A nudged CTM simulation for chemical constituent distribution during the stratospheric sudden warming observed by SMILES in 2010

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Stratospheric sudden warming (SSW) is highly nonlinear and drastic phenomena in winter stratosphere. During SSW, the distribution of chemical constituents, the associated chemical forcing, and the transport varies severely inside/outside the polar vortex. In this study chemical constituent distributions during the SSW in January 2010 are simulated by the MIROC3.2-Chemical Transport Model (CTM) nudged toward the ERA-Interim reanalysis data or the GEOS5 data, and dynamical and chemical budget of ozone amount in the Arctic polar vortex in the lower stratosphere are investigated. We discuss how the ozone amount in the Arctic polar vortex is determined in a 3-D framework. The analysis indicates that in the polar vortex during the SSW, lower stratospheric ozone concentration kept nearly a constant value in a balance between chemical ozone destruction and horizontal ozone influx from the outside of the vortex. In particular, a region with the strongest chemical ozone destruction is found near the east edge of the polar vortex in the middle of the SSW on 20-26 January, where the polar vortex extended southward and the air mass was exposed to the sunlight. At the same time, the air mass was mixed with the air mass with higher ozone concentration outside the polar vortex due to the rapid migration and extension. This region is also in a balance between strong chemical ozone destruction and strong horizontal ozone influx. The results are compared with the zonal-mean framework of the Transformed Eularian Mean. In the framework, ozone at the high latitudes increases due to enhanced downward advection, which corresponds to the ozone budget outside the polar vortex. The SMILES observation and the CTM calculation reveal the longitudinally dependent structure of chemical ozone destruction and ozone transport at the high latitudes during the SSW.

Comparison of Surface Ozone Concentrations at Two Different Population Density Areas in Malaysian Peninsula

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The Ozone (O_3) mixing ratios of some urban towns (Shah Alam, Cheras) and semi-urban (Putrajaya) in Malaysian Peninsula were measured based on a nine year (2003-2011) sample data obtained from the Malaysian Department of the Environment (DOE) through long-term monitoring conducted by Alam Sekitar Sdn. Bhd. The focus of this study was to determine the diurnal and monthly variations of ozone concentrations at the monitoring sites mentioned above. Results obtained reveal an interesting pattern in which the diurnal O_3 fluctuations are characterized by peak value of 59 ppbv for Shah Alam and 46 ppbv for Cheras. However, in the semi-urban area the maximum recorded diurnal O_3 was 50 ppbv. Furthermore, it was observed that during the night time that the pollution was at its minimum value. From the lunar calendar point of view, the maximum concentration of high surface O_3 concentration was recorded between January and April, while lowest surface O_3 concentration was observed between June and August. In-terms of sites, Shah Alam recorded the highest ozone concentrations. The comparisons of average and maximum concentrations of ozone for the three stations showed that the strongest significant correlation was recorded in the Cheras station with the value $R^2= 0.707$. The concentration of O_3 most likely to be influenced by the wind directions moreover during different monsoons seasons. By means of HYSPLIT back trajectory analysis (-72h), a trace air parcels movement was initiated on days with highest recorded O_3 concentrations to analyze the air-mass transport patterns.

Keywords: Surface ozone, Precursors, Monitoring stations, HYSPLIT trajectory analysis

A Bibliometric Analysis of Climate Engineering Research

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The past five years have seen a dramatic increase in the number of media and scientific publications on the topic of climate engineering, or geoengineering, and some scientists are increasingly calling for more research on climate engineering as a possible supplement to climate change mitigation and adaptation strategies. In this context, understanding the current state of climate engineering research can help inform policy discussions and guide future research directions.

Bibliometric analysis—the quantitative analysis of publications—is particularly applicable to fields with large bodies of literature that are difficult to summarize by traditional review methods. The multidisciplinary nature of the published literature on climate engineering makes it an ideal candidate for bibliometric analysis.

Publications on climate engineering are found to be relatively recent (more than half of all articles during 1988–2011 were published since 2008), include a higher than average percentage of nonresearch articles (30% compared with 8–15% in related scientific disciplines), and be predominately produced by countries located in the Northern Hemisphere and speaking English. The majority of this literature focuses on land-based methods of carbon sequestration, ocean iron fertilization, and solar radiation management and is produced with little collaboration among research groups.

This study (*WIREs Clim Change* 2013. doi: 10.1002/wcc.229) provides a summary of existing publications on climate engineering, a perspective on the scientific underpinnings of the global dialogue on climate engineering, and a baseline for quantitatively monitoring the development of climate engineering research in the future.

Effect of the 2009 Sarychev volcano eruption on chemistry of the lower stratosphere: balloon-borne observations and model calculations

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Total nitrogen species (NO_y) play an important role in the chemistry of the stratosphere. Nitrogen oxides (NO_x) are major catalysts responsible for significant ozone destruction in the middle stratosphere. In the gas phase, NO_x interact with the hydrogen and halogen species in catalytic cycles affecting ozone loss rates in the lower stratosphere. An important process between reactive and reservoir species deals with the heterogeneous reaction of the N_2O_5 reservoir on liquid sulfate aerosols. Including this hydrolysis reaction has been shown to better reproduce observations of NO_y partitioning and ozone loss at mid-latitude for background aerosol conditions (i.e. in volcanically quiescent periods) in the lower stratosphere. This is also true when sulphate aerosols from major volcanic eruptions, such as the Pinatubo in 1991, are present in the stratosphere on scales of years.

Over the past decade several "moderate" eruptions (i.e. about 10-20 times lower amounts of sulphur dioxide (SO_2) injected compared to the Pinatubo event) have affected and even modulated the sulphate aerosol content of the lower stratosphere with residence times of several months after each eruption. Here we investigate the impact of such a "moderate" volcanic eruption on the chemistry of the lower stratosphere particularly focussing on the partitioning of nitrogen species.

The Sarychev volcano eruption located in Kuril Islands (48.1°N, 153.2°E) injected about 0.9 Tg of SO_2 in June 2009 up to an altitude of 19 km. By mid-July 2009, the aerosol cloud had spread out over the northern hemisphere and remained detectable until late November 2009. A balloon campaign took place from the Kiruna/Esrange launching base (67.5°N, 21.0°E) in summer 2009 and allowed us to capture the aerosol plume from the Sarychev eruption. Balloon-borne aerosol counters and spectrometers (using in situ and solar occultation techniques) were deployed in the period from 7 August to 7 September to provide the aerosol concentrations and the vertical profiles of key chemical compounds.

Conducted in the Arctic later summer, the balloon-borne observations correspond to a period when N_2O_5 has sufficiently recovered so that its hydrolysis should constitute an important factor, providing chemical states comparable to mid-latitude conditions. Through the N_2O_5 hydrolysis, we show that the increase in the aerosol content has therefore significantly affected the NO_y partitioning in the lower stratosphere. We have made model calculations using a 3D Chemistry Transport model (CTM) and the simulations reveal the importance of implementing the observed aerosol surface area densities (SAD) into the CTM to reproduce the observations of NO_2 and of the NO_2/HNO_3 ratio. Calculated NO_2 amounts are decreased by several percents. Using a 1D version of the CTM forced by satellite observations of total NO_y minimizes spurious transport effects and consequently even better reproduce the measured profiles.

However, we show that computing the enhanced SAD from the eruption has nearly no impact on the simulated ozone amounts indicating that such moderate eruptions have negligible effects on ozone destruction in the lower stratosphere for extra-vortex conditions (at least for the investigated period) contrary to major eruptions.

A semi-empirical model of the stratosphere in the Antarctic climate system

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A simple climate model is being extended to include an interactive stratosphere such that computationally efficient projections of Antarctic ozone depletion can be generated. The stratospheric component of the interactive simple climate model is named SWIFT (Semi-empirical Weighted Iterative Fit Technique). SWIFT comprises a set of coupled first-order differential equations describing the time rate of change of key species in the polar stratosphere, including HNO_3 (total), HNO_3 (gas phase), HCl, $ClONO_2$, ClO_x and ozone. It describes the time evolution of these prognostic variables throughout the polar winter, starting from prescribed initial conditions. The equations for these species include terms for the overall net effect of the relevant chemical mechanisms rather than one term for each reaction, which is the approach usually used in models of stratospheric chemistry. SWIFT is driven by time series of FAP, the fractional area of the polar vortex experiencing temperatures below 195 K (the temperature threshold for polar stratospheric cloud formation), and FAS, the fractional area of the vortex exposed to sunlight. Both can be calculated from stratospheric temperature fields which are generated using a module called STePS (Stratospheric Temperature Pattern Scaling) that uses climate pattern scaling methods to generate temperature fields from CO_2 and vortex-averaged ozone concentrations. STePS begins with an *a priori* ozone field and then runs iteratively with SWIFT to produce physically consistent ozone and temperature fields that are also consistent with instantaneous equivalent effective stratospheric chlorine (EESC) and CO_2.

This presentation will describe the different new components of the interactive simple climate model (MAGICC6), how these components interact, and some initial results. A specific focus will be on the development and performance of the STePS module since this is the first time climate pattern scaling has been used to generate stratospheric temperature fields. The possible non-linear ozone and CO_2 dependence in STePS will be investigated. An outlook for the application of this interactive simple climate model for projecting the evolution of the Antarctic ozone hole for a range of greenhouse gas emission scenarios will also be presented.

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Unaccounted ozone source in the upper troposphere

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Convective redistribution of ozone and its precursors between the boundary layer (BL) and the free troposphere (FT) influences the photochemistry, in particular the ozone production in the middle and upper troposphere (UT). We present a case study of convective transport during the GABRIEL campaign over the tropical rain forest in Suriname in October 2005. During a measurement flight on October 12 the inflow and outflow regions of a cumulonimbus cloud (Cb) have been characterized, providing clear indications of convective transport. We identified a distinct layer between 9 and 11 km altitude with enhanced mixing ratios of CO, HO_x, acetone, acetonitrile and ozone.

The elevated O_3 in this layer contradicts the expectation that convection transports ozone poor air from the boundary layer to the outflow region. From budget analysis we calculate that entrainment of ozone rich air into the convective outflow accounts for 64 % (range: 34 % - 93 %) of the observed O_3. Furthermore, ozone is enhanced by 4-5 % by photochemical production in the outflow catalysed by lightning NO. The photochemical contribution is derived from steady state model calculations, using in-situ observations including the first reported HO_x measurements over the tropical rainforest. The unaccounted ozone in the outflow is most probably due to direct production from lightning by cold corona discharge. To close the ozone budget in the outflow region we deduce an O_3 production rate per flash which is an order of magnitude higher compared to previous estimates.

Stratospheric aerosol including volcanoes simulated with the EMAC model: effects on radiation, dynamics and heterogeneous chemistry

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The chemistry-climate model EMAC interactively simulates stratospheric and tropospheric chemistry, aerosols and dynamics, accounting for trace gas and particle emissions including volcanic SO_2. Aerosols are represented using a seven-mode submodel from which optical properties are calculated based on Mie theory, optionally feeding back into radiative heating rates and atmospheric dynamics. Further, the simulated sulphate aerosol affects stratospheric heterogeneous chemistry which influences ozone and also feeds back onto the dynamics. We present simulations that represent the major atmospheric perturbation due to the eruption of Mt. Pinatubo in 1991, and the period from 1999 to 2009 with background aerosol, mostly from COS oxidation, influenced by 6 medium-size volcano eruptions. The comparison with satellite observations demonstrates that the model is able to realistically simulate the formation of aerosol from SO_2 injections, including the lofting in the Brewer Dobson circulation together with enhanced tropical upwelling due to radiative heating perturbations. We show that the volcanic SO_2 injections (Brühl et al., Atmos. Chem. Phys. Disc. 13, 11395) and the oxidation of tropospheric COS (Brühl et al., Atmos. Chem. Phys. 12, 1239) are the predominant sources of the main stratospheric aerosol layer (Junge layer). The calculated radiative forcings of the troposphere by stratospheric aerosol (Figure 1) are consistent to those derived from observations.

The presentation will include a comparison with an EMAC reference simulation within CCMI (Chemistry-Climate Model Initiative) in which aerosol properties are prescribed based on satellite observations, and some evaluation of simulated middle atmosphere SO_2 against MIPAS satellite observations.

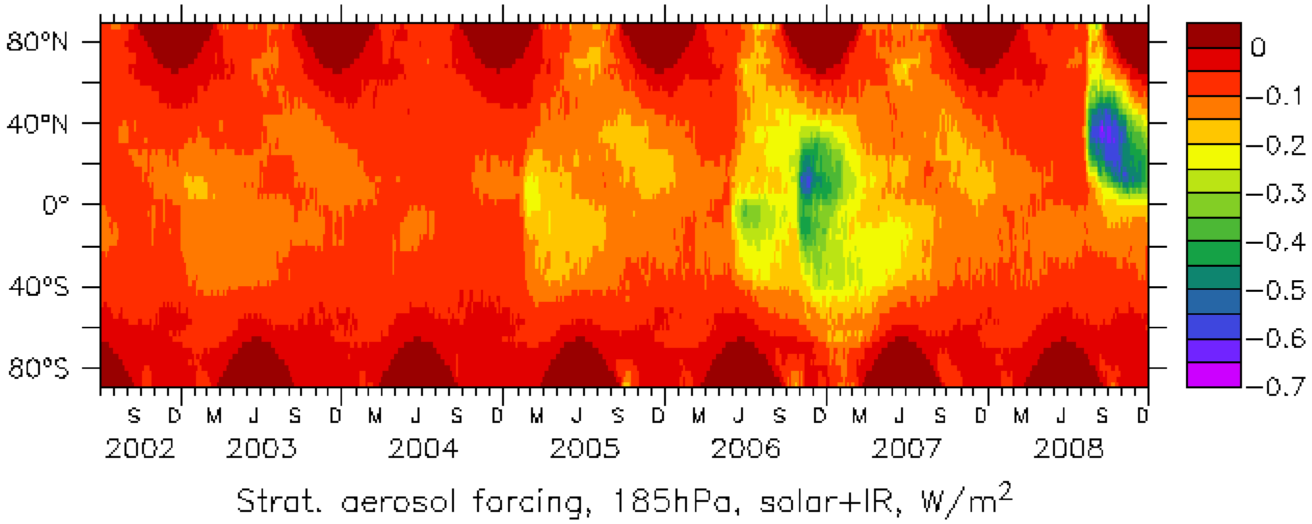

Figure 1: Simulated zonal average aerosol radiative forcing near the tropopause.

The Chemistry-Climate Modelling Initiative: Evaluation and preliminary projections from HAdGEM3 simulations

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The new joint IGAC/SPARC chemistry-climate modelling initiative (CCMI) has defined a range of coordinated reference and sensitivity simulations that will allow the community to perform quantitative inter-model comparisons and make multi-model projections, particularly of stratospheric ozone and climate. It is anticipated that results from these community simulations will provide supporting scientific material for the forthcoming WMO/UNEP stratospheric ozone assessment. The Met Office Hadley Centre is contributing to this coordinated effort using a version of its HadGEM3 climate model which includes a combined stratosphere-troposphere chemistry scheme developed under the United Kingdom Chemistry-Aerosol (UKCA) project. The future twenty-first century reference and sensitivity simulations are being run with a coupled ocean to enable the impact of stratospheric change on surface climate to be properly assessed. An evaluation of the model performance, particularly in the stratosphere, will be presented based on the CCMI historical reference simulation - REF-C1. Depending on the progress of the simulations preliminary projections from the REF-C2 scenario simulation will be presented too.

The Impact of Methane Clathrate Emissions on the Earth System

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Methane is locked in ice-like deposits called clathrates in ocean sediments and underneath permafrost regions. Clathrates are stable under high pressures and low temperatures, so in a warming climate, increases in ocean temperatures could lead to dissociation of the clathrates and release methane into the ocean and subsequently the atmosphere, where methane is both an important greenhouse gas and a key species in atmospheric chemistry. Clathrates in the shallower parts of the Arctic Ocean (around 300m depth) are predicted to be particularly important since clathrates at that depth are expected to start outgassing abruptly in the next few decades. We will present the atmospheric impact of such methane emissions using multi-century steady-state simulations with a version of the Community Earth System Model (CESM) that includes atmospheric chemistry. Our simulations include a plausible release from clathrates in the Arctic that increases global methane emissions by 22%, as well as a scenario with 10 times those clathrate emissions. The CESM model includes a fully interactive physical ocean, to which we added a fast atmospheric chemistry mechanism that represents methane as a fully interactive tracer (with emissions rather than concentration boundary conditions). The results indicate that such Arctic clathrate emissions (1) increase global methane concentrations by an average of 38%, non-uniformly; (2) increase surface ozone concentrations by around 10% globally, and even more in polluted regions; (3) increase methane lifetime by 13%; (4) increase the interannual variability in surface methane, surface ozone, and methane lifetime, and (5) show modest differences in surface temperature and methane lifetime compared to simulations in which the clathrate emissions are distributed uniformly.

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What are the drivers of interannual fluctuations of atmospheric methane concentrations ?

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Atmospheric methane (CH_4) concentrations have more than doubled since pre-industrial times, making CH_4 the second most important greenhouse gas after CO_2 in termes of radiative forcing. After a period of about 8 years with growth rates close to zero, methane conentrations are rising again since 2007. With a 25-times larger global warming potential than CO_2, methane will play an even more important role in global climate change if it continues to rise. Changes in the strength of different methane sources and/or in the main methane sink, the chemical oxidation by OH, are being discussed as potential reasons for this temporal development and the strong interannual variability in the methane growth rate.

We are in the process to perform simulations using the SOCOL (Solar Climate Ozone Links) chemistry-climate model, for the time period 1980-2010, nudging the model with prescribed meteorological fields to ensure best comparability with observational data sets. To investigate the relative importance of different source categories such as natural wetlands, rice paddies, ruminants, industry, etc., and sink processes, additional methane tracers have been included into SOCOL, which allow the tracking of methane emissions from different source categories and geographical regions. In addition, the impact of outstanding episodes like strong ENSO events or volcanic eruptions on atmospheric methane concentrations, either by changing methane emissions from, e.g., natural wetlands, or by changing chemical methane loss rates via OH are studied. These new simulations are an innovative way to obtain a better understanding of the atmospheric methane interannual variability, and first results of the source attribution will be presented.

The stratospheric water vapor feedback

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Variations in stratospheric water vapor play a role in the evolution of our climate. We show here that variations in water vapor since 2004 can be traced to tropical tropopause layer (TTL) temperature perturbations from at least three processes: the quasi-biennial oscillation, the strength of the Brewer-Dobson circulation, and the temperature of the troposphere. The connection between stratospheric water vapor and the temperature of the troposphere implies the existence of a stratospheric water vapor feedback, which we estimate to have a strength of +0.1 W/m^2/K and is potentially responsible for up to 0.4 K (10%) of the equilibrium climate sensitivity. Analysis of a set of CCMVal models shows that, as a group, they agree well with the observations.

Enhancing Integrated Earth Observations of the Composition of the Atmosphere: The Role of the CEOS Atmospheric Composition Constellation

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The Committee on Earth Observation Satellites (CEOS) coordinates civil space-borne observations of the Earth. Participating agencies strive to enhance international coordination and data exchange and to optimize societal benefit. In recent years, CEOS has collaborated closely with the Group on Earth Observations (GEO) in implementing the Global Earth Observing System of Systems (GEOSS) space-based objectives. To support these activities, CEOS has developed the concept of virtual, space-based constellations. A virtual constellation is a coordinated set of space and/or ground segment capabilities from different partners that focuses on observing a particular parameter or set of parameters of the Earth system.

The goal of the CEOS Atmospheric Composition Constellation (ACC) is to collect and deliver data to improve monitoring, assessment and predictive capabilities for changes in the ozone layer, air quality and climate forcing associated with changes in the environment through coordination of existing and future international space assets. In addition, ACC is supporting a new CEOS priority to contribute to the WMO Global Climate Observing System (GCOS) Implementation Plan through the development and harmonization of relevant fundamental climate records.

We report on current ACC activities, which include multiple projects of relevance to the community. A project to coordinate and enhance the science value of a future constellation of geostationary sensors measuring parameters relevant to air quality has made substantial progress in the past year, as the European Sentinel-4, Korean GEMS, and US TEMPO missions are all likely to fly in the 2018-2020 timeframe. Two recent ACC workshops have focused on activities relevant to the multiple, long-term total ozone data sets produced by space-based and ground-based observations. A consensus emerged that ACC would facilitate the collaborations among the various instrument groups to make a best-effort characterization of errors of the individual data sets and to enhance interaction between North American researchers and European ozone_cci project participants to bridge the gap between these two sets of total ozone measurements. ACC is also concerned with looming gaps in limb-scattering ozone profile measurements, recognizing the unexpected end of SCIAMACHY O_3 measurements, the aging of existing sensors, and uncertainty in US JPSS ozone limb measurement plans.

An ACC-led activity to establish a multi sensor volcanic eruption alert system saw the establishment of an Aviation Control Service (SACS) web service that provides an SO_2 alert service based on multi-sensor (OMI, GOME2, IASI, AIRS) measurements.

In addition, ACC plans to respond to the forthcoming CEOS Carbon Task Force report and its progress in composing a response to the GEO Carbon Strategy Report. ACC will play a role in implementing some of its atmospheric relevant recommendations (e.g., on the generation of GHG essential climate variables).

Heterogeneous Formation of Polar Stratospheric Clouds – Nucleation of Ice on Synoptic Scales

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Shortly after the discovery of the Antarctic ozone hole, anthropogenic halogen compounds were identified as important ozone depleting substances and the key role of polar stratospheric clouds (PSCs) in activating these substances was recognized. However, in spite of more than two decades of research, details of PSC formation remain uncertain. The nucleation mechanisms of nitric acid trihydrate (NAT) and ice particles have recently been put to test by the EU-project RECONCILE and the results of a detailed microphysical modelling study indicates that new heterogeneous nucleation mechanisms are required for both NAT and ice, namely freezing on solid nuclei immersed in liquid stratospheric aerosols, possibly of meteoric origin (Hoyle et al., 2013; Engel et al., 2013). A companion abstract by Hoyle et al. focuses on NAT nucleation. In this paper we examine this new mechanism for nucleation of ice.

During January 2010, ice PSCs were shown by the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) spaceborne lidar to have occurred on synoptic scales (~ 1000 km dimension). We demonstrate that the formation of these PSCs is not readily reconciled with expectations from the conventional understanding of PSC nucleation mechanisms and cannot be explained merely by homogeneous ice nucleation. Hence, a heterogeneous nucleation mechanism is required, which might occur on the surface of meteoritic particles. The efficiency of the ice nuclei seems surprisingly high, namely comparable to that of known tropospheric ice nuclei such as mineral dust particles. To gain model agreement with the ice number densities inferred from observations, the presence of small-scale temperature fluctuations, with wavelengths unresolved by the numerical weather prediction models, is an additional necessity. With the derived rate parameterization for heterogeneous ice nucleation we are able to explain and reproduce CALIPSO observations throughout the entire Arctic winter 2009/2010.

These new findings will be extremely important for improving global and regional scale climate and chemistry models. As a first step, we implemented the new parameterization for NAT nucleation into the Chemical Lagrangian Model of the Stratosphere (CLaMS), which so far represented NAT nucleation with the help of a constant nucleation rate. First results look promising and motivate to go a step further and improve also the ice nucleation scheme.

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Analysis of precipitation data from the NIWA-UKCA climate runs

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The atmosphere-ocean chemistry-climate model NIWA-UKCA has been run over the period of 1960 - 2100, following a mid-range Representative Concentration Pathway scenario, as well as with individual climate drivers, increases in greenhouse gases, and variations in ozone-depleting substances, removed. The simulations are aimed at improving understanding of the roles of these drivers in Southern-Hemisphere climate change.

We present an analysis of the NIWA-UKCA model runs, and a comparison to other available data such as observations, re-analyses, and the CMIP5 simulations. Precipitation data and other atmospheric fields are analysed to investigate shifts in the precipitation bands, which could indicate movement of the major tropospheric overturning cells. Using the sensitivity simulations, we will attribute such shifts to the major climate drivers causing climate change in the Southern Hemisphere. The simulations will contribute to the Chemistry-Climate Modelling Initiative (CCMI).

Update on the Soundings of Ozone and Water in the Equatorial Region (SOWER) through the Year 2014

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Water plays a crucial role on the earth's radiation budget and its stratospheric variation could drive global surface climate change on a decadal scale (Solomon et al., 2010). The Soundings of Ozone and Water in the Equatorial Region (SOWER), initiated in 1998, has contributed to establish the climatology and variabilities in the tropical stratospheric water concentrations (Fujiwara et al., 2010). It has been making recent efforts toward in situ observations of the air parcels advected horizontally in the Tropical Tropopause Layer (TTL) of the western Pacific trying to accumulate evidences of the cold-trap dehydration hypothesized by Holton and Gettelman (2001). Those instruments primarily used are NOAA FrostPoint Hygrometers (FPHs) and University of Colorado Cryogenic Frostpoint Hygrometers (CFHs) for water and ECC ozonesondes for ozone along with conventional radiosondes. The existence of ice particles and aerosols has been diagnosed by lidars simultaneously operated with sonde flights.

Series of SOWER data successfully characterized the nature of TTL dehydration. The findings include 80 % supersaturation in relative humidity with respect to ice (RH_{ice}) in subvisible cirrus clouds located near the cold point tropopause (CPT) at the temperature around 180 K and the evolution of the cold-trap dehydration effectively functioning between 360 K and 380 K isentropes while some moistening taking place before the air parcel reaches 400 K (Hasebe et al., 2013). The first attempts of the TTL water vapor 'match' suggest the threshold of nucleation being 146 ± 19 % in RH_{ice} with the typical dehydration time scale of about an hour in the lower TTL (Inai et al., 2013). Simultaneous observations by lidar and Optical Particle Counters near the CPT reveal cirrus clouds within the aerosol layer composed of liquid phase aqueous sulphuric acid particles. The estimated concentrations of cloud particles and aerosols are not consistent with the interpretation of cloud formation in liquid phase aerosol particles (Shibata et al., 2012).

SOWER will continue to work on TTL science through the year 2014. What is distinct from the previous activities is the data acquisition in January-February period correlative with aircraft observations cooperated by Airborne Tropical TRopopause Experiment (ATTREX), Convective Transport of Active Species in the Tropics (CONTRAST) and Coordinated Airborne Studies in the Tropics (CAST). Effort will be also made to better describe Lagrangian evolution of air parcels by improving data assimilation technique. Another direction SOWER intends to collaborate is the collection of constituent and aerosol samples in the TTL and the lower stratosphere. The analysis of data collected during the cruise in the equatorial eastern Pacific by Research Vessel Hakuho-Maru is under way.

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Where is the equator? The geography of atmospheric chemistry

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The general circulation of Earth’s atmosphere partially isolates the northern and southern hemispheres, enabling divergent composition and reactive chemistry. An important chemical difference between the hemispheres is the concentration of hydroxyl (OH). The main sink for methane and hydrofluorocarbons (HFCs), tropospheric OH controls the abundance of these greenhouse gases, as well mediating production of tropospheric ozone---a greenhouse gas and air pollutant. Observations of methyl chloroform, a synthetic gas that reacts mainly with tropospheric OH and has negligible residual sources, imply that there is about 10% more methane loss in the southern hemisphere. In contrast, atmospheric chemical transport models (CTMs) generally simulate greater OH concentrations and methane loss in the northern hemisphere. This apparent shortcoming of many models derives in large part from an inconsistent definition of the hemispheres. For model results, OH concentrations and methane loss are most commonly averaged over the geographic northern and southern hemispheres, with the geographic equator as the dividing line. For the observations, however, the hemispheres are defined by the atmosphere’s circulatory mixing barrier that rarely coincides with the geographic equator. Instead the barrier to interhemispheric mixing corresponds to the rising branch of the Hadley circulation and follows the seasonal migration of the sun.

In this work, we create artificial tracers in a CTM to locate the interhemispheric mixing barrier and define the circulatory (as opposed to geographic) hemispheres. The CTM is driven by assimilated meteorology from the European Centre for Medium-range Weather Forecasts (ECMWF; cycle 36, T42 resolution). The tracers are released in the high latitudes of the northern and southern hemispheres and the circulatory equator can be identified from either the tracer gradients or where the northern and southern hemispheric tracers are equal. We show that the circulatory equator usually lies north of the geographic equator and coincides with the intertropical convergence zone and major monsoons. Using this dynamic and time-varying definition of the equator, we average methane loss in the CTM over the circulatory hemispheres and compare the results to observations. Our method tests the simulated OH chemistry on hemispheric scales in a consistent way with observations. Our results suggest that the apparent discrepancy between the models and observations of methane loss is smaller than previously estimated.

Heterogeneous Formation of Polar Stratospheric Clouds - Nucleation of Nitric Acid Trihydrate (NAT) in the Arctic Stratosphere

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Satellite based observations during the Arctic winter of 2009/2010 provide firm evidence that, in contrast to the current theory, the nucleation of nitric acid trihydrate (NAT) in the polar stratosphere does not only occur on preexisting ice particles. In order to explain the NAT clouds observed over the Arctic in mid-December 2009, a heterogeneous nucleation mechanism is required for NAT, occurring on the surface of dust or meteoritic particles. For the first time, a detailed microphysical modelling of this NAT formation pathway has been carried out (Hoyle et al. 2013). Heterogeneous NAT formation was calculated along tens of thousands of trajectories, ending at Cloud Aerosol Lidar with Orthogonal Polarisation (CALIOP) observation points. Comparing the optical properties of the modelled NAT PSCs with these observations enables the thorough validation of a newly developed NAT nucleation parameterisation, which has been built into the Zurich Optical and Microphysical box Model (ZOMM). The parameterisation is based on active site theory and is simple to implement in models. It is shown that the new method is capable of reproducing both the size and the classification of observed PSCs very well (Fig. 1), despite the varied conditions experienced by air parcels travelling along the different trajectories. A companion study examines the heterogeneous nucleation of ice PSCs (Engel et al. 2013).

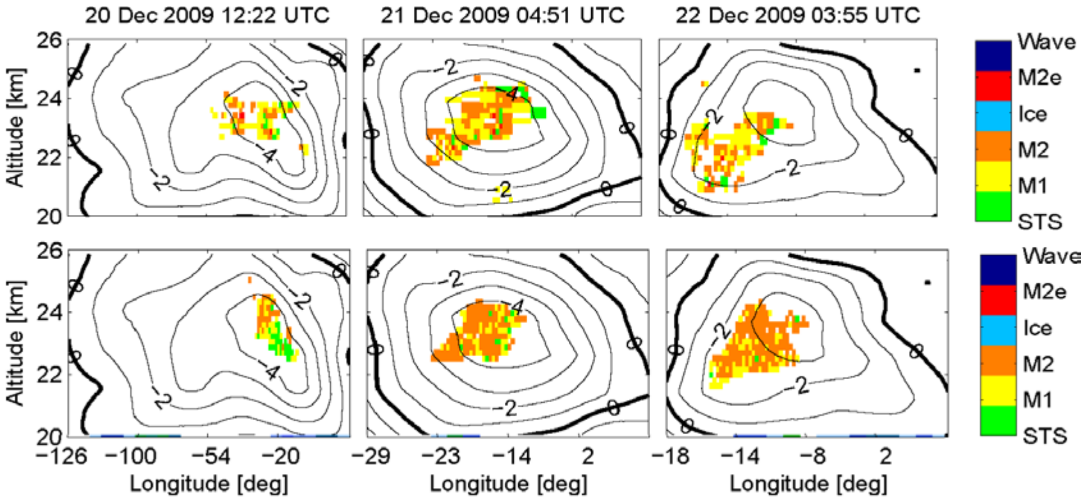

Figure 1. A comparison between the classifications of observed (upper row) and modeled (lower row) PSCs, along three different CALIPSO orbits. The colour coding indicates the classification of the cloud, ranging from STS, mixtures of NAT and STS (M1, M2, M2e) to ice and wave ice clouds.

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50 Years of Measurements of Tropospheric Ozone at Northern Midlatitude Site (51.83°N, 20.78°E) - Belsk, Poland; Dobson Umkehr Data Recalculated by the Neural Networks Method

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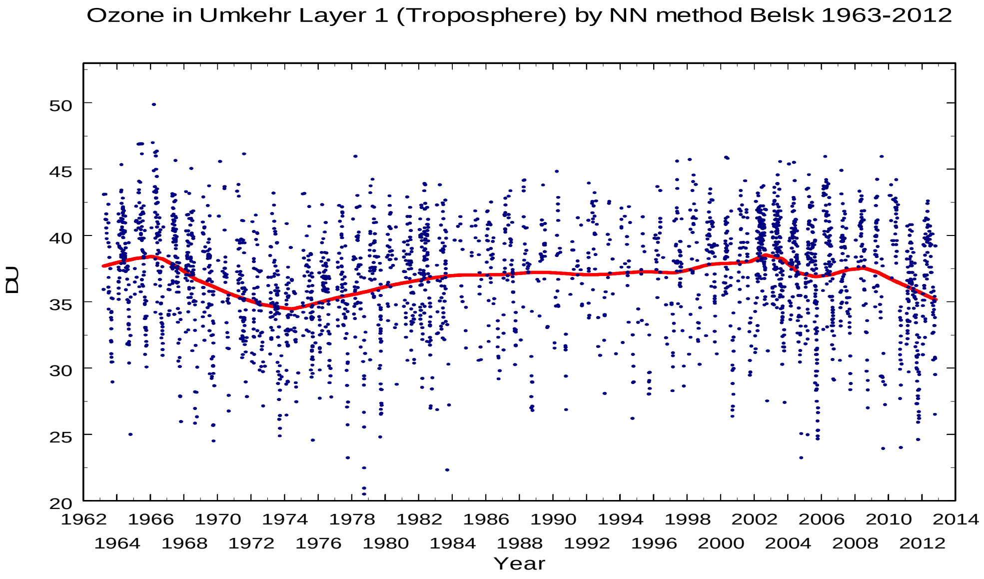
Atmospheric total ozone content and ozone profile measurements by the Umkehr method have been performed at the Central Geophysical Observatory of the Institute of Geophysics at Belsk, Poland since 1963. It is one of the longer series of measurements of the total ozone and the ozone vertical profile in the world, often used for the analysis of long-term changes in ozone because of recognized quality.

It is known that the Umkehr method gives relatively inaccurate results when determining the tropospheric ozone content. To improve this accuracy, the new method for determining the ozone content in the troposphere based on the neural networks (NN) method has been proposed (Jarosławski, 2013). The results of ozone soundings performed at Legionowo station (60 km from Belsk station) served as a reference dataset for NN training procedures. Thus, a precise, ozonesonde-like quality information about tropospheric ozone content for a pre-satellite and pre-ozonesonde period has been obtained. The results (daily means of ozone in Dobson Units (DU)) are shown in Figure 1. The information about tropospheric ozone trends since the seventies is consistent with the information from other sources: increase in the seventies and decrease after the maximum at around 2003. However, unexpectedly, another maximum in the tropospheric ozone content followed by the decrease of the ozone content during the sixties has been observed. This maximum is higher than the maximum observed in 2003.



Ozone in Umkehr Layer 1 (Troposphere) by NN method Belsk 1963-2012

References: Janusz Jarosławski (2013): Improvement of the Umkehr ozone profile by the neural network method: analysis of the Belsk (51.80°N, 20. 80°E) Umkehr data, International Journal of Remote Sensing, 34:15, 5541-5550.

Volcanic SO_2 emissions in the stratosphere from 1979 to 2009

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The climate effects of volcanic eruptions are well acknowledged. These effects are due to the production of a layer of sulphate aerosols in the lower stratosphere, which efficiently backscatters solar radiation, increases the planetary albedo, and causes cooling at the surface. For these radiative effects to accumulate, the aerosols must remain in the atmosphere for an extended period of time. The stratospheric aerosol e-folding lifetime is strongly dependent on the altitude of injection. The residence time of aerosols is about 1 week when the injection occurs only in the troposphere and varies from a few months for moderate eruptions to more than 1 year for major eruptions (volcanic explosive index ≥ 6).

The recurrent moderate eruptions have resulted in a net negative radiative forcing in the period subsequent to 2000, offsetting the positive radiative forcing owing to increased concentrations of well-mixed greenhouse gases and hence global warming. It is therefore important to monitor the volcanic SO_2 emissions to be able to forecast the physical properties of stratospheric aerosols and to quantify their radiative and chemical impact.

In the last years an effort have been made to archive all the volcanic SO_2 emissions in an unique database. This work was achieved in the framework of the AeroCom hindcast project. All the volcanic eruptions listed in the Global Volcanism Program's database provided by the Smithonian Institution were considered. This new database contains the amounts and altitudes of the SO_2 injected from explosive and effusive eruptions as well as degassing from 1 January 1979 to 31 December 2009. Each element of the database corresponds to the emission of SO_2 by a specific volcano on a specific day.

From 1979 to 2009 volcanic eruptions have injected 831Tg of SO_2 into the atmosphere, of which around 56Tg (~7%) reached the stratosphere. Stratospheric SO_2 emissions are essentially due to cataclysmic eruptions like El Chichón in 1982 (7.5Tg) or Mount Pinatubo in 1991 (18Tg). More regularly moderate eruptions (<2Tg) are likely to modulate the “background” aerosol layer in the lower stratosphere (Soufrière Hills, 2006, Kasatochi, 2008, Sarychev Peak, 2009). The Chemistry Climate Model LMDz-INCA has been used to quantify the impact of these volcanic emissions on the stratospheric aerosol distribution and to evaluate their radiative effects.

Impacts of Model Errors on Global Estimates of CO and CH_4 inferred from MOPITT and TES datasets

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Inverse modeling has been widely used to constrain emission estimates of pollutants and greenhouse gases. However, the inversion analyses are sensitive to systematic errors in the atmospheric models. In this work, we assimilate multi-spectral (near infrared and thermal infrared) retrievals of CO from MOPITT, using the GEOS-Chem model, to study the influence of vertical transport errors on inferred CO source estimates. We compare the source estimates obtained by assimilating the profiles and the surface level retrievals from June 2004 to May 2005. The discrepancies in the CO source estimates are also used to assess the uncertainty in the CH_4 emission estimates from Indonesian peat fires in a moderate El Nino during fall 2006, constrained from TES CH_4 observations. Our results demonstrate that the inferred source estimates for long-lived tracers, such as CO, CO_2 and CH_4, could be significantly biased in regions where strong emissions are co-located with significant vertical mass flux due to convection. Our results also show that the multi-spectral MOPITT data provide a valuable dataset for evaluating vertical transport and characterizing biases in models.

The Link between Springtime Total Ozone and Summer UV Radiation in Northern Hemisphere Extratropics

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The link between stratospheric ozone decline and ultraviolet (UV) radiation increase at the Earth's surface is well established. In the Northern Hemisphere extratropics stratospheric ozone is accumulated from autumn to spring as a result of transport from its source region in the Tropics. The amount of accumulated ozone varies from year to year due to natural dynamical variability and chemical destruction by natural and anthropogenic substances. Observational and modeling studies show that these total ozone anomalies persist in the extratropics from spring through summer. Here we analyze timeseries of ground–based UV measurements and satellite retrievals of total ozone and UV radiation and demonstrate that there is a strong link between springtime total ozone and summer UV anomalies in the Northern Hemisphere extratropics. In some regions, the interannual variability in springtime ozone abundance explains 20–40% of the summer UV variability, and this relation can be used for improving seasonal UV forecasts. Such possibility may have implications for agriculture. The advance information about summer UV radiation may also influence human behavior.

Using chemistry–transport models we estimate the influence of polar chemical ozone loss on the summer UV north of 35°N. We estimate that the massive Arctic ozone depletion 2011 increased the March–August cumulative erythemal clear–sky UV dose in the Northern Hemisphere extratropics by 3–4% compared to the climatology, with about 75% of the increase accumulated after the breakup of the polar vortex. This result provides relevant information for the Parties to the Montreal protocol. We suggest that the effect of seasonal ozone anomaly persistence should be included in assessment of the impacts of polar ozone losses.

Modulation of Southern Hemisphere climate drivers by large-scale geoengineering

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Geoengineering is increasingly being discussed as a tool to lessen the impacts of anthropogenic climate change through deliberate modification of the climate system. While it could play a key role in reducing the risk of dangerous climate change, it is not without significant regional risks of its own. To date, the effects of geoengineering on the Southern Hemisphere (SH) climate have not been well studied, nor have the impacts on SH climate drivers been considered.

Within the context of the Geoengineering Model Intercomparison Project (GeoMIP), we use the CSIRO Mk3L climate system model to investigate and quantify the potential response of the SH climate to large-scale geoengineering. In this study, we focus on two GeoMIP experiments: G3 and G4. These experiments explore the application of solar radiation management to counteract rising greenhouse gas (GHG) concentrations within the CMIP5 RCP4.5 emissions scenario. Experiment G3 simulates the gradual introduction of stratospheric sulphate aerosols during the period 2020 to 2070, with the aim of keeping the net radiative forcing constant. In contrast, experiment G4 simulates the sudden onset of geoengineering in the year 2020.

Two of the dominant drivers of SH climate variability are the Southern Annular Mode (SAM) and the Southern Oscillation (SO). Here we focus on how these might be modulated if geoengineering were to be implemented. We show that the SAM shifts towards a more neutral state, reversing the observed trend towards the positive phase over recent decades. As a result, there is a northward shift and weakening of the SH westerly winds. The SO also shifts towards a more neutral state, offsetting the El Niño-like response of the tropical Pacific to increasing GHGs. We see a consistent reduction in precipitation at most latitudes, including a weakening of the summer monsoon systems. The climatic impacts of geoengineering are greater in experiment G3 than in G4, reflecting the fact that the mean radiative forcing due to stratospheric sulphate aerosols is stronger in the former experiment than in the latter. Under both scenarios, any climatic impacts cease abruptly as soon as geoengineering is stopped.

Distribution and variation of biomass burning tracers CO, HCN, and CH3CN in UTLS

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By using the 3-D global chemical transport model GEOS-Chem simulations, we analyze the distribution and variation of biomass burning tracers CO, HCN, CH3CN in upper troposphere and lower stratosphere (UTLS). Model simulations well capture the main aspects of CO, HCN and CH3CN concentration in UTLS regions observed by satellite instruments. GEOS-Chem also simulates the satellite observed seasonal and inter-annual variabilities of these biomass burning tracers in UTLS. Associated with the analysis of ground-based observations (FTIR) and global biomass burning emission data (GFED), we explain that both seasonal and inter-annual variabilities are mainly induced by surface emissions, especially from tropical regions. On the other hand, the dynamical uplift due to Monsoon systems may also play an important role transporting the regional emissions to UTLS and affecting variations of biomass burning tracers in UTLS. In this study, we will focus on the UTLS distribution and variation of CO, HCN and CH3CN from emissions of different tropical regions such as S.E. Asia, Indonesia, Africa, and Southern America, respectively. The influences of dynamical transport by Monsoon systems especially the Asian Summer Monsoon will be analyzed as well.

Assessment of Short-Lived Unobserved Radicals at Lauder Using a Photochemical Steady-State Single-Column Model

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Lauder is a research station located on the South Island of New Zealand known for its clean air representative of southern mid-latitudes. Long-term records of various atmospheric chemical compounds have been measured here for up to three decades. The absence of short-lived pollutants means that relatively few constituents define the oxidation capacity at this site. Within this study we aim to use measurements of these species, together with other available data as needed, to infer short-lived species such as OH which are not commonly observed, since they are central to atmospheric chemistry and remain difficult to model correctly in global chemistry models. To this end we have constructed a photochemical steady-state single-column model for the Lauder site. Long-lived and stable compounds in this model are prescribed using Lauder observations; the model predicts short-lived constituents such as OH as inferred from the long-lived constituents. We discuss here first results from this activity, particularly regarding variations in photolysis rates and OH at the Lauder site.

Whole-atmosphere aerosol microphysics simulations of the Mt. Pinatubo eruption: evaluation of simulated aerosol properties, assessment of radiative effects and uncertainty quantification via Gaussian emulation

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The Mt Pinatubo volcanic eruption in June 1991 injected between 14 and 20 Tg of sulphur dioxide into the tropical stratosphere between about 21 and 28km altitude. Following chemical conversion to sulphuric acid, the stratospheric aerosol layer thickened substantially causing a strong radiative, dynamical and chemical perturbation to the Earth's atmosphere with effects lasting several years.

In this presentation we show results from model experiments to simulate the evolution of the stratospheric aerosol through the Pinatubo period, and quantify the associated perturbation to top of the atmosphere radiative fluxes. The simulations are carried out in the UK Chemistry and Aerosol composition-climate model (UKCA) which extends the high-top (to 80km) version of the HadGEM3 climate model. The HadGEM3-UKCA model uses the GLOMAP-mode aerosol microphysics module coupled with a stratospheric chemistry scheme including sulphur chemistry.

We evaluate simulated size-resolved stratospheric aerosol properties against observations from mid-latitude balloon-borne optical particle counter measurements. We also compare the model to a range of satellite measurements through the Pinatubo period and quantify simulated radiative flux perturbations (RFPs). We carry out multiple perturbed parameter experiments and use Gaussian emulation to quantify the uncertainties in the simulated RFPs to varying several uncertain parameters associated with how the sulphur is injected (magnitude, injection height) and with aerosol model processes (sedimentation, nucleation) and parameters (prescribed standard deviation for accumulation and coarse modes).

Relationship between Ozone Chemical Forcing and Wave Activities for the Period 1990-2011 using the MIROC3.2 nudged CTM

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It is well known that the formation, development and inter-annual variation of the ozone hole are related to the dynamics in winter polar stratosphere. Stratospheric sudden warming was detected in the Southern Hemisphere in 2002 for the first time and then the ozone hole area (defined by the area inside 220DU) was reduced to less than 5 million square kilometers. A similar reduction of ozone hole was also simulated by CCSR/NIES CCM with CCMVal-REF2 scenario, in which the wave number 2 was unusually developed. These suggest a possibility that ozone hole may suddenly be reduced in a specific year by the dynamics in the future, apart from the effect of the decrease in chlorine and bromine concentration in the atmosphere due to the halogen regulation. Thus, in order to speculate ozone hole trend and the variability in the course of the long-term climate change of the future, it is needed to clarify the relationship between wave activity and ozone hole in the past.

Relationships among the ozone hole indices (maximum ozone hole area and minimum total ozone), wave activity and temperature in the Southern Hemisphere were investigated using observation data. The indices were evaluated by the total ozone data of the TOMS Ver8 and OMI Level 3 Data. The meteorological fields used for analysis were ERA-interim and JRA25. Correlation between the indices and the upward component of EP-flux were evaluated for the period 1990-2012, when the ozone hole was developed enough and the effect of wave flux on ozone hole may be easy to detect. EP-flux was calculated for the two components that are dominant over the Southern Hemisphere high latitudes; the wave number 1 stationary component and the wave number 2 transient component, to estimate each wave effect on the year-to-year variation of ozone hole. The analysis shows that a high correlation was obtained between the ozone hole indices (maximum ozone hole area/minmum total ozone) and the EP-flux for stationary wave number 1 in August (-0.68/0.72) and September (-0.61/0.70). On the other hands, for transient wave number 2, a high correlation was obtained only in September (-0.62/0.48). The high correlation between the wave number 2 and ozone hole indices in September may suggest a large mixing of ozone and NOx between the inside and outside of the polar vortex. The high correlation between the wave number 1 and ozone hole indices in August and September may suggest a temperature effect.

In order to clarify each contribution of chemical and dynamical (transport and mixing) processes on the year-to-year variation of ozone hole, a nudged CTM calculation was performed using the MIROC3.2 CCM. The zonal wind velocity, the meridional wind velocity and the temperature were nudged toward ERA-interim data for the period 1979-2011. These meteorological quantities of ERA-interim data are well reproduced in the CTM. The year-to-year variation of ozone hole is also well reproduced. Thus similar correlations between ozone hole indices and the wave flux were obtained. We will discuss quantitative relationships among ozone hole, wave activity and chemical ozone forcing for the two wave components.

Quantifying the radiative impacts of CFCs on past changes in temperature and dynamics in the UTLS using a chemistry-climate model

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Although weak in concentration, halocarbons are potent greenhouse gases (GHGs) because they absorb and emit infrared radiation in the atmospheric window. Although the anthropogenic emission of CO_2 is the dominant contributor to middle tropospheric and surface warming, the fact that CO_2 cools the stratosphere means that CO_2-induced temperature trends in the upper troposphere and lower stratosphere (UTLS) are small and the sign of the trend uncertain. Since halocarbons warm the troposphere and lower stratosphere, their radiative impacts on the UTLS could be as large or larger as that of CO_2. Since tropical tropopause temperature determines the entry value of water vapour into the stratosphere, changes in halocarbons may also be important for stratospheric water vapour and ozone. The role of halocarbons in the UTLS region was discussed by Forster and Joshi (2005), who showed that they have contributed to a warming of the tropical tropopause of ~0.4 K over the past 50 years, dominating the effect of other well-mixed GHGs at those levels. Nevertheless, in spite of their important role in climate, virtually no work has been done to assess the radiative impacts of halocarbons in climate models.

Here we discuss simulations from the Canadian Middle Atmosphere Model (CMAM) that quantify the radiative impacts of halocarbons, in particular chlorofluorocarbons (CFCs), on past changes in temperature and dynamics in the UTLS region. We do this by comparing the radiative effects of CFCs to those of CO_2 and differencing simulations in which the concentrations of those two gases are held fixed in time and transiently evolving. We show that the 250-hPa temperature trends due to CFCs are approximately 40% as large as those due to CO_2 over the past 50 years, which is roughly consistent with estimates of the radiative forcing by halocarbons. In the UTLS region, however, the relative importance of CFCs is substantially larger, with the positive trends in both cold-point temperature and stratospheric water vapour being as large or larger than those due to CO_2. Although the positive trends in tropical upwelling in the lower stratosphere are dominated by the CO_2 changes, trends in *net* upward mass flux are of the same size for both gases. Negative trends in tropical lower stratosphere ozone are larger for CO_2 in accordance with the trends in tropical upwelling.

Forster, P. M., and M. Joshi, The role of halocarbons in the climate change of the troposphere and stratosphere, *Climatic Change*, **71**, 249-266, 2005.

Attribution of Ozone Changes: Nonlinear Interactions between Ozone Depleting Substances and Greenhouse Gases

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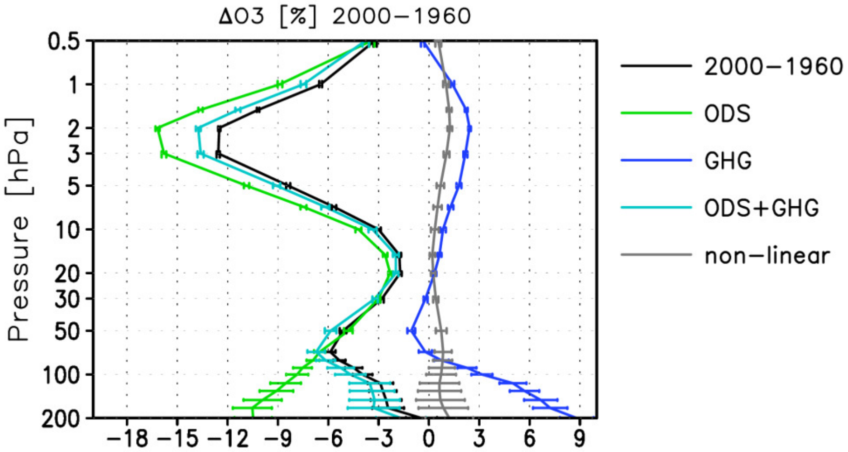
The attribution of ozone changes between 1960 and 2000 to anthropogenic external forcings, i.e. ozone depleting substances (ODSs) and well-mixed greenhouse gases (GHGs), is analyzed in a set of timeslice simulations with the Chemistry-Climate-Model EMAC. The appropriate definition of the sensitivity experiments allows the separation of the effects of ODSs and GHGs on the ozone change between 1960 and 2000 as well as the detection of non-linear interactions.

In former studies the separation of the GHG and ODS effects was often obtained by multiple linear regression based on the timeseries of the stratospheric chlorine burden and CO_2. As the halogen loading of the stratosphere itself is influenced by climate change, the resulting attribution of the ozone change to ODS is slightly imprecise. Furthermore, the increasing concentrations of the GHGs methane (CH_4) and nitrous oxide (N_2O) can modify the ozone chemistry. This is not taken into account if only the CO_2 induced temperature change is considered.

In this study we analyze the impact of the GHG induced stratospheric cooling on the chemical ozone destruction as well as the impact of increasing emissions of CH_4 and N_2O on a low level of atmospheric ODS concentrations. Analogically, the impact of increasing emissions of ODSs is investigated on a low level of GHG concentrations. The combination enables us to estimate the impact of non-linear interactions between increasing ODS and GHG concentrations, such as changes in the tropical upwelling and hence in the transport and the lifetime of ODSs in the stratosphere or the partitioning between radicals and reservoir species. It is found that due to climate change the ODS induced annual mean global mean ozone loss between 1960 and 2000 is significantly reduced in the upper (~1.3%) and lower stratosphere (0.74%) due to the interactions between ODSs and GHGs. The difference between the sum of the single forcings and the combined effect is in the range from -9 to -15 %.



Annual mean global mean relative ozone change between 1960 and 2000 (black) and the contributions from ODSs (green), GHGs (dark blue) and non-linear interactions (gray).

Relationship between PSC types and ozone destruction rate quantified from CALIPSO and MLS data

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The stratospheric ozone destruction in the Arctic has been smaller than that of Antarctic for years. The main cause of this can be attributed to its higher winter minimum temperature of ~10-20 K than that of Antarctic stratosphere owing to topography. The average winter minimum stratospheric temperature in the Arctic is just around the threshold temperature of polar stratospheric cloud (PSC) formation. This results in the appearance of intermittent PSC formation, which is the key factor of severe ozone depletion.

Several types of PSCs are reported. The major ones are nitric acid trihydrate (NAT), supercooled ternary solution (STS), and water ice. However, it still remains unknown whether different types of PSCs have different ability of chlorine activation and ozone destruction efficiency or not. In order to clarify the above question, we made satellite match analysis using CALIPSO and MLS data.

Pitts et al. [2009] and [2011] developed a method to categorize the PSC types from 532 nm backscatter ratio and depolarization data from CALIPSO data. They categorized the PSC types into 6 types; i.e., Mix1, Mix2, Mix2-enhanced, Ice, Wave-ice, and STS. Mix denotes the mixture of NAT and STS. We made satellite match analysis from the location of certain type of PSC categorized by CALIPSO. On the forward and backward trajectories, MLS measurement locations were searched within 150 km and +/- 3 hours difference. As a result, ozone destruction rate was estimated in terms of sunlit hours on the trajectory. We analyzed Antarctic winter/spring in 2007, and Arctic winter/spring in 2010 and 2011. It was found that the ozone destruction rate was the greatest for Mix, followed by STS and ICE PSCs. Hemispheric difference for ozone destruction rate was found.

Unravelling impact factors for past changes of the Brewer-Dobson Circulation from simulations with the CCM EMAC

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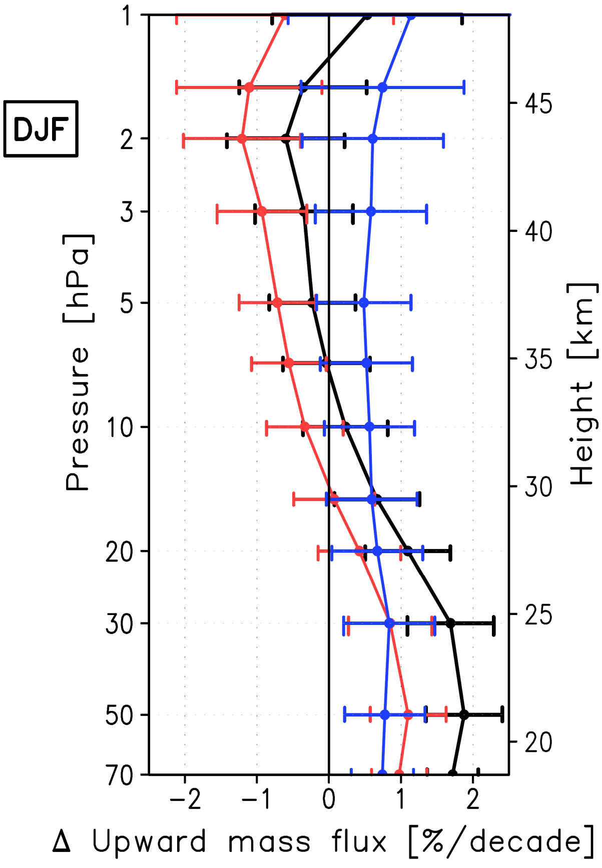
Model simulations indicate an acceleration of the BDC in the past with a direct impact on the exchange of air masses between the troposphere and the stratosphere (e.g., SPARC CCMVal, 2010; Garcia and Randel, 2008; Oman et al., 2009). However, most observational datasets of the recent past do not approve the model results. Bönisch et al. (2010) and Ray et al. (2010) indicate a strengthening of the BDC in its shallow branch and a weakening in its deep branch. In contrast, balloon-borne measurements and Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) satellite observations show no significant acceleration of the BDC at northern hemisphere mid-latitudes (Engel et al, 2009; Stiller et al., 2012).

In our study, the change of the BDC in the recent past is analyzed from sensitivity simulations with the Chemistry-Climate Model (CCM) EMAC, by using different diagnostics, like the tropical upward mass-flux and the mean age of stratospheric air (AoA).



As the sign of change considerably depends on the underlying forcings, namely GHG concentrations and ozone depleting substances (ODS) as well as prescribed sea surface temperatures (SSTs) and sea-ice concentrations (SICs), we separate their role from sensitivity simulations, covering at least 40 years each. The figure shows the changes in the tropical upward mass-flux due to the single forcings in Northern Hemisphere winter season (DJF). It indicates a strengthening of the BDC from 1960 to 2000 (black) in the lower and a weakening in the upper stratosphere with a change in sign around 10 hPa. While rising GHG concentrations (blue) cause a constant increase in tropical upward mass flux of about 1 %/decade, rising ODS concentrations (red) produce a reversal in sign.

The single portions arising from changes in GHGs & SSTs/SICs and ODS and the underlying mechanisms will be presented in this study.

The impact of new estimates of mixing ratio and flux-based halogen scenarios on ozone evolution

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The evolution of ozone in the 21st century has been shown to be mainly impacted by the halogen emissions scenario and predicted changes in the circulation of the stratosphere. New estimates of mixing ratio and flux-based emission scenarios have been produced from the SPARC Lifetime Assessment 2013. Simulations using the Goddard Earth Observing System Chemistry-Climate Model (GEOSCCM) are conducted using this new A1 2014 halogen scenario and compared to ones using the A1 2010 scenario. This updated version of GEOSCCM includes a realistic representation of the Quasi-Biennial Oscillation and improvements related to the break up of the Antarctic polar vortex. We will present results of the ozone evolution over the recent past and 21st century to the A1 2010, A1 2014 mixing ratio, and an A1 2014 flux-based halogen scenario. Implications of the uncertainties in these estimates as well as those from possible circulation changes will be discussed.

Seasonal Ventilation of the Stratosphere: Robust Diagnostics from One-Way Flux Distributions

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We present the first analysis of the seasonally varying ventilation of the stratosphere using one-way flux distributions. Robust transport diagnostics are computed using the state-of-the-art comprehensive GEOSCCM general circulation model subject to fixed present-day climate forcings. From the one-way flux distributions we determine the mass of the stratosphere in any given residence-time interval and stratospheric mean residence times. The seasonalities of the transport masses and mean residence times are quantified with respect to (1) when air enters the stratosphere at the tropical tropopause, (2) when air is in transit in the stratosphere, and (3) when air exits back to the troposphere. We find that the cross-tropopause flux of air back into the northern hemisphere (NH) high-latitude troposphere, with residence times less than three months, is 34% larger for air entering the stratosphere in summer than in winter. For months during late summer to early fall, air with residence times less than 6 months is about six times more likely to be flushed back into the troposphere than for months during winter and early spring. Integrated over all residence times, 51% and 39% of the annually-averaged mass in transit in the stratosphere that entered in the tropics will leave at midlatitudes in the NH and southern hemisphere after residing on average 5 to 6 years in the stratosphere. Most of this mass will eventually exit the stratosphere downstream of the Pacific and Atlantic storm tracks.

The climate response following injection of aerosol into the tropical stratosphere: The role of a well-resolved stratosphere

S. Osprey[1], L. Gray[1], A. Jones[2] and J. Haywood[2]

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Discussions of our response to climate change invariably involve issues of adaptation and mitigation. The former presupposes unavoidable climate consequences and recognises a need to lessen their impact. The latter attempts to lessen the effects of increasing greenhouse gases (GHG) by (1) reducing GHG emission, (2) creating CO2 sinks (e.g. carbon sequestration) or (3) blocking the effects of solar radiation (solar radiation management - SRM).

The SPICE project was set up to investigate the feasibility of implementing a practical method of SRM using the stratospheric injection of aerosols. SPICE remit includes: engineering design for the delivery of stratospheric aerosol, laboratory measurements for characterising the properties of optimal aerosol, and modelling studies looking into the parameterisation and impact of stratospheric aerosols within a state-of-the-art global climate model. The project has also pressed for the need for governance of geoengineering research.

Here we describe idealised experiments investigating the environmental impact following sulphate aerosol injection into the tropical low-mid stratosphere. We compare a climate geoengineering scenario (GeoMIP G4), which includes a constant injection rate of SO2 (S 5Tg/year) beginning at 2020, against a control simulation of increasing greenhouse gas forcing, as outlined by the CMIP5 RCP4.5 scenario. The well-documented stratosphere-resolving Hadley Centre model HadGEM2-CCS is used and compared against the standard HadGEM2-ES configuration, which has also been used in CMIP5 and previous geoengineering studies. Both models have significantly different representations of stratospheric processes, which are crucial for capturing the circulation and dynamical response to large perturbations of stratospheric aerosol.

Global Trends of $CHClF_2$ (HCFC-22) and CCl_3F (CFC-11) estimated from ACE-FTS, HIPPO and WACCM4

Mijeong Park[1], William J. Randel[1], Douglas E. Kinnison[1], Peter F. Bernath[2], Kaley A. Walker[3], Chris D. Boone[4], Elliot L. Atlas[5], Steven A. Montzka[6], and Steven C. Wofsy[7]

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Global trends of $CHClF_2$ (HCFC-22) and CCl_3F (CFC-11) are analyzed using the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) satellite measurements for 2004-2010 periods and compared with the flask sample data obtained during the HIAPER Pole-to-Pole Observations (HIPPO) campaign for 2009-2011. Global distributions and trends of HCFC-22 and CFC-11 are then compared to global chemistry transport model (Whole Atmosphere Community Climate Model, WACCM4) results. Positive and negative trends are observed from the HCFC-22 and CFC-11 distributions in the global upper troposphere, respectively. Measurements from HIPPO and WACCM4 simulations show general agreement with the trends estimated from ACE-FTS. Latitudinal gradients in global trends are revealed both in ACE-FTS and WACCM4. HCFC-22 trends have maxima in the Northern Hemispheric troposphere and tropical lower stratosphere. Trends in CFC-11 show less variability in the troposphere but relatively larger trends in the subtropical lower stratosphere possibly linked to large-scale circulation in the stratosphere. Global seasonal variabilities in WACCM4 HCFC-22 and CFC-11 have general agreement with ACE-FTS.

Simulation of major GHGs and ODSs using chemistry-transport model

P. K. Patra[1], B. Xiang[2], S. C. Wofsy[2], and ACTM, AGAGE, HIPPO and NOAA teams

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We simulated concentration of greenhouse gases (GHGs; e.g., CO_2, CH_4, N_2O, SF_6, HFC-134a) and ozone depleting substances (ODSs; e.g., CCl_4, CFC-11, CFC-12, HCFC-22, HCFC-141b, Halon-1211, Halon-1301, CH_3CCl_3) for the period of 1980-present. The transport and chemistry is calculated online using the CCSR/NIES/FRCGC atmospheric general circulation model (AGCM)-based Chemistry-Transport Model (ACTM) for two distributions of hydroxyl radical (TransCom-CH4 vs. CHASER), transport fields nudged to JRA-25 at horizontal resolutions of ~2.8x2.8 (or ~1.2x1.2) degrees and 67 pressure-sigma layers up to 90 km model top. The GHGs fluxes are either prepared using inversion modeling and/or emission inventories. The ODSs emission distributions are either used a one fixed map or temporally varying maps based on United Nations Environmental Programme (UNEP) country statistics. Net emissions for most species are scaled globally uniformly to reproduce observed growth rates, recorded by the measurement networks of the National Oceanic and Atmospheric Administration (NOAA) and the Advanced Global Atmospheric Gases Experiment (AGAGE) and its predecessors.

Uncertainties in chemistry-transport modeling of long-lived species arise from the parameterization of transport processes, surface fluxes and photochemical loss. We use multispecies measurements with extensive spatial and temporal coverage from five Hiaper Pole-to-Pole Observations (HIPPO) campaigns at regular latitude/longitude intervals along the flight tracks between USA – Arctic Ocean – Central Pacific (all altitudes) – Antarctica – Tropical Equatorial Pacific – USA. Species with unique behaviour have been chosen to characterize ACTM simulations, such as the vertical and latitudinal gradients in (1) SF_6 is used for diagnosing inter-hemispheric transport, (2) N_2O in the upper troposphere for evaluating the timing and strength of stratosphere-troposphere exchange, (3) CH_3CCl_3 and HFC-134a as diagnostic of the oxidizing capacity of the troposphere, i.e., budget of tropospheric OH (Figure 1). Following validation of large-scale model transport and tropospheric OH budget, we find the emission distributions of restricted ODSs prepared using UNEP country statistics are more suitable for ACTM to simulate the observed meridional gradients, compared to the simulation using fixed spatial pattern of emissions. Detailed discussions on emissions, instantaneous and total lifetimes will be presented at the workshop.

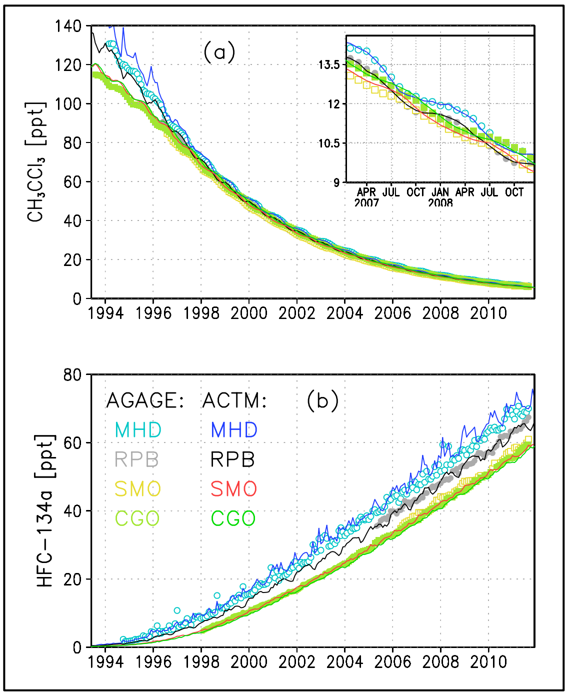

Figure 1: Time series of ACTM simulated and AGAGE measured concentrations of CH_3CCl_3 and HFC-134a at 4 sites, namely, Mace Head (MHD), Republic of Barbados (RPB), Samoa (SMO) and Cape Grim (CGO).

Radiative Forcing of Polar Stratospheric Clouds: Seasonal and Interannual Variability of PSC Optical Depth

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In addition to their critical role in ozone depletion, polar stratospheric clouds (PSCs) can also affect stratospheric radiation and dynamics. Earlier studies indicated that PSCs could significantly affect radiative heating rates, but the magnitude and even the sign of the effect varied greatly from study to study, depending on many factors, e.g. PSC optical depth and underlying tropospheric cloud cover (e.g., Pollack and McKay, 1985; Kinne and Toon, 1990; Rosenfield, 1993). A more recent study by Hicke and Tuck (2001), which assumed nominal PSC conditions of 100% cloud fraction and visible optical depth of 0.01 for non-ice PSCs and 0.04 for ice PSCs, suggested that PSCs could produce significant perturbations to the radiative heating rates in the Antarctic stratosphere. A comprehensive evaluation of the radiative effects of PSCs requires more accurate knowledge of PSC characteristics over the entire polar region and throughout complete seasons. With the advent of the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) mission in 2006, a more complete picture of PSC composition and occurrence is becoming available. The polarization-sensitive CALIOP (Cloud-Aerosol LIdar with Orthogonal Polarization) lidar system onboard the CALIPSO spacecraft is acquiring, on average, over 300,000 backscatter profiles per day at latitudes poleward of 55^o (including the polar night region up to 82^o), providing a unique opportunity to examine the distribution of PSC optical depth on vortex-wide scales and over entire PSC seasons.

In this paper, we describe an approach to calculate PSC optical depth from the CALIOP 532-nm attenuated backscatter measurements. We retrieve the PSC extinction profile downward from cloud top using a composition-dependent extinction-to-backscatter ratio and then integrate the extinction profile to derive PSC optical depth. We then examine this multi-year PSC optical depth record to determine the spatial and seasonal variability for the Arctic and Antarctic, respectively. Multi-year composites provide insight to the interannual variability. The long-term goal of this study is to use this comprehensive PSC optical depth database in combination with the standard CALIOP database of tropospheric cloud optical depth and radiative modeling to provide realistic estimates of the radiative impact of PSCs.

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Stratospheric water vapor in coupled models: assessing the feedback strength

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Tropospheric water vapor feedback is a major feedback to climate change that is largely driven by the increased carrying capacity of water vapor in air as temperature increases. Stratospheric water vapor (SWV) can also vary as climate changes, but from different mechanisms less well understood, such as changes in the tropical cold point temperature, sedimentation of particles in the tropical tropopause region, and transport pathways to the stratosphere. We investigate the magnitude of the SWV climate feedback process using the CMIP (3 and 5) model repositories. A large range of climate feedback strengths due to this process is found in the models, with magnitudes similar to those estimated for the ice-albedo feedback. The mid- and high-latitude lowermost stratosphere is found to be the critical region in controlling the magnitude of the feedback in models. We calculate the SWV radiative effect using a coupled climate model (CESM-WACCM) run with constant sea surface temperatures and the imposed SWV changes found in RCP85 experiments of this model. This isolates the overall effect of SWV changes more accurately than using offline radiative transfer calculations and only stratospheric temperature adjustment (which was used in our earlier work). The implications of these results for the SWV feedback process will be discussed.

Climate Change Scenario over Bangladesh using by High resolution AGCM

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[3]Meteorological Research Institute, Tsukuba, Japan

Abstract

Rainfall and mean surface air temperature were simulated using Meteorological Research Institute (MRI) global 20-km mesh Atmospheric General Circulation Model (AGCM), called MRI-AGCM. Through calibration and validation the MRI-AGCM was adapted for Bangladesh for generating rainfall and temperature scenarios. The model generated rainfall was calibrated with ground-based observed data in Bangladesh during the period of 1979-2006. The Climate Research Unit (CRU) data is also used for understanding of the model performance. Better performance of MRI-AGCM obtained through validation process made confidence in utilizing it in the future rainfall projection for Bangladesh. Rainfall and mean surface air temperature projection for Bangladesh is experimentally obtained for the period of 2075-2099. This work finds that the MRI-AGCM simulated rainfall and temperature are not directly useful in application purpose. However, after validation and calibration, acceptable performance is obtained in estimating annual rainfall and mean surface air temperature in Bangladesh. Change of rainfall is projected about 0.64 % in Monsoon season (JJAS), 1.90 % in Post-monsoon season (ON) and 13.46 % in winter season (DJF) during the period of 2075-2099. Similarly, change of mean surface air temperature is projected about 2.5 degrees Celsius for the same period.

Key words: Rainfall, temperature, forecast, climate change, simulation.

Stratosphere-Ocean Coupling

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Various independent studies reported that the stratosphere and the ocean are coupled on decadal time scales. In particular, the Meridional Overturning Circulation of the North Atlantic is involved in this coupling, but other ocean phenomena like variations in Arctic and Antarctic sea ice coverage, ENSO, or the Pacific Decadal Oscillation may also be affected. Better understanding this co-variability between the stratosphere and the ocean may provide important clues on the dynamics of climate and be relevant for the decadal climate prediction problem. I will give an overview of recent findings from my own and other works and pose new and outstanding questions. I will discuss the different views that exist about the structure and physical mechanisms behind stratosphere-ocean coupling and explain which of the two sub-systems is actually driving which. A related question concerns the nature and origin of the unprecedented stratospheric low-frequency variability in recent observations. I will discuss possible drivers for this variability and show how well it is simulated in models.

How will air pollution and climate change impact global tropospheric ozone in the 21st century?

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While stratospheric ozone is essential for shielding life on Earth from harmful ultraviolet radiation, tropospheric ozone is a greenhouse gas and an air pollutant, but also sustains the oxidative capacity of the atmosphere (through the production of hydroxyl radicals, OH). As well as contributing to air pollution and climate change, global tropospheric ozone is, in turn, also affected by these phenomena. Through the 21st century, tropospheric ozone will be affected by emissions of ozone precursor compounds – nitrogen oxides and volatile organic compounds, which undergo chemical reactions to produce ozone – and by a number of factors related to climate change, such as temperature, humidity, and stratosphere-troposphere exchange events. Examples are the marine troposphere in the tropical Western Pacific, where elevated UV levels and humidity are thought to contribute to a “hole” in the tropospheric ozone and OH columns, or the Amazon Basin, where isoprene emissions lead also to very low ozone and OH columns in the troposphere (Figure 1).

To elucidate the impacts of air pollution and climate change on tropospheric ozone abundances in the 21st century, particularly in the sensitive regions shown in Figure 1, simulations were performed with the SOCOL (Solar Climate Ozone Links) chemistry-climate model. 21 ozone tracers have been introduced into the SOCOL model, which show the transport of ozone within and between the stratosphere and troposphere. In addition, important chemical reactions pertaining to the ozone budget are tracked over latitude, longitude, pressure and time. Coupled together, these diagnostic tools allow a comprehensive investigation into the evolution of the tropospheric ozone layer in a changing climate.

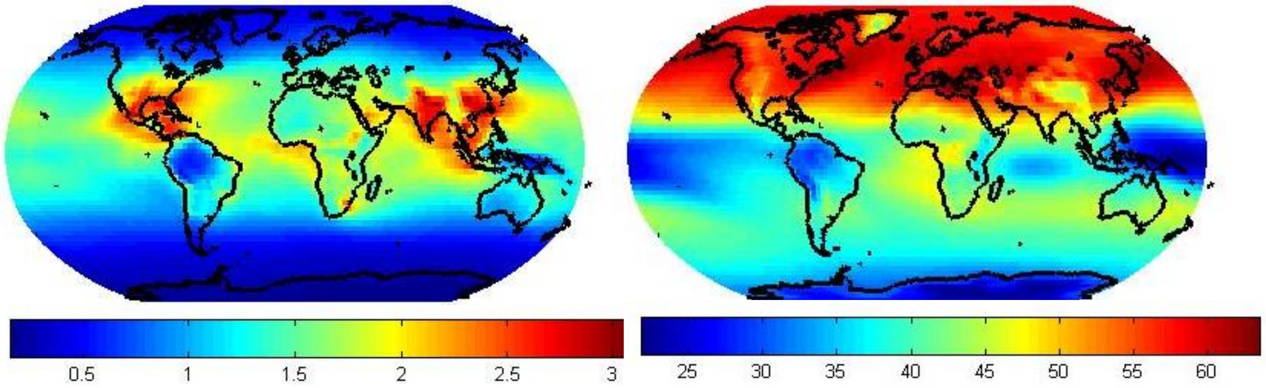

Figure 1. Annual-mean tropospheric OH and ozone simulated by SOCOL under year 2000 conditions (T42 horizontal resolution). Left: Tropospheric column OH (10^{12} molecules cm^{-2}). Right: Tropospheric column ozone (DU).

The SPARC Stratospheric Sulfur and Its Role in Climate (SSiRC) Activity

Markus Rex[1], Stefanie Kremser[2], Larry Thomason[3], Claudia Timmreck[4], and Jean-Paul Vernier[5]

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The stratospheric aerosol layer is a key component of the climate system. It affects the radiative balance of the atmosphere directly through interactions with solar and terrestrial radiation, and indirectly through its effect on stratospheric ozone. Because the stratospheric aerosol layer is prescribed in many climate models and Chemistry-Climate Models (CCMs), model simulations of future atmospheric conditions and climate generally do not account for the interaction between the sulfur cycle and changes in the climate system. The current understanding of how the stratospheric aerosol layer may be affected by future climate change and how the stratospheric aerosol layer may drive climate change is, therefore, very limited. To facilitate improved understanding of relevant processes and to better understand the role of stratospheric sulfur for climate change, the WCRP's (World Climate Research Programme) SPARC (Stratospheric Processes and their Role in Climate) has organized a new activity entitled Stratospheric Sulfur and Its Role in Climate (SSiRC). The scientific and programmatic goals of SSiRC include: (i) providing a coordinating structure for the various individual activities already underway in different research centres; (ii) encouraging and supporting new instrumentation and measurements of sulfur containing compounds, such as COS, DMS, and non-volcanic SO_2 in the UT/LS globally; and (iii) initiating new model/data inter-comparisons. The presentation will discuss on-going activities and key results from the SSiRC workshop (October 2013).

Is There a Hole in the Global OH Shield Over the Tropical Western Pacific Warm Pool?

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[3]California Institute of Technology, Pasadena, CA, USA
[4]IFM-GEOMAR, Kiel, Germany

Hundreds of organic species are emitted into the atmosphere mostly from biogenic processes. The rapid breakdown by reactions with OH radicals prevents most of them from reaching the stratosphere. Hence, the omnipresent layer of OH in the troposphere shields the stratosphere from these emissions and is particularly relevant for those species that do not photolyse efficiently. A strong source of OH in clean tropical air are reactions involving ozone. Hence the OH concentration is closely coupled to ozone abundances.

The Western Pacific warm pool is key for troposphere to stratosphere exchange. We report measurements of 14 ozonesondes launched during the Transbrom ship cruise through the center of the warm pool in October 2009. During a 2500km portion of the ship track between 10S and 15N we found ozone concentrations below the detection limit of the sondes throughout the troposphere. Based on modelling and measurements of OH on the ER-2 during the STRAT campaign we suggest that there also is a pronounced minimum in the tropospheric columns of OH over the West Pacific.

We show that this increases the lifetime of chemical species and amplifies the impact of surface emissions on the stratospheric composition. Specifically, emissions of biogenic halogenated species from natural sources and from kelp and seaweed farming can have a larger effect on stratospheric ozone depletion. Also, increasing anthropogenic emissions of SO2 in South East Asia or from minor volcanic eruptions can play a larger role for the stratospheric aerosol budget, a key element for explaining the recently observed decrease in global warming rates.

Variability of Tropospheric Methane above the Mediterranean Basin inferred from Satellite and Model Data

P. Ricaud[1], B. Sic[1], L. El Amraoui[1], J.-L. Attié[1,2], B. Josse[1], V. Marécal[1], P. Huszar[1,3], J. Parmentier[1], N. Jaidan[1], D. Saint-Martin[1], M. Michou[1], R. Abida[1], R. Zbinden[2], D. Hauglustaine[4], S. Szopa[4], F. Carminati[1,5] and J. Warner[5]

[1]Météo-France/CNRS, Toulouse, France; [2]Laboratoire d'Aérologie/CNRS, Toulouse, France; [3]Charles University, Prague, Czech Republic; [4]LSCE/CNRS, Saclay, France ; [5]University of Maryland, College Park, Maryland, USA

The CHARMEX (Chemistry and Aerosol Mediterranean Experiment) Project is the atmospheric chemistry component of a large multidisciplinary Mediterranean regional programme is being carried out by French laboratories together with other international partners. It intends, among other things, to quantify processes explaining the temporal evolution of chemical compounds and aerosols in the troposphere above the Mediterranean Basin (MB). The Work Package "Variabilities, Recent and Future Trends" uses a large set of data (satellite, ground-based, sondes, aircraft, models and assimilation) to 1) estimate the variabilities and recent trends of several species (e.g. O_3, CO, N_2O, etc.) and aerosols, 2) evaluate the synoptic-scale circulation that control their transport, and 3) estimate the future chemical climate over the MB by 2100. The present study will focus on the analysis of the variability of methane (CH_4) from the lowermost to the uppermost troposphere as observed by satellites (AIRS, IASI and GOSAT) from 2008 to 2011 and as calculated by models (MOCAGE, CNRM-CCM and LMDz-ORINCA) from 2000 to 2010. The impact of the Asian Monsoon Anticyclone onto the CH_4 fields in the Eastern MB in summer will be highlighted.

Chemical Climate Evolution above the Mediterranean Basin

P. Ricaud[1], N. Jaidan[1], P. Huszar[1,2], D. Saint-Martin[1], M. Michou[1], L. El Amraoui[1], J.-L. Attié[1,3], B. Josse[1], V. Marécal[1], R. Abida[1], R. Zbinden[3], D. Hauglustaine[4], S. Szopa[4] and the ACCMIP modellers

[1]Météo-France/CNRS, Toulouse, France; [2]Charles University, Prague, Czech Republic; [3]Laboratoire d'Aérologie/CNRS, Toulouse, France; [4]LSCE/CNRS, Saclay, France

The CHARMEX (Chemistry and Aerosol Mediterranean Experiment) Project is the atmospheric chemistry component of a large multidisciplinary Mediterranean regional programme is being carried out by French laboratories together with other international partners. It intends, among other things, to quantify processes explaining the temporal evolution of chemical compounds and aerosols in the troposphere above the Mediterranean Basin (MB). The Work Package "Variabilities, Recent and Future Trends" uses a large set of data (satellite, ground-based, sondes, aircraft, models and assimilation) to 1) estimate the variabilities and recent trends of several species (e.g. O_3, CO, N_2O, etc.) and aerosols, 2) evaluate the synoptic-scale circulation that control their transport, and 3) estimate the future chemical climate over the MB by 2100. The present study will focus on the analysis of the model results from the ACCMIP project concentrating on the time evolution of O_3, CO, NO_2, CH_4 and CO_2 from the surface to the upper troposphere above the Mediterranean Basin from 2000 to 2100 according to different emission scenarios.

On the relationship between age-of-air changes and changes in residual circulation and eddy mixing

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Changes of the Brewer-Dobson circulation, observed in terms of Age-of-Air (AoA), appear to contradict model simulations that predict an acceleration of the residual circulation for increased levels of greenhouse gases (*Engel et al., 2009*). There is, however, increasing evidence that wave-driven eddy mixing effects must be included in the interpretation of observations.
We analyze AoA variations in terms of the underlying dynamic and transport processes, based on a multi-annual simulation with the Lagrangian transport model CLaMS driven by ERA-Interim meteorological data. CLaMS realistically simulates transport and mixing of tracers in the lower stratosphere. Simulated AoA changes for the time period from 2002 to 2011 reveal a complicated and inhomogeneous pattern, reflecting some important structures observed by MIPAS-Envisat (*Stiller et al., 2011*). The forcing of AoA changes is further analyzed within the isentropic zonal mean framework (similar to the Tranformed Eulerian Mean framework), which allows for separation of the effects of residual circulation and wave-driven eddy mixing. The time-mean effect of eddy mixing causes an increase of AoA in the tropics and subtropics equatorwards of about 40°, and a decrease of AoA at higher latitudes. We find a large degree of cancellation between the effects related to the residual circulation and wave-driven eddy mixing, with age-of-air variations turning out to be small residuals in a delicate balance between both effects. This implies that there is no strong relation between the acceleration of the residual mean circulation and changes of AoA for relatively short time scales (~10yr). In particular, the stratospheric region around 40° latitude turns out problematic for interpreting AoA changes, as the effects of residual circulation and eddy mixing both reverse sign around this latitude.

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Diurnal variations in stratospheric O_3 and HCl as observed by the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES)

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Considerable uncertainties remain in the global pattern of diurnal variation in stratospheric O_3, particularly lower to middle stratospheric O_3, which is the principal contributor to total column O_3. The Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) attached to the Japanese Experiment Module (JEM) on board the International Space Station (ISS), was developed to gather high-quality global measurements of stratospheric ozone at various local times. Using the SMILES dataset, as well as data from nudged chemistry-climate models (MIROC3.2-CTM and SD-WACCM), we show that the SMILES observational data have revealed the global pattern of diurnal O_3 variations throughout the stratosphere. We also found that these variations can be explained by both photochemistry and dynamics (i.e., vertical transport of background O_3 by tidal vertical wind). The peak-to-peak difference in the stratospheric ozone mixing ratio (total column ozone) reached 8% (1%) over the course of a day.

These diurnal variations need to be considered for (1) trend analysis and for (2) bias correction. For (1), we note that long-term trends of vertical profile of O_3 were estimated using merged data set from different satellite measurements at various local times (e.g., SBUV(/2)). The potential impact of diurnal variations on such analysis will be estimated using the present SMILES results. For (2), it was pointed out that SAGE II sunrise and sunset O_3 measurements differed from each other by up to 10% between 35—55 km (Kyrola et al., 2013). Our results indicate that the bias may be largely explained by diurnal variations.

In addition, the importance of tidal vertical wind on variations in atmospheric minor constituents indicates that long-lived species such as O_3 (in the lower stratosphere) and HCl can be, in turn, used to estimate the tidal vertical wind whose amplitude is too small (1 mm s^{-1}) to be directly measured. We thus estimated the tidal vertical in the lower stratosphere, using HCl as well as O_3 from SMILES measurements. It is found that the estimated magnitude shows a roughly good agreement with that from reanalysis data.

High water vapor and associated signatures from MLS in the mid-latitude summer lowermost stratosphere: Implications for posited ozone destruction

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Aura Microwave Limb Sounder 100-hPa, 83-hPa and 68-hPa H_2O measurements show that the Asian monsoon anticyclone (AMA) and North American monsoon anticyclone (NAMA) are unusually wet, both in mean values and in outliers reaching 18 ppmv. The 9-year MLS record, with associated constituent measurements, provides context for the report by Anderson, et al. (A2012) of high values of convectively-injected water vapour (H_2O) from in situ measurements in the North American (NA) summer lowermost stratosphere (LMS). A2012 contends that in such cold/wet conditions, heterogeneous chemistry on binary water-sulfate aerosols will activate chlorine, leading to catalytic ozone destruction. In this work, we examine the statistics of extreme AMA and NAMA H_2O observations within the global context of the MLS record. We discuss the implications of MLS vertical resolution on observation of ~1-km layers of enhanced H_2O such as are reported in A2012, and discuss the usefulness of MLS hydrochloric acid (HCl), ozone (O_3) and chlorine monoxide (ClO) measurements in putting observational limits on chlorine activation and ozone loss. Monsoon LMS parcels that are cold enough ($T < \sim 205$ K) to be candidates for aerosol growth and chlorine activation typically have much less chlorine, in the form of HCl, to activate and much less O_3 to destroy than is used in A2012 as model initial conditions, regardless of whether they are wetter or drier than A2012's activation threshold. Parcels that are wet enough for activation will have been diluted with tropospheric air, and so will have lower O_3 and HCl than dry parcels of similar temperature. While cooling of the stratosphere with climate change may lead to greater chlorine activation and ozone loss in the future, A2012 almost certainly overstates the potential for chemical chlorine loss in the NA summer LMS under current climate conditions.

Detection of Climate Engineering Activities by the Global Observing System Is Limited by Earth System Variability

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Proposals for engineering the climate system to counteract anthropogenic climate change are gaining momentum. A growing body of literature addresses scientific, technical, environmental, ethical, legal and governance issues. However, it largely ignores the question: "Could the global observing system detect either a planned, publicized or an independent, undisclosed climate engineering effort?" The answer depends on: (1) high-precision, globally-complete, long-term observations, and (2) the background variability of the climate system.

For proposals to increase solar radiation reflection (albedo), a decade-long observational record, combined with low persistence and no trend in albedo, facilitate detection of large interventions. But for proposals to reduce atmospheric CO_2, detection is much more challenging due to sparse sampling of the observing system and to long-term trends and interannual and decadal variability. These findings have implications for the climate observing system for future detection of climate engineering, as well as for the broader climate engineering debate.

This study uses standard statistical tests and existing observational records to estimate detection limits for hypothetical and idealized (abrupt, sustained) albedo interventions (i.e., “solar radiation management”). Detection is easier for larger regions with low climate variability and for abrupt (vs. gradual) interventions, given a sufficiently long observational record before and after intervention. At the global scale, with a 5-year prior record, abrupt albedo increases $< \sim 0.002$ are unlikely (95% confidence) to be detected within a year. A 0.002 change is comparable to ~ 0.7 Wm^{-2} reduction in radiative forcing, exceeds year-to-year global albedo variability by a factor of ~3, and exceeds the hoped-for albedo increase for most proposed schemes.

The spatial pattern of detection limits at 1º latitude × 1º longitude ($\sim 10^{10}$ m^2) resolution reflects the pattern of interannual albedo variability and ranges over an order of magnitude from 0.01 (e.g., northern Africa) to 0.10 (e.g., the equatorial Pacific), highlighting the sensitivity of detection to the choice of test location. Although albedo increases in these small regions may be detectable, they would change global-average albedo by only ~0.000001, three orders of magnitude less than the global detection limit.

ENSO variability and future change of ozone in troposphere and lower stratosphere

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Ozone is one of important greenhouse gases as well as atmospheric pollutant and UV shield. This study is concerned with its inter-annual variability and long-term change. A dominant driver of ozone variability on inter-annual scale is the El Niño Southern Oscillation (ENSO). Oman et al. (2013) examined ozone variability associated with ENSO for a test of chemistry-climate model (CCM). Long-term change in ozone can be influenced by emission of ozone precursors and depletion substances as well as climate change. The change was investigated under model inter-comparison projects (e.g. Young et al., 2013). The modelling studies generally used CCMs with about 300 km horizontal resolution. However, it is quite unclear whether such CCM resolution appropriately simulates the ozone variability and change. This study evaluates ENSO-related ozone variability during 2004–2009 using the MIROC-ESM-CHEM model with two different horizontal resolutions (about 300 km and 120 km). We also investigate future change in ozone between 2000s and 2100s and compare simulations of the two resolutions. As for the sensitivity to horizontal resolution, this study particularly focuses on impacts on ozone transport process, employing an O_3-tracer-transport (tagged tracer) model version. Our result shows that there are no crucial differences in ENSO-related ozone variability between the two resolution simulations. However, in fact, high-resolution model (HM) shows larger anomalies than med-resolution model (MM) by a factor of five. The anomalies in HM agree with the observations better than those in MM. Future changes of ozone in the two simulations under RCP6.0 scenario are little different except in the lower stratosphere of northern hemisphere, where HM shows about two times larger increase in ozone than the change simulated by MM. To investigate process of future ozone change, we also quantify individual impacts of transport and chemistry using a sensitivity simulation with 2100s climate and 2000s chemical field. The simulation indicates that future change in ozone is largely driven by chemistry process in the lower stratosphere. Transport causes increase in tropospheric ozone burden, which cancels out 23% of the decrease due to chemistry.

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Direct and Indirect Effects of Solar Variations on Stratospheric Ozone and Temperature

Lin Shang

Abstract: We have used a fully coupled chemistry-climate model (WACCM) to investigate the relative importance of the direct and indirect effects of 11a solar variations on stratospheric temperature and ozone. Although the model does not contain a quasi-biennial oscillation (QBO) and uses fixed sea surface temperature (SST), it is able to produce a second maximum solar response in tropical lower stratospheric (TLS) temperature and ozone of approximately 0.5 K and 3%, respectively. In the TLS, the solar spectral variations in the chemistry scheme play a more important role than solar spectral variations in the radiation scheme in generating temperature and ozone responses. The chemistry effect of solar variations causes significant changes in the Brewer-Dobson (BD) circulation resulting in ozone anomalies in the TLS. The model simulations also show a negative feedback in the upper stratosphere between the temperature and ozone responses. A wavelet analysis of the modeled ozone and temperature time series reveals that the maximum solar responses in ozone and temperature caused by both chemical and radiative effects occur at different altitudes in the upper stratosphere. The analysis also confirms that both the direct radiative and indirect ozone feedback effects are important in generating a solar response in the upper stratospheric temperatures, although the solar spectral variations in the chemistry scheme give the largest solar cycle power in the upper stratospheric temperature.

Key words: solar cycle, stratospheric ozone, stratospheric temperature, chemistry, radiation

Motivation and importance: The main focus is to quantify the effect of ozone feedbacks to 11-year solar variations and we attempt to distinguish the relative importance of spectral variations of the radiation scheme and spectral variations of the chemistry scheme in generating solar responses in stratospheric temperatures and ozone.

Impact of Coagulation Efficiency in Simulations of Mt Pinatubo Eruption Using a Coupled Sulfate Aerosol-Chemistry-Climate-Model

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The eruption of Mt. Pinatubo in June 1991 emitted 7–11 Mt sulfur into the lower stratosphere. It is an important analogue and benchmark for geoengineering. However, current modeling approaches are unsuccessful to accurately predict the evolution of the stratospheric aerosol burden. Most modeling attempts yield shorter residence times of the volcanic aerosols in the stratosphere compared to satellite observations. In this study, we coupled a sulfate aerosol module to the global chemistry-climate model (CCM) SOCOL. The aerosol module AER (Weisenstein et al., 1997; SPARC, 2006) includes comprehensive sulfur chemistry and microphysics, in which the particles are size-resolved by 40 size bins spanning radii from 0.39 nm to 3.2 um. Radiative forcing is calculated online from the aerosol module according to Mie theory. We put particular emphasis on investigating the aerosol particle coagulation efficiency described by a Lennard-Jones potential (Narsimhan and Ruckenstein, 1985) in the free molecular regime, where very low efficiency values may prevail. The results show that after an eruption the simulated stratospheric aerosol burden with the reduced coagulation efficiency values agrees well with the high-resolution infrared radiation sounder (HIRS) measurements and Laramie in situ measurements. Although the coagulation efficiency is largely uncertain, this suggests that the sedimenting large particles form more slowly due to the reduced coagulation process under stratospheric conditions, and therefore stratospheric aerosols have a longer residence time than derived in previous studies. This study represents the first steps to properly predict large volcanic eruptions and also potential geoengineering measures against dangerous climate change. Further efforts will be necessary to narrow the uncertainty of the coagulation efficiency, particularly under stratospheric conditions.

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Global Sulfur Budget and Sensitivity Studies of Anthropogenic SO_2 Emissions: Results from a Coupled Sulfate Aerosol-Chemistry-Climate-Model

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The stratospheric aerosol layer affects the Earth's radiation balance and plays a significant role in the global climate change. However, uncertainties of current modeling studies are large in part because of model inadequacies, e.g. unresolved particles size distributions or lack of radiative coupling between the aerosol module and an underlying chemistry-climate model (CCM). In this study, we coupled the AER (Weisenstein et al., 1997; SPARC, 2006) stratospheric sulfate aerosol module into our global CCM SOCOL. The aerosol module comprises sulfur chemistry in both gas and aqueous phases, as well as comprehensive microphysics, including homogeneous nucleation, condensation, evaporation, coagulation and sedimentation. The particle size distribution is resolved by 40 size bins spanning radii from 0.39 nm to 3.2 um and the size-dependence of the particle concentration (weight percent of sulfuric acid) is fully taken into account. Radiative forcing input data required by the CCM SOCOL are calculated online from the aerosol module according to Mie theory. In order to simulate the global sulfur budget for volcanically quiescent conditions, we apply emission flux boundary conditions of the year 1990. Model results for the aerosol burden in the Junge layer compare well with measurements and earlier model simulations reported in SPARC (2006). Sensitivity studies are conducted for the present condition by changing anthropogenic SO_2 emissions in East Asia and India. The results suggest that anthropogenic SO_2 emissions in East Asia and India may influence stratospheric aerosols more pronouncedly compared to emission changes in Europe and USA. This indicates that the deep convection in the Western Pacific region might magnify the impact of sulfur emissions in the tropics and provide a significant portion to the stratospheric aerosol burden in addition to carbonyl sulfide (OCS) emissions and SO_2 emissions from small volcanic eruptions. This study improves our knowledge about future changes and impacts of stratospheric aerosols during volcanic quiescent periods.

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Middle atmospheric sciences using data from the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES)

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The Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) aboard the Japanese Experiment Module (JEM) of the International Space Station (ISS) made atmospheric measurements of minor species in the stratosphere and mesosphere for about six months from October 2009 to April 2010. High-sensitivity measurements of SMILES had been performed by a receiver using superconductor- insulator-superconductor (SIS) mixers, cooled to 4.5 K by a compact mechanical cryocooler. Mission objectives are: i) Space demonstration of 4-K mechanical cooler and super-conductive mixer for the submillimeter limb-emission sounding in the frequency bands of 624.32- 627.32 GHz and 649.12- 650.32 GHz, and ii) global measurements with its high sensitivity for atmospheric minor constituents in the middle atmosphere (O_3, HCl, ClO, HO_2, HOCl, BrO, O_3 isotopes, HNO_3, CH_3CN, etc), contributing to the atmospheric sciences. Thus global and vertical distributions of about ten atmospheric minor constituents related to the ozone chemistry are derived. See Kikuchi et al. (2010) in more detail about the SMILES mission.

In this presentation, we will introduce an overview of the SMILES measurements and show some observational results in association with middle atmospheric chemistry and dynamics. To support the SMILES observational results, we also used outputs from nudged chemistry-climate models (MIROC3.2-CTM and SD-WACCM) in a complementary way. We first performed several validation studies to evaluate the SMILES performance for the target minor species such as O_3, ClO, and HCl, and found the results are reasonable (e.g., Imai et al, JGR, 2013). As to useful scientific usages of the SMILES data, one of the most unique characteristics of the SMILES measurements is that the data from SMILES can be used to capture the diurnal variation of atmospheric minor constituents such as O_3, ClO, HO_2 and BrO, since the ISS took the non-sun-synchronous orbit. For ozone we clearly found the diurnal variations in the stratosphere (Sakazaki et al., JGR, 2013) and in the mesosphere (Smith et al., JGR, 2013). These results demonstrate that the SMILES high sensitivity measurements are expected to provide further insights into atmospheric chemistry and dynamics.

Lower stratospheric correlation between O_3 and HCl as observed by SMILES in the southern high latitudes

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Ozone (O_3) in the upper troposphere (UT) has an effect on radiative forcing. One of poorly constrained source of tropospheric O_3 is the stratosphere. Influx of O_3 from the stratosphere has been estimated as 550±140 Tg O_3 a year on the basis of aircraft observations (Olsen et al., 2001). The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) has inferred it as a range of 401-663 Tg/yr (Young et al., 2013). Marcy et al. (2004) have suggested that measurements of HCl in the UT can be used to calculate how much O_3 was transported from the lower stratosphere (LS). Using the correlation between O_3 and HCl in the LS, a fraction of the source of the stratosphere has been quantified in the UT air. Here, we will present the lower stratospheric correlation between O_3 and HCl or the O_3/HCl ratio as observed by the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) (Kikuchi et al., 2010). We focus on the high-latitudes between 50°S and 66°S (the limit of the SMILES measurement) and time periods of November (in 2009), February, and April (in 2010), when SMILES mainly covered the Southern Hemisphere (S.H.). We have analyzed the version 2.4 of Japan Aerospace Exploration Agency (JAXA)'s Level 2 data (the latest available, to date, from http://smiles.tksc.jaxa.jp) The preliminary results have revealed that the O_3/HCl ratios at 450 K level in potential temperature outside the polar vortex in Nov. (1500-2000) are higher than those at 420-430 K level in Feb. and Apr. (no polar vortex). The lower values in Feb. and Apr. are likely to be due to mixing of air inside the vortex, where the ratio is very low (< 500), and outside the vortex. This result suggests that the seasonal variation of the O_3/HCl correlation in the S.H. should be taken care for evaluating the contribution of stratospheric O_3 in the UT. We will also examine the stratospheric fraction in the UT at lower latitudes (< 50°S) where the air is transported from the LS at the above high-latitudes.

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Representation of the solar signal by radiation codes of the ECHAM family

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Solar radiation is the main input source of energy to the Earth’s atmosphere and in many respects defines its composition, photochemistry, temperature profile and dynamics. Variations in solar irradiance affect different spectral bands differently, and recent observations show that variability in the UV spectral bands is particularly high and can be several times higher than previously expected. But model studies of the impact of the spectral solar irradiance variability fail to show a clear response in middle stratospheric temperature and composition, likely due to the radiation codes implemented in the models. Accurate methods of solving radiative transfer equation are too computationally expensive, therefore different methods were designed to provide a consensus between accuracy and efficiency. In this work three generations of ECHAM (4, 5 and 6) radiation schemes are investigated, and improvements of the ECHAM5 solar radiation code are presented using available parameterizations of the heating rate due to absorption by O2 and O3. Offline calculations results are compared to the high resolution model libRadtran as reference, revealing a much better representation of the heating rates in the mesosphere due to implemented O2 absorption and better response of heating rates to solar irradiance variability due to implemented O3 absorption without significant increase of the computation time.

Validation of the photolysis rate response to the solar irradiance variability

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Substantial differences between the reconstructed and measured by SIM and SOLTICE instruments spectral solar irradiance (SSI) evolution during the declining phase of 23rd solar cycle have substantial implications for the modeling of the middle atmosphere state. Different magnitude and spectral composition of the SSI changes leads not only to a substantial alteration of the heating rates but also affects photolysis rates which drive atmospheric chemistry and regulate the ozone distribution in the atmosphere. The global ozone abundance is maintained by the ozone production, destruction and transport by air motions, but in the tropical stratosphere above ~30 km the ozone concentration depends primarily on the photochemical processes. The ozone can be produced here only through the photolysis of the molecular oxygen, therefore the oxygen photolysis plays a crucial role in the atmospheric chemistry. The spectral composition and the magnitude of the SSI changes are very important because they define not only the magnitude but also the sign of the direct ozone response. In order to estimate the response of the most important photolysis rates to the difference in the available SSI data sets we have calculated JO2, JO3, JO3*, and JNO2 for August 2004 and 2008 using the SSI data obtained from two different reconstructions NRL, COSI and two composites based on SORCE data. Using libRadtran software as reference we performed evaluation of the ability of two methods being used in atmospheric models for photolysis rates calculation (common used “lookup” table method and on-line method based on Fast JX code) to follow SSI changes.

VSLS Bry Estimation from JEM/SMILES BrO Observation

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Very Short Lived Source (VSLS) gases of Bry have been investigated, since it affects the chemistry of the stratosphere such as the long term recovery of the Ozone layer. VSLS Bry can be estimated from the difference of observed and model BrO value and the Bry/BrO calculated by the model. But previous VSLS Bry estimation spread over 4-8 ppt, due to the large uncertainty of the BrO observation. In this paper, we will report the SMILES (Superconducting Submillimeter-wave Limb Emission Sounder) BrO observation, and VSLS Bry estimation using SMILES L2 and Specified Dynamics WACCM Model (nudged to GEOS-5 data) calculation.
SMILES operated on the ISS from Oct. 12, 2009 to Apr. 21, 2010. SMILES measured BrO in the bands A and C. Both Band A and C L2 BrO showed strong systematic bias due to interference of the overlapping species (Fig. 1), but it can be corrected using night time mean value since BrO should be 0 below 37 km at night. After the bias correction, both band A and C BrO can be usable for scientific applications.

The VSLS Bry can be estimated, $Bry_{VSLS} = (Bry_{waccm})/(BrO_{waccm})\bullet(BrO_{smiles}) - Bry_{waccm}$, where SD-WACCM included no VSLS Bry for this study. SMILES zonal mean BrO has significant standard deviation, for example, 4.3 pptv to the mean value 10.7 ppt at the 31 km in the equatorial region during Jan. 10-20, but its standard error is enough small 0.4 ppt die to the large number SMILES observation, and the estimated VSLS Bry is ~4 pptv (Fig. 2).

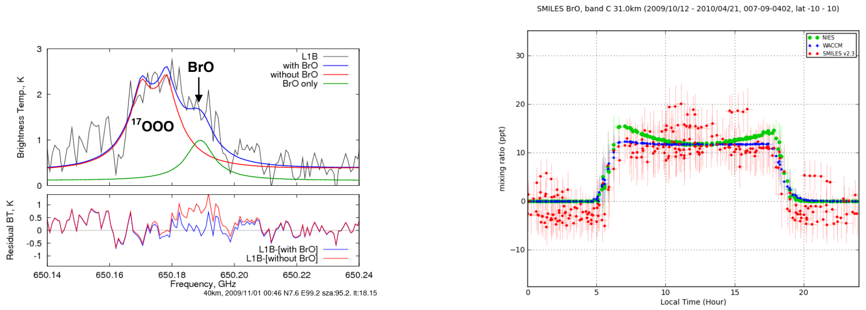

Fig. 1 Band C BrO spectral fitting at 40 km and Diurnal variation of Band C BrO at 31 km.

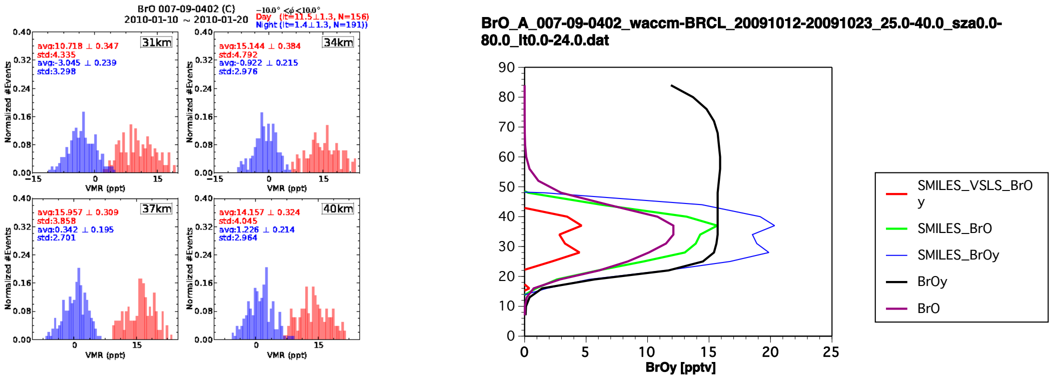

Fig. 2 Day-Night Histogram of SMILES Band C BrO at 31-40 km and the estimated VSLS Bry from Band A BrO at 25N-40N in Oct. 12-23, 2009.

The role of oceanic halogen and sulfur compounds for the middle atmosphere

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The decline of anthropogenic chlorine in the stratosphere within the 21st century will increase the relative importance of naturally emitted, halogenated very short lived substances (VSLS) from the ocean on stratospheric ozone destruction. Halogenated VSLS play a significant role in present day ozone depletion, in particular in combination with enhanced stratospheric sulfate aerosol loading. There is a need to better understand how much of the observed stratospheric halogen and sulfur aerosol originates from natural sources, in particular from oceanic emissions, and how this will change and affect the middle atmosphere in a future climate.

In this study we present current and future emission scenarios of brominated and iodinated halocarbons and their contribution to the stratospheric halogen loading based on transport calculations with the Lagrangian particle dispersion model FLEXPART. Following the same approach we will use dimethylsulphide (DMS) emission scenarios to assess the impact of naturally emitted sulfur on the stratospheric aerosol loading. In addition to current state-of-the-art determined emission maps based on parameterized flux calculations, we apply the newly developed inversion framework FLEXINVERT to derive optimized fluxes from given atmospheric observations. This combined approach will advance our understanding of oceanic VSLS and sulfur emissions and their temporal and spatial variability. We will assess the impact of identified highly localized sources, in particular in the tropical West Pacific, which are shown to be correlated with events of very efficient transport from the surface to the cold point tropopause. Finally, the ozone-depletion potential (ODP) weighted emissions of VSLS will be compared to those of other ozone depleting substances to give an estimate of the overall impact of VSLS on the ozone layer for current day conditions and future scenarios.

FinROSE chemistry transport model simulations of the variability and trends of water vapour in the Arctic stratosphere

L. Thölix, L. Backman, R. Kivi and A. Karpechko

Water vapour is a chemically and radiatively important trace gas in the upper troposphere-lower stratosphere (UTLS). Its quantity modifies the stratospheric radiative balance, and is an important driver of climate change. Stratospheric water vapour also affects stratospheric ozone through odd-hydrogen chemistry and by formation of polar stratospheric clouds (PSCs) leading to ozone depletion. The main sources of stratospheric water vapour are intrusion from the troposphere via thetropical tropopause and production through oxidation of methane in the upper stratosphere.

The observational data of UTLS water vapour is relatively sparse. There is a very steep gradient in the water vapour profile over the tropopause and the stratosphere is relatively dry, which makes accurate observations difficult to obtain. Modelling the extratropical stratospheric water vapour is also challenging, e.g. due to temperature dependent processes (dehydration) in the tropical tropopause. Accurate measurements of stratospheric water vapour have been made above Sodankylä, in northern Finland, since early 2000s using balloon borne research grade hygrometers.

The Arctic polar vortex in the 2009/2010 winter was very cold and polar stratospheric clouds were formed. The cold pool in the stratosphere over Northern polar latitudes was large and stable and for the first time large scale persistent dehydration was observed in the Arctic stratosphere. During the LAPBIAT atmospheric sounding campaign in Sodankylä in January 2010 accurate stratospheric water vapour profiles were measured. The water vapour profiles indicated formation of polar stratospheric ice clouds and dehydration. In this study chemistry transport model simulations covering several Arctic winters, including 2009/2010, were performed with FinROSE-ctm using ERA-Interim winds and temperatures. Dehydration events and trends in Arctic stratospheric water vapour were analysed based on observations and model results.

The FinROSE-ctm is a global middle atmosphere model that produces the distribution of 30 long-lived species and tracers and 14 short-lived species. The chemistry describes around 110 gas phase reactions, 37 photodissociation processes and the main heterogeneous reactions related to aerosols and polar stratospheric clouds.

The dynamical response to volcanic aerosol: climate model sensitivity to prescribed volcanic forcing set

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Stratospheric sulfate aerosols produced by major volcanic eruptions modify the properties of the atmosphere through their reflection of solar radiation and absorption of infrared radiation. The impact of volcanic aerosol heating in the lower stratosphere on the polar vortex is a key part of theories connecting volcanic aerosol with changes in surface dynamics which give rise to changes in climate indices such as the North Atlantic Oscillation and Northern Annular Mode, and the “winter warming” phenomenon, which has been observed for many eruptions of the past. However, climate model simulations of the historical period, like those in the CMIP3 and CMIP5 projects, which are forced with prescribed volcanic aerosol data sets, generally do not capture this observed dynamical response to volcanic aerosol. We investigate this issue by performing a sensitivity study using the CMIP5 model MPI-ESM and performing ensemble simulations of the 1991 Pinatubo eruption using four different prescribed volcanic aerosol forcing sets. Two forcing sets are based on satellite observations of Pinatubo aerosol: the Stenchikov et al. (1998) data set, which was used in MPI-ESM CMIP5 historical simulations, and the Chemistry-Climate Modeling Initiative data set based on updated retrievals of the satellite measurements. Two other aerosol forcing sets are products of independent simulations with the coupled aerosol-climate model MAECHAM5-HAM. By assessing the modeled response to each of the prescribed aerosol forcing sets, we aim to identify the important mechanisms connecting volcanic aerosols and atmospheric dynamics, and assess the sensitivity of these mechanisms to variations in the prescribed volcanic forcing.

Polar Stratospheric Ozone in a Changing Climate: Closing the Knowledge Gaps

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The ban of CFCs by the 1987 Montreal Protocol and its subsequent amendments will eventually lead to the recovery of the stratospheric ozone layer. However, due to these substances' long lifetimes, significant ozone reductions still occur over both poles in spring today, and are expected to do so for some decades. Over the same time, temperature, circulation patterns and chemical composition in the stratosphere may undergo significant changes as a result of climate change (and quite dramatic changes could arise should humans turn to geo-engineering ventures to mitigate climate change). To predict the future evolution of the ozone layer and the timing of complete recovery as accurately as possible, a correct representation of all relevant dynamic and chemical processes in prognostic climate models (such as chemistry climate models, CCM) is needed.

Five years ago, details of some important processes were not completely understood. For example, large uncertainties in the ClOOCl photolysis rate – the most critical kinetic parameter determining catalytic ozone loss in polar winter/spring – and serious inconsistencies between laboratory data and atmospheric observations triggered the establishment of a SPARC initiative on *Halogen Chemistry and Polar Ozone*. This initiative prompted or endorsed a number of studies and activities that focused on this issue and other processes relevant to polar ozone depletion. One such activity was the international project RECONCILE that employed new laboratory studies, field observations and modelling studies to address several open questions related to polar loss.

As a result of these efforts (i.e. RECONCILE and other projects as well as studies by individual groups), a number of questions were answered and uncertainties could be significantly reduced over the past five years. The uncertainty in the ClOOCl photolysis rate has been reduced to about 20 %, and consistency between laboratory and field studies established. Heterogeneous nucleation of polar stratospheric clouds (PSCs) on solid particles has been unambiguously demonstrated, and an enhanced scheme of PSC microphysics has improved the representation of denitrification in chemical transport models (CTM).

Based on the findings on PSC formation and catalytic ozone chemistry, kinetic and photochemical rate constants have been updated and a novel non-equilibrium PSC scheme was incorporated in the CCM LMDz Reprobus as part of RECONCILE. The more realistic description of these key processes is expected to enhance the robustness of the past and future ozone trends calculated by the LMDz CCM. Indeed, for past ozone trends, the RECONCILE upgrade results in a better fit to observations for Antarctica. For the Arctic, however, no significant difference between the simulations is discerned from the large dynamic variability.

This presentation will review the progress in our understanding of polar ozone loss made in the past five years within RECONCILE and other activities and summarize remaining uncertainties and knowledge gaps.

Multi-model assessment of the sensitivity of the tropical upper troposphere ozone towards regional biogenic emissions estimates

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The occurrence of low ozone events in the tropical upper troposphere (TUT), which can reach values of just a few ppb, has been shown to be sensitive to both convective transport and variations in the tropospheric lifetime of ozone in the lower troposphere. In that biogenic emissions are the dominant source of ozone precursors in the tropics means simulating such events is likely to be sensitive towards the seasonality and regional variability in biogenic emission estimates. Here we analyse the daily variability in ozone simulated in the TUT from a multi-model ensemble containing four independent members for the year 2005 to investigate (i) the uncertainty introduced across models towards capturing such low events, (ii) the accuracy to which the model ensemble can capture the vertical distribution of tropical ozone by comparison against measurements (iii) the sensitivity of such events towards different biogenic emission estimates and (iv) to differentiate which tropical region imposes the strongest signal on ozone in the TUT with respect to the seasonality and magnitude of natural emissions for the most important tropical regions.

How is chlorine activation affected by the composition of Polar Stratospheric Clouds and background aerosol particles?

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We investigate how important details of the microphysics of Polar Stratospheric Clouds (PSCs) and background aerosol particles are for the representation of polar ozone loss in chemistry transport models. For this purpose, the Lagrangian Chemistry and Transport Model ATLAS was applied to simulate the stratospheric chemistry in the Arctic winter 2009/2010. After a validation of the model results against measurements by the satellite-borne Microwave Limb Sounder (MLS), a number of sensitivity runs were performed. Thus, the efficiency of chlorine activation on different types of liquid aerosols versus activation on nitric acid trihydrate (NAT) clouds was explored. Moreover, the effects of particle composition and denitrification on ozone loss were analysed.

It is shown that even large changes in the underlying assumptions regarding detailed activation surfaces have only a small impact on the modelled ozone loss. Differences in column ozone between the various sensitivity runs remain below 10% at the end of the winter. Chlorine activation on liquid aerosols alone is able to explain the observed magnitude and morphology of the mixing ratios of active chlorine, reservoir gases and ozone. This is even true for cold binary aerosols (no uptake of HNO_3 from the gas-phase allowed in the model). This demonstrates that the heterogeneous chlorine activation in the polar stratosphere is dominated by the temperature dependence of the heterogeneous reaction rate constants rather than by the composition of the solid or liquid aerosol particles.

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Studies on mesoscale chemical and dynamical structures in the Arctic winter/spring 2010 UTLS region with MIPAS-STR

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The FTIR Limb-Sounder MIPAS-STR (Michelson Interferometer for Passive Atmospheric Sounding – STRatospheric Aircraft) deployed aboard the high-flying research aircraft M55-Geophysica allows for vertically and horizontally resolved measurements of ozone relevant minor species and long-lived tracers (e.g. O_3, HNO_3, $ClONO_2$, CFCs), temperature and cloud parameters in the UTLS region (Upper Troposphere/Lower Stratosphere) in the vertical range from ~5 to 20 km. MIPAS-STR was operated during the RECONCILE campaign (Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions) in Arctic winter/spring 2010, aiming on improving our understanding of Arctic ozone loss and its climate interactions. For different RECONCILE flights in the phase between end of January and begin of March 2010 it is shown that with MIPAS-STR mesocale chemical and dynamical structures with vertical extensions in the order of one kilometer and horizontal extensions of several tens of kilometers can be resolved. Retrievals have been improved for the conditions of optically partially transparent polar stratospheric clouds and give insight into the chemical and dynamical situation within and below these clouds, a region which is hardly accessible with comparable satellite-borne techniques. The high vertical and horizontal resolution of the MIPAS-STR measurements allows for detailed comparisons with high-resolution chemistry models of the atmosphere. Involving simulations of the chemistry transport model CLaMS (Chemical Lagrangian Model of the Stratosphere) and the MIPAS-STR measurements, a study on denitrification in Arctic winter/spring 2009/10 was carried out. The investigation of measured and modelled patterns from vertical redistribution of HNO_3 through denitrification shows enhanced agreement if reduced settling velocities are considered for nitric acid trihydrate particles in the model domain. The results suggest that large potential NAT-particles observed by in-situ instruments during RECONCILE (von Hobe et al., 2012 and references therein) might be explained by significantly aspheric NAT particles characterised by reduced settling velocities.

Overall, the MIPAS-STR measurements give detailed insight into the properties of the Arctic winter/spring 2010 UTLS region and provide the basis for further studies with atmospheric chemistry models.

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Introduction and evaluation of the ACCESS-UKCA chemistry-climate model

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ACCESS is the Australian Community Climate Earth-System Simulator (www.accessimulator.org.au, Bi et al., in press), and forms the basis for current and future Australian contributions to CMIP and IPCC activities. ACCESS is coupled to a detailed interactive chemistry and aerosol scheme (UKCA, O'Connor et al., 2013). Other components of ACCESS include the CABLE land surface model (Wang et al., 2011), and an ocean model. CABLE has recently been coupled to the UKCA chemistry scheme.

An evaluation of ACCESS with UKCA tropospheric chemistry is presented here. Comparisons are made against both in-situ and satellite observations of chemical species in order to test the performance of ACCESS-UKCA. The model simulations were conducted at 1.875 degrees x 1.25 degrees resolution, with 38 vertical levels extending to 42 km in height, with prescribed sea-surface temperatures. Anthropogenic emissions are derived from the ACCMIP dataset. We run ACCESS-UKCA in both free-running and nudged modes, making use of ECWMF's ERA-Interim reanalyses. Nudged simulations allow meaningful comparisons against specific observations at a point in space and time, while free-running simulations allow evaluation of the performance of the model from a climatological perspective.

The aim of the evaluation is to benchmark this first iteration of the ACCESS-UKCA chemistry-climate model. An updated version of ACCESS will extend simulations up to 85 km in height, and make use of an explicit isoprene chemistry mechanism. Future work will see UKCA used within the fully-coupled atmosphere-ocean configurations for chemistry-climate process studies and long-term climate simulations.

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Recent Global Emission Patterns of Refrigerants HCFC-22 and HFC-134a

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HCFC-22 (CHClF2) and HFC-134a (C2H2F4) are two major refrigerant gases currently used worldwide. They replaced CFC-12 (CCl2F2), a historically used refrigerant which has strong ozone depleting potential (ODP) and therefore was regulated by the Montreal Protocol to be phased-out globally by 2010. The production of HCFC-22 started in the 1930s and its emissions have been rising since then, with a recent speed up around 2005. The emissions of HFC-134a have been rising steadily since the early 1990s. Neither of them showed an emission surge around 2010 that would have been expected from the phase-out and the replacement of CFC-12 by that year. Because both species are potent greenhouse gases (GHGs), understanding their global emission distributions and the temporal variability is of great environmental importance and also supports mitigation policy decisions. In this work, we studied a set of atmospheric observation data with extensive sampling over the Pacific Ocean in a three-year span around 2010 (HIPPO). A three-dimensional global chemistry transport model (ACTM) was constrained by the long-term observations from global surface sites (AGAGE, NOAA) and was validated using chemical and transport tracers. By comparing the ACTM simulations to the HIPPO observations and by conducting the “tagged-tracer” inversions, we found increasing HFC-134a emissions in the tropical and Southern Hemisphere regions and decreasing Northern Hemisphere mid-latitude emissions, different from the existing production/consumption based emission inventories (GEIA, UNEP, UNFCCC).

Trends and Variability of Ozone in the Southern Hemisphere Middle and High Latitudes between 1997 and 2010

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It is well established that stratospheric ozone changes significantly affect tropospheric ozone, mainly through the moderation of stratosphere-troposphere exchange (STE). Here we present results from simulations covering the year 1997 to 2010 using NIWA-UKCA, a coupled stratosphere-troposphere chemistry-climate model. We compare modelled ozone to ground-based ozone measurements taken at Lauder, New Zealand, and at Arrival Heights, Antarctica, both clean-air sites, to derive modelled and observed trends and variability of Southern-Hemisphere mid- to high latitudes ozone over this period. Linkages between changes in stratospheric and tropospheric ozone will be explored, and we will attribute observed changes. Ozone measurements used in this study include long-term ozone sonde data and Dobson total-column ozone.

Multi-model assessment of the impact of biogenic emissions on the composition of the troposphere in the Southern Hemisphere

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Biogenic emissions act as dominant sources of ozone precursors, carbon monoxide (CO) and non-methane hydrocarbons, in the troposphere in both the tropics and the Southern Hemisphere (SH). The various approaches adopted for estimating global biogenic emission fluxes result in large regional and seasonal uncertainties, in particular for those provided for isoprene and the monoterpenes whose annually integrated totals differ by ~100 Tg C yr^{-1}. Here we present the first results of a multi-model study focussing on whether current large-scale atmospheric chemistry models can capture the seasonal distribution of CO and formaldehyde (HCHO) in the SH. We use four independent chemistry-climate and chemical transport models which are driven by different meteorological fields for the period 2004-2008, with each model using biogenic emission estimates available from both the LPJ-GUESS and MEGAN-v2.1 inventories. For the first time we exploit high-precision ground-based FTIR measurements of CO and HCHO columns at four locations in the SH, namely Darwin, Wollongong, Lauder, and Arrival Heights. By comparing against these measurements and those available from other monitoring networks we show that although the seasonal signal is simulated quite well, there is generally an over-estimation in the tropospheric CO burden in the SH across models. In addition we quantify the sensitivity of the inter-annual distribution of SH CO and HCHO towards both direct (via emissions) and indirect (via changes in oxidative capacity) influences.

Global VSLS Emission Estimates based on in-situ Measurements for the Past and Future

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The HalOcAt data base initiated through SOLAS / COST Action 735 and SOPRAN currently contains about 200 contributions, comprising roughly 55,400 oceanic and 476,000 atmospheric measurements from all oceanic depths and atmospheric heights. These data include 19 different halocarbon compounds and were collected between 1989 and 2011 (https://halocat.geomar.de). Based on available surface data of the HalOcAt database global marine and atmospheric surface maps of bromoform ($CHBr_3$), dibromomethane (CH_2Br_2) and methyl iodide (CH_3I) are calculated in order to derive finally global air – sea flux estimates from a bottom up approach (Ziska et al., 2013 *ACPD*). For chemistry transport model studies the data were interpolated onto a 1°x1° grid, while missing grid values were extrapolated with different regression techniques based on the observed biogeochemical distribution of the VSLS. Global air-sea fluxes were calculated with climatological input data. All three compounds exhibit enhanced flux strengths in different oceanic regimes of the tropics. In contrast to recent studies, negative fluxes (sinks in the ocean) occur in each climatology. Future emission VSLS scenarios are calculated with CCMVal 2 RefB2 and CMIP5 RCP8.5 model simulations, including a full interactive ocean. For this approach the present climatological oceanic and atmospheric concentrations are combined with projected oceanic hydrological and meteorological parameters. Finally, the role of climate change on future VSLS emissions will be assessed.

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