

Atmospheric Halogens: Model simulations of VSLS and long-lived ODS

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In this talk I will deal both with numerical simulations of ozone depleting substances and their observations. I will also talk about emissions of inorganic halogens, from the ocean and from snow packs. A unifying theme will be chemistry-climate interactions.

I will describe long-term measurements of a number of VSLS species, which started in Borneo in 2008. Model-based estimates of their emission strengths, and possible transport to the TTL, will be discussed.

The UM-UKCA chemistry-climate model has been run to consider a wide range of future ODS scenarios (including changes in climate and changes in tropospheric ozone precursors). Latest results will be presented.

Separating the influences of climate and chemistry on future lifetimes of ozone-depleting substances: no indication of much shortened lifetimes

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Knowledge of the lifetimes of ozone-depleting substances (ODSs) is used not only to estimate their present emissions and future abundances, but also to calculate their ozone-depletion and greenhouse warming potentials, information that is essential for guiding policy making on the control of these substances. In this study, the SOCOL (Solar Climate Ozone Links) Chemistry-Climate Model is used to estimate the atmospheric lifetimes of several key ODS species for two periods: year 2000 (current conditions) and year 2100 (future conditions). Several time-slice simulations are used to separate the different influences of chemistry and climate on future ODS lifetimes, including the effect of using different emissions scenarios for ODSs (RCP2.5 as well as the widely-used WMO 5-A3 scenario (2011)). Interestingly, our results indicate that the effects of chemistry and climate act in opposite directions, but are of similar magnitude: the future reduction in ODS emissions leads to an increase in ODS lifetimes, largely through the resultant increase in stratospheric ozone, which reduces photochemical destruction rates in the stratosphere and thus ODS lifetimes lengthen; in contrast, climate change acts to reduce ODS lifetimes through the effect of an enhanced Brewer-Dobson Circulation and faster transport through the atmosphere. While the circulation effect by itself would lead to a shortening of lifetimes by between 3-13%, the changed chemistry compensates to a large degree (by between 2-11%), and future lifetimes of ODSs are estimated to be only 1-4% shorter than at present. On a per-molecule basis, this finding indicates the continued effectiveness of these important ozone depleting and greenhouse gases, and puts previous reports into perspective that have suggested much shortened lifetimes.

Future Arctic Temperature and Ozone: The Role of Stratospheric Composition Changes

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Multi-decadal simulations with the ECHAM/MESSy Atmospheric Chemistry (EMAC) chemistry climate model were analyzed to examine the role of changing concentrations of ozone depleting substances and greenhouse gases (GHGs) on Arctic springtime ozone. The focus is on potential changes in the meteorological conditions relevant for Arctic ozone depletion. It is found that with rising GHG levels the lower Arctic stratosphere will significantly cool in early winter, while no reliable temperature signal is identified later in winter or spring. A seasonal shift of the lowest polar minimum temperatures from late to early winter in the second part of the 21st century occurs. However, Arctic lower stratosphere temperatures do not seem to decline to new record minima. The future Arctic lower stratosphere vortex will have a longer lifetime, as a result of an earlier formation in autumn. No extended vortex persistence is found in spring due to enhanced dynamical warming by tropospheric wave forcing. Because of the dominant early winter cooling, largest accumulated areas of potential polar stratospheric cloud formation (A_{PSC}) are projected for the middle of the 21st century. A further increase of A_{PSC} towards the end of the 21st century is prevented by increased dynamical polar warming. The model suggests that within the next few decades there is a chance of low Arctic springtime ozone in individual years, as was observed in March 2011, however there is no indication of a formation of regular Arctic ozone holes comparable to the Antarctic. Towards the end of the 21st century further rising GHG levels will lead to an increase of Arctic springtime ozone.

Cloud/aerosol Interactions inferred from Satellite Observations

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Feingold and Stevens (2009) describe the cloud aerosol interaction problem as a buffered system. The outputs of such systems involve a complex set of feedbacks that buffer the cloud and precipitation responses to given aerosol perturbation. The nature of this buffering as it applies to cloud albedo changes in response to aerosol changes is examined through the use of satellite data applied to the study of low clouds. The analysis reveals a similar picture - that the observed correlations between albedo and aerosol of low clouds are a complicated function of opposing meteorological influences. These buffering influences of processes will be demonstrated pointing to the cloud water budgets as the controlling effects on albedo and impacts on changing water balance assumptions in global models will be explored. Implications of these findings on the estimate of indirect radiative effects of low clouds, on aerosol influences on convective clouds and on precipitation will be described.

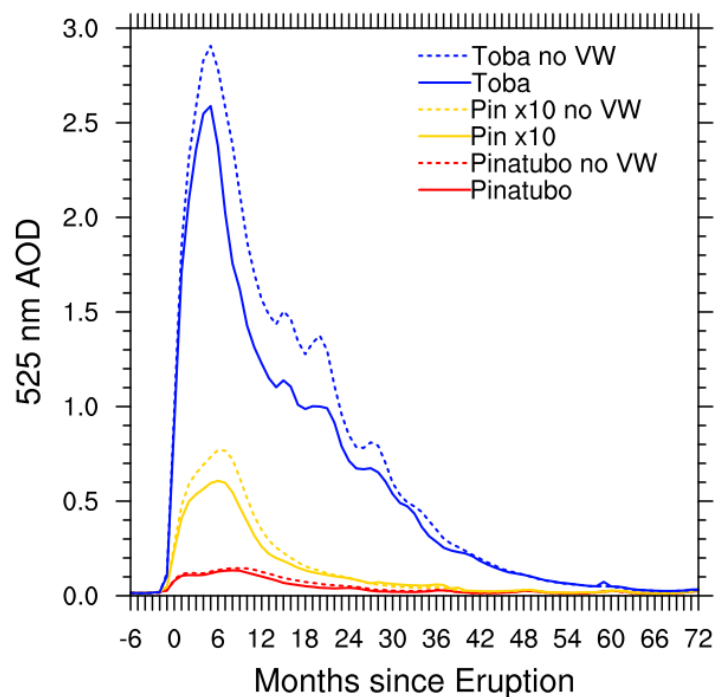
Aerosol Size Distributions After Large Volcanic Eruptions Evolve In a Complex Manner

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Large volcanic eruptions such as the eruption of Toba 74,000 years ago may cause severe climatic perturbations. There remain many uncertainties regarding the magnitude and details of climate perturbations after such eruptions due in part to unknowns in the evolving aerosol size distributions. We utilize a sectional aerosol microphysical model coupled to a whole atmosphere general circulation model (WACCM/CARMA) and include van der Waals forces in our coagulation scheme to study the evolving aerosol size distributions after three eruptions ranging in size from Pinatubo to Toba.

Simulations find aerosol optical depth (AOD) after large eruptions such as Toba to increase less than linearly due to particle growth and sedimentation. Despite a 100-fold increase in sulphur dioxide injection, AOD increases by only a factor of 20 (Figure 1). Inclusion of van der Waals forces in our coagulation scheme (solid lines) increases the rate of coagulation and particle size, reducing AOD by about 15% compared to the scheme without van der Waals forces (dotted lines) [English *et al.*, 2013]. Global-average AOD remains elevated for 1, 2, and 4 years respectively for the three eruptions. Simulations of Toba with our sectional model predict aerosol effective radii larger than those predicted by a modal model [Timmreck *et al.*, 2010] and by a bulk aerosol modal [Robock *et al.*, 2009].



We then fit lognormal size distributions to our bin model for a direct comparison to modal models and find mode width to vary from 1.2 to 1.8 temporally and spatially, suggesting that two-moment modal models with a fixed mode width may not be accurately capturing the complexity of an evolving aerosol size distribution. These simulations suggest the importance of including van der Waals forces in the coagulation scheme and using sectional models to study large perturbations to stratospheric aerosols.

English, J. M., O. B. Toon, and M. J. Mills: Microphysical simulations of large volcanic eruptions: Pinatubo and Toba, *J. Geophys. Res. Atmos.*, 118, 1880–1895, doi:10.1002/jgrd.50196.
Robock, A., C. M. Ammann, L. Oman, D. Shindell, S. Levis, and G. Stenchikov (2009), Did the Toba volcanic eruption of ~74 ka B.P. produce widespread glaciation?, *J. Geophys. Res.*, 114, D10107, doi:10.1029/2008JD011652.
Timmreck, C., H.-F. Graf, S. J. Lorenz, U. Niemeier, D. Zanchettin, 90 D. Matei, J. H. Jungclaus, and T. J. Crowley (2010), Aerosol size confines 91 climate response to volcanic super-eruptions, *Geophys. Res. Lett.*, 37, 92 L24705, doi:10.1029/2010GL045464.

Stratospheric implications of climate engineering through solar radiation management

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In the context of global warming, various climate engineering (or “geoengineering”) techniques have been proposed as a “last resort”, in case mitigation efforts fail or the consequences of the warming prove more severe than expected. Among the suggested approaches filed under “solar radiation management” (SRM), the arguably most discussed one is to emit sulfur into the stratosphere and thereby mimic the effects of large volcanic eruptions. Such a deliberate global-scale manipulation of the radiative budget of the Earth may allow a counterbalancing of global mean temperature increase due to the effects of continued greenhouse gas emissions, but may also result in undesirable side effects for crucial parts of the Earth system.

This presentation will provide an overview on the current state of knowledge concerning effects of SRM methods. One focus is on the question what climate would result from SRM. Results presented are in particular based on climate model simulations performed within the Geoengineering Model Intercomparison (GeoMIP) project. One of the expected effects of SRM is that if a certain temperature increase would be compensated, the according precipitation increase would be overcompensated. Here it will be discussed why this overcompensation effect would be stronger for stratospheric sulfur emissions than for a simple reduction of solar irradiance at the top of the atmosphere. Other questions that will be discussed are how stratospheric circulation is expected to respond to SRM and to what extent experiences from volcanic eruptions can be used to estimate effects of SRM.

Recent anthropogenic increases in SO₂ from Asia have minimal impact on stratospheric aerosol

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There has been significant debate over why the Earth has not experienced more extensive global surface warming from increases in greenhouse gases since the year 2000. The outcome of this research helps to elucidate the answer by examining the variability of the stratospheric aerosol layer, which is mainly comprised of sulfuric acid droplets produced from natural and anthropogenic precursors, and is known to have significant impacts on decadal climate variability. The layer plays a key role in climate by reflecting sunlight away from the Earth before it can act to heat the atmosphere. Observations suggest stratospheric aerosol increased by 4-10% per year from about 2000 to 2012. While we know from the observational record that aerosol has increased, the relative importance of various sources of this change are unknown. Recent observations suggest that volcanic eruptions have played a significant role in the heightened level of aerosol by injecting large amounts of sulfur and aerosol into the stratosphere. However, these observations alone cannot rule out the possible influence of increased anthropogenic emissions of sulfur from the large industrial expansion of China and India over the same period. This increase may have an especially large effect due to the proximity of China and India to the Asian monsoon region, a major path of surface pollution into the stratosphere. To disaggregate the aerosol layer's natural and anthropogenic sources, a global climate model coupled to a detailed model of sulfate aerosol was used to examine the relative influence of volcanic and anthropogenic emissions on the global variability of stratospheric aerosol. This is the first study to apply such a model to this problem and to isolate the separate impacts of major anthropogenic and natural sources. The results show that human activity had little impact on the aerosol budget of the stratosphere since 2000, and, consequently, the layer's impact on climate. Instead, the results suggest that the coincidental string of a series of moderate volcanic eruptions since 2000 may have prevented as much as 25% of the surface warming expected from greenhouse gas emissions during this period. Though current changes in the distribution of anthropogenic sulfur emissions apparently had little impact on the stratosphere, historical emission trends suggest that since the industrial revolution global sulfur emissions have increased the amount stratospheric aerosol significantly. The historical trend implies that projected future reductions in sulfur emissions, created by the cleaner burning of fossil fuels over the 21st century, may enhance future global warming due to greenhouse gases.

Near-term Climate Forcing

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Mitigation measures for shorter-lived pollutants that have a near-term climate forcing such as methane, black carbon and ozone have been proposed by organisations such as the Climate and Clean Air Coalition. To understand the effectiveness of this approach we need to quantify the climate effects of these pollutants and understand how they behave on different timescales.

The global climate impact of short-lived species depends where they are emitted from, and the regional climate impacts even more so due to the heterogeneity of the forcing patterns. We might expect the benefits of mitigation measures (say of black carbon) to be greater than the global average over the region implementing the measures, but how much greater? I will show how regional climate metrics can be used to relate continental emission reductions to latitudinal changes in surface temperature. Once calculated these climate metrics can be used to estimate the temperature impacts of any mitigation scenarios such as those generated under the UNEP Assessment on Tropospheric Ozone and Black Carbon.

The science behind the effects of near-term climate forcers is relatively new and is advancing rapidly. The impacts of near-term climate forcers on precipitation patterns are even more complex than the impacts on temperature. Multi-model studies are being carried out to identify whether there are robust relationships that can be quantified.

Chemical/dynamical interactions and consequences for climate

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Most climate models use prescribed distributions of chemical species and do not capture potentially important climatic effects of spatial and temporal inhomogeneities of chemical substances. For example, past research found that prescribing zonal asymmetries in stratospheric ozone leads to significant differences in simulated climate and climate trends over the Southern Hemisphere. Here, we examine the effects of fully interactive chemistry on the simulation of climate. This is accomplished by comparing simulations from CMIP5 models with interactive chemistry against parallel simulations from models in which the concentration of chemical species are prescribed.

We find that the inclusion of interactive chemistry leads to a robust increase in climate variability over the extra tropics of both the northern and southern hemisphere. There is large consistency in the spatial and temporal structure of this increase amongst the different models: it has a pronounced seasonal cycle with a maximum in spring and summer, is largest in the stratosphere, and also impacts the surface. Under anthropogenic ozone depletion there is a strong amplification of the increase over the Southern Hemisphere. During mid-winter there is indication for a slight reduction of variability, in particular over the Northern Hemisphere. We hypothesize that interactions between ozone and other chemical species, shortwave radiation, and the atmospheric circulation create a positive feedback during spring and summer. Photochemical ozone depletion leads to a further increase in the strength of this feedback.

We discuss the consequences of our findings for the design of modern climate models, the prediction of natural and anthropogenic climate variations, and the detection of anthropogenic climate change.

Tropospheric Ozone Response to Variability in the Stratospheric Circulation: The Role of ENSO and the QBO and Relationship to Long-Term Changes

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Chemistry-climate models robustly predict increases in the large-scale stratospheric circulation and stratosphere-troposphere exchange (STE) in response to increasing greenhouse gases. The impact of these stratospheric dynamical changes on “background” levels of tropospheric ozone, knowledge of which is critical to developing effective air quality and climate policies, has heretofore been unconstrained. Our previous work has shown that current variability in the stratospheric circulation and stratosphere-to-troposphere ozone flux driven in part by El Niño /Southern Oscillation (ENSO) provides a “natural experiment” that may reduce uncertainties in predictions of the tropospheric ozone response to future changes in stratospheric transport. Using six years of measurements from the Tropospheric Emission Spectrometer (TES) and Microwave Limb Sounder (MLS) onboard NASA’s Aura satellite, we found that interannual variability in the stratospheric circulation of $\sim\pm 40\%$ leads to changes of $\sim\pm 2\%$ in northern midlatitude tropospheric ozone (equaling $\sim 1/2$ the total observed interannual variability). Here, we combine these measurements with two longer time series of stratospheric observations from the GOZCARDS and SWOOSH merged datasets and three chemistry-climate models. We further examine the relationship between ENSO and the QBO in driving changes in the stratospheric circulation and STE and assess the relationship between the tropospheric ozone response to current stratospheric variability and its response to long-term changes in stratospheric transport.

The Impact of Polar Stratospheric Ozone Loss on Southern Hemisphere Stratospheric Circulation and Surface Climate

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We present results of a study using a fully coupled chemistry-climate model, UM-UKCA, to assess the impacts of lower stratospheric ozone loss on Southern Hemisphere stratospheric circulation and climate. Two simulations were run, both using perpetual year 2000 boundary conditions. The first used a full stratospheric chemistry scheme, while in the second heterogeneous chemistry on Polar Stratospheric Clouds is suppressed. By comparing the two integrations we are able to quantify the climate and dynamical impacts of polar stratospheric ozone depletion.

As expected, modelled heterogeneous processes result in severe ozone depletion in the spring time Antarctic polar vortex. However, from December to March ozone mixing ratios at ~10hPa are increased due to changes to the stratospheric circulation. Statistically significant ozone changes extend beyond the polar vortex. While global total column ozone is greatly reduced throughout the year, with decreases of more than 15DU at the equator, from November to February column ozone around 60°N increases.

Ozone changes drive significant changes to high latitude Southern Hemisphere climate. Here we assess the impact of stratospheric ozone loss on temperature, zonal wind speed and the residual mean vertical circulation and discuss the physical mechanisms behind these changes. The response of all three is seasonal, with statistically significant changes in austral spring and summer, and no significant changes in winter. Polar ozone loss causes the lower polar stratosphere to cool from October to March due to decreased shortwave heating, with maximum cooling of 12K during November and December at 70hPa. The polar vortex accelerates and its breakup is delayed by approximately 2 weeks. This alters the propagation and breaking of planetary waves in the stratosphere, resulting in increased wavebreaking in the stratosphere between 50°-70°S and increased downwelling over the pole. Increased downwelling over the pole causes the middle and upper stratosphere to warm from November to February due to increased dynamical heating, with maximum warming in December at ~10hPa. Increased dynamical heating mitigates the cooling of the lower stratosphere and acts as a negative dynamical feedback. The lower stratosphere temperature and zonal wind response propagate down into the troposphere and to the surface in December and is consistent with observed trends. We present a preliminary assessment of the causes and feedbacks of the surface response to stratospheric ozone depletion within the model.

The hiatus in global surface warming over 1998-2011

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During the past 15 years, global-mean surface temperature (GMST) has shown a comparatively small warming trend, termed a hiatus. However, almost all CMIP5 historical simulations show a GMST trend over the past 15 years that is higher than the observed trend. The present study investigates possible causes and the spatial structure of the surface-warming hiatus by performing 24 ensemble simulations with the Max Planck Institute Earth System Model (MPI-ESM).

We examine the conjecture (Solomon et al. 2011) that background stratospheric aerosol forcing caused the hiatus, by using MPI-ESM with two different datasets of stratospheric aerosol loading. In one set of experiments, we perform 12 simulations with CMIP5 historical forcing (augmented for the period 2006–2011 by RCP4.5). The volcanic aerosol data from Stenchikov et al. (1998) are used before year 1999, augmented for the period 2000–2011 with the volcanic aerosol data of 1999. In the second set of experiments, we replace the stratospheric aerosol forcing of the CMIP5 historical simulations with the near-global satellite stratospheric aerosol data from CCMI (Chemistry-Climate Model Initiative, Eyring et al., 2013) over 1990–2011. We also perform 12 simulations in this set of experiments.

Our results show a negative radiative forcing due to the stratospheric aerosol increase over 1998–2011 of about -0.07 Wm^{-2} , reducing the recent global warming by about $0.02 \text{ }^\circ\text{C}$. Our simulations with CCMI stratospheric aerosol forcing show almost the same GMST trend as our CMIP5 historical simulations. Hence, ignoring the stratospheric aerosol increase after year 2000 does not cause the overestimation of GMST trend over the past 15 years in MPI-ESM.

We find a very strong cooling trend over the Northern-Hemisphere continental region over 1998–2011 in all observational and reanalysis data, which might compensate surface warming over other regions and thus lead to a hiatus in GMST increase. This strong continental-cooling trend, which mainly happens in Northern-Hemisphere winter and spring, is linked to a negative NAO (North Atlantic Oscillation) and probably also by dramatic Arctic summer sea-ice loss.