochemical ozone production rate should also increase over this time period. However, more systematic measurement and modeling studies are needed to evaluate the importance of convective transport of precursor gases during the whole spring. An intensive aircraft sampling of trace gases and aerosols to be conducted in April-May, 2001 will significantly enhance the understanding of the points mentioned above.

BIBLE-A, BIBLE-B: ITCZ

Latitudinal variations in the mixing ratios of ozone and precursor gases above 8 km were obtained during the transit flights from Japan to Australia along the $140 \pm 5^\circ E$ meridian in September 1998, as shown in Figure 2. Both O_3 and NO_y showed abrupt decreases at 23°N, corresponding to the changes from midlatitude to subtropical air masses, according to trajectory analysis. The O_3 and NO_y reached minimum values of 20 ppbv and 30 pptv, respectively, at 0-7°N. Tropical Rainfall Measuring Mission (TRMM) and meteorological data indicate that the ITCZ was located in this region. Large-scale convection over the clean tropical ocean transported boundary layer air poor in O_3 and NO_y to the upper troposphere. Because the dominant component of NO_y in the maritime lower troposphere should be HNO_3 [Kondo *et al.*, 1997],

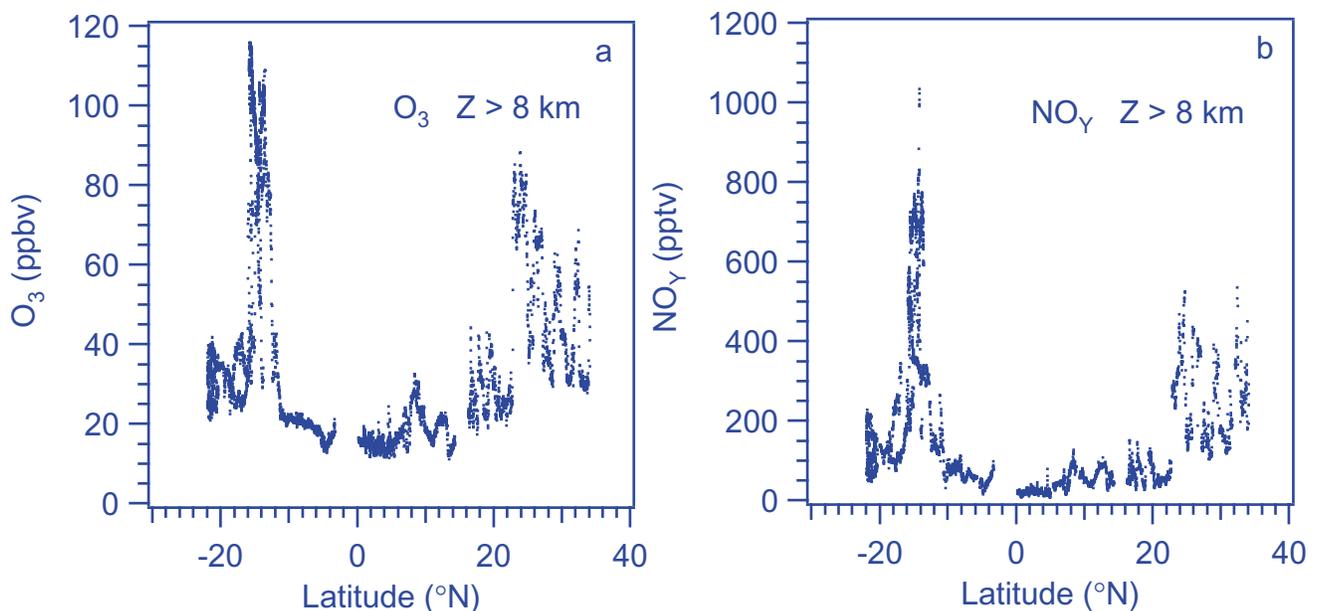


Figure 2: Latitudinal distribution of O_3 (a) and NO_y (b) obtained during transit from Japan to Darwin in September 1998 during BIBLE-A.

heterogeneous removal of HNO_3 during upward transport might have further decreased NO_y . Similarly, low NO_y and O_3 values were observed near the ITCZ and SPCZ in February during PEM-West B [Kawakami *et al.*, 1997]. The data from the other transit flights from Indonesia to Japan basically showed similar features, although the boundary between different air masses shifted somewhat.

Similar transit flights were made in August-September 1999 during BIBLE-B. Distributions of trace gases in the mid-latitude and subtropical regions above 8 km were quite different during this period as compared with 1998 mainly due to the differences in meteorological conditions. In addition, lightning activity over the western Pacific strongly influenced the level of reactive nitrogen in subtropical air masses. These results indicate significant variability in trace gases and aerosols in the upper troposphere over the western Pacific, even during the same season. The data obtained during BIBLE will improve our understanding of the transport of Asian continental air to the western and central Pacific Ocean.

Biomass burning in Indonesia and Africa/South America

Biomass burning activities over Indonesia were lower than average in October 1998. In 1997, biomass burning activity was quite high [Fujiwara *et al.*, 1999; Kita *et al.*, 2000]. This was because the regrowth of burned biomass was comparatively slow in 1998, and therefore the amount of fuel in 1998 was much smaller than that of 1997. In addition, La Niña prevailed during this period causing higher convective activity leading to higher humidity and a higher precipitation rate. Although there was some influence of biomass burning or urban pollution on CO, NMHCs, and reactive nitrogen, the enhancements in 1998 were much lower compared with those in 1997. Lightning activity seems to have elevated concentrations of reactive nitrogen in the upper troposphere. However, the production of reactive nitrogen by lightning was sometimes mixed with the effects of biomass burning or urban pollution, judging from the CO and NMHC data. Enhanced reactive nitrogen either from lightning or biomass burning did not lead to significant increases in ozone in the vicinity of Indonesian islands because of the limited time for photochemical ozone production. By contrast, trajectory analyses showed that in the absence of intense convection, ozone was produced photochemically at a rate of about 2 ppbv/day in air masses with moderately high ozone precursors during the transport from the Indonesian region to the Indian Ocean. Considering that BIBLE-A was conducted in a year of very low biomass burning, the obtained data will provide a good baseline for assessing the impact of biomass burning in future studies.

In the upper troposphere in the subtropical region over northern Australia, O_3 and NO_y mixing ratios were en-

hanced, reaching as high as 100 ppbv and 1000 pptv, respectively (Figure 2). Trajectories show that these air masses were from Africa/South America or Indonesia. During PEM-Tropics, enhanced values of O_3 , HNO_3 , CO, NMHC were observed mostly between 3 and 7 km over the subtropical central Pacific Ocean in September [e.g., Gregory *et al.*, 1999]. The present observations indicate that the biomass burning plumes over Africa/South America can be transported to above 8 km.

Further information, including highlights of the BIBLE campaigns, can be obtained at <http://www.eorc.nasda.go.jp/AtmChem/GLACE/bible/BIBLE.html>.

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Cape D'Aguilar (Hok Tsui) atmospheric research station in Hong Kong

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In 1993 the Hong Kong Polytechnic University (HKPU) established an atmospheric research station at the south-eastern tip of Hong Kong in coastal South China (Figure 1). The primary objectives for the studies there are to monitor the composition of the atmosphere, document important trends, and study the processes affecting the fate and transport of air pollution in eastern Asia. Situated on the boundary between the Asian continent and the South China Sea, the Cape D'Aguilar (Hok Tsui) station receives continental outflow in winter and maritime inflow in summer, making it suitable for studying both natural and anthropogenic trace gases and aerosols. Through long-term observation it is possible to study the impact of the increasing air pollution, associated with the rapid Asian economic growth, on the chemistry and climate in eastern Asia, particularly in the subtropical part of eastern Asia.

Measurement activities

Since the establishment of the station, a suite of chemically active trace gases and aerosols have been measured, including O_3 (surface and total column), CO, NO, NO_x , NO_y , SO_2 , C_2 - C_6 non-methane hydrocarbons (NMHC), mass and chemical composition of total and suspended particulate (TSP and RSP), and absorbing and scattering coefficients of aerosols. In addition to the efforts carried out at the research station, HKPU scientists have been collaborating with the Hong Kong Observatory to study ozone vertical structure by launching ozonesondes. The station has also accommodated visits of local and overseas scientists for study of acid rain and long-term aerosol trends.

International collaboration

The Cape D'Aguilar research station has been actively involved in several major international atmospheric chemistry studies. Shortly after its establishment, the station took part in the IGAC/NASA PEM-West B study in spring 1994. It has been a part of the IGAC/APARE surface monitoring network, and a regional station in the WMO-GAW network. The station is also involved in the China-MAP project, serving as the southernmost site in the air chemistry network in eastern China; the station's scientists also have been measuring trace gases in the Yangtze Delta region of China since June 1999. The research station will



Figure 1. Location of Hong Kong in eastern Asia.

play an active role in the upcoming IGAC/NASA TRACE-P and ACE-Asia studies in spring 2001.

Atmospheric research and some research highlights

Trace gases and gas phase chemistry studies

Measurement of key trace gases has been a major focus of the research program at the station. This is partly because of the very limited investigations that have previously been conducted in Asia on reactive trace gases that control the atmospheric oxidizing capacity. Among the gases under study, ozone and CO have been continuously measured since 1994. Their seasonal trends have been established, showing minima in summer and maxima in winter, or in the case of ozone in late-autumn [Lam *et al.*, 1998] as indicated in Figure 2a for 1994-1996. Typical summer-time levels of 10-20 ppbv for O_3 and 90-100 ppbv for CO have been observed (Figure 2), and these levels are considered to represent the background levels in the South China Sea air. A similar summer-low and winter-high pattern has been indicated by C_2 - C_6 NMHC measurements using an on-site gas sampling and GC/FID system from October 1995 to March 1997 [Cheung, 1999] and from studies of the chemical composition of aerosols [Cheng *et al.*, 2000]. The Asian monsoonal circulation has been considered to the primary cause of such a seasonal pattern. Monsoon winds transport clean, humid, maritime air to the site in summer and pollutant-laden, dry air masses from the Asian continent in winter. Studies of the summer-time background levels of oxides of nitrogen and sulfur dioxide have been complicated by suspected emissions from ships brought to the site by southwesterly winds.

Reactive nitrogen compounds, NO_x and NO_y , have been measured, with the application of research-grade instrumentation, to study ozone chemistry and transport processes. During the PEM-West B mission in February-

March 1994, NO_y and several other gases including O_3 , CO , SO_2 , and NO were measured to characterize the air masses under the condition of strong continental outflow [Wang *et al.*, 1997]. Later in 1997 and 1999, NO and NO_2 were measured using a set of high-sensitivity and high-selectivity instruments (modified EcoPhysics CLD 770 AL ppt chemiluminescent detector and PLC 760 photolytic converter) during the photochemically active summer and autumn seasons. NO_y has been measured since September 1999 using catalytic conversion and chemiluminescent detection techniques (TEI 42C-Y Trace Level).

The extensive trace gas (and aerosol) measurements from the HK station have been compared with data collected in limited locations of East Asia as well as other parts of the globe to help characterize the chemical environment of the study site. Autumn is a season of particular interest because the research station is subjected to minimal influence of local emissions but strongly impacted by air masses from the western Pacific, coastal regions of

mainland China, and the island of Taiwan. After applying a wind filter to remove the impact of urban plumes from Hong Kong, the levels of the trace gases in that season have been found to be comparable to those of a "polluted" rural atmosphere in the industrialized eastern US or Europe. Figure 3 shows diurnal variations of median concentrations of NO_x measured in October and November 1997 for major types of air masses determined by backtrajectory analysis. Model calculations using observed chemical precursors and meteorological parameters show strong *in situ* ozone production and abundant radicals in the air masses sampled. The results also suggest that the period from late autumn to early winter is a strategic time for studies of photochemical processes that affect the removal of pollutants emitted in northeast Asia and, further, that the subtropical South China Sea is a suitable place for such studies. Detailed results of ozone chemistry will be included in two forthcoming manuscripts [Wang *et al.*, manuscripts in preparation, 2000].

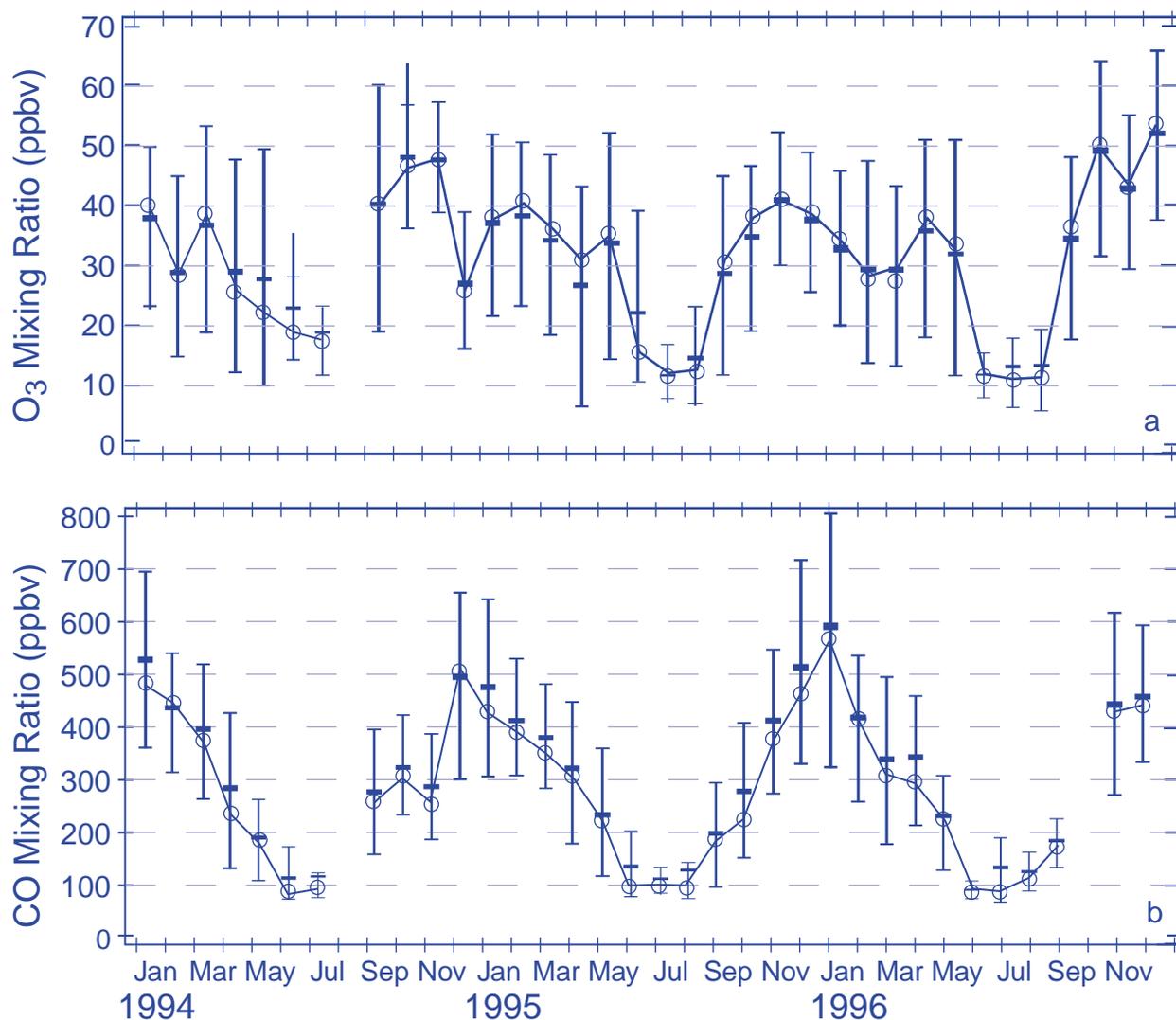


Figure 2. Monthly median (circle), mean (thick bar), 33 and 67 percentiles (whisker) of surface ozone (a) and CO (b) for 1994-1996.

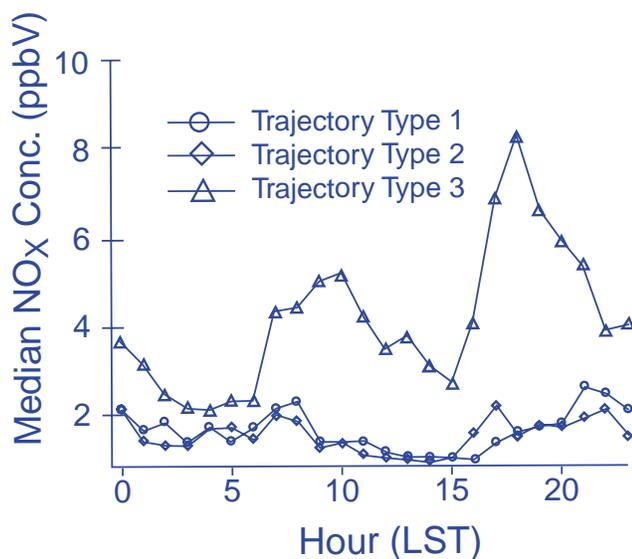


Figure 3. Diurnal variation of median NO_x concentrations for several types of air mass observed in October and November 1997. Type (1): marine western Pacific, (2): weak continental outflow, and (3): strong continental outflow. The large-scale air masses are classified using 10-day back trajectories calculated by Dr. Joyce Harris of the NOAA/CMDL.

Aerosol chemical properties

From May 1993 to March 1996, TSP and RSP aerosol particles were collected with the aim of contributing to the overall understanding of the aerosol levels and their origins in an upwind location of Hong Kong. A high volume air sampling protocol using Whatman EPM-2000 glass microfiber filters and Whatman 41 cellulose filters was used to collect the aerosol for analysis. Inorganic compounds including major ions and various trace elements were measured by ion chromatography, neutron activation analysis, and X-ray fluorescence spectrometry. Detailed results have been reported in *Lam et al., 1997* and *Cheng et al., 2000*.

The mean mass concentrations of TSP and RSP in the Cape D'Aguiar coastal area are 69 and 39 $\mu\text{g m}^{-3}$ respectively. The average ratios of Cl^-/Na^+ in both TSP and RSP slightly deviate from the corresponding ratios in sea water. The annual mean sulfate concentration in TSP was 14 $\mu\text{g m}^{-3}$. The $\text{SO}_4^{2-}/\text{NO}_3^-$ ratios are 3.28 for TSP and 4.17 for RSP. The non-sea salt sulfate in RSP accounts for 93% of the total sulfate indicating a substantial anthropogenic origin. Trace elements such as Se, Zn, Pb and As are highly enriched over the concentrations expected from the observed concentrations of crustal material or atmospheric sea salt.

Aerosol optical and sizing measurements

Aerosol optical properties have been monitored from September 1997 to April 1999. The light scattering coefficients have been measured using a M903 integrating

nephelometer (Radiance Research, USA) and the light absorption coefficients have been measured using a particle soot absorption photometer (PSAP, Radiance Research). Particle size distributions have been measured using a LASAIR 510 (Particle Measuring Systems, Inc. USA) and an eight-stage Andersen non-viable ambient sizing sampler. The single-scatter albedo, ω , was calculated from the measurements and compared with the critical value, ω_{crit} , estimated for the region. During summer, ω was found to be less than ω_{crit} , indicating that aerosol population may exert a warming effect in summer. On the other hand, ω was found to be larger than ω_{crit} in winter, suggesting a possible cooling effect of aerosols in winter. During the winter, the number concentration of aerosol fine particles with diameters less than 2.5 μm was higher than that of coarse particles by a factor of 10⁴. The variations of the scattering coefficients have been matched with the concurrent variations of particle sizes and it was found that the correlation was best for particle size range of 0.5 to 1.0 μm . Detailed results are being included in a manuscript [*Man et al., manuscript in preparation, 2000*].

Ozone vertical distributions

Ozonesondes have been launched in Hong Kong in collaboration with the Hong Kong Observatory. Data analyses of the ozone profiles have been performed, with a focus on the seasonal cycle in tropospheric ozone [*Chan et al., 1998*]. Results show that the tropospheric ozone column has an obvious maximum in spring and a minimum in summer. The former is a common feature at many locations in the Northern Hemisphere. Tropospheric ozone has an especially strong influence on the seasonal cycle of total ozone at Hong Kong. The seasonal cycle of ozone mixing ratio below 2 km is bimodal with ozone peaks in spring and autumn. A frequently observed feature in late autumn and winter is a relative minimum ozone mixing ratio (as low as 30-40 ppbv) in the upper troposphere (from about 9 to 16 km). Trajectory analysis shows this relative minimum in ozone mixing ratios is associated with air masses transported from the tropical region. It is proposed that the East Asia local Hadley circulation is responsible for this feature.

Also, high ozone (80-138 ppbv) events in the lower troposphere (2.5-4.5 km) over Hong Kong have been captured by ozonesonde launches during late winter and spring. Back trajectories showed air masses arriving at the altitude of the ozone peak had passed over continental Southeast Asia, which is where the bulk of biomass burning occurs at this time of the year. With the increased relative humidities accompanying these events, we suggest that the ozone-rich air in the lower troposphere in springtime is the result of photochemical production rather than the result of transport from the upper troposphere of higher latitudes [*Liu et al., 1999*]. Further analysis of ozonesonde data obtained during the NASA PEM-

SUPPLEMENT: Correction to previous issue

Editor's Note: Due to a copyediting error, some references for the methyl bromide features were omitted from the last issue of *IGACTivities* (January 2000, #19). A complete reference list follows.

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West B in February 1994 using the NOAA AVHRR satellite image data on global fire strongly suggest that biomass burning in SE Asian countries such as Vietnam and Thailand strongly influences the ozone climatology in Southeast Asia and Hong Kong [Chan *et al.*, 2000]. Also, strong ozone enhancements, with mixing ratios up to 100 ppb and an enhancement layer up to 10 km thick, have been observed over Hong Kong during the strong El Niño event in December 1997. Our analysis suggests that these conditions are related to the Indonesian fires in SE Asia at that time.

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PEACAMPOT and PEACAMPOT II campaigns

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The Pacific coast of East Asia, like Europe and North America, is a region in which human activities impose a heavy load on the atmosphere. Air pollutants released from this region are known to affect not only the Asian continent but also a large area of the Pacific. Moreover, emissions of SO₂ and NO_x, which in western Europe and North America have been decreasing or at least remaining steady since 1980, are still growing in Asia and will likely continue to do so in the 21st century. Although the Pacific coast of East Asia has a significant influence on regional and global environments, the atmospheric chemistry of this area has not been thoroughly or systematically monitored and thus is not fully understood. Scientific data on the pollutants transported from Asia to Japan and the Pacific Ocean are very important in analyzing the present status of the atmosphere of this area and in developing international cooperation on air pollution controls.

Experimental methods

The atmosphere was sampled from an aircraft (either a Cessna 404 or a Fairchild Swearingen SA-226 [Merlin-IV]). Ambient air was introduced into analyzers from a glass manifold connected to a 9.5-mm Teflon tube that

led from an inlet set either on the nose of the Cessna or on a window forward of the engine air intake of the Merlin. Aerosols were collected on Fluoropore filters with a high-volume sampler (Kimoto) through a 25-mm stainless steel tube set either on the copilot's window on the Cessna 404 or on the window forward of the engine air intake of the Merlin.

Sulfur dioxide was measured on board with a TECO Model 43S pulse fluorescence SO₂ monitor. A nitrogen oxide analyzer based on the NO₂ chemiluminescence method, which itself is based on the reaction of NO + O₃ → NO₂^{*}, was modified for use in aircraft observations. The main modifications were the introduction of the sample air intake method (800 SCCM) using a mass flow meter, a method to generate ozone for reaction from pure oxygen, and reduction of the absolute pressure of the reactor (from 5.7 to 5.2 kPa) to improve the efficiency of the chemiluminescence. A metallic molybdenum reducing agent (with a reaction temperature of 320°C) supported on a carrier was used to convert NO_x to NO. In the experiment in February 1999, the position of the converter was changed to make it possible to measure NO_γ in place of NO_x. The resulting data were stored in a TEAC DR-F3 digital data recorder at 10 s intervals. PAN in air was collected by a U-shaped trap packed with 0.2 g Teflon beads and chilled with granulated dry ice. Each aerosol sample filter was divided into 2 equal parts in our laboratory, and each part underwent ultrasonic extraction in 10 mL of distilled water. Cations and anions in the extracted solutions were analyzed by ion chromatography. The ambient temperature and relative hu-

midity were measured with a Väisala HMP 133Y thermometer-hygrometer.

PEACAMPOT campaign

The National Institute for Environmental Studies (NIES) created the “Perturbation by the East Asian Continental Air Mass to the Pacific Oceanic Troposphere” (PEACAMPOT) program as part of its “Study on Transport and Changes in Qualities of Acidic and Oxidizing Substances in East Asia” project, which is funded by the Japan Environment Agency, for promoting comprehensive studies of the global environment. NIES has been chemically monitoring the atmosphere over East Asia in cooperation with other institutes under international programs.

Aircraft and intensive ground-based observations were started in 1991 as a five-year project under an initiative taken by NIES. Figure 1 shows the area covered by PEACAMPOT.

In 1991, observations were made as part of an international research project based on the IGAC/APARE program, in cooperation with the PEM-West program of NASA (USA). In 1992, we conducted aerial observations over the Sea of Japan off the San'in Region of Japan and over the East China Sea off the southern coast of Cheju Island, Korea. These were made at four different altitudes to measure the vertical distribution of pollutants more thoroughly than in the previous year. At the same time, we made intensive ground-based observations in the Oki Islands and in the Happo-one Mountains, Japan.

In 1993, we made our observations in concert with Phase B of PEM-West. Aerial observations were made in early

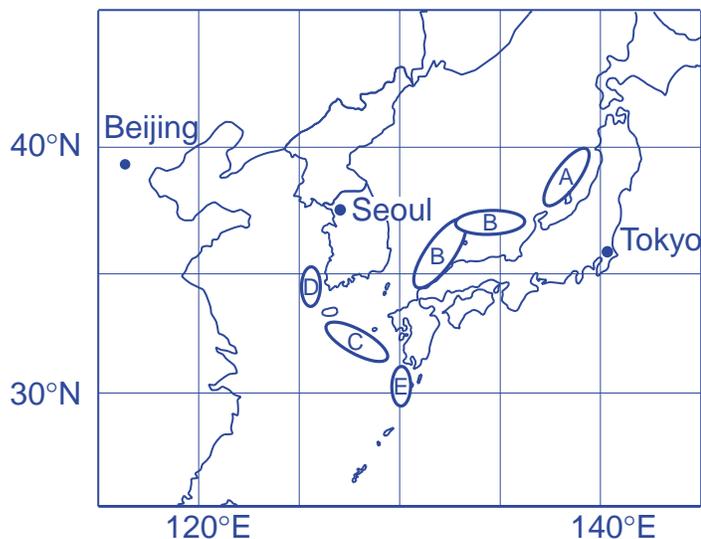


Figure 1. Observation area for the PEACAMPOT campaign. A: North of the Sea of Japan, B: southwest of the Sea of Japan, C: East China Sea, D: Yellow Sea, E: west of Yaku Island.

spring. Intensive ground-based observations were conducted in the Oki Islands and in the Happo-one Mountains. At the same time, the Korean Institute of Science and Technology (KIST) conducted intensive ground-based observations on Cheju Island and gave us their data.

In 1994, intensive aerial and ground-based observations were conducted in and over Yaku Island, Japan, with the same objective as before. Yaku Island is an area where nature has been preserved, hence Yaku Island became a World Heritage Area. Nevertheless, it is being jeopardized by environmental degradation, especially the decline of forests. Thus, it was a matter of great interest to learn how the atmospheric pollutants transported from Asia affect the natural environment of the island.

In 1995, the final year of the project, observations focused mainly on the influence of slightly polluted areas in north-eastern Asia. We made both ground-based and aerial observations in the northern Sea of Japan. The main objective was to see how air masses are transported not from areas with large-scale pollution sources, such as China and Korea, but from areas with few pollution sources, such as eastern Russia.

All data have been compiled and published as data books [Hatakeyama, 1993, 1994, 1995, 1996a, 1996b] and data CDs [Hatakeyama, 1997, 1998]. We drew four main conclusions:

1. The air over the seas between Japan and the Asian continent is strongly affected by anthropogenic emissions, given the vertical distribution of PAN (Figure 2). The vertical gradient in pollutant concentrations was reported to be gentle over a polluted continent [Meyrahn *et al.*, 1984; central Europe] but steep over clean oceans [Singh and Salas, 1983]. The slope in Figure 2 is similar to that reported for the polluted continent.
2. A high concentration of SO₂ was transported over the western part of the Sea of Japan via the Korean peninsula. This distance is very short.
3. The concentration of sulfate was higher over the East China Sea, where air pollutants spent a relatively long time and were oxidized well.
4. The air mass transported from northern Eurasia was less polluted than that from other areas mentioned above. Findings 2-4 are already reported [Hatakeyama *et al.*, 1995].

PEACAMPOT II campaign

From 1996 to 1998, the second phase of PEACAMPOT (PEACAMPOT II) was conducted as part of a project of the Global Environment Research Fund of the Environment Agency of Japan called “Studies on the Development of a Comprehensive Model of Atmosphere and Soil and an International Cooperative Field Survey to Clarify the Budget of Environment-Acidifying Substances in East

Asia." The target area was confined to the northern East China Sea to determine the transport of air pollutants from central China. Samples were taken in almost the same area as before (Figure 3). Ground-based observations were made on Fukue Island.

On 2 February 1999, we found very high concentrations of SO_2 over the observation area (Figure 4). SO_2 was high not only at low altitude (~300 m) but also at high altitude (~2300 m). NO_x , aldehydes, gaseous HCl, and gaseous HNO_3 showed very similar variations. Sulfate and ammonium showed similar variations (data not shown). Thus, it can be said that the air sampled on that day was highly polluted and well mixed. The vertical distribution of the PAN/ NO_y ratio also clearly showed the mixing of air (Figure 5). On 4 and 6 February, the ratio showed a remarkable difference between upper and lower altitudes. In contrast, it showed little dependence on altitude on 2 February, which indicates that the air was thoroughly mixed within the lower troposphere.

A low pressure system originating near Taiwan passed over this area very quickly on 1 February, and the weather was very windy on 2 February. The system moved along the southern coast of Japan to reach the northwest Pacific; it would have caused mixing. Uno *et al.* [1998] pointed out that just such a weather pattern should cause a large-scale, long-range transport of polluted air. The results of 2 February support this contention very well. Thus, we found one more path for the

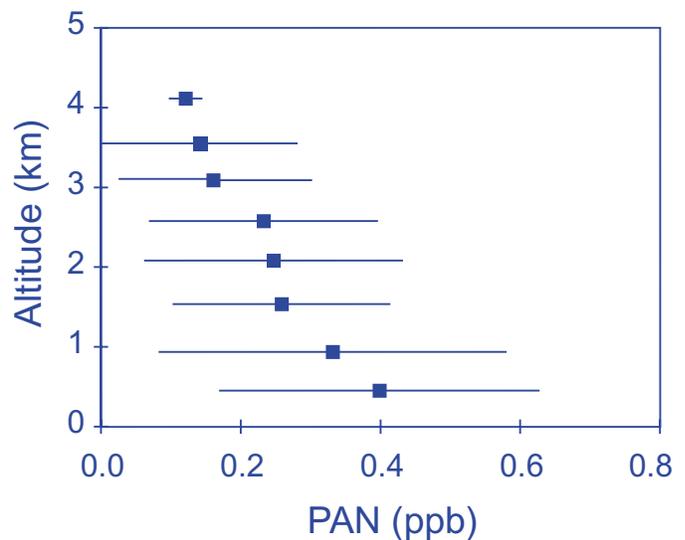


Figure 2. Vertical distribution of PAN over the seas between Japan and the Asian continent. Horizontal bars show 1 standard deviation. Adapted from data in Watanabe *et al.* [1998] and Hatakeyama [1998].

long-range transport of an air mass from Asia in addition to the three transport patterns we found in PEACAMPOT [Hatakeyama *et al.*, 1997] (findings 2-4 of the previous section). This other path is from central China (around Shanghai) via the northern East China Sea to Japan.

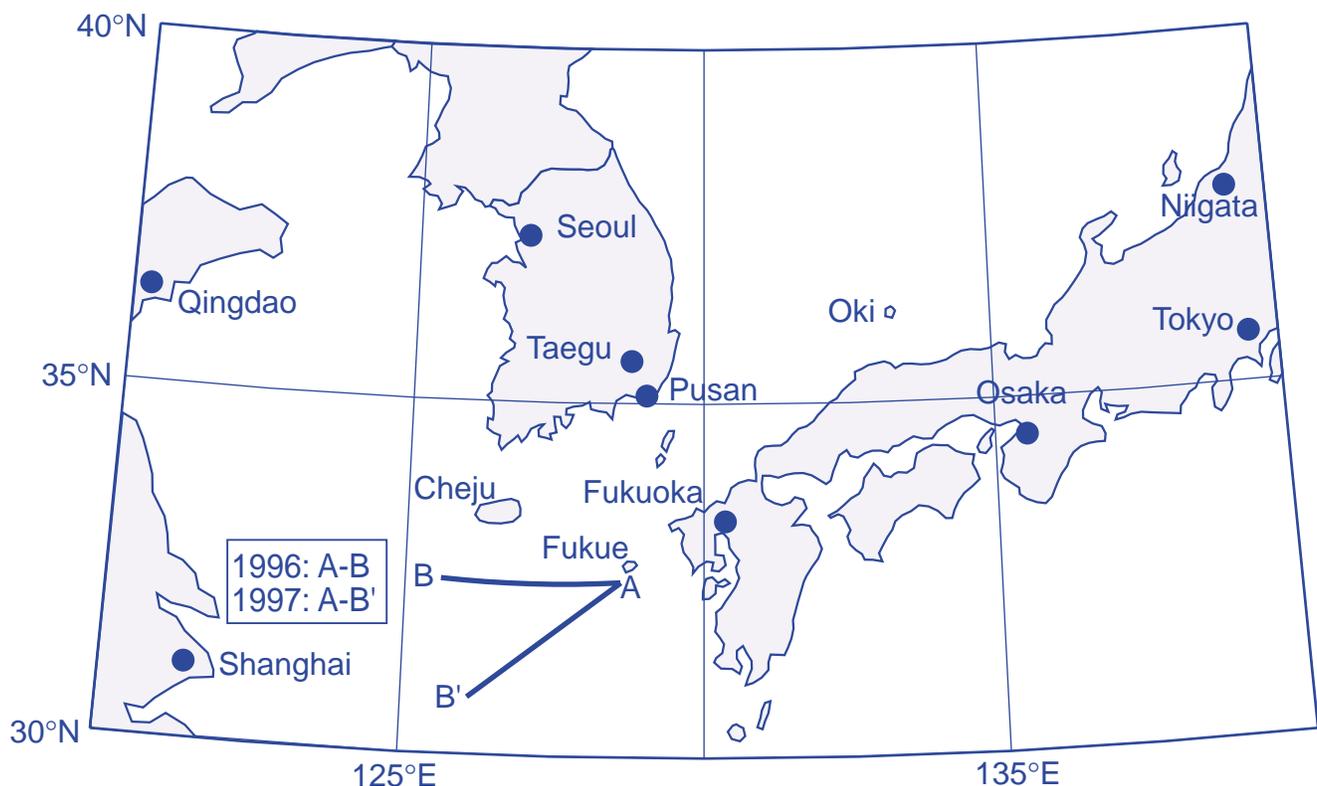


Figure 3. Flight plans for PEACAMPOT II.

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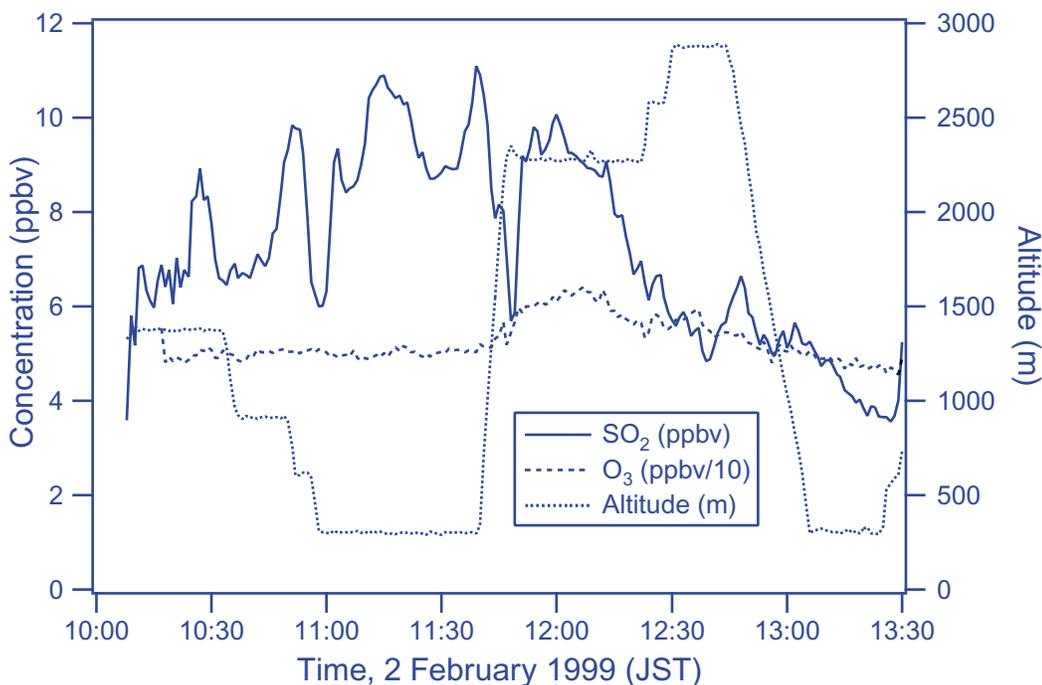


Figure 4. Altitude of measurements and concentrations of SO₂ and ozone measured on 2 February 1999.

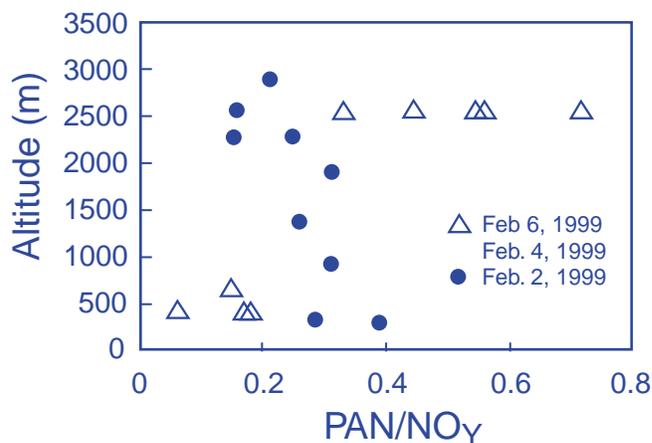


Figure 5. Vertical distribution of PAN/NO_y ratio.

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7th IGAC Scientific Conference

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Introduction

The International Global Atmospheric Chemistry (IGAC) Project is a Core Project of the International Geosphere-Biosphere Programme (IGBP) in cooperation with the Commission on Atmospheric Chemistry and Global Pollution (CACGP) of the International Association of Meteorology and Atmospheric Science (IAMAS). Since 1993, IGAC scientific conferences have been convened to provide an international forum for exchange of data and presentation of research results. With increasing amounts of data becoming available, the atmospheric chemistry community is devoting significant effort to an in-depth analysis of recent scientific findings and integration of diverse information into the broad context of Earth science.

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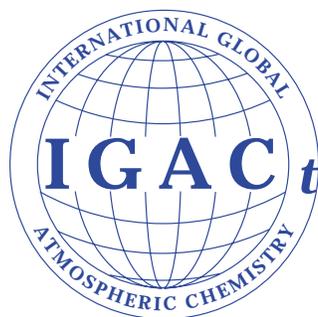
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