

solas event report

Report 07 | November 2017

Session on:

“Good Hope for Earth Science: Atmospheric Chemistry and Physics for the 21st Century

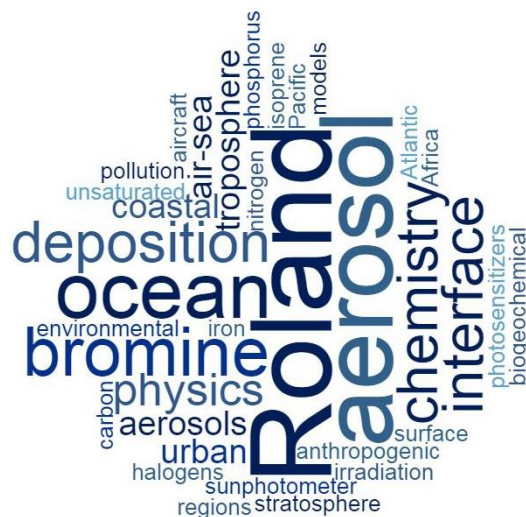
SOLAS sub-session: A tribute to Prof. Roland von Glasow”

**28 August - 1 September, 2017
Cape Town, South Africa**

From 28 August until 1 September 2017, the session M01 “Atmospheric Chemistry and Physics for the 21st Century” has been organised under the auspices of the International Commission on Atmospheric Chemistry and Global Pollution (iCACGP) at the International Convention Center in Cape Town, South Africa.

The sessions on August 28 and 29 were dedicated to SOLAS as a tribute to Prof. Roland von Glasow, who served on the Scientific Steering Committee of SOLAS (2008-2013), and was an active Commission Member of iCACGP when he suddenly passed away in 2015. Roland’s leadership and increasingly important contributions to the SOLAS community were honoured during the presentation of the two invited speakers Prof. Rainer Volkamer and Prof. Maria Kanakidou, who outlined the scientific path of Roland and presented recent results as a continuation of research topics initiated by him.

Roland studied Physics at the University of Mainz (Diplom in 1997), followed by a PhD in Atmospheric Physics at the Max Planck Institute for Chemistry (2001) working on tropospheric



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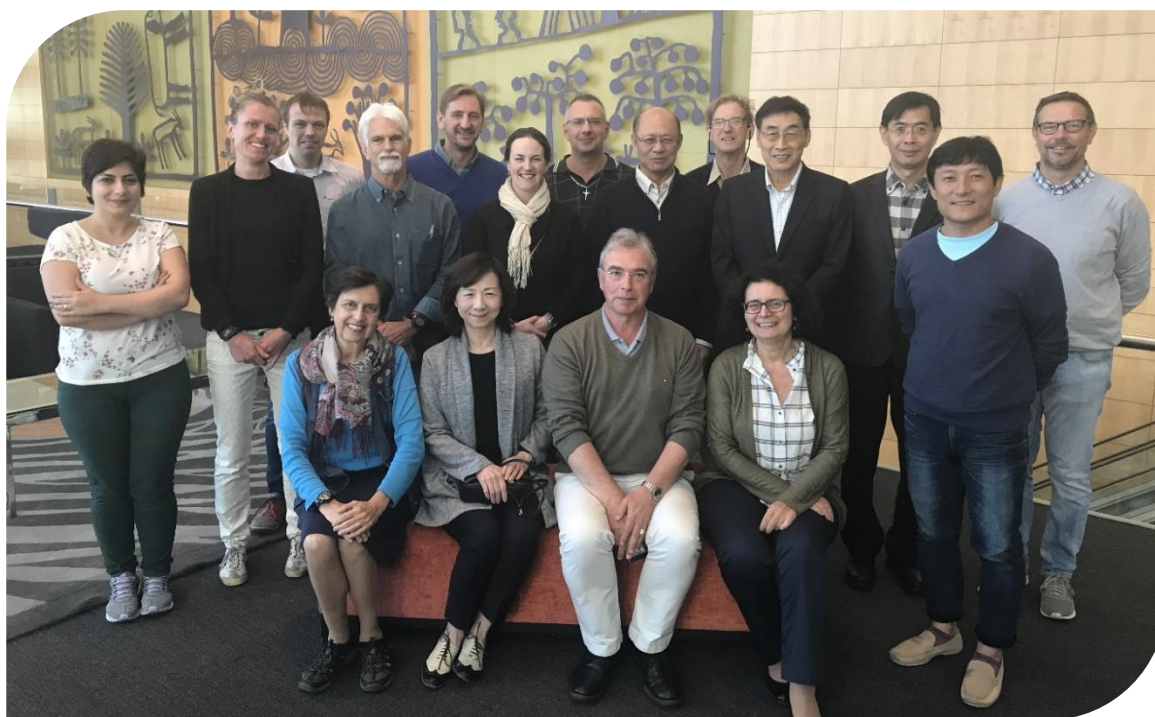


Figure 1: The iCACGP members and delegates, who organised the SOLAS sub-session. © Maria Kanakidou
Front row (sitting) from left to right: Anne Thompson, Sachiko Hayashida, John P. Burrows, Maria Kanakidou
Back row (standing) from left to right: Soheila Jafariserajehlou, Fiona Tummon, Sven Krautwurst, Russell R. Dickerson, Rainer Volkamer, Robyn Schofield, Stuart Piketh, Tong Zhu, James R. Drummond, Shaw Chen Liu, Charles C.-K. Chou, Hiroshi Tanimoto, Christian George.

halogens, a topic that he has developed and extended throughout his scientific career as is exemplified by his role as co-chair of the SOLAS/ International Global Atmospheric Chemistry (IGAC) task “Halogens in the Troposphere” (since 2005). After a Postdoc at Scripps Institution of Oceanography, San Diego, USA (2001-2003), he returned to Germany to head a Junior Research Group (Emmy Noether) at the Institute for Environmental Physics, University of Heidelberg, Germany (2004-2007). Roland joined the University of East Anglia (UEA), Norwich, UK, in 2007, where he was promoted to Professor in Atmospheric Sciences in 2012. At UEA he conceived and directed the Centre for Ocean and Atmospheric Sciences in 2013, unifying physical and biogeochemical sciences under one umbrella. Today Roland’s work continues at UEA within “The Roland von Glasow Air-Sea-Ice Chamber” built following his ideas. He was a member of numerous scientific committees (including SOLAS and iCACGP), generously giving his time and sharing his enthusiasm for research. From

2009-2012 he co-chaired the International Geosphere-Biosphere Program (IGBP) Fast Track Initiative on “Air-sea interactions in megacities and the coastal zone” to provide an integrated view of the human impacts on the Atmosphere-Land-Marine Ecosystem. Inspired by Roland, SOLAS researchers are organising a workshop on 26 October 2017 in Gothenburg, Sweden, to develop a research theme that assesses the effects of commercial shipping on the ocean-atmosphere system.

The SOLAS part of session M01 highlighted two aspects of Roland’s work on “Halogens in the Troposphere” and “Coastal Megacities”, and was attended by ~50 participants from five continents. On 28 August, the “Roland von Glasow Memorial Lecture: Tropospheric Halogen Sources from Sea Spray Aerosol” by Prof. Volkamer presented new field measurements that inform the bromine conundrum, i.e. the fact that atmospheric models often predict more bromine oxide (BrO) radicals compared to surface measurements over remote oceans. This phenomenon had been puzzling

Roland since the early days of his career. Roland had highlighted the important role of aerosol acidity, and its changes with altitude that accelerate the heterogeneous recycling of bromine in the free troposphere. New measurements of gas- and aerosol bromide probed for the first time the bromine hot-spots over the tropical Pacific Ocean as part of the Tropical Ocean Troposphere Exchange of Reactive Halogens and Oxygenated Volatile Organic Compounds (OVOC) project cruise, and identify a missing gas-phase process that converts BrOx (= Br + BrO) into inorganic bromine. The field measurements also identify unaccounted sources of marine OVOC, incl. glyoxal, aliphatic aldehydes and ketones, providing evidence that a more vigorous chemical coupling between the marine organic carbon cycle and tropospheric halogens might be responsible for the bromine conundrum. Sea salt aerosols are a source of bromine in the free troposphere, and possibly the upper troposphere and lower stratosphere (UTLS). The chemical detail identified by Roland's early work appears to be relevant to understanding the global distribution of halogens, yet it is missing from most atmospheric models today.

Following the first presentation, Prof. Monica Rhein spoke about "Ocean ventilation changes and impact on oxygen and anthropogenic carbon distributions in the North Atlantic". She discussed the changes in global decline in oxygen in the ocean of the recent past, and in particular in the tropics and North Pacific and Atlantic Ocean. These changes have been attributed to reduced ventilation. Prof. Rhein also presented results from the Labrador Sea on temperature and salinity changes that appear to be linked to global warming, and results on the use of Helium and Neon isotopes to calculate melt water fractions from the Greenland Ice Sheet into the Labrador Sea.

Dr. Christian George presented about "Photosensitized chemistry at the air-sea interface: Biology vs Chemistry", challenging the audience to re-think air-sea exchange processes. He stressed that the interface between the atmosphere and

the ocean is a good place for chemistry to take place, due to the enrichment of surfactants and photosensitizers that can harvest solar photons within the actinic flux region of the sun, and convert them into chemical energy that is available to trigger photochemical reactions. He presented results from laboratory experiments about the products of photosensitized chemistry of alcohols and fatty acids at the interface, which upon irradiation appear to dehydrate and form unsaturated products such as isoprene, and unsaturated OVOC. This chemistry presents an abiotic pathway to isoprene formation from organic matter that originates from cell lysis in the surface ocean, and accumulates at the air-sea interface. On 29 August, the "Human driven changes in the marine atmosphere and in the nutrient deposition to the global ocean. A tribute to Roland von Glasow" by Prof. Kanakidou first presented the wide range of chemical, physical and biological interactions between land-ocean and atmosphere at the coastal interface that is vulnerable to climate change as reviewed in the conceptual paper issued from the fast track initiative workshop on "Air-sea interactions in megacities and the coastal zone" led by Roland. Then focus was put on the impact of anthropogenic emissions that are intensively occurring over urban areas but having regional and global impacts due to atmospheric transport, and how these emissions impact atmospheric deposition of nutrients to the ocean. New global model results from the Pollution Alters Natural aerosol composition: Implications for Ocean Productivity, Climate and air quality (PANOPLY) project and the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection (GESAMP) Working Group 38 (on atmospheric input of chemicals to the ocean) have been presented for nitrogen, phosphorus and iron deposition. In particular, the impact of cloud and aerosol water acidity and multiphase chemistry on the production of organic ligands (represented in the model by oxalic acid) from soluble carbonyls, and on solubilising nutrients (namely iron and phosphorus) present in atmospheric aerosol, has been shown to be

significant. The global modelling studies evaluated the large contribution of the organics to the global atmospheric deposition of these nutrients, producing the first global deposition maps of organic nitrogen and organic phosphorus, in agreement with existing observations. They have revealed the potentially large contribution of bio-aerosols as a source of bioavailable nitrogen and phosphorous to the ocean, which under certain circumstance is of comparable importance with soluble dust inputs of phosphorous. This fraction has been neglected in most global modelling studies until now.

Following the presentation of Prof. Kanikadou, Dr. Joseph Adesina discussed the "Contribution of marine aerosols to the total columnar aerosol loading over Ascension Island". A cluster method was used to interpret sunphotometer measurements of wavelength dependent aerosol optical depth, single scattering albedo, and their seasonal variations to identify the types of aerosols (marine, dust, urban and biomass burning, and continental average) that influence the air column above five stations in Southern Africa and Ascension Island, on a seasonal basis. Significant differences of the aerosol sources were corroborated by back trajectory analysis that point to different source regions from the West African continent, that influence the air column above Ascension in different seasons.

Finally, Dr. Hans Schlager, presented on "Aircraft observations of sulfur dioxide (SO_2) transport to the UTLS from major source regions". The High Altitude and Long Range Research Aircraft (HALO) and Falcon aircrafts operated by Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, Germany, were used to measure SO_2 and aerosol size distributions with special attention to separating the nucleation mode sized particles (2-4 nm diameter) from Aitken mode

and larger diameters. SO_2 concentrations above 10 km typically ranged from 10-100 pptv, though concentrations up to 1 ppbv were observed. The back-trajectory analysis suggests an important role of the warm conveyor belt for lofting air masses containing SO_2 ; the subsequent oxidation to form sulfuric acid provides a possible mechanism for the initial cluster formation from air pollution. Dr. Schlager also discussed tracer release studies from urban areas as a technical approach to use aircraft observations to normalize dilution of air during transport downwind.

We thank the workshop sponsors: the International Association for the Physical Sciences of the Oceans (IAPSO), the International Association of Meteorology and Atmospheric Sciences (IAMAS), the International Association of Geomagnetism and Aeronomy, the International Union of Geodesy and Geophysics (IUGG), iCAGGP and SOLAS, whose support allowed us to have such a fruitful and warm session to recall Roland.

Organising Committee:

Volkamer Rainer (rainer.volkamer@colorado.edu), University of Colorado, Boulder, United States

Kanakidou Maria (mariak@uoc.gr), University of Crete, Heraklion, Greece

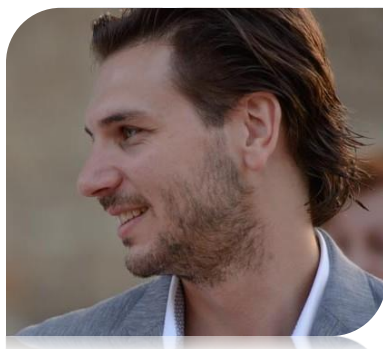
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The outcomes of the session contribute to the Cross-Cutting Theme 'Integrated Topics' (polar oceans and sea ice), and Core Theme 5 'Ocean biogeochemical control on atmospheric chemistry' of the SOLAS 2015-2025: Science Plan and Organisation.

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Stelios Myriokefalitakis holds a Ph.D in atmospheric chemistry from University of Crete, Greece. In 2014, he moved to Columbia University, New York, United States of America, to conduct research in coupled climate modelling as a Fulbright Visiting Scholar. From 2016, Stelios is a Marie-Curie Fellow at Utrecht University, Netherlands, investigating the effect of air-pollution on ocean biogeochemistry.

Modelling the impact of air pollution on iron deposition fluxes over the marine environment

Myriokefalitakis, S.

Institute for Marine and Atmospheric Research (IMAU), Utrecht University, Utrecht, Netherlands

s.myriok@uu.nl

Oceans modulate the carbon cycle by contributing to the removal of atmospheric carbon dioxide (CO_2) via physical, chemical, and biological processes. Atmospheric deposition has been suggested as an important source of nutrients for the marine environment in the open ocean. The overall impact of aerosol deposition on the carbon cycle is likely causing an increased CO_2 uptake in the current climate (Krishnamurthy *et al.*, 2009). Atmospheric composition, however, has been heavily perturbed by human activities and considerable uncertainty remains in our understanding of the impact of atmospheric deposition on the marine limitations. My current work is focused on the iron (Fe) deposition fluxes calculations in an Earth System Model (ESM).

Fe is considered as a key micronutrient that significantly modulates the gross primary production in High-Nutrient-Low-Chlorophyll oceans, where macronutrients like nitrate are abundant but primary production is limited by Fe scarcity. Aeolian dust is the principal source of Fe to the surface open ocean, followed by Fe-containing aerosols from biomass burning and fossil-fuel combustion emissions. However, Fe can only be utilised by phytoplankton in a dissolved form (aqueous, col-

loidal, or nanoparticulate). Atmospheric processes related to air-quality can convert Fe to become more soluble in the atmosphere. Indeed, strong acids (e.g., sulfuric acid, nitric acid) and organic ligands (e.g., oxalic acid) that coat deliquesced minerals, eventually transform part of the contained insoluble forms of Fe (e.g., ferric oxide) into soluble forms (e.g., Fe(II), inorganic soluble species of Fe(III), and organic Fe-complexes) during atmospheric processing. State-of-the-art chemistry-transport modelling studies (e.g. Luo *et al.*, 2008; Johnson and Meskhidze, 2013; Myriokefalitakis *et al.*, 2015; Wang *et al.*, 2015; Ito and Shi, 2016) currently calculate an oceanic dissolved Fe deposition flux of $\sim 0.1\text{--}0.3 \text{ Tg Fe yr}^{-1}$, clearly supporting the view that air-quality affects its deposition pattern over oceans. Furthermore, it is demonstrated that the future air-quality may decrease soluble Fe deposition by roughly 25% (Myriokefalitakis *et al.*, 2015), owing to the projected reduction in mineral-Fe dissolution ($\sim 55\%$).

Although current offline studies successfully reproduce the main aspects of present-day dissolved Fe concentrations compared to in-situ observations, giving thus high-confidence to our

understanding on Fe-release processes, ESM simulations are required to estimate the holistic impact on the global marine primary productivity (Figure 2).

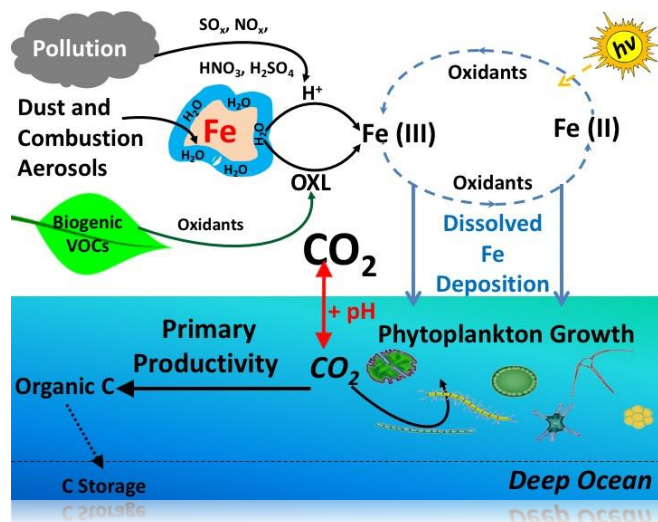


Figure 2: Concept of Fe-cycle development in the Earth System Model-EC-Earth.

For this, the state-of-the-art ESM EC-Earth has been extended with an on-line atmospheric Fe dissolution scheme, taking into account dust minerals and combustion aerosols, and it is extensively evaluated against Fe-containing aerosol observations. Currently, we develop the coupling of the new Fe deposition fields to the ocean biogeochemistry component of EC-Earth. Overall, this will allow us to investigate in more detail the potential ocean biogeochemistry perturbations due to changes in Fe deposition for the past and future atmosphere.

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Joseph Ayodele Adesina received his master's degree in Physics in Nigeria in 2010 and moved to South Africa in 2012 for his Ph.D. He investigated aerosol characteristics over different regions of southern Africa - using sunphotometer and satellite measurements.

Intercomparison and assessment of long-term (2004–2013) multiple satellite aerosol products over two contrasting sites in South Africa

Adesina, A.J.^{1*}, Kumar, K.R.², Sivakumar, V.³, and Piketh, S.J.¹

¹North West University, Potchefstroom, South Africa

²Nanjing University of Information Science and Technology, Nanjing, China

³University of KwaZulu-Natal, Durban, South Africa

*Joseph.adesina@nwu.ac.za

To establish a long-term database for climatological studies, an intercomparison of aerosol optical depth (AOD) values derived from different satellite sensors is needed. The advantage of this is an improvement in accuracy, while such a coverage cannot be achieved by single sensors (Prasad and Singh, 2007; Zhang and Reid, 2010). The study compares the variation of monthly AOD values retrieved from the two sampling stations at Skukuza and Richards Bay (SKZ and RBAY) respectively in South Africa, during the study period 2004–2013 from MODerate resolution Imaging Spectroradiometer (MODIS, on satellites Aqua and Terra) and Multi-angle Imaging Spectro-Radiometer (MISR). The common feature observed at both locations is that the three satellites noticed high AOD from August to October, which is considered as a seasonal phenomenon of the region (Queface *et al.*, 2011; Adesina *et al.*, 2014). In SKZ, all the sensors noticed high aerosol loading (> 0.25) during late winter and springtime for the years 2005, 2008, and 2010 (Figure 3). Similar variations of high AOD have

been clearly depicted at RBAY for the same years, with more pronounced AOD observed from MODIS-Aqua (Figure 3). This suggests the better performance of MODIS-Aqua over coastal sites compared to MODIS-Terra, which is in agreement with Chu *et al.* (2002).

The seasonal mean AOD is highest in spring (0.16 ± 0.05) followed by summer (0.11 ± 0.04) and autumn (0.10 ± 0.04), while the lowest was observed during the winter (0.09 ± 0.04). In springtime, the highest AOD is attributed to increased absorbing aerosols emitted from biomass burning, agricultural residues burning to clear harvest and forest fires over the study region and its surroundings. High AOD was also observed in summer months because high temperature and humidity are beneficial for the formation and hygroscopic growth of aerosols. In addition, strong atmospheric convection enhances the vertical transportation of near surface particles in summer. In addition, high temperatures in summer favour the photochemical reactions, leading to the production of secondary aerosols.

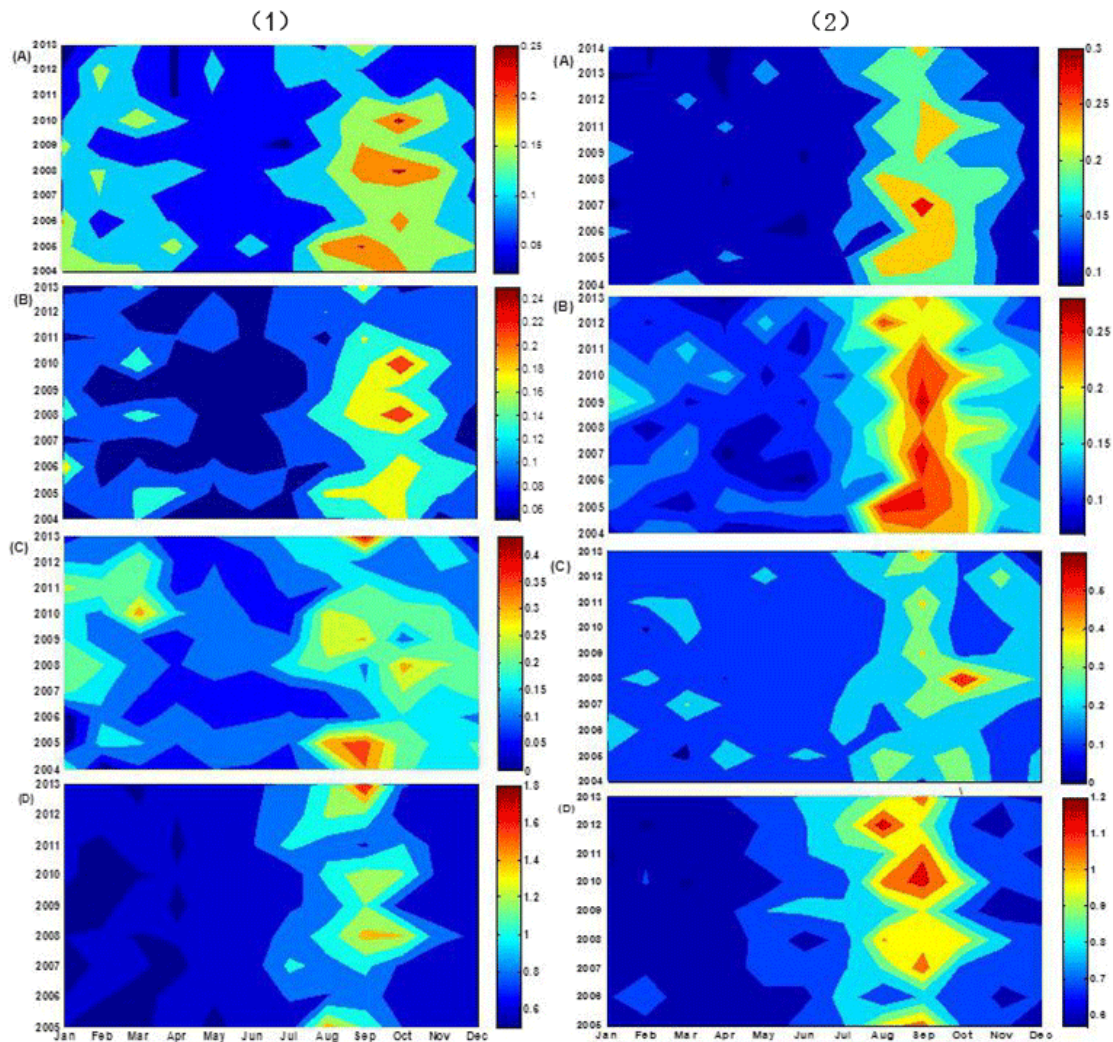


Figure 3: Averaged inter-annual variations of AOD retrieved from (A) MODIS-Terra, (B) MODIS-Aqua, (C) MISR, and ultra violet aerosol index variations from (D) Level-2 OMI at study sites (1) SKZ and (2) RBAY.

Further, the frequent occurrence of long-range transport of dust from north-western South Africa lead to an increase in AOD during spring and summer seasons (Kumar *et al.*, 2013). The annual and seasonal variability of ultra violet aerosol index retrieved from the Level-2 Ozone Monitoring Instrument data for the two study regions during 2005–2013, showed a significant increase at SKZ, having an increment of $+0.009 \text{ yr}^{-1}$ and RBAY with $+0.006 \text{ yr}^{-1}$. Overall, the performance of MISR and MODIS sensors based on the validation showed that MISR is better correlated with the AEROSOL ROBOTIC NETWORK (AERONET) sun-photometer than the MODIS products at SKZ and RBAY.

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Acknowledgements

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Theodore Konstantinos Koenig completed his Bachelors of Science at California Institute of Technology, Pasadena, United States of America, in 2012, majoring in Chemistry with minors in Geological and Planetary Science and History. Since 2012, he has pursued a Ph.D in Chemistry at the University of Colorado, Boulder, United States of America, focusing on passive remote sensing of trace gasses, particularly halogen oxides, from aircraft and mountaintops.

Inorganic bromine and iodine in the remote marine troposphere

Koenig, T.K.

University of Colorado Boulder, Boulder, United States of America

theodore.koenig@colorado.edu

Oceans and coastal marine ecosystems are the primary sources of halogens to the atmosphere. The halogens bromine and iodine have a number of important impacts in the troposphere. Both catalytically destroy ozone, and modify oxidative balance and capacity, e.g. hydrogen oxide radicals and nitrogen oxides. In addition, bromine is the primary reactant which oxidizes atmospheric mercury and reacts with some oxygenated organic compounds, and iodine contributes to particle nucleation and growth (Sherwen *et al.*, 2016). These effects in turn have important impacts on climate and human health. Halogens are emitted as organic and inorganic chemical species from biotic and abiotic sources which were particularly important in preindustrial times (Parella *et al.*, 2011). One of the sources of halogens that is of particular interest is from sea salt aerosol (SSA). Heterogeneous reactions can activate and liberate halogens to the gas phase from SSA in the free troposphere just as what they do at the sea surface. Biogeochemical control of atmospheric chemistry extends from the surface ocean through the marine boundary layer and free troposphere into the lower stratosphere. Historically, inorganic bromine and iodine in the free troposphere and stratosphere have been be-

lieved to be derived from the processing of organic species. However, recent modelling studies (Long *et al.*, 2014) suggest that much of the bromine in the mid and upper troposphere can instead be explained by this sea salt source. Aircraft measurements over the Western and Eastern Pacific during the Troposphere Exchange of Reactive Halogen Species and Oxygenated VOC (TORERO) and Convective Transport of Active Species in the Tropics (CONTRAST) campaigns (Volkamer *et al.*, 2015; Wang *et al.*, 2015; Dix *et al.*, 2016; Koenig *et al.*, 2017) confirm that inorganic bromine concentrations peak near the altitudes of outflow from deep marine convection (around 12 km), and suggest that SSA sources influence the composition of the free troposphere.

There are very few measurements that constrain the vertical distribution of inorganic bromine and iodine in the atmosphere. Aircraft studies such as those cited above are ideal for investigating vertical distributions, but they are expensive, offer only snapshots in time, and do not constrain the seasonal and interannual variation. Surface stations on remote tropical mountaintops on oceanic islands provide a platform to observe the tropical free troposphere and lower stratosphere, while



Figure 4: (A) MODIS imagery of La Réunion Island in the southwest Indian Ocean from June 11, 2017. Arcs indicate the viewing direction of the BIRA operated Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) in Le Port on the coast and the University of Colorado Boulder MAX-DOAS at Maïdo Observatory at 2.16 km. B) The Maïdo Observatory. The MAX-DOAS telescope is mounted on the roof with the rest of the instrument below it. During the intensive phase other instruments will be on the roof, or use inlets extending above the roof, or horizontally from the observatory. C) The University of Colorado Boulder MAX-DOAS telescope. The telescope can move in the vertical plane, which is perpendicular to photo, to gather profile information by probing different altitudes.

maintaining access to the boundary layer.

The Volkamer group at University of Colorado Boulder has been operating an instrument at Mauna Loa Observatory (19.5°N, 155.6°W, 3.40 km altitude) on Hawai'i for a period in January to June 2014, and again since February 2017 to present. Another one is operating at Maïdo Observatory (21.1°S, 55.4°E, 2.16 km altitude) on La Réunion since February 2017. We are further collaborating with the Royal Belgian Institute for Space Aeronomy (BIRA/IASB) who operates an instrument at sea level on La Réunion (Figure 4). Utilizing recent advances in profile retrieval methods (Coburn *et al.*, 2016), state-of-the-art retrievals are being developed for both sites to retrieve profiles with five degrees of freedom distributed among the boundary layer, free troposphere and lower stratosphere.

From March to May 2018 I will be participating in an intensive operating phase at Maïdo Observatory. In addition to our passive remote sensing

instrument we will have an in situ sensor to better characterize halogens and small OVOC at the site. Other instruments will also sample aerosol when the observatory is in the free troposphere to characterize the potential SSA contribution and processing. Other instruments present will further characterize organic species, the interplay of the marine boundary layer and free troposphere, and aerosol nucleation events which have been observed.

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近海海洋环境科学国家重点实验室 (厦门大学)

State Key Laboratory of Marine Environmental Science (Xiamen University)



Contact

SOLAS International Project Office

GEOMAR Helmholtz Center for Ocean Research Kiel, Germany
State Key Laboratory of Marine Environmental Science, Xiamen University, China

solas@geomar.de

Editors:
Jessica Gier
Li Li