Quantifying the influence of meteoric smoke particles and fragments on stratospheric aerosol and chemistry

Dr Wuhu Feng[1,2,3], Prof. John Plane[3] and Dr Graham Mann[2]

http://homepages.see.leeds.ac.uk/~earfw

https://john-plane.leeds.ac.uk/

https://environment.leeds.ac.uk/see/staff/1395/dr-graham-mann

Contact emails: w.feng@leeds.ac.uk; J.M.C.Plane@leeds.ac.uk; G.W.Mann@leeds.ac.uk

[1] National Center for Atmospheric Science (NCAS), University of Leeds, UK

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

[3]School of Chemistry, University of Leeds, UK

External Collaborators:
Prof. Daniel Marsh (NCAR, Boulder and Leeds): WACCM chemistry-climate modelling
Prof. Martyn Chipperfield (Leeds): Stratospheric chemistry and PSC modelling
Dr. Michael Pitts (NASA Langley Research Center): CALIOP satellite lidar PSC measurements
Dr. Josef Höffner (Leibniz Institute of Atmosheric Physics): Ground-based lidar observations

Scientific Background and Motivation

A large amount of total cosmic dust enters the Earth's atmosphere every day (Plane, 2012; Gardner et al., 2014, Carrillo-Sánchez, et al., 2016) as small meteoroids at different mass, size and entry velocity (11.2-72 km/s) (Klekociuk et al., 2005, Carrillo-Sánchez, et al., 2015). The meteoroids will fragment if they experience pressures higher than their tensile strength during atmospheric entry (Plane, 2012; James et al., 2018), with high-resolution video cameras showing ~90% of large meteoroids undergo fragmentation (Subasinghe et al., 2016). Meteoric ablation also occurs as cosmic dust particles enter the atmosphere at high velocity, with intense flash heating through collisions with air molecules, followed by rapid evaporation of metal atoms and oxides once the particles melt (Plane et al., 2015).

A variety of metals (e.g. Na, Fe, Mg, K, Ca, Ni, and Si) are produced as the meteoroids ablate in the mesosphere and lower thermosphere (MLT, ~70-120 km), generating layers of metal atoms between 80 and 105 km. These meteoric metal layers can be observed by the ground-based lidar (laser radar) techniques, as well as by satellite-borne optical spectroscopy. Metallic ions are also measured by mass spectrometry on sub-orbital rockets, each metal provide a unique tracer of the physics and chemistry of the atmosphere at the interface with geospace (Plane et al., 2015).

The metal vapors generated by this meteoroid ablation (principally Fe, Mg, and Si) then oxidize and condense into tiny particles with the size typical around 0.5-2 nm in radius (Plane et al. 2018). These are termed Meteoric Smoke Particles (MSPs), and are produced by polymerization of metal-containing molecules (e.g. FeOH, $Mg(OH)_2$,$MgCO_3$, $NaHCO_3$ and $Si(OH)_2$), forming within the relatively long-lived reservoir species on the underside of the metallic layers (Plane et al., 2015). The MSPs are observed by the satellite measurement SOFIE spectrometer on the AIM satellite (see Hervig et al., 2009), and are transported down to the stratosphere inside the polar vortex (Bardeen et al., 2008).

Perhaps the dominant influence from the meteoric material is the way both types of meteoric particle (MSPs and meteoric fragments, MFs) may trigger the nucleation of polar stratospheric clouds (PSCs) (Figure 1, James et al., 2018). This heterogeneous nucleation of PSCs on meteoric particles has been inferred from a range of different observations from recent field campaigns (see e.g. Hoyle et al., 2013; Steiner et al., 2020).

It has long been hypothesised (e.g. Cadle and Kiang, 1977; Rosen et al., 1978; Turco et al., 1981) that stratospheric aerosol particles nucleate also heterogeneously on MSPs, and lab experiments at Leeds measuring the dissolution of MSP analogues in sulphuric aerosol in the lab (Saunders et al., 2012) and also the uptake of nitric acid (Frankland et al., 2015) and H2O2 (James et al., 2016). The Leeds stratospheric composition modelling group recently adapted the interactive stratospheric aerosol configuration of the UK composition-climate model (Dhomse et al., 2014; Mann et al., 2015) to represent this heterogeneous nucleation of MSP-sulphuric particles (Brooke et al., 2017; Dhomse et al., 2020).

This PhD project will involve global chemistry-climate model experiments to understand how meteoric particles (both MSPs and MFs) affect the stratospheric aerosol layer, forming two distinct sub-classes of stratospheric sulphuric aerosol particles, these preferentially freezing to form nitric acid PSCs in the cold Arctic, with subsequent effects on polar ozone loss via heterogeneous chemistry. The project will assess the simulated mix of stratospheric aerosol and PSCs for recent Arctic winters to satellite measurements from the CALIOP lidar, which provides an unique record of PSC occurrence for the period 2006-present (Pitts et al., 2018), with recently also a new CALIOP stratospheric aerosol type product (Kar et al., 2019).

The composition-climate models include the influences from dynamics, transport, microphysics, photochemistry, radiation and their influences on stratospheric ozone depletion (Chipperfield et al., 2005; Feng et al., 2011). A current community research focus is to understand why satellite observations show HCl is essentially completely depleted inside the dark, midwinter Antarctic polar vortex but most models significantly overestimate HCl in this region (e.g., SPARC 2010; Grooβ et al., ACP, 2018). The project will also investigate the influence of MSPs on Antarctic PSCs and explore whether the uptake of HCl on MSPs and/or meteoric fragments could potentially be causing this apparent discrepancy.

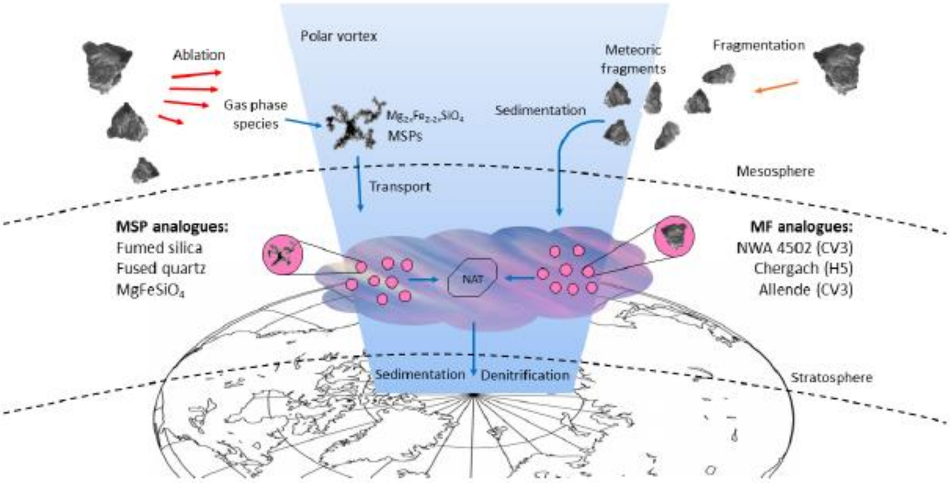

Figure 1. The pathways of two kinds of meteoric material through the atmosphere: meteoric smoke particles (MSPs) and meteoric fragments (MFs). both may heterogeneously nucleate nitric acid trihydrate (NAT) in polar stratospheric clouds. (James et al., 2018).

Objectives:

The goal of this project is to answer a number key questions: How do MSPs form and how are they transported to the stratosphere; and where they deposit at the surface? Why do current global models (e.g. Brooke et al., 2017) fail to capture the observed surface depostion of MSPs? How do MSPs and fragments affect stratosphere aerosol, PSCs and chemistry, and thus climate change in the middle atmosphere? Which important processes are missing in the current whole atmosphere models?

In this project, you will work with scientists at Leeds to apply global earth system models (WACCM(-X) and UKESM) with new capability for meteoric particles, and then evaluate the new models compared to lidar and satellite measurements. Specific goals will include:

1) Learning to run the existing WACCM (WACCM-X) which has a self-consistent treatment of MSPs explicilty through metal chemistry, and the interactive stratospheric aerosol configuration of UKESM, which uses a prescribed climatology of MSPs from WACCM with the GLOMAP aerosol microphysics module.
2) Developing the sulfur chemistry and sectional aerosol microphysics in WACCM-CARMA, adding the meteoric fragments and the meteoric sulfur scheme (Gomez Martin et al., 2017) and current WACCM (-X) models with different metal chemistry and D region chemistry (Plane et al., 2015; Feng et al., 2017; Kovacs et al., 2016);
3) Collecting available lidar/satellite data, including from one project collaborator, Dr. Josef Höffner at the Leibniz-Institute of Atmospheric Physics, Germany, who plans to make continous lidar measurements of MSPs in the stratosphere using the VAHCOL (Vertical And Horizontal COverage by Lidar) system at Davis station in Antarctica (Viehl et al., 2016) , and then comparing with the WACCM-CARMA and UKESM models;
4) Performing long-term simulations to investigate the impact of stratosphere aerosol characteristics and chemical compostions with the inclusion of MSPs/fragments in the WACCM and UKESM models.

Potential for high impact outcome:

This project addresses the coupling of the atmosphere to the geospace environment by developing self-consistent models for us to better understand the meteor astronomy, chemistry and transport processes that control atmospheric composition and aerosol in the middle atmosphere. The project is therefore likely to have significant impacts in a number of fields, including global atmospheric modelling, aeronomy, and aerosols.

Training:

The student will work under the supervision of Dr Wuhu Feng, Professor John Plane and Dr Graham Mann. This project will provide a high level of specialist scientific training in: (i) the application of a world-leading atmospheric chemistry-climate models; (ii) analysis and synthesis or large datasets; (iii) use of advanced High Performance Computing facilities (e.g. the UK national supercomputer archer.ac.uk, and the Leeds Advance Computing arc.leeds.ac.uk). The successful PhD student will have an opportunity to visit Germany and NCAR for collecting observations and model development, as well as training organised by the Doctoral Training Programme, the National Centre for Atmospheric Science, and attendance at national/international conferences.

References:

Bardeen, C. G., Toon, O. B. et al. (2008), Numerical simulations of the three-dimensional distribution of meteoric dust in the mesosphere and upper stratosphere, *J. Geophys. Res.*, vol. 113, D17202, doi:10.1029/2007JD009515.

Brooke, J. S. A., W. Feng et al. (2017), Meteoric smoke deposition in the polar regions: A comparison of measurements with global atmospheric models, *J. Geophys. Res.*, vol. 122, 11,112–11,130.

Cadle, R. D. and Kiang, C. S. (1977), Stratospheric Aitken particles, *Rev. Geophys. and Space Phys.*, vol. 15, no. 2, 195-202.

Carrillo-Sánchez, J. D. et al.(2015), On the size and velocity distribution of cosmic dust particles entering the atmosphere, *Geophys. Res. Lett.*, 42, 6518–6525, doi:10.1002/2015GL065149.

Carrillo-Sánchez, J. D. et al. (2016), Sources of cosmic dust in the Earth's atmosphere, *Geophys. Res. Lett.*, 42, 6518–6525, doi:10.1002/2015GL065149.

Chipperfield, M.P., W. Feng, and M. Rex, (2005), Arctic Ozone Loss and Climate Sensitivity: Updated Three-Dimensional Model Study, *Geophys. Res. Lett.*, 10.1029/2005GL022674.

Dhomse, S. S., K. M. Emmerson, G.W. Mann et al. (2014), Aerosol microphysics simulations of the Mt. Pinatubo eruption with the UM-UKCA composition-climate model, Atmos. Chem. Phys., vol. 14, 11,221-11,246, http://www.atmos-chem-phys.net/14/11221/2014/

Dhomse, S. S., Mann, G. W. et al. (2020), Evaluating the simulated radiative forcings, aerosol properties and stratospheric warmings from the 1963 Agung, 1982 El Chichón and 1991 Mt Pinatubo volcanic aerosol clouds, accepted for *Atmos. Chem. Phys.*, Sept 2020, https://doi.org/10.5194/acp-2020-344

Feng, W., et al., (2011): Modelling the effect of denitrification on polar ozone depletion for Arctic winter 2004/2005, *Atmos. Chem. Phys.*, 11, doi:10.5194/acp-11-6559-2011

Feng, W., et al, (2017), Impacts of a sudden stratospheric warming on the mesospheric metal layers, *J. Atmos. Sol.-Terr. Phys.*,http://dx.doi.org/10.1016/j.jastp.2017.02.004.

Frankland, V. L. et al. (2015), The uptake of HNO_3 on meteoric smoke analogues, *J. Atmos. Sol.-Terr. Phys.*, https://doi.org/10.1016/j.jastp.2015.01.010.

Gardner, C. S., et al. (2014), Inferring the global cosmic dust influx to the Earth's atmosphere from lidar observations of the vertical flux of mesospheric Na, *J. Geophys. Res. Space Physics*, doi:10.1002/2014JA020383.

Gomez Martin et al. (2017), Impacts of meteoric sulfur in the Earth's atmosphere, *J. Geophys. Res. Atmos.,* 122, doi:10.1002/2017JD027218.

Grooβ, J.-U., et al., (2018), On the discrepancy of HCl processing in the core of the wintertime polar vortices, Atmos. Chem. Phys., 18, doi:10.5194/acp-18-8647-2018.

Havnes, O., et al. First detection of charged dust particles in the Earth's mesosphere, *J. Geophys. Res.*, 101(A5), 10839– 10847, doi:10.1029/96JA00003.

Hervig, M.E, et al. (2009), First satellite observations of meteoric smoke in the middle atmosphere. *Geophys. Res. Lett*. **36**, https://doi.org/10.1029/2009gl039737.

Hervig, M.E., et al. (2012), The content and composition of meteoric smoke in mesospheric ice particles from SOFIE observations. *J. Atmos. Sol. Terr. Phys.*, **84–85**, 1–6 (2012).

Hervig, M.E., et al., (2017), Constraints on meteoric smoke composition and meteoric influx using SOFIE observations with models, *J. Geophys. Res.,*. https://doi.org/10.1002/2017JD027657

Hoyle, C. R. et al. (2013), Heterogeneous formation of polar stratospheric clouds – Part 1: Nucleation of nitric acid trihydrate (NAT), *Atmos. Chem. Phys.*, 13, 9,577-9,595, http://www.atmos-chem-phys.net/13/9577/2013/

James, A.D., et al.(2016), The Uptake of HO2 on Meteoric Smoke Analogues, *J. Geophys. Res. Atmos.*, 121, doi:10.1002/2016JD025882.

James, A. D. et al. (2018), Nucleation of nitric acid hydrates in PSCs by meteoric material, *Atmos. Chem. Phys*., 18, 4519-4531, https://doi.org/10.5194/acp-18-4519-2018

Kar et al. (2019), CALIPSO level 3 stratospheric aerosol profile product: version 1.00 algorithm description and initial assessment , *Atmos. Meas. Tech.*, 12, 6173-6191, https://doi.org/10.5194/amt-12-6173-2019.

Klekociuk, A., et al., (2005), Meteoritic dust from the atmospheric disintegration of a large meteoroid. *Nature* **436,** 1132–1135 (2005). https://doi.org/10.1038/nature0388.

Kovacs, T. et al., (2016), D-region ion-neutral coupled chemistry (Sodankyla Ion Chemistry, SIC) within the Whole Atmosphere Community Climate Model (WACCM 4) - WACCM-SIC and WACCM-rSIC, *Geosci. Model Dev.*, doi:10.5194/gmd-9-3123-2016.

Mann, G. W. et al. (2015), Evolving particle size is the key to improved volcanic forcings, *Past Global Change*, vol. 23, no. 2, 52-53.

Pitts, M. C., Poole, L. R. et al. (2018), Polar stratospheric cloud climatology based on CALIPSO spaceborne lidar measurements from 2006 to 2017, *Atmos. Chem. Phys.*, vol. 18, 10,881-10,913, https://doi.org/10.5194/acp-18-10881-2018.

Plane, J.M.C. (2012), Cosmic dust in the Earth's atmosphere, *Chem. Soc. Rev.*, vol. 41, 6507-6518, doi:10.1039/c2cs35132c.

Plane, J.M.C., W. Feng and E. Dawkins (2015), The mesosphere and metals: Chemistry and changes, *Chem. Rev*., doi:10.1021/cr500501m.

Plane, J.M.C. et al, (2018), Impacts of cosmic dust on planetary atmospheres and surfaces, *Space Sci Rev.* 214, 23, https://doi.org/10.1007/s11214-017-0458-1.

Rosen, J. M. et al. (1978), A steady-state stratospheric aerosol model, *J. Atmos. Sci.*, vol. 35, 1304-1313.

Saunders, R. W., Dhomse, S. et al. (2012), Interactions of Meteoric Smoke Particles with sulphuric acid in the Earth's stratosphere, *Atmos. Chem. Phys.,* 12, 4,387-4,398, doi.org/10.5194/acp-12-4387-2012.

SPARC, 2010: SPARC CCMVal Report on the Evaluation of Chemistry-Climate Models. V. Eyring, T. Shepherd and D. Waugh (Eds.), SPARC Report No. 5, WCRP-30/2010, WMO/TD – No. 40, available at www.sparc-climate.org/publications/sparc-reports/

Steiner, M., et al.(2020), Evaluation of polar stratospheric clouds in the global chemistry-climate model SOCOLv3.1 by comparison with CALIPSO spaceborne lidar measurements, *Geosci. Model Dev. Discuss.*, https://doi.org/10.5194/gmd-2020-102.

Subasinghe, D. et al., (2016), Physical characteristics of faint meteors by light curve and high-resolution observations, and the implications for parent bodies, *Monthly Notices of the Royal Astronomical Society*, vol. 457, Issue 2 doi:10.1093/mnras/stw019

Turco, R. P., Toon, O. B. et al. (1981), Effects of meteoric debris on stratospheric aerosols and gases, *J. Geophys. Res.*, vol. 86, no. C2, 1,113-1,128.

Viehl, T. P., J. M. C. Plane, J.M.C., Feng, W. and Höffner, J (2016), The photolysis of FeOH and its effect on the bottomside of the mesospheric Fe layer, Geophys. Res. Lett., 43, 1373-1381, doi:10.1002/2015GL067241.

Voigt, C. et al., (2000), Nitric Acid Trihydrate (NAT) in Polar Stratospheric Clouds, 290 (5497), 1756—1758, *Science*, 10.1126/science.290.5497.1756.