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Book of Abstracts – Oral Presentations

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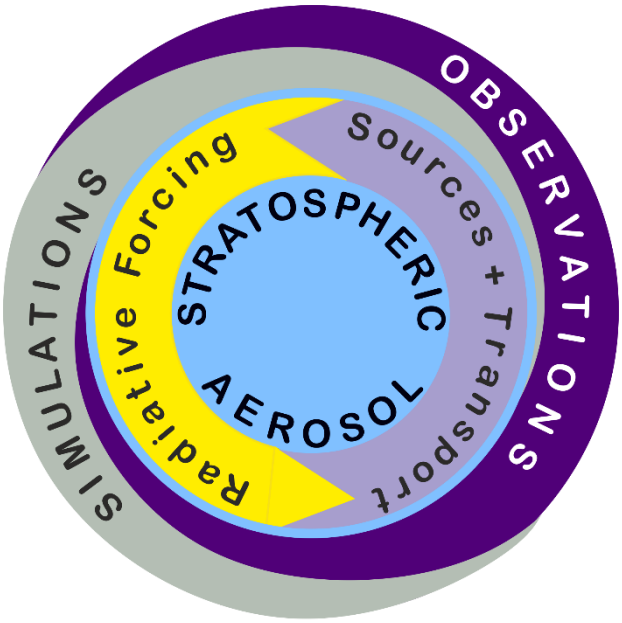

SSiRC bridges observations and models and connects scientists from different fields to gain quantitative understanding of stratospheric aerosol processes from emissions to radiative forcing.

Stratospheric Warming and Ozone Destruction by the 2019-2020 Australian Wildfire Smoke

Pengfei Yu, Sean M. Davis, Owen B. Toon, Robert W. Portmann, Charles G. Bardeen, John E. Barnes, Hagen Telg, Christopher Maloney, and Karen H. Rosenlof

Australian wildfires burning from December 2019 to January 2020 injected massive smoke into the stratosphere. A comparison of numerical simulations by CESM-CARMA model (Yu et al., 2015) to satellite observations of the plume rise suggests that the smoke mass contained 2.5% black carbon. Model calculations project a 1-2 K warming in the stratosphere of the Southern Hemisphere (SH) midlatitudes for more than 6 months following the injection of black-carbon containing smoke. The projected temperature anomalies are observed by satellites data reported in Rieger et al. (2021). Assuming that smoke particles coat with sulfuric acid in the stratosphere and have similar heterogeneous reaction rates as sulfate aerosol, model simulations by CESM-CARMA suggest the smoke may cause ozone loss of 10-20 Dobson units (i.e. 4-6%) from August to December in mid-high southern latitudes. Similar ozone anomalies are observed in SH midlatitudes by satellites (Santee et al., 2022). Both the satellite observations and model simulations suggest that the reactive nitrogen is depleted due the Australian fire and the hydrolysis of N2O5 partly leads to the ozone loss in the mid-latitudes in SH (Solomon et al., 2022)

Multi-satellite views of Stratospheric Injections of Massive Carbonaceous Aerosol Plumes from Canadian (2017) and Australian (2020) PyroCb events.

Omar Torres[1]

P.K. Bhartia[1], Gassan Taha[2], Hiren Jethva[2], Sampa Das[3], Peter Colarco[1], Ali Omar[4], Nickolay Krotkov[1], Changwoo Ahn[5]

[1]NASA Goddard Space Flight Center, Greenbelt, MD, 20771

[2]Morgan State University, Baltimore, MD

[3]University of Maryland, College Park, MD

[4]NASA Langley Research Center, Hampton, VA, USA

[5]Science Systems and Applications Inc., Lanham, MD USA

Abstract

Carbonaceous aerosol plumes associated with wildfires in British Columbia in August 2017, and in Southeast Australia in January 2020 reached the stratosphere a few days following the onset of the fires. Near-hourly observations by the EPIC sensor onboard the DSCOVR satellite, CALIOP on the CALIPSO platform, and OMPS-LP on the SNPP satellite observed the spatial and temporal evolution of the 2017 British Columbia aerosol plume. EPIC-based aerosol mass estimates indicate that 268±40% Kt of aerosol material was injected in the stratosphere by the 2017 pyroCb event. About 10% of this aerosol mass reached the stratosphere on August 13, whereas the remaining 90% arrived in the stratosphere over the following two days. A similar analysis of S5P-TROPOMI observations following the 2019-2020 chain of pyroCb's in Australia yielded a stratospheric aerosol mass of 546±40% Kt, which is about twice as much as produced by the 2017 pyroCb event. Unlike during the 2017 pyroCb, when the aerosol plume reached the upper troposphere and subsequently ascended to the lower stratosphere, the Australian plume seemed to have been directly injected in the stratosphere. The lifetimes and aerosol mass of these events are comparable to those of moderate volcanic eruptions.

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Acknowledgements

TROPOMI at TROPOMI/Sentinel-5P Near UV Aerosol Optical Depth and Single Scattering Albedo L2 1-Orbit Snapshot 7.5 km x 3 km

EPIC at https://eosweb.larc.nasa.gov/project/dscovr/dscovr_epic_l2_aer_01

OMPS-LP at https://snpp-omps.gesdisc.eosdis.nasa.gov/opendap/SNPP_OMPS_Level2/OMPS_NPP_LP_L2_AER675_DAILY.1.5/contents.html

CALIPSO at https://www-calipso.larc.nasa.gov/products/lidar/browse_images/production/

Abstract Title: Radiative impacts of the Australian bushfires 2019–2020 and Hunga Tonga volcano eruption 2022: comparison of extreme fires and volcanic eruptions

Presenting Author title and name: Dr Pasquale Sellitto[1]

Co-author Names: The ASTuS team [B. Legras[2], A. Podgajen[2], J. Cuesta[1], C. Duchamp[2], R. Belhadji[1], C. Kloss[3], J.B Renard[3], P. Billant[4], A. Limare[5], T. Roberts[3], A. Nies[3], D. Dritschel[6], G. Berthet[3], F. Jégou[3]] and E. Carboni[7], R. Siddans[7], S. Khaykin[8], M. Boichu[9]

[1]Laboratoire Interuniversitaire des Systèmes Atmosphériques, IPSL, UPEC, CNRS, Créteil, France

[2]Laboratoire de Météorologie Dynamique, IPSL, CNRS, ENS-PSL/Ecole Polytechnique/Sorbonne Univ., Paris, Fr
[3]Laboratoire de Physique et Chimie de l'Environnement et de l'Espace, CNRS, Univ. Orléans, Orléans, France

[4]LadHyX, CNRS, Ecole Polytechnique, Palaiseau, France
[5]Institut de Physique du Globe de Paris, Paris, France
[6]Mathematical Institute, Univ. St Andrews, UK
[7]Rutherford Appleton Laboratory, Oxford, UK

[8]Laboratoire Atmosphère, Observations Spatiales, UVSQ, CNRS, Sorbonne Univ., St Quentin, France

[9]Laboratoire d'Optique Atmosphérique, Villeneuve d'Ascq, France

Abstract (100 - 500 Words)

Two major stratospheric-perturbing events occurred in the last few years: Australian fires 2019-2020 and the Hunga Tonga volcanic eruption 2022. As a consequence of extreme heat and drought, record-breaking wildfires developed and ravaged south-eastern Australia during the fire season 2019–2020 and produced extreme pyro-Cb activity on January 2020; this is considered the largest fire-induced perturbation to the stratosphere on records [Khaykin et al. 2020]. The Hunga Tonga volcano violently erupted on January 15th, 2022, producing the largest volcanic perturbation into the stratosphere, and the highest-altitude one, since the Pinatubo eruption in 1991. Through their large-scale and long-lived stratospheric perturbation, these events had large impacts on the radiative balance [Sellitto et al., 2022a; Sellitto et al., 2022b; Sellitto et al., 2022c]. Here we discuss the radiative forcing of these events in terms of top-of-the-atmosphere and surface imbalances and their in-plume localized heating/cooling, through a hybrid observations-modelling method. The fundamental differences of fire and volcanic perturbations, due to fundamentally different aerosol optical properties of biomass burning and volcanic aerosols, as well as the concurrent radiative impacts of co-emitted species like water vapour, are also discussed. The role of the in-plume absorption within Australian fires smoke plume to the generation of ascending smoke vortices in the stratosphere is also presented.

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[3] Sellitto, P., Belhadji, R., Cuesta, J., Podglajen, A., and Legras, B.: Radiative impacts of the Australian bushfires 2019–2020 – Part 2: Radiative heating and the formation of ascending smoke vortices, in preparation, 2022b.

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Acknowledgements

We acknowledge support of the Agence Nationale de la Recherche through grant ASTuS ANR-21-CE01-0007-01, support from CNES via TOSCA/IASI action, support from AERIS ICARE Data Center, the resources of the IPSL Meso Center, ESA, NOAA, Copernicus Climate Service and ECMWF.

Cloud/Aerosol Categorization of Stratospheric Aerosol Observations from Stratospheric Aerosol and Gas Experiment (SAGEIII/ISS)

Presenting Author title and name: Mahesh Kovilakam[1,2]

Co-author Names: Larry Thomason[2]

[1]SSAI, Hampton, VA USA

[2]*NASA Langley Research Center, Hampton, VA USA*

Abstract:

The Stratospheric Aerosol and Gas Experiment (SAGE III/ISS) aboard ISS began its mission in June 2017. SAGEIII/ISS is an updated version of SAGEIII-Meteor instrument that makes observations of stratospheric aerosol extinction coefficient at wavelengths that range between 385 and 1550 nm with a near global coverage between 60S-60N. While SAGEIII/ISS makes reliable and robust solar occultation measurements in stratosphere—similar to its predecessors, interpreting aerosol extinction measurements in the vicinity of tropopause and in the troposphere have been a challenge for all SAGE measurements. Here, we study the challenges associated with the discrimination of aerosols and clouds from the extinction measurements. Additionally, recent volcanic/PyroCb events make it more challenging to separate aerosols from clouds. Here, we describe the methods implemented to categorize Clouds and aerosols using available SAGEIII/ISS aerosol measurements. Cloud categorization is developed based on a method proposed by Thomason and Vernier (2013) with some modifications that now incorporates the influence of recent volcanic/PyroCb events and a new method of locating aerosol centroid based on k-medoid clustering. We use version 5.2 of SAGE III/ISS extinction coefficients for the analysis. The current algorithm now classifies standard (background) and non-standard (enhanced) aerosols in the stratosphere and identify enhanced aerosols and aerosol/cloud mixture in the tropopause region. Extinction coefficient measurements from SAGE series of observations make an important contribution in the GloSSAC data base and therefore, the impact of cloud-filtered aerosol extinction coefficient measurements on the latest version of GloSSAC (version 2.1) is also discussed.

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Abstract Title: Radiative forcing from volcanoes and the 2019/20 Australian forest fires using the GCM EMAC and satellite data with different approaches

Presenting Author title and name: Dr Christoph Brühl[1]

Co-author Names: Dr Jennifer Schallock[1], Matthias Kohl[1], Landon Rieger[2]

[1] *Max Planck Institute for Chemistry, Mainz, Germany*
[2] *U Saskatchewan, Saskatoon, Canada*

Abstract

We present transient simulations for radiative forcing with the chemistry climate model EMAC for the period 2019 to 2020 using 3D-OSIRIS observations to derive volcanic SO2 injections (Schallock et al.) and injections of organic aerosol by pyro-cumulonimbus based on 3D-perturbations in the lower stratosphere. Alternatively, the estimates for the smoke injections by the 2 pyro-cumulonimbus events of Peterson et al. are used. We show that the latter can explain only a part of the large perturbations in January and February seen by OSIRIS and OMPS-LP but we are able to reproduce lofting of the plumes by radiative heating. This includes some sensitivity studies on uncertainties due to temporal and spatial distribution of "point sources" for smoke particles and volcanic SO2 which might be of interest also for ISA-MIP.

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Observed Raikoke Volcanic SO_2/Sulfate Anticyclonic Contained Circulations

Michael D. Fromm, NRL

George P. (Pat) Kablick III, NRL

Isabelle Taylor, Oxford University

Don Grainger, Oxford University

Abstract:

Maps of the Raikoke UTLS sulfur plume (gaseous and particulate) contain evidence of multiple distinctive, quasi-circular plume sub-elements with diameter < ~300 km. These quasi-circular SO_2/sulfate entities are tracked for up to two months. We find that they are defined not only by their shape/size but also a discernable anticyclonic circulation. In addition, they embody a marked signal of window IR absorption, detectable for at least one month. The term we apply to this phenomenon is "SO_2/sulfate anticyclonic contained circulation," abbreviated SSACC. Using high temporal resolution satellite data and trajectories we track two SSACCs and establish their genesis as early as 24 June 2019, two days post eruption. The circulation aspect is gleaned from geostationary-based visible-image animations and confirmed via a novel application of high-resolution radiosonde position data. We also show that the SSACCs undergo diabatic lofting, rising above initial altitude by as much as ~10 km. The SSACC discovery represents a new and distinct manifestation of an enclosed, anticyclonically circulating stratospheric plume, joining the recent identification of similar qualities in stratospheric wildfire smoke plumes.

Abstract Title: Improved estimation of volcanic SO2 injections from satellite retrievals and Lagrangian transport simulations: The 2019 Raikoke eruption

Zhongyin Cai1,2 , Sabine Griessbach1 , Lars Hoffmann1

Co-author Names: Zhongyin Cai1,2 , Sabine Griessbach1 , Lars Hoffmann1

[1]Affiliation 1: Jülich Supercomputing Centre (JSC), Forschungszentrum Jülich, Jülich, Germany
[2]*Affiliation 2: Institute of International Rivers and Eco-security, Yunnan University, Kunming, China*

Abstract (100 - 500 Words)

Monitoring and modelling of volcanic plumes are important for understanding the impact of volcanic activity on climate and for practical concerns, such as aviation safety or public health. In a case study of the eruption of the Raikoke volcano (48.29°N, 153.25°E) in June 2019 we investigated the SO2 injections into the upper troposphere and lower stratosphere and their subsequent long-range transport and dispersion by means of Lagrangian transport simulations.

For the estimation of an altitude-resolved SO2 injection time series we used SO2 retrievals from AIRS (Atmospheric Infrared Sounder) and TROPOMI (TROPOspheric Monitoring Instrument) together with a backward trajectory approach. We applied a scaling factor to the initial estimate of the SO2 mass and added an exponential decay to simulate the time evolution of the total SO2 mass. By comparing the SO2 mass from our estimate with TROPOMI retrievals, we show that the volcano injected 2.1 +- 0.2 Tg SO2 and the e-folding lifetime was about 13 to 17 days. The reconstructed SO2 injection time series were consistent between the AIRS nighttime and the TROPOMI daytime measurements.

We compared the forward transport simulations based on the altitude resolved emission time series with a constant SO2 injection rate. The results show that the modelled SO2 change, driven by chemical reactions, captures the SO2 mass variations from the TROPOMI retrievals. Moreover, the forward simulations reproduce the SO2 distributions in the first ~10 days after the eruption. However, diffusion in the forward simulations is too strong to capture the internal structure of the SO2 clouds, which is further quantified in the simulation of the compact SO2 cloud from late July to early August.

Our study demonstrates the potential of using combined nadir satellite retrievals and Lagrangian transport simulations to further improve SO2 time- and height-resolved injection estimates of volcanic eruptions.

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Acknowledgements

Simulation and Evaluation of the Stratospheric Aerosol Layer Since 2017 in the NASA GEOS Earth System Model

Presenting Author: Peter Colarco[1]

Co-author Names: Sampa Das[1,2], Ghassan Taha[1,2], Nick Gorkavyi[3], Parker Case[4], Omar Torres[1]

[1]NASA Goddard Space Flight Center, Greenbelt, Maryland, USA
[2]University Space Research Association, Columbia, Maryland, USA

[3]Science Systems and Applications, Incorporated, Lanham, Maryland, USA
[4]University of Colorado, Boulder, Colorado, USA

Abstract

The stratospheric aerosol layer has been significantly perturbed by a number of events in the past several years. Beginning with the major British Columbia pyro-Cb event in August 2017, we simulate the stratospheric aerosol loading up to the present day, including the injections of sulfate aerosol precursors from recent volcanic eruptions (i.e., Ambae, Ulawun, and Raikoke) and smoke from additional pyro-Cb events, especially the late 2019/early 2020 Australia wildfires. Simulations are performed in the NASA Goddard Earth Observing System (GEOS) Earth system model with an online aerosol module that is radiatively coupled to the atmospheric dynamics. Results of our simulations are compared to observations from various space-based remote sensing instrument, including from the Ozone Monitoring Profiler Suite (OMPS) Limb Profiler (LP) measurements that use the limb scattering technique to retrieve the vertical profile of aerosol extinction. We estimate the radiative forcing of these aerosol perturbations and discuss uncertainties in our results due to assumptions of aerosol optical properties and composition.

Abstract Title: *in situ* Observations of Aerosol Size Distribution in the Stratosphere and Upper Troposphere from 2021 DCOTSS Mission: Potential Influence of Volcano and Wildfires

Presenting Author title and name: Yaowei Li[1]

Co-author Names: John Dykema[1], Frank Keutsch[1], Jean-Paul Vernier[2], Mike Fromm[3], David Peterson[4], Jessica Smith[1], Jasna Pittman[1], Bruce Daube[1], Steven Wofsy[1], DCOTSS Team

[1] John A. Paulson School of Engineering and Applied Sciences, Harvard University, USA
[2] National Institute of Aerospace, and NASA Langley Research Center, USA

[3] US Naval Research Laboratory, Washington, DC, USA
[4] US Naval Research Laboratory, Monterey, CA, USA

Abstract (100 - 500 Words)

In the upper troposphere and lower stratosphere (UTLS), aerosols modify radiative forcing by scattering or absorbing solar radiation, as well as affecting the nucleation of ice clouds. Also, stratospheric aerosols affect ozone chemistry by modulating the heterogenous reaction rates of halogen species on the particle surface, such as the chlorine activation reactions. Detailed representation of aerosol size distribution is critical in understanding the radiative and chemical impacts of aerosols in the UTLS. The 2021 Dynamics and Chemistry of the Summer Stratosphere (DCOTSS) airborne mission obtained a diverse set of in-situ measurements of aerosol size distribution in the stratosphere and upper troposphere over North America. With the NASA ER-2 high-altitude aircraft platform, aerosol size distribution (130-3000 nm) up to 510 K potential temperature level (22 km) was isokinetically sampled and measured by an optical particle spectrometer (DPOPS instrument). Measurements show a persistent stratospheric aerosol layer between 400-450 K potential temperature (18-20 km) during July and August 2021, which was not present a year before according to balloon-borne measurements (NOAA CSL). Preliminary satellite analysis indicates that this aerosol layer might result from volcanic eruptions which took place on April 2021 from La Soufriere in St. Vincent. A few wildfire smoke encounters from pyrocumulonimbus (pyroCb) events occurred in the upper troposphere. Concurrent measurements of aerosol size distribution and gas tracers (e.g., CO, H_2O, O_3, etc.) within the smoke plumes will be presented.

Acknowledgements

We thank Emrys Hall, Elizabeth Asher, Michael Todt, and Troy Thornberry for providing the balloon-borne data. This work was supported by the NASA grant 80NSSC19K0326 for DCOTSS mission.

A new challenge in stratospheric chemistry: wildfire smoke

Although pyrocumulonimbus towers have long been known to reach the stratosphere and inject particulate matter, their chemical significance has only recently been recognized. I will summarize multiple observations of aerosol and NO2 concentrations from three independent satellite instruments that identify decreases in stratospheric NO2 concentrations following major Australian 2019 through 2020 wildfires. The data confirm that N2O5 hydrolysis did occur on these smoke particle surfaces, with important implications for both composition and ozone depletion. However, additional and more important processes are evident in available satellite data. Those findings will be discussed, and avenues for improvements in understanding will be identified. The results indicate that increasing wildfire activity in a warming world can be expected to slow the recovery of the ozone layer for at least the next several decades.

Tracking the Jan 2022 Hunga-Tonga aerosol cloud using space-based observations

Presenting Author: Dr. Ghassan Taha[1,2]

Co-authors: Robert Loughman[3], Peter Colarco[2], Tong Zhu[4]

[1]Morgan State University, Baltimore, MD USA
[2]NASA Goddard Space Flight Center, Greenbelt, MD USA

[3]Hampton University, Hampton, USA.
[4]Science Systems and Applications, Inc., Lanham, MD USA

Abstract

On January 15, the submarine Hunga Tonga-Hunga Ha'apai volcanic eruption lofted materials high in the upper stratosphere, reaching a record-breaking altitude of ~55 km, unprecedented in the satellite observations era. Within two weeks, the bulk of the injected materials, between 20 – 30 km altitude, circulated the globe, as observed by OMPS LP and CALIPSO. Volcanic emissions can influence the earth's radiative balance and play a role in the chemical and dynamical processes related to ozone destruction in the stratosphere. Initial analysis indicates that this effect may not be as significant as the Pinatubo eruption because of its relatively low sulfur dioxide contents, estimated to be 0.4 Tg.

We use space-based observations of OMPS LP, CALIPSO, and SAGE III to monitor the Hunga-Tonga volcanic plume evolution and transport at different altitudes as it circulates the globe. While the main aerosol layer remains trapped in the tropical pipe, small parts of it have already made it to higher latitudes in the SH. The poleward and downward transport of the aerosol is expected to further increase during the SH winter, which is almost certain to influence this year's ozone hole.

Early evolution of the Hunga-Tonga stratospheric aerosol plume from lidar observations at La Reunion island:

Presenting Author title and name: Dr Alexandre Baron[1]

Co-author Names: Patrick Chazette[2], Nelson Bègue[1], Sergey Khaykin[3] & Valentin Duflot[1,4]

[1]Laboratoire de l'Atmosphère et des Cyclones (LACy), UMR 8105, Université de La Réunion - CNRS - Météo-France, Saint-Denis de La Réunion, France
[2]*Laboratoire des Sciences du Climat et de l'Environnement, LSCE-IPSL, UMR 8212, Université Paris-Saclay - CEA - CNRS - UVSQ, Gif-sur-Yvette, France*
[3]*Laboratoire Atmosphère Milieux Observations Spatiales, LATMOS-IPSL, UMR 8190, Sorbonne Université - UVSQ - CNRS, Paris, France*
[4] *Observatoire des Sciences de l'Univers de La Réunion (OSU-R), UAR 3365, Saint-Denis de La Réunion, France*

Abstract

Explosive volcanism periodically induces atmospheric disturbances up to the stratosphere [1]. These injections of massive amount of ash and aerosol gas precursors, perturb locally the physico-chemical balance of the low et middle stratosphere, in particular the ozone concentration via heterogeneous chemistry on particles. Exceptional eruptions can have a significant influence at the global scale as shown by the Earth radiative budget following the eruptions of El Chichon in 1982 and Mount Pinatubo in 1991 [2].

On January 15, 2022, the Hunga-Tonga volcano erupted in the Tonga archipelago (20.5°S, 175.4°W). The Plinian eruption was of a rare intensity, especially because of the depth of the underwater caldera. The first estimates indicate a power equivalent to ~10 Tg of TNT, probably the most powerful since the eruption of Krakatoa in 1883. This short but intense explosion whose pressure wave was observed all around the globe injected about 0.4 Tg of SO_2 into the stratosphere (to be compared to the 10 to 14 Tg injected during the Mount Pinatubo eruption [3]). The Volcanic Stratospheric Plume (VSP) quickly moved westwards and then overflew the island of La Réunion (21°S, 55°E), located at ~12000 km away from Tonga.

In order to monitor the evolution of the VSP, lidar observations were performed at the Observatoire de Physique de l'Atmosphère de La Réunion (OPAR). This observatory is equipped with lidars capable of stratospheric aerosols measurements at the wavelengths of 355 and 532 nm. First observations were performed every night from 19 to 27 January 2022 when the first passage of the VSP occurred. The plume structures appeared to be highly variable along time, with altitudes decreasing along time from 36 km to 19 km above the mean sea level while plume thicknesses were ranging from ~1 km to more than 3 km. Remarkable aerosol optical depth were associated with these stratospheric aerosol layers, up to 0.8 at 532 nm on January 21.

The temporal evolution of the VSP structure and optical properties will be described and discussed in the light of previous findings [4–6] and in conjunction with available spaceborne observations.

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Acknowledgements

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Abstract Title: The dispersion and evolution of the Hunga Tonga plume in the stratosphere

Presenting Author: B. Legras[1]

Co-author Names: The ASTuS team [P. Sellitto[2], A. Podgajen[1], J. Cuesta[2], C. Duchamp[1], R. Belhadji[2], C. Kloss[3], J.B Renard[3], P. Billant[4], A. Limare[5], T. Roberts[3], A. Nies[3], D. Dritschel[6], G. Berthet[3], F. Jégou[3]] and E. Carboni[7], R. Sidans[7], S. Khaykin[8], M. Boichu[9], L. Blarel[9], F. Ploeger[10], J.-U. Grooβ[10], N. Begue[11]

[1]Laboratoire de Météorologie Dynamique, IPSL, CNRS, ENS-PSL/Ecole Polytechnique/Sorbonne Univ., Paris, Fr
[2]Laboratoire Interuniversitaire des Systèmes Atmosphériques, IPSL, UPEC, CNRS, Créteil, France
[3]Laboratoire de Physique et Chimie de l'Environnement et de l'Espace, CNRS, Univ. Orléans, Orléans, France

[4]LadHyX, CNRS, Ecole Polytechnique, Palaiseau, France
[5]Institut de Physique du Globe de Paris, Paris, France
[6]Mathematical Institute, Univ. St Andrews, UK
[7]Rutherford Appleton Laboratory, Oxford, UK

[8]Laboratoire Atmosphère, Observations Spatiales, UVSQ, CNRS, Sorbonne Univ., St Quentin, France

[9]Laboratoire d'Optique Atmosphérique, CNRS, Université de Lille, Lille, France

[10]Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research (IEK-7), Jülich, Germany

[11]Laboratoire de l'Atmosphère et des Cyclones, Univ. La Réunion, CNRS, La Réunion, France

Abstract (100 - 500 Words)

The Hunga Tonga eruption of 15 January 2022 has sent a massive plume in the stratosphere with a large amount of water vapour which mainly detrained between 26 and 32 km and has been persisting until the end of March. The evolution of the aerosols and water vapour in the Hunga Tonga stratospheric plume has been studied using several satellite instruments (CALIOP, OMPS, IASI, MLS, AEOLUS), AERONET, LOAC measurements and Lagrangian trajectories. Among the two main initial components of the plume at 30 km and 28 km, the conversion of sulphur dioxide primary emissions to secondary sulphate aerosols is completed in 5 days for the first and takes one more week for the second. Sulphur dioxide persists for a longer time at 20-22 km. We discuss the role of water vapour in causing these differences. Ash particles which have been massively washed out during the first hours following the eruption might be present but only as ultra-thin submicronic particles. Due to the fast cooling induced by water vapour longwave emission, the plume descends rapidly until the first week of February after which the cooling decreases due to the dispersion by the meridional shear and the plume almost stalls for more than a month. The plume extends gradually to the tropical pipe but, for a few expelled filaments, stays mostly confined within the 30S-10N band due to the zonality of the summer stratospheric circulation at 25km and the weakness of wave mixing. The first month after the eruption is characterized by a number of dynamically induced compact patches between 20S and 30S. In the second period, the enhanced mixing induces a growth of particles by a seemingly combined effect of water uptake and coagulation. In the diluted phase, slow sedimentation is compensated by net radiative heating.

Acknowledgements

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support from AERIS ICARE Data Center, the resources of the IPSL Meso Center, ESA, NOAA, Copernicus Climate Service and ECMWF.

Balloon-borne observations of stratospheric aerosol following the Hunga Tonga eruption: rapid response measurements and regular soundings

Elizabeth Asher[1,2]

Michael Todt[1,2], Troy Thornberry[2], RuShan Gao[2], Stephanie Evan[3], Jerome Brioude[3], Dale Hurst[1,4], Emrys Hall[1,4], Allen Jordan[1,4], Patrick Cullis[1,4], Kensy Xiong[1,4], Penny Smale[5], Richard Querel[5]

[1] Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, USA

[2] NOAA Chemical Sciences Laboratory, Boulder, USA

[3] Laboratoire de l'Atmosphère et des Cyclones (LACy), CNRS, Université de La Réunion, Saint-Denis, France

[4] NOAA Global Monitoring Laboratory, Boulder, USA

[5] National Institute of Water & Atmospheric Research (NIWA), Lauder, New Zealand

The stratospheric aerosol layer is regularly perturbed by volcanic eruptions of varying magnitudes. On January 15th, 2022 a short but explosive eruption of the Hunga Tonga – Hunga Ha'apai submarine volcano in the South Pacific (20.5° S, 175.4° W) injected ash and ~ 0.4 Tg SO_2 as high as ~30 km. Meteorological balloons carrying lightweight sondes provide one of the only means for in situ sampling of the middle stratosphere and can be deployed on short notice. Here, we present recent observations of aerosol number density and size distributions over La Réunion (21° S, 55.4° E) and Lauder, NZ (45.0° S, 169.7° E) from the Baseline Balloon Stratospheric Aerosol Profiles (B^2SAP) project. We characterize aerosol size distributions from multiple aerosol layers in the early stages of the plume following the eruption as part of the Tonga Volcano Rapid Response Experiment (TR^2EX), analyse detailed snapshots of the evolution and spread of volcanic stratospheric aerosol, quantify the role of the Hunga Tonga's water vapor signature on stratospheric aerosol surface area, and discuss the eruption's impact on aerosol optical properties and chemistry. This series of in situ measurements will provide tight constraints on models simulating the impact of the unique Hunga Tonga eruption on climate and stratospheric chemistry.

Acknowledgements

We would like to thank Jean-Marc Metzger, Paul Walter, Sergio Alvarez, and Alex Baron for their help in launching sondes throughout the night during the initial, targeted campaign at the Maido observatory January 21 – 25th, 2022. We would also like to thank Karen Rosenlof, Melody Avery, and many others for their help in deducing the best times to launch to sample the volcanic plumes based on model simulations and CALIPSO data.

Aerosol characterization within the stratospheric plume from the volcanic eruption at Hunga Tonga January 15th 2022

Corinna Kloss[1]

Nelson Bègue[2], Jean-Baptiste Renard[1], Pasquale Sellitto[3,4], Gwenaël Berthet[1], Alexandre Baron[2], Bernard Legras[5], Richard Siddans[6], Marion Ranaivombola[2], Tjarda Roberts[1], Alexander Nies[1], Aurelien Podglajen[5], Sergey Khaykin[7], Valentin Duflot[2], Guillaume Payen[2], Fabrice Jégou[1]

[1] Laboratoire de Physique et Chimie de l'Environnement et de l'Espace, CNRS, France

[2] *Laboratoire de l'Atmosphère et des Cyclones, Université de La Réunion, France*

[3] *Laboratoire Interuniversitaire des Systèmes Atmosphériques, Université de Paris, France*

[4] *Istituto Nazionale di Geofisica e Vulcanologia, Italy*

[5] *Laboratoire de Météorologie Dynamique, CNRS, Paris, France*

[6] UK Research and Innovation, Rutherford Appleton Laboratory, UK

[7] *Laboratoire Atmosphères, CNRS, France.*

Abstract (100 - 500 Words)

The Hunga Tonga eruption on January 14th and 15th 2022 injected gas, steam and ash plumes into the Upper Troposphere and Lower Stratosphere up to altitude of ~50 km, highly discussed within the SSiRC volcano emailing list. Space-borne observations show that the eruption had the strongest impact on the stratospheric Aerosol Optical Depth since the Pinatubo eruption in 1991. We performed three successful measurement flights with the Light Optical Aerosol Counter (LOAC) within the fresh stratospheric Hunga Tonga plume on January 23rd, 25th and 26th on La Réunion island (21°S). Here, we will present first aerosol size distribution, concentration and typology analysis of the Hunga Tonga plume based on LOAC (version 1.5, Light-weight Optical Aerosol Counter) observations in comparison with aerosol lidar information at OPAR (L'observatoire de physique de l'atmosphère de La Réunion). Lidar and LOAC aerosol observations agree well in terms of plume (peak) altitude identification and timing, but derived extinction tend to show significantly lower values for the aerosol counter. LOAC results show exclusively small aerosol particles (<1 µm in diameter) within the stratospheric plume, indicating freshly nucleated aerosol particles. While satellite observations only identify sulfate particles, LOAC typology information show indications of a small absorbing component for particles < 0.5 µm

Furthermore, we bring our local microphysical LOAC observations in the global context with space-borne observations. Satellite observations are analyzed for the plumes' transport, the global distribution, stratospheric aerosol load and overall microphysical properties..

Interactive stratospheric aerosol simulations of the Hunga-Tonga aerosol cloud re: stronger than expected observed mid-visible stratospheric AOD

Sandip Dhomse[1,2]

Graham Mann[1,3], Sarah Shallcross[1,3], Nicholas Bellouin,[4] Luke Abraham,[5,6] , Ros Hatcher[4,7], Grenville Lister[4,7], John Barnes[8]

[1]School of Earth and Environment, University of Leeds, Leeds, UK,

[2]National Centre for Earth Observation, University of Leeds, Leeds, UK,

[3]National Centre for Atmospheric Science, University of Leeds, Leeds, UK, [4]Department of Meteorology, University of Reading, Reading, UK, [5]Department of Chemistry, University of Cambridge, Cambridge, UK

[6]National Centre for Atmospheric Science, University of Cambridge, Cambridge, UK,

[7]National Centre for Atmospheric Science, University of Reading, Reading, UK,

[8]NOAA Earth System Research Laboratories' Global Monitoring Laboratory, Boulder, USA

Abstract

In this presentation we present initial findings from a series of interactive stratospheric aerosol simulations of the Hunga-Tonga volcanic aerosol cloud with the UM-UKCA composition-climate model. The model experiments apply the same version of the UM-UKCA model used to simulate Agung, El Chichon and Pinatubo aerosol clouds (Dhomse et al., 2020), those runs aligned with the Historical Eruption SO2 emissions Assessment experiment within ISA-MIP (Timmreck et al., 2018).

The "standard" Hunga-Tonga UM-UKCA experiment emits 0.4Tg of SO2 at 29-31km, within a 24-hour period, matching the detrainment duration specified for the ISA-MIP HErSEA experiment protocol.

Following the stronger than expected mid-visible backscatter ratios (BSR) measured by CALIOP, for transects intercepting the Hunga-Tonga plume, and very high BSR values (values in the range 100-

200) observed also from Reunion Island, we also ran UM-UKCA simulations with Hunga-Tonga SO2 emission more than observed, scaling-up by a factor 2, 3 and 4 (0.8, 1.2 and 1.6 Tg of SO2).

As a 1st order metric to evaluate the strength of the zonally-dispersed Hunga-Tonga aerosol cloud, we assess the magnitude and timing of maximum zonal-mean stratospheric 550nm AOD in the southern tropics. Most of interactive stratospheric aerosol simulations for Pinatubo eruption (e.g. Dhomse et al., 2014; Sheng et al., 2015; Mills et al., 2016) consistently over-predict stratospheric AOD compared observation based data sets. Dhomse et al. (2020) indicated that these models may be missing an important removal process such as heterogeneous uptake of SO2 onto fine ash particles, the process recently demonstrated to have removed ~43% more sulphate than SO2-only simulations for the Kelut aerosol cloud (Zhu et al., 2020).

In contrast, for the Hunga-Tonga UM-UKCA simulations with the observed 0.4Tg of SO2, the model tends to under-predict the zonal mean mid-visible stratospheric AOD observed from OMPS-LP, generating maximum zonal-mean 550nm strat-AOD ~0.02-0.03, the observations suggesting this metric exceeded 0.05 consistently between 10 ^{o}S and 20^{o}S from late February into early March (pers. comm. Sergey Khaykin). Comparing the scaled-up SO2 simulations, we find the "factor-3

increased SO2 emission" experiment (1.2 Tg) to best match the observed stratospheric AOD, zonal-mean values in the Southern tropics at ~0.06, but peaking slightly earlier than observed, in late February 2022.

The apparent discrepancy between the observed SO2 and maximum zonal-mean stratospheric AOD might be due the fact that Hunga-Tonga emitted substantial particulate matter within the intensely explosive submarine eruption. Hence evolution of "primary volcanic aerosol" cloud might have affected by marine aerosol from vaporized seawater within the eruptive column (e.g. sea-salt aerosol), an unusual amount of (or influence from) co-emitted ultra-fine ash particles, or "in-plume oxidised sulphate" already converted from SO2 at the time of detrainment (e.g. via aqueous-phase oxidation within water droplets within the plume).

We note these experiments apply the same "free-running approximate QBO-match" initialisation technique applied for the UM-UKCA HErSEA experiments within Dhomse et al. (2020). The approach has a key benefit of enacting a heating effect fully consistent with the emitted SO2 (it does not require nudging to re-analysis fields), and also allows to project forward in time for the full removal of the aerosol, and thereby predict a total volcanic forcing estimate. However, our current approach may not capture initial wind-shear, hence plan repeat the simulations in "pre-nudged accurate-QBO free-running" approach. We will show that nudged simulations to improve the plume shear structure within UM-UKCA interactive Pinatubo aerosol simulations, compared to lidar measurements from Mauna Loa (Barnes and Hofmann, 1997) as highlighted in Shallcross, 2021 (PhD thesis). We will also compare to UM-UKCA Hunga-Tonga experiments which emit these SO2 amounts in just 1 hour, then more consistent with the 15th Jan 2022 eruption.

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Abstract Title: Last Millennium Volcanic Eruptions in the UK Earth System Model: Comparison to PMIP4 Reconstructions and Proxy Records

Presenting Author title and name: Dr Lauren Marshall[1]

Co-author Names: Prof Anja Schmidt[1,2*], Dr Andrew Schurer[3], Dr Luke Abraham[1], Prof Rob Wilson[4], Prof Kevin Anchukaitis[5], Prof Gabi Hegerl[3], Prof Ken Carslaw[6], Prof Piers Forster[7]

[1] Department of Chemistry, University of Cambridge, Cambridge, UK
[2] Department of Geography, University of Cambridge, Cambridge, UK
[3] School of Geosciences, University of Edinburgh, Edinburgh, UK

[4] School of Earth and Environmental Sciences, University of St Andrews, St Andrews, UK
[5] School of Geography, Development and Environment, University of Arizona, city, USA
[6] School of Earth and Environment, University of Leeds, Leeds, UK

[7] Priestley International Centre for Climate, University of Leeds, Leeds, UK

** Now at: Institute of Atmospheric Physics (IPA), German Aerospace Center (DLR), Oberpfaffenhofen, Germany, and Meteorological Institute, Ludwig Maximilian University of Munich, Munich, Germany*

Abstract

Large-magnitude volcanic eruptions have been a major driver of climate variability over the last millennium, but a long-standing mismatch exists whereby the volcanic cooling simulated by climate models is larger than that reconstructed from tree-rings. Here, using the UK Earth System Model (UKESM) and explicitly accounting for aerosol microphysical processes, we investigate the volcanic forcing and climate response for three large-magnitude tropical eruptions (1257 Samalas, 1458 unidentified, 1815 Mt. Tambora) and three unidentified extratropical eruptions (1729, 1329 and 1269) that reflect typical eruptions during the last millennium as identified by ice core sulfate records. Our simulated volcanic forcing differs considerably from the reconstructed forcing used in the Paleo Model Intercomparison Project Phase 4 (PMIP4), with large differences in the spatial distribution of the aerosol for the tropical eruptions. We find good agreement between our UKESM simulated cooling and the most up-to-date hemispheric mean summer temperature tree-ring reconstruction, especially for 1257 Samalas, but the cooling appears to be overestimated when using the PMIP4 forcing in the same model. Our study highlights the importance of the initial dispersion of aerosol for the climatic impact and the importance of detailed aerosol modelling in reconciling simulations and climate records throughout the last millennium.

Acknowledgements

This work was funded by the Natural Environment Research Council grant VOL-CLIM (NE/S000887/1).

Abstract Title: Global Cooling Pause after Tropical Volcanic Eruptions in CMIP5

Presenting Author title and name: Dr Fei Liu [1]

Co-author Names: Chen Xing[1], Bin Wang[2], Deliang Chen[3], Jian Liu[4], and Bin Liu[4]

[1] Earth System Modeling and Climate Dynamics Research Center, Nanjing University of Information Science and Technology, Nanjing 210044, China
[2] *Department of Atmospheric Sciences and Atmosphere-Ocean Research Center, University of Hawaii at Manoa, Honolulu, HI 96825, USA*
[3] *Department of Earth Sciences, University of Gothenburg, 405 30 Gothenburg, Sweden*

[4] Key Laboratory of Virtual Geographic Environment of Ministry of Education, School of Geography Science, Nanjing Normal University, Nanjing 210023, China

We investigate how well the changes in global surface air temperature (SAT) after five major tropical volcanic eruptions during 1870-2005 were simulated in 97 historical and 58 Atmospheric Model Intercomparison Project (AMIP) runs participating in phase five of the Coupled Model Intercomparison Project (CMIP5). In observations there was a pause in global cooling during the first post-eruption boreal winters, represented by a 0.1 K average global mean SAT recovery which, consisting of El Niño-like tropical warming and Eurasian warming, was against the 3-year global cooling after the five eruptions. This pause was simulated by the multi-model ensemble (MME) mean of AMIP runs, while that of historical runs didn't show it due to absent El Niño in the first post-eruption winter. In historical runs, simulation of El Niño was influenced by the initial ocean condition (IOC). An El Niño-like response was simulated when the IOC was not in an El Niño state, but it was much weaker compared to observations. Despite being reproduced by the MME mean of both AMIP and historical runs, the Eurasian warming response was not so strong as in observations. This is because the simulated positive polar vortex response, an important stratospheric forcing for Eurasian warming, was very weak, which suggests the CMIP5 models, and even the Climate Forecast System model, underestimate impacts of volcanic aerosols on the stratosphere. Most of coupled models failed to replicate both El Niño and enhanced polar vortex responses, indicating an urgent improvement of air-sea interaction and stratospheric processes in the models.

This work is supported by the Natural Science Foundation of China (41975107), the Public Science and Technology Research Funds Projects of Ocean (201505013), the National Science Foundation of the US (AGS-1540783), the Strategic Priority Research Program of Chinese Academy of Sciences (XDA20060401), and the National Natural Science Foundation of China (41971108). We acknowledge the World Climate Research Programme's Working Group on Coupled Modelling, which is responsible for CMIP, and we thank the climate modeling groups for producing and making available their model outputs. This paper is ESMC Contribution No. 297.

Size-Dependent Aerosol Climate Forcing and Chemistry in the Lower Stratosphere

NOAA Chemical Sciences Laboratory

Measurements of the size and composition of particles in the lower stratosphere show frequent perturbations not only from moderate volcanic eruptions but also from fires and other sources. These measurements have implications for the climate forcing and chemical impacts of aerosol particles in the lower stratosphere in the present-day stratosphere as well as the implications for volcanic or intentionally added material.

Some of the impacts of particles in the lower stratosphere are climate forcing, surface area for heterogeneous chemistry, infrared heating, production of diffuse light, and possible effects on photolysis. Sulfuric acid particles in the background stratosphere are near the optimal diameter for climate forcing. Mixed tropospheric particles that make their way into the stratosphere are less efficient at climate forcing but have significant surface area for possible heterogeneous chemistry that can affect ozone.

Possible material to be deliberately added to the stratosphere is often compared to the Mount Pinatubo eruption. Sulfuric acid particles after the Mount Pinatubo eruption had relatively little surface area compared to their climate impact. Added material could therefore easily have more impact on heterogeneous chemistry than an analogy to volcanic eruptions would suggest. Infrared heating is more important for aerosol in the lower stratosphere than it is in the lower troposphere. There is no optimum size for added material that simultaneously minimizes all potential side effects such as heterogeneous chemistry and reduction of direct sunlight.

Abstract Title: Potential impact of the 1991 Mt Pinatubo eruption on the mesosphere inferred from HALOE temperature data and UA-ICON simulations

Presenting Author title and name: Sandra Wallis[1,]

Co-author Names: Christoph Hoffmann[1], Hauke Schmidt[2], Christian von Savigny[1]

[1]*University of Greifswald, Felix-Hausdorff-Str. 6, 17489 Greifswald*
[2]*Max Planck Institute for Meteorology, Bundesstrasse 53, 20146 Hamburg*

Abstract

The 1991 eruption of Mt. Pinatubo was the strongest eruption of the last 100 years and emitted between 10-20 Mt SO_2 into the lower stratosphere. This event triggered a global surface cooling of about 0.4-0.5 K and a heating of the tropical lower stratosphere of approximately 2.5-3.5 K. The impact of this eruption on temperature of the mesosphere and mesopause region is, however, less investigated. There are very few data sets of middle atmospheric temperature covering the period around or after the eruption. One of these data sets is provided by the solar occultation instrument HALOE on the Upper Atmospheric Research Satellite (UARS) that started its scientific observations four months after the Pinatubo eruption. Our analysis revealed anomalous positive temperatures at some latitudes in the upper mesosphere during the first months of the HALOE time series. After accounting for the seasonal variations and the solar cycle we fitted an exponential decay function with an e-folding time of 6 months to the HALOE temperatures. This approach suggests a maximum volcanic warming of approximately 6 K in the upper mesosphere. It indicates that HALOE probably measures (only) the decay of a mesospheric perturbation that was potentially forced by the Pinatubo eruption and suggests a rapid response of the mesosphere to the volcanic event. The mechanism that allows a strong tropical volcanic eruption to have an impact on the mesosphere was further investigated by simulations using the upper-atmosphere icosahedral non-hydrostatic (UA-ICON) general circulation model.

Acknowledgements

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Abstract Title: Is it possible to determine aerosol optical depth from historic colour paintings?

Presenting author: Christian von Savigny[1]

Co-author Names: Anna Lange[1], Christoph Hoffmann[1], Alexei Rozanov[2]

[1] Institute of Physics, University of Greifswald, Greifswald, Germany
[2] Institute of Environmental Physics, University of Bremen, Bremen, Germany

Abstract

The idea of estimating stratospheric aerosol optical depth from the twilight colors in historic paintings – particularly under conditions of volcanically enhanced stratospheric aerosol loading – is very tantalizing, because it would provide information on the stratospheric aerosol loading over a period of several centuries. This idea has in fact been applied in a few studies in order to provide quantitative estimates of the aerosol optical depth after some of the major volcanic eruptions that occurred during the past 500 years. Based on radiative transfer simulations with the SCIATRAN model we critically review this approach and come to the conclusion that the uncertainties of the estimated aerosol optical densities are so large that the values have to be considered highly questionable. We show that several auxiliary parameters – which are typically poorly known for historic eruptions – can have a similar effect on the red-green color ratio as a change in optical depth typically associated with eruptions such as, e.g. Tambora in 1815 or Krakatao in 1883. Among the effects considered here, uncertainties in the aerosol particle size distribution have the largest impact on the color ratios and hence the aerosol optical depth estimate. For solar zenith angles exceeding 80°, uncertainties in the stratospheric ozone amount can also have a significant impact on the color ratios. In addition, for solar zenith angles exceeding 90° the color ratios exhibit a dramatic dependence on solar zenith angle, rendering the estimation of aerosol optical depths essentially impossible. We also discuss potential color changes over time due to ageing effects, and whether the initial colors of a painting can be expected to be realistic.

Title: Opportunities and challenges in quantifying the climate response to a volcanic eruption in near real-time

Authors: Anja Schmidt[1,2,3] and Thomas J. Aubry[4,5]

Affiliations:

1 Institute of Atmospheric Physics (IPA), German Aerospace Center (DLR), Oberpfaffenhofen, Germany

2 Meteorological Institute, Ludwig Maximilian University of Munich, Munich, Germany

3 Yusuf Hamied Department of Chemistry, University of Cambridge, Cambridge, United Kingdom

4 Sidney Sussex College, Cambridge, UK

5 Department of Geography, University of Cambridge, Cambridge, UK

Abstract: During eruptive crises, direct hazards posed by volcanic eruptions such as ash fall or tsunamis are a top priority in terms of scientific response and hazard management by authorities. The climate response to an eruption with potential subsequent impacts on local to global scales is another aspect that needs to be considered during an eruptive crisis. There are, however, significant challenges in quantifying the climate response to an eruption in near real-time mainly because of i) the significant computational costs of running complex climate models, which are traditionally used to quantify volcanic climate impacts; and ii) the uncertainties associated with near real-time estimates of volcanic emissions and plume height.

We will discuss the opportunities and challenges that arise from using relatively simple models that can predict the perturbations to aerosol optical depth and to Earth´s energy balance as well as surface temperatures based on the mass, height and latitude of a volcanic sulfur dioxide (SO_2) injection. We showcase how these models have been applied to recent volcanic eruptions such as 2019 Raikoke and 2022 Hunga Tonga-Hunga Ha'apai. We will also present ideas for the development of a webtool, which allows users to calculate the climate response to an eruption in near real-time.

Abstract Title: Modelling volcanic emissions: Vertical distributions and 3D fields – an evaluation using satellite data

Presenting Author title and name: Matthias Kohl[1]

Co-author Names: Christoph Brühl[1], Andrea Pozzer[1], Jennifer Schallock[1]

[1]Max Planck Institute for Chemistry, Mainz

Abstract (100 - 500 Words)

Sensitivity studies of SO_2 emissions following volcanic eruptions are presented using the chemistry climate model EMAC. A new submodel for Explosive Volcanic ERuptions (EVER) was developed, capable of emitting different vertical distributions of trace gases and aerosol species over variable time spans. Emissions of a specified mass of SO_2 in different vertical distributions as well as 3D emissions derived from satellites as performed by Schallock et. al. (2021) are intercompared and evaluated using satellite observations from MIPAS and OSIRIS. Based on the emission inventory by Schallock et. al. (2021) and additional satellite information from IASI, a setup for SO_2 emissions from volcanic eruptions since 2008 was developed that can be used in future studies. Finally, an outlook on potential studies on volcanic ash and tropospheric-stratospheric exchange is given.

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Abstract Title: Impacts of tropical versus extratropical volcanic eruptions with sulphur and halogen injections to the stratosphere

Presenting Author title and name: Zhihong Zhuo[1]

Co-author Names: Herman Fuglestvedt[1], Matthew Toohey[2], Kirstin Krüger[1] [1]Affiliation 1 Department of Geosciences, University of Oslo, Oslo, Norway

[2]Affiliation 2 Institute of Space and Atmospheric Studies, University of Saskatchewan, Saskatoon, Canada

Abstract (100 - 500 Words)

Explosive volcanic eruptions cause significant surface cooling and stratosphere warming due to increased sulphate aerosols in the stratosphere. Tropical volcanic eruptions were widely understood to have a larger climate impact than extratropical volcanic eruptions. However, recent analyses of tree ring northern hemisphere (NH) temperature reconstructions and aerosol climate model simulations revealed stronger hemispheric cooling and larger hemispheric stratosphere aerosol optical depth (SAOD) relative to estimated sulphur emissions from past NH extratropical volcanic eruptions compared to tropical eruptions[1].

Here we newly investigate how the co-injection of sulphur and halogens affect the impacts of tropical versus extratropical eruptions using a fully coupled Earth System Model with an interactive ocean. For this, we have performed ensemble simulations based on the Community Earth System Model version 2 (CESM2)[2] with the version 6 of the Whole Atmosphere Community Climate Model (WACCM6)[3]. CESM2-WACCM6 goes up to ~140 km altitude and includes prognostic stratospheric aerosol and chemistry[4]. We simulate explosive eruptions at 15 N and 64 N in January injecting 17 Mt of SO_2 together with halogens at 24 km altitude, taking different initial conditions, i.e., the quasi-biennial oscillation, El Niño–Southern Oscillation and polar vortex phases into account. The halogen masses are based on Central American Volcanic Arc petrological measurements[5], scaled to Pinatubo strength, and assume a 10% injection efficiency to the stratosphere[6,7]. The model results indicate the same maximum global mean SAOD, but a larger NH extratropical mean SAOD after NH extratropical eruptions compared to tropical eruptions. The NH extratropical eruptions lead to larger maximum NH extratropical mean surface cooling and ozone depletion compared to tropical eruptions. Overall, our CESM2-WACCM6 model results confirm the tree ring and model results for the surface temperature and SAOD from Toohey et al (2019)[1], but show however differences in the magnitude and time evolution of aerosol size, aerosol burden and SAOD for tropical and extratropical eruptions.

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Interactive Stratospheric Aerosol models response to different amounts and altitude of injection of SO_2 during the Pinatubo eruption

Presenting Author title and name: Ilaria Quaglia[1,2]

Co-author Names: Claudia Timmreck[3], Ulrike Niemeier[3], Daniele Visioni[2], Giovanni Pitari[1], Christoph Brühl[4], Sandip Dhomse[5], Henning Franke[3,6], Anton Laakso[7], Graham Mann[5,8], Michael Mills[9], and Timofei Sukhodolov[10]

[1]*Department of Physical and Chemical Sciences, Università dell'Aquila, 67100 L'Aquila, Italy*

[2]*Sibley School of Mechanical and Aerospace Engineering, Cornell University, Ithaca, NY, USA*

[3]*The Atmosphere in the Earth System, Max Planck Institute for Meteorology, Hamburg, Germany*

[4]*Max Planck Institute for Chemistry, Mainz, Germany*

[5]School of Earth and Environment, University of Leeds, Leeds, U.K

[6]*International Max Planck Research School on Earth System Modelling, Hamburg, Germany*

[7]*Finnish Meteorological Institute, Kuopio, Finland*

[8]*UK National Centre for Atmospheric Science, University of Leeds, Leeds, UK*

[9]*Atmospheric Chemistry Observations and Modeling, National Center for Atmospheric Research, Boulder, CO, USA*

[10]*Physikalisch-Meteorologisches Observatorium Davos World Radiation Center, Davos, Switzerland*

Abstract (100 - 500 Words)

Previous model intercomparison studies showed model discrepancies in reproducing the climatic impact of large explosive volcanic eruptions within the VolMIP Tambora-ISA experiment [1, 2], and Stratospheric Aerosol Injection strategies [3], calling into question the reliability of model simulations for future scenarios. Here, we analyse the well-known model disparities in the simulation of the evolution of the aerosol plume by comparing model results with several observations, in order to investigate which are, for each model, the main mechanisms that cause these differences.

The analysis concerns the uncertainties in the initial emission of the Pinatubo eruption in 1991, following the protocol of the Historical Eruptions SO2 Emission Assessment experiments (HErSEA), in the frame of the model intercomparison project ISA-MIP [4].

Six climate models with interactive aerosol microphysics (ECHAM6-SALSA, EMAC, ECHAM5-HAM, SOCOL-AER, ULAQ-CCM, UM-UKCA) performed different simulations of the volcanic aerosol cloud exploring the uncertainties in the amount of SO_2 injected, that ranges between 5 and 10 Tg-S, and the altitude of injection, estimated between 15-20 km.

We find that the common and main weakness of all models consist in reproducing the persistence of the sulfate aerosols in the stratosphere, which results in the overestimation of the initial burden and in a shorter e-folding time after reaching the maximum value, compared to the observations. The vertical distribution of the effective radius a few months after the eruption agrees with measured values in the Tropics and over Laramie, showing that 7 Tg-S of SO_2 to be injected in order to have a comparable radius produces a number of particles too high in the initial phase. In the following

months, the e-folding time of the sulfate burden simulated by the models ranges between 10 and 14 months, depending on the simulation, compared to 20 and 21 months of HIRS and SAGE II measurements, respectively. Most models also show a stronger transport in the northern hemisphere, at the expense of the observed tropical confinement.

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The SSiRC Data Rescue Initiative:

Recovering lost or forgotten stratospheric lidar and searchlight datasets to provide new constraints for stratospheric change and volcanic impacts

Juan Carlos Antuña-Marrero[1], Graham Mann[2], John Barnes[3], Sarah Shallcross[2],

Sandip Dhomse[2], Abel Calle[1], Victoria Cachorro[1], Terry Deshler[4], Li Zhengyao[2]and Nimmi Sharma[5]

[1]Group of Atmospheric Optics (GOA-UVa), University of Valladolid, Spain *email: antuna@goa.uva.es*

[2]School of Earth and Environment, University of Leeds, Leeds, UK.

[3]NOAA ESRL Global Monitoring Laboratory, Boulder, CO, USA

[4]Laboratory of Atmospheric and Space Physics, University of Colorado, Boulder, CO, USA,

Abstract: *Climate models simulating the climatic effects of volcanic eruptions in the so-called "post-in-situ pre-satellite era" (~1957-1979) rely primarily on stratospheric aerosol optical depth (sAOD) values derived from ground based observations of solar radiation: direct solar beam, diffuse and total radiation, stellar extinction and lunar brightness during eclipses [1,2]. However, the vertical profile of the stratospheric aerosol layer was measured from the ground via active remote sensing instruments since the early 1960's, both lidar [3,4], and searchlight [5,6], and also from high altitude balloon [7,8,9].*

The Data Rescue Activity of the Stratospheric Sulfur and its Role in Climate (SSiRC) (http://www.sparc-ssirc.org/ Activities à Data Rescue) has thus far focused mainly on lidar and searchlight datasets within major volcanic periods, aiming to provide new constraints for interactive stratospheric aerosol models and improve the fidelity of volcanic forcings in climate model. The activity also identified and recovered a dataset of balloon borne solar extinction soundings from 1965-67 [10], also identifying recoverable datasets for U-2 aircraft impactors [10,11] and ozonesondes profiles [12,13,14] during the 1963-65 post-Agung period.

Here we present, two recovered active remote sensing datasets that measured the progression of the vertical profile of the northern hemisphere dispersed portion of the Agung volcanic aerosol cloud during 1963-65.

Firstly, a total of 66 nights of lidar profiles of attenuated backscatter at 694nm measured at Lexington (42°N), Massachusetts between January 1964 and July 1965 were recovered from Tables within an MIT PhD thesis processed to derive aerosol extinction profiles ( ) at 532 nm. Another 9 lidar profiles of attenuated backscatter at 694nm measured at College (64°N), Alaska in July and August 1964 [3,4] have been recalibrated to produce at 532 nm. Both lidar datasets have been published as peer-reviewed journal article [15,20], and in a public data repository [16].

Secondly, a searchlight dataset of the Agung aerosol cloud is also presented, recovered by re-digitising Figures within an "Atlas of aerosol attenuation" report [17] documenting observations at White Sands (32°N), New Mexico. The dataset of 105 profiles on 36 nights between December 1963 and December 1964 have been recalibrated, contemporaneous measurements of tropospheric aerosol and stratospheric ozone used to recover with particular emphasis on the tropospheric and stratospheric aerosol phase functions (APF) [18]. The original APF, used from the surface to 35km, was replaced in the troposphere by the APF from the White Sands AERONET site [19] and in the

stratosphere by an AFP computed from four particle size distributions measured by an aircraft at 18km between April and August 1963 around 32°N [11].

Accounting accurately for variations in tropospheric aerosol and ozone leads to substantial change in the re-calibrated searchlight dataset. Maximum values of in the original and the recalibrated dataset were $5.7x10^{-3}$ km^{-} and $8.2x10^{-3}$ km^{-1} respectively, a 42% increase. The original sAOD monthly mean was 0.027, the recalibrated 0.033, a 23% of increase. Recalibrated sAOD reasonably agree with sAOD measured at Mauna Loa, 20°N and Mt Locke, Texas, 31°N [20].

The already rescued and recalibrated datasets provide new information about the spatiotemporal structure and optical properties of the 1963 Agung SA at a subtropical, a midlatitude and a polar site in the northern hemisphere.

[5]Department of Physics and Earth Sciences, Central Connecticut State University, CT, USA

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Abstract Title: Sensitivity of a Fresh Volcanic Cloud to Mass of Volcanic Debris, Height of Injection, Presence of Ash, and Water Vapor

Prof. Georgiy Stenchikov[1]

Alex Ukhov[1]

[1]Division of Physical Sciences and Engineering, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia

Abstract

Global climate models largely miss the initial evolution of multicomponent (SO2, SO4, ash, H2O) fresh volcanic clouds due to insufficient spatial resolution and a lack of relevant physics.

We modified a regional WRF-Chem model to study the dispersion of a volcanic cloud in a latitude belt with 25-km grid spacing explicitly accounting for SO2, ash, sulfate, water vapor, and hydrometeors radiative feedbacks, assuming various parameters of injection.

We found that volcanic clouds' AOD and spatial-temporal evolution are sensitive to the initial concentrations and vertical distributions of volcanic materials, mainly volcanic ash and SO2. The volcanic eruption of a different magnitude from the Pinatubo size to one-two orders of magnitude more minor eruptions, like Raikoke, Kelud, or Hunga-Tonga, exhibit qualitatively different radiative feedbacks. The effect of injected water vapor is controlled by the temperature at the injection height. E.g., volcanic water released at the cold point above the tropical tropopause converts to ice and deposits almost wholly. The altitude of a matured volcanic cloud depends not only on the height of injection but is defined by radiative heating and lofting of a fresh cloud.

Our results show that the initial evolution of volcanic debris is essential, as a freshly developed volcanic cloud's vertical structure, composition, and altitude affect its long-term change. Utilizing a more realistic description of the volcanic cloud's initial stage improves overall volcanic cloud simulation.

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Simulating major volcanic aerosol clouds as global dust veils

Graham Mann[1,2], Geraint Vaughan[3], Sarah Shallcross[1,2], Juan Carlos Antuna-Marrero[5], Sandip Dhomse[1,4], Anja Schmidt[6,7], Ryan Neely[1,2], Ken Carslaw[1], Nicolas Bellouin[8], David Winker[9], Stuart Young[10]

[1]*School of Earth and Environment, University of Leeds, Leeds, UK,*
[2]*National Centre for Atmospheric Science, University of Leeds, Leeds, UK,*
[3]*National Centre for Atmospheric Science, University of Manchester, Manchester, UK,*
[4]*National Centre for Earth Observation, University of Leeds, Leeds, UK,*
[5]*Departamento de Física Teórica, Atómica y Óptica, Universidad de Valladolid, España,*
[6]*Departments of Chemistry, University of Cambridge, Cambridge, UK,*
[7]*Department of Geography, University of Cambridge, Cambridge, UK,*
[8]*Department of Meteorology, University of Reading, Reading, UK,*
[9]*NASA Langley Research Center, Hampton, Virginia, USA,*
[10]*CSIRO Oceans and Atmosphere Flagship, Aspendale, Victoria, Australia.*

Abstract

Lidar observations from tropical aircraft surveys 4 weeks after the Pinatubo eruption [1,2] show the lowermost extent of the highly heterogeneous layers present at this time were substantially depolarising (5-10%), indicating the presence of solid particles within the predominantly liquid sulphuric acid droplet aerosol. Moderate depolarisations (a few percent) were also present throughout the highly backscattering layers detected.

Ground-based lidar measurements from Aberystwyth, U.K. [3] from August 1991 to March 1992 provide a potentially unique record of the progressing morphology of the Pinatubo cloud, as it dispersed to the Northern Hemisphere. Whereas initial 1-2km-depth layers of the cloud had little depolarisation, in September a 4-6km-depth cloud in September showed a clear signal of moderate (1-2%) depolarisation.

Frequent lidar measurements from Aberystwyth were made throughout October and November, the soundings detecting the deepening of the mid-latitude-dispersed cloud to 10km vertical extent, measured also from Northern Hemisphere mid-latitude balloon soundings [4] and captured in interactive stratospheric aerosol model simulations [5]. Whereas the volcanic aerosol cloud essentially doubled in vertical extent, the depolarising lowermost portion remained contiguous within a few km depth, suggesting a continuing export from the base of the tropical volcanic aerosol reservoir with different (mixed-phase) particle morphology.

The Aberystwyth measurements show a slow descent of the moderate-depolarisation base-portion of the dispersing cloud, a steady shallowing eventually progressing to the 10km depth cloud becoming a clear non-depolarising signal in March 1992.

Impactor samples from the NASA ER-2 and DC-8 surveys of the Northern Hemisphere mid-latitude lower stratosphere in Jan and Feb 1992 [6] measured the latter phase of this steady progression in Pinatubo aerosol particle morphology. Impactor samples from the ER-2 flight altitude (18-20km) showed the cloud there consisted of sulphuric acid droplets, whilst those from the DC-8 altitude (10-12km) show the lowermost plume contained also internal mixtures of ash and sulphuric acid.

Overall, the measurements suggest that after the Pinatubo cloud had matured into a deep tropically-confined volcanic aerosol reservoir, its lowermost portion retained a mixture of ash and sulphuric acid particles for around 9-12 months after the eruption.

This presentation will explain the adaptation of the interactive stratospheric-tropospheric aerosol model UM-UKCA [5,7,8] to represent major volcanic aerosol clouds consistently with this conceptual picture of a mixed plume of ash-sulphuric and pure sulphuric aerosol particles.

Lidar measurements from CSIRO Aspendale [9,10] show the base of the Southern Hemisphere dispersed Pinatubo cloud was similarly depolarising. Furthermore, we note that U-2 aircraft surveys of the El Chichon (e.g.[11,12]) and Agung aerosol clouds (e.g. [13,14]) suggest the ash may also have remained present for a similar timescale. The findings also link to the new SSiRC data-rescue activity, which is beginning to recover measurement datasets of in-situ and remote sensing aerosol measurements within major volcanic clouds (http://www.sparc-ssirc.org Activities à Data Rescue).

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Abstract Title: A rapid response to the Hunga-Tonga eruption; the Tonga Rapid Response Experiment (T^2REx)

Dr. Karen H. Rosenlof[1] and the T^2REx team

[1]NOAA Chemical Sciences Laboratory, Boulder, CO USA

Abstract (100 - 500 Words)

On January 15, the Hunga Tonga - Hunga Ha'apai volcano north of Tongatapu in the south west Pacific erupted injecting material into the stratosphere to altitudes up to or higher than 30 km. Estimates are that it injected 0.4 Tg of sulfur dioxide into the stratosphere and 50 to 100 Tg of water vapour. The mass injected was much smaller than that of Pinatubo, but the water injection was unprecedented over the satellite era.

There have not previously been in situ measurements made in a volcanic plume immediately after the eruption. To parameterize the processes involved in the conversion of SO_2 to sulfate aerosol, it is advantageous to get measurements shortly after eruption and ideally extend those measurements in time as the plume evolves. To achieve that goal, NOAA CSL and collaborators rushed to get people and instruments to a site downwind of the eruption to sampling the volcanic plume. The Hunga-Tonga volcano is at ~20 S. The wind was blowing the stratospheric plume towards the Maïdo climate observatory 8000 miles to the west, at ~21 S on Réunion Island.

In just a few days, instruments were prepared, travel arrangements were made, and our colleagues on Réunion Island were able to make baseline balloon and lidar measurements and arrange supplies for multiple balloon launches when the plume was within reach. The plume and the US scientists arrived at approximately the same time, and measurements were made over the course of 5 nights. In this presentation, I will provide an overview of the measurements taken and highlight some of the modeling studies presently underway.

Abstract Title: Balloon-based SO_2 measurements during the Hunga Tonga eruption

Presenting Author title and name: Paul Walter[1]

Co-author Names: James H. Flynn[2], Sergio L. Alvarez[2], Jerome Brioude[3], Stephanie Evan[3], Jean-Marc Metzger[4], Gary A. Morris[5], Subin Yoon[2], Alex Kotsakis[6], Elizabeth Klovenski[2], Mark D. Spychala[7], Ernesto Coralles[8], Alfredo Alan[8], Jorge A. Diaz[9], and Jonathan Harnetiaux[10]

[1]St. Edward's University, Austin, TX, 78704, USA
[2]*Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, 77004, USA*
[3]*Laboratoire de l'Atmosphère et des Cyclones, UMR8105, CNRS, Université de La Réunion, 97744 Saint-Denis, France*
[4]*UAR 3365 – OSU Réunion, Université de La Réunion, Saint-Denis, Réunion, France*
[5]*NOAA Global Monitoring Laboratory, Boulder, CO, 80305, USA*
[6]*ERT, Inc., Laurel, MD, 20707, USA*
[7]*Hamelmann Communications, Pagosa Springs, CO, 81147, USA*
[8]*GasLAB, CICANUM. Universidad de Costa Rica, San José, Costa Rica*
[9]*INFICON, East Syracuse, NY, 13057, United States*
[10]*En-Sci, Westminster, CO, 80234, USA*

Abstract (100 - 500 Words)

A novel technique has been developed to measure sulfur dioxide (SO_2) using a modification of the existing electrochemical concentration cell (ECC) ozonesonde technology. The previous sonde-based method to measure SO_2 (i.e. the dual-sonde approach) has large uncertainties in the stratosphere that would limit its effectiveness in measuring SO_2 from an explosive volcanic eruption. Due to that and other limitations, several modifications were made to create a single-sonde system that would directly measure SO_2 (i.e. the SO_2 sonde). These modifications included (1) a positively biased ECC background current, (2) the addition of an O_3 removal filter, and (3) the addition of a sample dryer. The SO_2 sonde measures SO_2 as a reduction in the cell current. Field tests in 2018 included lower tropospheric measurements near Kīlauea Volcano (before and during the 2018 eruption in the Lower East Rift Zone), Costa Rica's Turrialba Volcano, and anthropogenic plumes from the Athabasca Oil Sands region of Alberta, Canada. The SO_2 sonde was deployed to La Réunion as part of the Tonga volcano Rapid Response Experiment (TR^2Ex). During 21-25 January 2022, the SO_2 sonde sampled SO_2 in the stratospheric Hunga Tonga volcanic plume during four free-release balloon flights. Two of those soundings also had ozonesondes, and notches in the ozone data were observed in the plume. The altitudes of those plumes ranged from 19 to 30 km. We will overview the SO_2 sonde measurements during the initial TR^2Ex deployment and compare the SO_2 sonde and ozonesonde measurements.

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MLS observations of SO_2 and H_2O from the Hunga Tonga eruption

Hugh C Pumphrey (University of Edinburgh)

The MLS team (NASA JPL)

The eruption of Hunga Tonga - Hunga Ha'apai on 15 January 2022 deposited large amounts of water vapour and sulfur dioxide in the stratosphere. We report on measurements of these species made by the MLS instrument on the Aura platform following the eruption. The total mass of SO_2 deposited in the stratosphere was 410Gg, in agreement with early reports from nadir-sounding instruments. The SO_2 was deposited at altitudes higher than any other eruption since the launch of Aura in 2004 and has decayed with an e-folding time of 10 days: a greater rate than for most other eruptions. The total mass of water vapour deposited in the stratosphere was 145 Tg. No event of this magnitude has been observed in the H_2O data during the entire MLS mission. The water vapour mixing ratios immediately after the eruption were sufficiently large as to prevent the MLS retrieval software working properly and many profiles are flagged as unusable. By three weeks after the eruption, the number of bad profiles had returned to the pre-eruption level, but the excess mass of water vapour had not changed by an observable amount.

Abstract Title: In-situ observations of water vapor and ozone following the Hunga Tonga eruption

Presenting Author title and name: Stephanie Evan[1]

Co-author Names: Jerome Brioude[1], Jean-Marc Metzger[2], Karen Rosenlof[3], Holger Vomel[4], Ryan M. Stauffer[5], Kevin Lamy[1], Paul Walter[6], James H. Flynn[7], Sergio L. Alvarez[7].

[1]Laboratoire de l'Atmosphère et des Cyclones, UMR8105, CNRS, Université de La Réunion, 97744 Saint-Denis, France

[2]UAR 3365 – OSU Réunion, Université de La Réunion, Saint-Denis, Réunion, France

[3]NOAA Global Monitoring Laboratory, Boulder, CO, 80305, USA

[4] National Center for Atmospheric Research, Boulder, Colorado, 80301, USA

[5] NASA Goddard Space Flight Center, Greenbelt, Maryland, United States of America

[6]St. Edward's University, Austin, TX, 78704, USA

[7]Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, 77004, USA

Abstract

The eruption of the submarine volcano Hunga Tonga on 15 January 2022 injected a large amount of water vapor directly into the stratosphere. There have not previously been in situ measurements made in a volcanic plume immediately after an eruption. Here we report observations of balloon-borne instruments of water vapor and ozone over the southwestern Indian Ocean a week after the eruption. Data from cryogenic frost point hygrometer (CFH), Compact Optical Backscatter and AerosoL Detector (COBALD) and electrochemical concentration cell (ECC) ozonesonde instruments were used to characterize the volcanic plume before/after the eruption. Extremely high CFH water measurements (up to 300ppmv) in the stratosphere were observed at Reunion Island on 22 January. In situ water vapor observations and Lagrangian modeling is employed to estimate the mass of water vapor injected in the stratosphere by the eruption. Possible ozone loss was also observed in the ozonesonde data in the week following the eruption. Ozonesonde data from Réunion Island taken in the months before and after the eruption support the observations of ozone loss. Observations of water vapor and ozone shortly after the eruption presents a challenge to remote sensing instruments and we present here water vapor/ozone observations following the eruption never reported before.

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Fast circumglobal transport of stratospheric aerosol and moisture produced by Hunga Tonga eruption

Presenting Author: Sergey Khaykin[1]

Co-author Names: F. Ploeger[2], J.-U. Grooβ[2], A. Podglajen[3], F. Tence[1], A. Baron[4], N. Begue[4], S. Bekki[1], V. Duflot[4,5], S. Godin-Beekmann[1], J. Jumelet[1], C. Kloss[6], B. Legras[3], R. Querel[7], J.-B. Renard[5], T. Sakai[8], P. Sellitto[9,10], G. Taha[11,12]

[1]Laboratoire Atmosphère, Observations Spatiales, UVSQ, CNRS, Sorbonne Univ., St Quentin, France

[2]Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research (IEK-7), Jülich, Germany

[3]Laboratoire de Météorologie Dynamique, IPSL, CNRS, ENS-PSL/Sorbonne Univ., Paris, France

[4]Laboratoire de l'Atmosphère et des Cyclones, Univ. La Réunion, CNRS, La Réunion, France

[5]UMS 3365 – OSU Réunion, Université de La Réunion, Saint-Denis, Réunion, France
[6]Laboratoire de Physique et Chimie de l'Environnement et de l'Espace, CNRS, Univ. Orléans, Orléans, France

[7]National Institute of Water & Atmospheric Research (NIWA), Lauder, New Zealand

[8]Meteorological Research Institute (MRI-JMA), Tsukuba, Japan

[9]Laboratoire Interuniversitaire des Systèmes Atmosphériques, IPSL, UPEC, CNRS, Créteil, France

[10]Istituto Nazionale di Geofisica e Vulcanologia, Osservatorio Etneo, Catania, Italy

[11]Morgan State University, Baltimore, MD USA

[12] Atmospheric Chemistry ad Dynamics Laboratory, NASA Goddard Space Flight Center, Maryland, USA

Abstract

The eruption of the underwater Hunga Tonga volcano in Central Pacific on 15 January 2022 was associated with a powerful blast that injected sulphur and water deep into the stratosphere. Here we study the early evolution and global transport of aerosol and moisture plumes produced by this eruption using satellite and ground-based observations coupled with a chemistry-transport simulation. In particular, we use aerosol extinction and backscatter measurements by OMPS-LP and CALIOP as well as ground-based lidar observations at various stations: La Reunion in the tropical Indian ocean, Lauder in New-Zealand and Dumont d'Urville in Antarctica. The water vapour measurements by Aura MLS are used to describe the transport of moist plumes and to quantify the large-scale perturbation of stratospheric water budget. Additionally, we use meteorological radiosoundings at various tropical locations in support of the MLS detections of water vapour enhanced layers.

A detailed insight into the global transport of moist and aerosol-rich plumes is provided by CLaMS chemistry-transport model simulation initialized by MLS H_2O data before and after the eruption. The results of the simulation, spanning 10 weeks are found in good agreement with the observations and reveal a fast poleward transport of weak plumes, which is confirmed by early detections of aerosol layers in the lower stratosphere above New Zealand and Antarctica 3-4 weeks after the eruption. Both MLS and CLaMS show that the upper stratospheric moist plumes, carried by strong zonal winds, have circumnavigated the globe in only 8 days. The bulk of aerosol and moisture is found in the

middle stratosphere, where it was subject to northbound transport and crossed the equator in about one week after the eruption.

Finally, using satellite records of aerosol extinction, we show that the stratospheric AOD perturbation by Hunga Tonga eruption exceeds the previous XXI century record produced by the Black Summer Australian wildfires in 2019/2020 and represents a 30-yr high. The perturbation of stratospheric water budget is estimated at around 75 Mt, which translates into a global anomaly in stratospheric water vapour column of about 12%. The planetary scale perturbation of stratospheric aerosol and water budget may have substantial repercussions for atmospheric radiative balance and chemical composition.

Abstract Title: Monitoring stratospheric aerosols in South America using a ground-based lidar system after the Hunga Tonga Eruption

Presenting Author title and name: Dr. Eduardo Landulfo[1]

Co-author Names: Fábio J. S. Lopes[1], Juan Carlos Antuña[2], René Arredondo[3,4], Ghassan Taha[5,6]

[1]Affiliation 1 Instituto de Pesquisas Energéticas e Nucleares, IPEN, Brazil

[2]*Affiliation 2 Group of Atmospheric Optics (GOA-UVa), University of Valladolid,Valladolid, Spain*

[3]*Affiliation 3 Instituto Geofísico del Perú, Lima, Peru*

[4]*Affiliation 4 Departamento de Radiación Solar y Óptica Atmosférica, INSMET*

[5]*Affiliation 5 Morgan State University, Baltimore, MD, USA*

[6]*Affiliation 6 NASA GSFC, Greenbelt, MD, USA*

Abstract

The volcanic plume transport from the Hunga-Tonga Hunga Ha'apai (HTHH) volcano, a submarine volcano in the South Pacific erupted on December 20, 2021 and its plumes soon traveled around the globe towards South America. The HTHH's volcanic plume in its initial blast reached above 50 km into the mesosphere, and a lower plume concentrated around 25-30 km. The lidar system at São Paulo, Brazil, part of Latin-America LIdar NETwork (LALINET)[1], the SPU-Lidar station, was employed to measure profiles of the particle extinction and backscatter coefficients as well detect the plume geometrical features such as base/top heights and plume thickness. The lidar system operated at 1064, 532 and 355 nm and the backscatter signal from these channels enabled us to get the volcanic optical properties. The HTHH plumes were mostly detected between 24 to 27 km (a.g.l.) and 18 km (a.g.l.). The results to be shown evaluate the temporal evolution of ash/plume optical properties such as aerosol optical depth (AOD) and lidar ratio (LR) for each different layer, in the stratosphere as the plume and volcanic ash settling took about several weeks. The transmittance method[2] was applied to extract volcanic layers optical properties in the stratosphere and correlative sunphotometer data also located at São Paulo was taken into account. To validate and inspect the plume evolution CALIOP[3], AEOLUS[4] and OMPS LP[5] data were also employed. The event is a strong indicator of the potential to extend monitoring activities within Latin America Lidar Network up to UTLS and thus enhance the network capability and open new pathways into exploring atmospheric couplings between the troposphere and atmosphere[7].

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Hunga-Tonga: a water rich stratospheric volcanic eruption – what does it mean to stratospheric chemistry and aerosol optics

Yunqian Zhu[1,2], Charles Bardeen[1], Michael Mills[1], Simone Tilmes[1], Lynn Harvey[2], Taha Ghassan[3], Corinna Kloss[4], Jadwiga Richer[1], Sasha Glanville[1], Pengfei Yu[5], Doug Kinnison[1], Karen Rosenlof[6], Brian Toon[2,7]

The 2022 Honga-Tonga Ha'apai Hunga volcanic eruption injected SO2 and massive water into the stratosphere. The water injection results in a short SO2 lifetime, enhanced ice cloud during the first couple of days, and a long-lasting enhanced SO4 extinction. H2O fast converts to OH and depletes SO2 showing a consistent SO2 lifetime with satellite observations. Sulfate swelled with water also explained the large extinction observed by OMPS in a month. We conduct a seasonal forecast and predict a 30% ozone depletion in October caused by this eruption.

The outflow of Asian biomass burning carbonaceous aerosol into the upper troposphere and lower stratosphere in spring: radiative effects seen in a global model

Prashant Chavan[1,2], Suvarna Fadnavis[1*], Tanusri Chakroborty[1], Christopher E. Sioris[3], Sabine Griessbach[4], and Rolf Müller[5]

[1]Indian Institute of Tropical Meteorology, Center for climate change, MoES, India

[2]Department of Atmospheric and Space Sciences, Savitribai Phule Pune University, Pune, India

[3]Air Quality Research Division, Environment and Climate Change, Toronto, Canada

[4]Forschungszentrum Jülich GmbH, Jülich Supercomputing Center, Jülich, Germany

[5]Forschungszentrum Jülich GmbH, IEK7, Jülich, Germany

Biomass burning (BB) over Asia is a strong source of carbonaceous aerosols during spring. From ECHAM6–HAMMOZ model simulations and satellite observations, we show that there is an outflow of Asian BB carbonaceous aerosols into the upper troposphere and lower stratosphere (UTLS) (black carbon: 0.1 to 6 ng m^{-3} and organic carbon: 0.2 to 10 ng m^{-3}) during the spring season. The model simulations show that the greatest transport of BB carbonaceous aerosols into the UTLS occurs from the Indochina and East Asia region by deep convection over the Malay Peninsula and Indonesia. The increase in BB carbonaceous aerosols enhances atmospheric heating by 0.001 to 0.02 K d^{-1} in the UTLS. The aerosol-induced heating and circulation changes increase the water vapor mixing ratios in the upper troposphere (by 20–80 ppmv) and in the lowermost stratosphere (by 0.02–0.3 ppmv) over the tropics. Once in the lower stratosphere, water vapor is further transported to the South Pole by the lowermost branch of the Brewer–Dobson circulation. These aerosols enhance the in-atmosphere radiative forcing (0.68±0.25 to 5.30±0.37 W m^{-2}), exacerbating atmospheric warming, but produce a cooling effect on climate (top of the atmosphere – TOA: −2.38±0.12 to −7.08±0.72 W m^{-2}). The model simulations also show that Asian carbonaceous aerosols are transported to the Arctic in the troposphere. The maximum enhancement in aerosol extinction is seen at 400 hPa (by 0.0093 km^{-1}) and associated heating rates at 300 hPa (by 0.032 K d^{-1}) in the Arctic.

publication.

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Interannual variability of aerosols in the UTLS and its connection to anthropogenic sources and Asian summer monsoon variability: Results from AeroCom models and observations

Presenting Author title and name: Mian Chin[1]

Co-author Names: Huisheng Bian[1,2], Qian Tan[3], Valentina Aquila[4], Xiaohua Pan[1,5], Ghassan Taha[1,2], Landon Rieger[5], Adam Bourassa[5], Doug Degenstein[5], John Burrows[6], Alexei Rozanov[6], Jayanta Kar[7], Jean-Paul Vernier[7], Simon Carn[8], Toshihiko Takemura[9], Hitoshi Matsui[10], Kostas Tsigaridis[11], Susanne Bauer[11], Paul Ginoux[12], Suvarna Fadnavis[13], Anton Laakso[14]

[1]*NASA Goddard Space Flight Center, USA*
[2]*University of Maryland at Baltimore County, USA*
[3]*Bay Area Environmental Research Institute, USA*

[3]*American University, USA*

[4]*ADNET, USA*

[5]*University of Saskatchewan, Canada*

[6]*University of Bremen, Germany*

[7]*NASA Langley Research Center, USA*

[8]*Michigan Tech University, USA*

[9]*Kyushu University, Japan*

[10]*Nagoya University, Japan*

[11]*NASA Goddard Institute of Space Studies, USA*

[12]*Geophysical Fluid Dynamics Laboratory, USA*

[13]*Indian Institute of Tropical Meteorology, India*

[14]*Finnish Meteorological Institute, Finland*

Abstract

We present results from a suite of AeroCom model experiments on the two-decadal variations of UTLS aerosols and factors controlling such variability. The model simulations from 8 AeroCom models are compared to four satellite products of aerosol extinction profiles, including OSIRIS, CALIOP, SCIAMACHY, and OMPS LP, during 2002-2018 in the upper troposphere, near the tropopause, and in the lower stratosphere. We attribute the change of UTLS aerosols to emissions from anthropogenic, volcanic, and biomass burning sources using model experiments tagging different emission sources. Further, investigate the connections of UTLS aerosol interannual variability to the variability of Asian summer monsoon strength and other climate indicators through a common transport tracer implemented in the models.

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Aerosol Effects on Clear-Sky Shortwave Heating in the Asian Monsoon Tropopause Layer

Author Name: Ms Jie Gao [1]

Co-author Names: Jonathon S. Wright[1]

[1]Affiliation 1: Department of Earth System Science, Ministry of Education Key Laboratory for Earth System Modelling, and Institute for Global Change Studies, Tsinghua University, Meng Minwei Science and Technology Building, Beijing 100084, China

Abstract (100 - 500 Words)

The Asian Tropopause Aerosol Layer (ATAL) has emerged in recent decades[1] to play a prominent role in the upper troposphere and lower stratosphere above the Asian monsoon[2,3,4]. Although ATAL effects on surface and top-of-atmosphere radiation budgets are well established[5,6,7], the magnitude and variability of ATAL effects on radiative transfer within the tropopause layer remain poorly constrained. Here, we investigate the impacts of various aerosol types and layer structures on clear-sky shortwave radiative heating in the Asian monsoon tropopause layer using reanalysis products and offline radiative transfer simulations[8,9]. ATAL effects on shortwave radiative heating based on the MERRA-2 aerosol reanalysis are on the order of 10% of mean clear-sky radiative heating within the tropopause layer, although discrepancies among recent reanalyses and hindcasts of ATAL amplitude and composition suggest that this ratio could be as small as 5% or as large as 40%. ATAL effects peak between 150 hPa and 70 hPa (370 K - 420K potential temperature) along the southern flank of the anticyclone. Both clear-sky and all-sky shortwave heating are at a minimum in this vertical range, which is situated between the positive influences of monsoon-enhanced water vapour and clouds and the negative influence of the 'ozone valley' in the monsoon anticyclone[10,11,12]. ATAL effects also extend further toward the west, where diabatic vertical velocities remain upward despite descent in pressure coordinates. ATAL-related temperature and radiative heating perturbations thus have implications for both isentropic and diabatic transport within the monsoon anticyclone[12,13,14,15], long recognized as an important pathway for air entering the stratosphere.

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Atmospheric OCS Observations From 22 Globally Disperse NDACC FTIR Stations Since 1986: Implications for Stratospheric Trends

James W. Hannigan, Ivan Ortega, Shima Bahramvash Shams

Atmospheric Chemistry, Observations & Modeling

National Center for Atmospheric Research

Boulder, CO USA

& the NDACC IRWG Community

Carbonyl sulfide (OCS) is a non-hygroscopic trace species in the atmosphere and a large sulfur reservoir maintained by both direct oceanic, geologic, biogenic, and anthropogenic emissions and the oxidation of other sulfur-containing source species. It is the largest source of sulfur transported to the stratosphere during volcanically quiescent periods. Data from 22 ground-based globally dispersed stations are used to derive trends in total and partial column OCS. Middle infrared spectral data are recorded by solar-viewing Fourier transform interferometers that are operated as part of the Network for the Detection of Atmospheric Composition Change between 1986 and 2020. Vertical information in the retrieved profiles provides analysis of discreet altitudinal regions. Trends are found to have well-defined inflection points.

In two linear trend time periods ∼2002 to 2008 and ∼2008 to 2016 tropospheric trends range from ∼0.0 to (1.55 ± 0.30%/yr) in contrast to the prior period 1986 to 2002, where all tropospheric trends are negative. Regression analyses using a range of proxies for the free tropospheric data show the strongest correlation, consistently and by far with anthropogenic emissions.

The stratospheric OCS partial column is regressed with simultaneously measured N_2O to derive trends accounting for dynamical variability at selected sites from 2001-2016. There is overall, a small but increasing trend in the stratosphere seen in the longest time series except at MLO 19.5°N and WLG at −34.4°S. Globally, northward of MLO and southward of WLG stratospheric trends have been increasing since 2001, 0.12%–0.32%/yr and 0.27%–0.79%/yr, respectively. This may infer an excess of stratospheric sulfur over time and that the limiting factor to conversion to sulfate aerosol may not be sulfur derived from OCS.

Stratospheric lifetimes are derived and range from (54.1 ± 9.7)yr in the sub-tropics to (103.4 ± 18.3)yr in Antarctica.

The trends in nearly all altitudes and stations was increasing during the period 2008–2016. But more recently, the trend in the free tropospheric data during the short period of 2016 to 2020 is decreasing at all stations and may have implications for future stratospheric loading. These unique long-term measurements provide new and critical constraints on the global OCS budget.

Abstract Title: Model calculations of the contribution of tropospheric SO2 to the stratospheric aerosol layer

Presenting Author title and name: Chiranjeevi Nalapalu[1]

Co-author Names: Dr. Ingo Wohltmann[1], Prof. Markus Rex[1]

[1]Affiliation: Alfred Wegener Institute Helmholtz Centre for Polar and Marine Research (AWI), Potsdam, Germany

Abstract (100 - 500 Words)

The stratospheric aerosol layer is important for stratospheric chemistry, climate change and in geo-engineering. Yet the processes governing the transport of sulfur to the stratosphere are poorly quantified. We present model calculations of the chemistry of sulfur dioxide (SO2) and its transport to the stratosphere and perform numerous sensitivity runs to assess the range of uncertainty of these calculations. The transport model is based on backward trajectories from the ATLAS model driven by either ECMWF ERA 5 or CAMS data. Sensitivity experiments explore the sensitivity to changes in OH, H2O2, DMS, cloud water, cloud pH value and in the driving analysis data.

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AWI INSPIRES - INternational Science Program for Integrative Research in Earth Systems

The Stratospheric Aerosol processes, Budget and Radiative Effects (SABRE) Airborne Science Mission

Eric Jensen[1,2] Troy Thornberry[1]

[1]NOAA Chemical Sciences Laboratory, USA

[2]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, USA

Stratospheric aerosols are an important component of Earth's albedo, and therefore energy balance, and they also provide surface area for heterogeneous chemistry, which can lead to stratospheric ozone loss. Acquiring an extensive database of detailed stratospheric aerosol, trace gas and dynamical observations is needed to establish the baseline state and background variability of the stratospheric aerosol. This extensive dataset will be essential for (1) developing a complete understanding of stratospheric dynamical and chemical processes that determine aerosol microphysics, radiative properties and heterogeneous chemistry, and (2) evaluating the stratospheric response to natural and anthropogenic perturbations including climate change, volcanic eruptions, pyrocumulonimbus, aircraft and rocket exhaust, and potential climate intervention activities.

The Stratospheric Aerosol processes, Budget and Radiative Effects (SABRE) mission, a component of the NOAA Earth Radiation Budget (ERB) program, is an extended airborne science campaign to study the formation, transport, chemistry, microphysics and radiative properties of aerosols in the upper troposphere and lower stratosphere (UTLS). During February, 2022, we successfully conducted the first SABRE flight series with six WB-57 sorties flown from Ellington Field, Houston. The WB-57 payload included 19 instruments, providing measurements of aerosol size distribution, composition, and extinction, as well as both long-lived tracer species and photochemically important trace gases in the subtropical and midlatitude UTLS region. The primary purpose of this initial flight series was to provide an opportunity to test a number of newly developed instruments that will be important for addressing the SABRE science questions in subsequent deployments. Overall, the payload performance was quite good, and in addition to accomplishing the intended instrument testing and evaluation, we acquired an initial science dataset for the SABRE program.

The planned SABRE deployments over the next few years will provide additional measurements with this payload in high latitude and tropical regions. Ultimately, SABRE will provide a dataset of UTLS aerosol and trace gas properties in different regions and seasons that can be used to understand stratospheric aerosol physical and chemical processes, evaluate/improve stratospheric aerosol representations in climate models, and provide a basis for better informed policy decisions regarding anthropogenic impacts on the stratosphere.

Abstract Title: Ablation rates of organic compounds in cosmic dust, meteoric fragmentation and atmospheric impacts

Presenting Author title and name: John M. C. Plane[1]

Co-author Names: David L. Bones[1], Juan Diego Carrillo Sánchez,[1,2] Alexander James[1], Alexander N. Kulak,[1] Simon D. A. Connell[3], Graham W. Mann[4]

[1]School of Chemistry, University of Leeds, Leeds, U.K.

[2] Current address: Department of Physics, Catholic University of America, Washington DC and ITM Physics Laboratory, NASA/Goddard Space Flight Center, Greenbelt MD, U.S.A.

[3]School of Physics and Astronomy, University of Leeds, Leeds, U.K.

[4]School of Earth and Environment, University of Leeds, Leeds, U.K.

Cosmic dust particles are produced in the solar system from the sublimation of comets as they orbit close to the sun, and also from asteroidal collisions between Mars and Jupiter. Recent advances in interplanetary dust modelling provide much improved estimates of the fluxes of cosmic dust particles into planetary (and lunar) atmospheres throughout the solar system. Combining the dust particle size and velocity distributions with new chemical ablation models enables the injection rates of individual elements to be predicted as a function of location and time. This information is essential for understanding a variety of atmospheric impacts, including: the formation of layers of metal atoms and ions; the production of meteoric smoke particles; ice cloud nucleation in the mesosphere and lower stratosphere; perturbations to atmospheric gas-phase chemistry; the injection of bio-available phosphorous and organics; and the effects of the surface deposition of micrometeorites and cosmic spherules.

Cosmic dust consists of mineral grains that are held together by a refractory organic “glue”. It has been proposed that loss of the organics leads to fragmentation of cometary dust particles into micron-sized fragments. *If this happens*, there are several important implications: 1) slow-moving particles, particularly from Jupiter Family Comets, will be undetectable by radar, so that the total dust input to the atmosphere may be considerably larger than current estimates of 20 – 50 tonnes per day; 2) meteoritic fragments appear to be a key agent for freezing stratospheric droplets in the polar lower stratosphere, producing polar stratospheric clouds which activate chlorine and cause ozone depletion; and 3) the measured accumulation rates of meteoric smoke particles, micrometeorites and cosmic spherules in the polar regions may be better explained.

At Leeds we have developed a new experimental system for studying the pyrolysis of the refractory organic constituents in cosmic dust during atmospheric entry. The pyrolysis kinetics of meteoritic fragments (CM2 and CV3 carbonaceous chondrites, radius = 36 – 100 mm) was measured by mass spectrometric detection of CO_2 and SO_2 at temperatures between 625 and 1300 K, with most carbon being lost between 700 and 800 K. The complex time-resolved kinetic behaviour is consistent with two organic components – one significantly more refractory than the other, which probably correspond to the insoluble and soluble organic fractions, respectively. The measured temperature-dependent pyrolysis rates were then incorporated into the Leeds Chemical Ablation Model (CABMOD), which demonstrates that organic pyrolysis should be detectable using a high performance/large aperture radar. Atomic force microscopy was used to show that although the residual meteoritic particles became more brittle after organic pyrolysis, they also became slightly harder, withstanding stresses that are at least 3 orders of magnitude higher than would be encountered during atmospheric entry. This suggests that most cosmic dust particles (radius < 100

μm) will not fragment during entry into the atmosphere as a result of organic pyrolysis, although a subset of slow-moving, low density particles mostly from Jupiter Family Comets could fragment.

Acknowledgements

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A 3-D modelling study of the seasonal funnelling of meteoric smoke particles into the Arctic polar vortex

Dr. Kamalika Sengupta[1],

Graham Mann[1], Ralf Weigel[3], James Brooke[2], Sandip Dhomse[1], Stephan Borrmann[3], John Plane[2]

[1]School of Earth and Environment, University of Leeds

[2]School of Chemistry, University of Leeds

[3]IPA - Institut für Physik der Atmosphäre [Mainz]

Abstract:

In-situ measurements have shown a steady influx of meteoric smoke particles in the lower stratosphere through the polar vortex in the Arctic winter. This source of meteoric particles may have significant implications for the formation of polar stratospheric clouds and with future changes in stratospheric circulation could be linked with ozone recovery. In this study we investigate the effects of the wintertime influx of meteoric smoke cores on the Arctic stratospheric aerosol layer.

We simulate two types of stratospheric aerosol particles - pure sulphuric acid particles and sulphuric acid particles with meteoric smoke core. Our model is able to reproduce the vertical profile of stratospheric particles measured in-situ during the EUPLEX 2003 campaign. We show that the wintertime influx of sulphuric acid particles with meteoric smoke cores into the lower stratosphere subsequently leads to an increase in the concentration of pure sulphuric acid particles at the onset of spring. We quantify the contribution of meteoric smoke particles to the total particle number concentration during winter through to spring and put the 2003 winter in context with a 5-year climatology over the Arctic.

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Acknowledgements:

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computational support from the UK national high performance computing service, ARCHER, for which access was obtained via the UKCP consortium and funded by EPSRC grant ref EP/K013564/1.

Abstract Title: Acid-Processed Meteoric Smoke can Cause Explain Observed Nucleation in Polar Stratospheric Cloud.

Presenting Author title and name: Dr Alexander D. James[1]

Co-author Names: Finn Pace[1], Sebastien N. F. Sikora[2], Benjamin J. Murray[2], Graham W. Mann[3], John M. C. Plane[1]

[1]Affiliation 1 School of Chemistry, University of Leeds, UK
[2]*Affiliation 2* School of Earth and Environment, University of Leeds, UK
[3]*Affiliation 3* Institute for Climate and Atmospheric Science, University of Leeds, UK

Please expand for additional authors

Abstract (100 - 500 Words)

Nitric Acid Trihydrate (NAT) crystal formation in the absence of water ice is important for a subset of Polar Stratospheric Clouds (PSCs) and thereby ozone. However, nucleation of these crystals is not well understood.

It has been suggested previously that either fragmented meteoroids or meteoric smoke particles (MSPs), or possibly both, are important as heterogeneous nuclei. The role of H_2SO_4, which is present in liquid PSCs, in these nucleation processes has not been investigated in the laboratory previously. It is known that metal-containing Meteoric Smoke Particles (MSPs) are processed, partially dissolving whilst some components re-precipitate within H_2SO_4 droplets, producing silica and alumina smoke particles of potentially altered size distributions. We recently found experimentally that size of MSPs is critical to their nucleation activity, with nano-particulate fumed silica a poor promoter of nucleation, whilst micron scale fused quartz was found to be effective. Both materials have similar chemical and structural (crystallographically amorphous) structure. Here we explore this size dependence of nucleation effectiveness quantitatively for the first time.

In this study we find experimentally that meteoric fragments sometimes retain their nucleation activity in ternary acid solution. However, combining sedimentation modelling with our results and recent experiments on fragmentation of incoming meteoroids suggests that fragments are unlikely to be competitive atmospheric nucleating particles.

In this study we also develop a model using Classical Nucleation Theory (CNT) where we account for surface curvature of primary grains. This model is able to explain the discrepancy in nucleation effectiveness of fumed silica and fused quartz, by treating them as having the same nucleating activity (contact angle) but differing particle size (or equivalently surface curvature), assuming interfacial energies which are physically reasonable given literature measurements. Here we use this CNT model to present evidence that nucleation of NAT on (10s nms) MSP analogues is effective enough to explain observed NAT crystal number concentrations in PSCs (without ice).

Acknowledgements

This work forms part of the MeteorStrat project funded by the UK Natural Environment Research Council (grant number NE/R011222/1).

New insights into PSCs from the nearly 17-year CALIPSO data record

Michael Pitts[1] and Lamont Poole[2]

[1]*NASA Langley Research Center, Hampton, VA USA*
[2]*Science Systems and Applications, Inc., Hampton, VA USA*

Polar stratospheric clouds (PSCs) play important roles in stratospheric ozone depletion during winter and spring at high latitudes (e.g., the Antarctic ozone hole). PSC particles provide sites for heterogeneous reactions that convert stable chlorine reservoir species to radicals that destroy ozone catalytically. PSCs also prolong ozone depletion by delaying chlorine deactivation through the removal of gas-phase HNO_3 and H_2O by sedimentation of large nitric acid trihydrate (NAT) and ice particles. A more complete picture of PSC processes on vortex-wide scales has emerged from the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) instrument on the CALIPSO satellite that has been observing PSCs at latitudes up to 82 degrees in both hemispheres since June 2006. This nearly 17-year data record has fundamentally improved our knowledge of PSC spatial and temporal distributions, composition, and formation processes. In this presentation, we provide a brief overview of the CALIOP PSC data record and highlight some of the recent advances in our understanding of PSC processes.

Cirrus perturbations from volcanism and the (un)surprising fate of organics in the stratosphere

Johan Friberg[1,2]

Bengt G. Martinsson[1] and Moa K. Sporre[1]

[1]Department of Physics, Lund University, Sweden
[2]SIGHT, The Royal Swedish Academy of Sciences, Sweden

We find evidence of seasonal volcanic impact on midlatitude cirrus clouds that may cool the climate. Stratosphere-troposphere-transport (TST) of air increases the sulfate load in the upper troposphere (UT). Its influence is largest in spring when the TST flow across the tropopause is at its maximum. In Friberg et al.[1] we found that the MODIS cirrus reflectance co-varied with the sulfate load in the extra-tropical transition layer (ExTL). In our most recent letter [2] we report that several cirrus data products from satellites co-vary with the aerosol load in the ExTL (retrieved by CALIOP). Northern Hemispheric cirrus have fewer ice crystals, less ice-mass, and smaller coverage in springtime during years with volcanically elevated ExTL aerosol load. This can be viewed as a general thinning of cirrus in periods of volcanic impact. No influence was found in the Southern Hemisphere, indicating a hemispheric difference in cirrus response to volcanic aerosol. Cirrus clouds have a net warming effect on climate. Hence, we expect the observed cirrus cloud thinning to cool the climate.

Moreover, we used the CALIOP instrument to study wildfire impact on the stratosphere. Limb-viewing satellite sensors like SAGE and OMPS-LP suffer from saturation effects for dense aerosol layers, such as fresh smoke clouds. The nadir viewing lidar-based CALIOP sensor has an advantage over these instruments due to its short line-of-sight, and can retrieve signals also in the densest smoke layers (AOD>3). Additionally, we have developed methods to account for attenuation in order to capture extinction coefficients of all smoke layers [3].

In CALIOP data, we find a rapid decrease in smoke AOD in the weeks following smoke injections to the stratosphere, not resolved by other satellite instruments [3]. The effect is visible after both the fires in eastern North America Aug 2017, and East Australia Dec 2019 / Jan 2020, and is accompanied by clear changes in aerosol particle sizes and properties (color and depolarization ratios). We estimate the half-life of the smoke AOD to be 10 days (e-folding: 14 days). The same numbers were reported in a model estimate of photolytic loss of organics in the mid-troposphere [4], suggesting photolytic loss as cause of the rapidly decreasing smoke load. We find that 80-90% of the smoke was depleted within a month of injection, after which the remaining aerosol survived until transported to the troposphere with the general circulation. These findings may prove important for simulations of organic aerosol in the stratosphere as well as in the troposphere.

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Abstract Title: Sensitivity of longwave radiative forcing and total radiative heating rates of lower stratospheric aerosol due to organic carbon content

Presenting Author title and name: John Dykema[1]

Co-author Names: Yaowei Li[1], Frank Keutsch[1], Pengfei Liu[2], Terry Deshler[3]

[1] John A. Paulson School of Engineering and Applied Sciences, Harvard University, USA
[2] Earth and Atmospheric Sciences, Georgia Institute of Technology, USA

[3] Laboratory for Atmospheric and Space Physics, University of Colorado, USA

Abstract (100 - 500 Words)

Stratospheric aerosols act as direct radiative forcing agents through scattering or absorption of solar radiation, as well as indirect agents by affecting the nucleation of ice clouds when sedimenting out of the stratosphere. Composition measurements and modeling indicate that lower stratospheric aerosol includes significant organic carbon content. The exact range of complex refractive index and mixing state of these organic-containing aerosols is unknown. Detailed representation of complex refractive index and morphology is necessary to calculate the radiative impacts of these lower stratospheric aerosols. To bound the range of uncertainty in these radiative impacts, a recently published analysis evaluated the influence of complex refractive index and morphology on shortwave radiative forcing. This study found that for Northern hemisphere midlatitudes the uncertainty in optical properties of organic-containing lower stratospheric aerosols is significant, particularly during periods of moderate or quiescent volcanic activity. Complementary work for the longwave radiative forcing and combined radiative heating rates will be presented. These longwave results are obtained with a similar approach, utilizing a combination of existing and new laboratory measurements to develop plausible estimates for complex refractive index and morphology. These aerosol properties are coupled with long-term in situ size measurements from balloons to obtain profiles of radiative fluxes necessary for heating rate and radiative forcing calculations.

Acknowledgements

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The NOAA B^{2}SAP project: toward systematic in situ observations of stratospheric aerosol size distribution

Troy Thornberry[1]

Elizabeth Asher[1,2], Michael Todt[1,2], Dale Hurst[1,3], Emrys Hall[1,3], Allen Jordan[1,3], Kensy Xiong[1,3], Penny Smale[4], Richard Querel[4]

[1]NOAA Chemical Sciences Laboratory, Boulder, Colorado, USA

[2]Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado Boulder, Boulder, Colorado, USA

[3]NOAA Global Monitoring Laboratory, Boulder, Colorado, USA

[4]National Institute of Water & Atmospheric Research (NIWA), Lauder, New Zealand

Abstract (100 - 500 Words)

Variations in stratospheric aerosol loading impact the climate system by altering Earth's albedo and therefore radiative balance. Stratospheric aerosols also provide surface area for heterogeneous chemical reactions that can lead to ozone loss. The chemical and radiative effects of stratospheric aerosols depend on particle size distribution, but models vary significantly both in how they parameterize aerosol microphysical processes and how they simulate size distributions, resulting in divergent predictions of stratospheric size distributions and the time evolution of radiative impacts from stratospheric aerosol perturbation events. Regular measurements of stratospheric aerosol number and size distribution, their variability and response to perturbation are needed to constrain the stratospheric aerosol processes in climate models that are used to assess the impacts of changes to stratospheric aerosol from natural and anthropogenic emissions and the efficacy and consequences of potential climate intervention strategies.

The Baseline Balloon Stratospheric Aerosol Profiles (B^{2}SAP) project, funded through the NOAA Earth's Radiation Budget (ERB) Initiative, is planned as a global network of systematic balloon sondes to measure water vapor, ozone, and aerosol number and size distributions up to the middle stratosphere (~28km). The project has been conducting launches approximately every two weeks from Boulder, Colorado and at a lower frequency from Lauder, New Zealand over the past two years, as well as recent launches from Réunion Island in response to the Hunga Tonga – Hunga Ha'apai eruption. We are working to expand the B^{2}SAP network in the near future to include additional sites at a range of different latitudes. The B^{2}SAP aerosol measurements will provide a database that will be useful for constraining stratospheric aerosol processes in global models and validating and improving satellite-based retrievals of stratospheric aerosol properties. Here we present results from the past two years of launches from Boulder and Lauder, which reveal both natural aerosol variability and the scale to which natural events, such as the Raikoke volcanic eruption and Australia's Black Summer pyrocumulonimbus super outbreak, can alter the composition of the stratosphere.

Acknowledgements

We would like to acknowledge the NOAA Earth's Radiation Budget Initiative for support of the B^{2}SAP project and funding from the NASA SAGE III/ISS Project for funding validation sonde launches.

Abstract Title: Temporal Variability of the size distribution of stratospheric aerosol from 2002 to 2005 from SAGE III-M3M-Measurements

Presenting Author title and name: Felix Wrana[1]

Co-author Names: Terry Deshler[2], Ulrike Niemeier[3], Larry Thomason[4], and Christian von Savigny[1]

[1] Institute of Physics, University of Greifswald, Germany

[2] Department of Atmospheric Science, University of Wyoming, USA

[3] Max Planck Institute for Meteorology, Hamburg, Germany

[4] Langley Research Center, NASA, Hampton, USA

We present the temporal evolution of size distribution parameters of stratospheric aerosol in the time frame from 2002 to 2005 based on the measurements of the Stratospheric Aerosol and Gas Experiment (SAGE) III on Meteor-3M (M3M). The occultation measurements of SAGE III-M3M roughly cover a range between 40°N and 80°N in the northern hemisphere and between 30°S and 60°S in the southern hemisphere.

The retrieval method, which is explained in detail in Wrana et al. (2021, Atm. Meas. Tech.)[1] , makes use of the spectral dependency of the extinction coefficients provided in the SAGE III-M3M data set to determine the median radius and mode width of a monomodal lognormal size distribution. Both parameters can be retrieved simultaneously due to the use of three wavelength channels of the broad spectral range covered by SAGE III. Based on the results other parameters, like the effective radius and the particle number density, were determined.

In the talk we show the seasonal variability of the size distribution parameters in the northern and southern hemisphere that can be observed in the data set. Furthermore, we discuss the possible occurrence of polar stratospheric clouds (PSCs) as well as an observed reduction of the average size of the stratospheric aerosol in the mid latitudes of both hemisphere in 2005, which may have been caused by the Manam eruption in 2004. For validation, we show a comparison of the size distribution parameters of our data set with collocations with the OPC measurements in Kiruna, Sweden, by the University of Wyoming.

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Measurements of Tropical Tropopause Layer and Lower Stratospheric Aerosol Size Distributions from Drifting Super Pressure Balloons During the Stratéole 2 Field Campaign

Lars Kalnajs and Terry Deshler

Laboratory for Atmospheric and Space Physics University of Colorado at Boulder, Boulder CO, 80303, USA

Stratéole 2 is an international long duration super pressure ballooning project that has deployed dozens of balloons at the upper edge of the tropical tropopause layer and the lower tropical stratosphere (18 – 21km) where they can fly for months at a time. LASP has built several instruments that have flown on these balloons, including a variant of our standard LASP Optical Particle Counter (LOPC). Over two field campaigns (2019 and 2021) we collected several months of in situ stratospheric aerosol size distributions at balloon flight level as well as profiles below the balloon flight level using a smaller OPC on a reel down platform.

Measurements in 2019 show trends in the aerosol extinction at flight level that are consistent with decreasing stratospheric aerosol loading in the aftermath of the Raikoke eruption and are consistent with the satellite aerosol extinction trend. Both the 2019 and 2021 campaigns periodically observed short duration enhancements of super micron particles, with size distributions extending above 16 micrometers. Similarly sized particles were also observed with a different OPC in 2010 on a long duration balloon flight at 20 km in the tropics. In all cases such large particles comprise well less than 1% of the observations. The nature and composition of these particles is unknown, the particles are observed significantly above the tropopause, at temperatures that are inconsistent with the formation of ice, and are broadly distributed around the tropical belt. These large particles have a sufficient aerosol extinction to be detected by limb sounding, however they appear to occur in vertically thin layers, below the vertical resolution of satellite sensors. There are few observations capable of resolving these particles in this hard to reach region of the atmosphere. Thus they remain a mystery for now.

In Situ Observations of the Concentrations of Total Aerosol and Optical Aerosol in the Austral Fall Polar Stratosphere, Results from the ASPEN 2019 Field Campaign

Dr. Terry Deshler, Lars Kalnajs, J. Douglas Goetz

Laboratory for Atmospheric and Space Physics, University of Colorado at Boulder, Boulder, CO, USA

The stratospheric aerosol is primarily known for the optically visible particles, diameters larger than several hundred nanometers. Such particles are measured remotely with lidars, solar photometers, limb scatterers, and in situ with optical particle counters (OPCs). These particles affect the radiation and chemical balance of the stratosphere, and must be accounted for in all global climate chemistry models. Underpinning these particles are the non-visible seeds upon which most of the stratospheric aerosol form. Particles in the size range of 10 nm. Observation of these small particles are only available from in situ instruments which supersaturate the air stream to grow all particles, bigger than a few nanometers in the air sample, to optically detectable sizes (1,2). Often these particles are known as condensation nuclei (CN). The source of these particles, upon which the stratospheric aerosol rely, is less well known. Meteoritic material (3), along with primary particle formation above the tropical tropopause (4), after large volcanic eruptions (5,6), and in the spring time polar stratosphere (7,2) have been implicated. Evidence for new particle formation from the polar regions was first observed in spring in the northern mid latitudes (7) as polar stratospheric air moved equatorward. The most likely source of these polar CN is from the condensation of sulfuric acid as air subsides in the polar vortex in spring, and such CN layers have been observed frequently in the Antarctic (8,2), and less frequently in the Arctic (9). Modeling work (10,11) has established sulfuric acid condensation as the primary mechanism, as air subsides and cools in the polar votex. Volatility measurements have confirmed this composition (2). Further modeling work (11) illustrated the spread of these CN to lower latitudes and at the same time indicated that a similar layer of CN may form in austral fall over Antarctica. In 2019 the ASPEN field campaign made in situ measurements of optical aerosol and of CN from late April to mid-June above McMurdo Station, Antarctica, 78°S, to test this prediction. Five CN soundings, two with a heated inlet, and four OPC flights were completed. While there was no evidence of a significantly enhanced population of CN, in contrast to the model predictions, there was a tendency for the CN mixing ratio to generally increase with altitude from a bit above the tropopause to the top of the soundings at 35 km. This and other results from these measurements will be presented.

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Stratospheric profiles of extinction coefficient and effective radius from SCIAMACHY limb observations

Christine Pohl[1]

Alexei Rozanov[1], Elizaveta Malinina-Rieger[2], Terry Deshler[3], Felix Wrana[4], Ulrike Niemeier[5], and John P. Burrows[1]

[1]Institute of Environmental Physics, University of Bremen, Germany
[2]Climate Research Division, Environment Canada, Victoria, Canada
[3]Department of Atmospheric Science, University of Wyoming, USA
[4]Institute of Physics, University of Greifswald, Germany
[5]Max Planck Institute for Meteorology, Hamburg, Germany

Abstract

Stratospheric aerosols, mainly from volcanic eruptions and biomass burning, interact with stratospheric trace gases and change the Earth's radiation budget. The amplitude of change depends on the amount of aerosols. The exact determination of the vertical and spatial distribution of stratospheric aerosols is therefore essential for the consideration of trace gas trends and the climatic development of the Earth. We have improved the retrieval algorithm of the particle size distribution (PSDs) from SCIAMACHY (Scanning Imaging Absorption spectroMeter for Atmospheric ChartograpHY) limb observations by including the actual effective Lambertian surface albedo derived from SCIAMACHY nadir observations. The retrieval algorithm is based on an optimal estimation method that assumes a fixed number density profile. From the retrieved PSDs, the extinction coefficients and effective radii are calculated and compared with other satellite observations (e.g., SAGE) and model simulations (e.g., ECHAM). By taking the actual effective Lambertian surface albedo into account, the SCIAMACHY retrieved extinction coefficient and effective radius are considerably improved.

ESM differences in stratospheric transport and aerosol microphysics as inferred by the GeoMIP G6Sulfur experiment:

Presenting Author title and name: Dr Daniele Visioni[1]

Co-author Names: Ben Kravitz[2], Douglas G. MacMartin[1], Simone Tilmes[3]

[1]Sibley School for Mechanical and Aerospace Engineering, Cornell University, Ithaca, NY, USA
[2]Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, IN, USA
[3]Atmospheric Chemistry, Observations, and Modeling Laboratory, National Center for Atmospheric Research, Boulder, CO, USS

Abstract (100 - 500 Words)

The Geoengineering Model Intercomparison Project (GeoMIP) has developed, as part of Phase 6 of the CMIP project, a new suite of experiments aimed at better understanding the inter-model differences in simulating continued sulfate injections in the stratosphere [1]. The G6sulfur experiment prescribes the injection of SO_2 between 10N and 10S in the lower stratosphere in order to offset part of the radiative forcing produced by the greenhouse gases in the SSP5-8.5 scenario by producing sulfate aerosols.

The continued injection of SO_2 for a number of years allows us to determine the long-term differences in the participating Earth System Models: we show how large inter-model differences still remain in the resulting aerosol distribution even in the new CMIP6 models and try to separate the role of stratospheric transport and of the different aerosol microphysics. By analyzing a subset of models with a prescribed stratospheric aerosol distribution we also show how differences in the aerosol forcing calculations and aerosol-cloud interactions can still result in different surface climate even given the same stratospheric aerosol optical depth.

Lastly, we show how G6sulfur simulations with fully coupled atmospheric chemistry can be used to determine the potential disruption of stratospheric ozone caused by the increased surface area density, and by the dynamical changes. In particular, we demonstrate that the differences in the transport of the aerosols at high latitudes strongly influence the projected impacts on the total ozone column over the poles [2].

Looking at the future, we discuss new simulations that can more systematically ascertain sources of models' disagreement and potential areas of improvement.

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Climate impacts from sulfate aerosol injection and the dependence on the injection latitude

Dr Ewa M. Bednarz[1]

Daniele Visioni[1], Yaga Richter[2], Andy Jones[3], Jim Haywood[3,4], Ben Kravitz[5,6], Douglas MacMartin[1]

[1]Sibley School of Mechanical and Aerospace Engineering, Cornell University, Ithaca, NY, USA.
[2]Climate and Global Dynamics Laboratory, National Center for Atmospheric Research, Boulder, CO, USA

[3]UK Met Office, Exeter, UK

[4]University of Exeter, Exeter, UK

[5]Department of Earth and Atmospheric Sciences, Indiana University, Bloomington, IN, USA

[6]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, USA

Abstract

Stratospheric Aerosol Injection (SAI) is a proposed geoengineering method aiming to mitigate some of the negative impacts of rising greenhouse gas levels by reducing surface temperatures. Yet, despite offsetting global mean surface temperature, various studies demonstrated that SAI could influence the recovery of stratospheric ozone and have important impacts on atmospheric circulation at tropical, mid- and high latitudes, thereby potentially playing an important role in modulating regional and seasonal climate variability. However, so far most of the assessments of such an approach have come from climate model simulations in which SO_2 is injected in the lower stratosphere in the equatorial region. Since it has been demonstrated that the choice of injection location could lead to different climate impacts, a systematic assessment would be extremely important but has so far been missing.

Here we address this using the CESM2(WACCM6) model, a comprehensive Earth System Model with prognostic aerosol microphysics and interactive stratospheric chemistry. We perform a set of five sensitivity experiments with constant point injections of 12 Tg-SO_2/yr at 22 km at 30°S, 15°S, 0°, 15°N and 30°N. We demonstrate the strong dependence of the simulated changes in tropospheric circulation, including changes in the Hadley Circulation and tropospheric jets, and in the associated precipitation patterns on the latitude of SAI. We then examine the latitude dependence of the stratospheric responses, focusing on the impacts on stratospheric ozone and the modes of high latitude variability. Lastly, we verify our results in a multi-model framework, comparing the CESM2 simulations with similar but shorter runs carried out with the UKESM1 and GISS-E2.1-G models. Overall, our results contribute to an increased understanding of the underlying physical processes and aim to reduce some of the sources of uncertainty in model projections of climate impacts from SAI.