

Ozone over the Oceans – TOAR-II Focus Working Group

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Scope within TOAR-II

Oceans cover approximately 70% of the Earth's surface. Much of the world population lives in coastal areas, and air quality in these regions is impacted by air masses transported across the open ocean. Despite its importance, there are large gaps in our understanding of the distribution, trends and processes involving tropospheric ozone over the open ocean. This is in part due to the logistical challenge of taking measurements, especially long-term measurements, in such an environment; however, several projects have been focused over the years on the marine boundary layer (MBL) using mobile platforms (e.g. ships, aircrafts) and coastal observatories [e.g., Sommariva & von Glasow, 2012]. These datasets can be used to improve our understanding of ozone chemistry in these remote, but important parts of the planet.

Ozone formation and loss over the open ocean is inextricably linked to the nitrogen cycle and halogen chemistry, and both have considerable uncertainties particularly in terms of their sources and sinks [Monks et al., 2015]. Emissions of NO_x in the MBL (e.g. from ships) and long-range transport of nitrogen reservoir species (e.g. organic nitrates) are poorly characterized, while recent studies have suggested that photolysis of particulate nitrate may be a source of NO_x in remote environments [Reed et al., 2017].

In the troposphere, halogens are known to be both sinks (via direct reactions of Cl, Br and I with O_3) and sources (via oxidation of VOCs by Cl and Br) of ozone [Dix et al., 2013; Simpson et al., 2015]. In addition, recent work has shown that ozone deposition to the surface can be a source of active iodine in the marine boundary layer [Carpenter et al., 2013]. Several global models – such as CAM-Chem, TOMCAT, GEOS-Chem, CHASER – include these processes with varying degrees of completeness [Hossaini et al., 2015; Sherwen et al., 2016; Iglesias-Suarez et al., 2020; Sekiya et al., 2020]. However, hardly any of the recent chemistry-climate Climate Model Intercomparison Project (CMIP-6) model runs, designed to inform the IPCC assessments, include halogen chemistry at all, when calculating ozone's radiative effect.

Omissions of important chemical processes can lead to significant errors and/or high uncertainties in the results, which inevitably affect the outcomes and the recommendations of the climate assessments and/or air quality control policies that rely on these models for input. The overall aim of the working group (WG) is to evaluate the current knowledge of ozone chemistry over the oceanic regions, and to improve the representation of this important part of the planet in global models.

Objectives of the Working Group

The main objective of the WG is to assess the tropospheric ozone budget over the oceans, looking in particular at each process that affect its concentration (from chemical production and loss, to stratosphere/troposphere exchange to deposition) and how this has changed over the recent years. A major area of interest is the impact of halogen chemistry on the tropospheric O_3 budget, in order to provide up-to-date information on this subject to the TOAR-II assessment report, as well as to the upcoming IPCC AR7. Other important aspects to be considered by the WG are the emissions of NO_x and the impact on ozone concentrations of long range transport of NO_y species in the remote marine environment (e.g., from biomass burning sources [Verreyken et al., 2020]).

The focus of the WG is on the oceanic regions and on the lower troposphere (marine boundary layer), a largely undersampled part of the planet. Polar regions are included, but only to the extent of the oceanic environment. Coastal areas are also included, although the main focus is on the open ocean and filtering may be necessary to exclude the effect of local and/or continental emissions. Likewise, the free troposphere is included but only to the extent of its impact on the MBL.

As TOAR-II is an “observation-based” assessment, this work should include comparisons/evaluation of O_3 and NO_x data which are already present in the TOAR database, as well as additional O_3 datasets collected over the oceans, together with the associated halogen/ NO_x / NO_y observations, as available. Model simulations are key to synthesize the outcome of the assessment and inform its integration in the overall TOAR and IPCC assessments. Hence, global models will be used for comparisons of ozone fields, trends and budget (e.g., CTM/reanalysis with and without halogens), NO_y distribution and speciation, and comparison with the observations at global scale.

Some questions that should be considered by the WG:

1. What are the ozone distribution and trends over the open ocean?
2. How do halogen species (Cl, Br, I) influence the global distribution of ozone over the oceans? Some of these species act as net sources and some act as net sinks. Deposition of ozone has also been shown to be a potential source of iodine in the MBL with significant impacts on the overall ozone budget [Carpenter et al., 2013], so the available database of iodide in seawater should be considered for inclusion in the models [Chance et al., 2014, 2019].
3. Can the models explain the distribution of NO_x and NO_y in remote regions? How does this affect the ozone budget?
4. Ship plumes are sources of NO_x and potentially of inorganic halogens: how do they impact ozone in the remote atmosphere?

Data availability

TOAR is committed to open data practices and all working groups follow the COPDESS (Coalition on Publishing Data in the Earth and Space Sciences) principles: <http://www.copdess.org/statement-of-commitment/>.

Several datasets of O₃ and NO_x relevant to this WG are already available in the TOAR database. Most of these data are collected at coastal sites (e.g. Cape Grim, Mace Head), and islands (e.g. Cape Verde, Okinawa) and are somewhat limited in what they can tell us about what happens on the open ocean. Influences by local sources and/or continental air masses will need to be accounted for: this may require information and data on local meteorology.

Measurements from a number of ship- and aircraft-based intensive field campaigns can fill in the gaps and provide a better understanding of the conditions far from the coasts [e.g., Lelieveld et al., 2004; Watanabe et al., 2005; Volkamer et al., 2015; Kanaya et al., 2019]. Ozone and NO_x are almost always available, and in some of these field campaigns, halogen species and NO_y species have also been measured [e.g., Lee et al., 2010]. Examples of available datasets that can be contributed to the TOAR database as part of this work:

- R/V Mirai (Japan) and R/V Polarstern (Germany) research cruises (mostly O₃, may include NO_x and IO/BrO).
- NOAA-PMEL (USA) and NERC (UK) research cruises (RHAMBLE, ACE, ICELOT, VOCALS field campaigns).
- Flights of US and British research aircrafts (e.g., TORERO, CONTRAST, CAST, ACSIS field campaigns) and ozonesondes data (e.g., SHADOZ).
- “Opportunistic” observations on commercial ships and aircrafts (e.g., IAGOS)

The datasets shall be harmonized and made available through a link from TOAR portal or added to the TOAR database itself (after the database will be able to accept moving coordinates).

Expected outcomes

The main outcome of the WG work will be a paper to be published in the TOAR-II Community Special Issue, which will provide new information for the overall TOAR-II assessment report. The paper will build upon the recent reviews on tropospheric ozone and halogens [Monks et al., 2015; Simpson et al., 2015] to include

the most up-to-date science, as well as novel analysis of ozone trends and budgets derived from the additional data collected by the WG, as explained above.

Another key objective is to feed the results of this work to the Climate Model Intercomparison Project (CMIP), in order to provide an assessment on the role of halogen chemistry on tropospheric ozone for its consideration in the CMIP7 model runs that will inform the upcoming IPCC AR7.

Additional data from ship-based and aircraft-based campaigns (including NO_x, NO_y and halogen species) will be collected and harmonized for future inclusion into the TOAR database.

Collaboration and sharing of information/data with other related TOAR-II Focus WGs will be pursued, as appropriate. Potential overlap can be found, for example, with the Oceania, with Tropospheric Ozone Precursors (TOP), and with the Ozone in the Tropics (OPT) focus working groups.

Proposed timeline

Early January 2021: complete WG proposal, start recruiting core members.

January 2021: finalize WG proposal and core membership before TOAR-II workshop (25 Jan-3 Feb).

February-June 2021: data collection, goal setting, action items listed, start analysis.

December 2021: data addition/examination, discussion of initial results from analysis.

January-June 2022: data submission & discussion of further results from observation-model comparisons.

December 2022: summarize analysis, discussion mainly on processes, define structure of the paper.

January 2023: Start writing draft of paper. Aiming for submission to the TOAR-II Community Special Issue by September 2023.

Statement

The WG members are aware that WG participation is voluntary and that no funding is provided by TOAR-II.

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