

SPECIAL PROJECT PROGRESS REPORT

Progress Reports should be 2 to 10 pages in length, depending on importance of the project. All the following mandatory information needs to be provided.

Reporting yearJuly 2016 to July 2017.....

Project Title:Development and testing of a microphysical aerosol scheme in the IFS.....

Computer Project Account: SPGBWOOD (phase 2).....

Principal Investigator(s): Dr Graham Mann.....
.....

Affiliation: NCAS-Climate, University of Leeds

Name of ECMWF scientist(s) collaborating to the project (if applicable) Johannes Flemming, Richard Engelen, Zak Kipling, Melanie Ades, Angela Benedetti.....
.....

Start date of the project: Jan 2015 (SPGBWOOD phase 1 ran Jan12 to Dec14)...

Expected end date: Dec 2017

Computer resources allocated/used for the current year and the previous one
(if applicable)

Please answer for all project resources

		Previous year		Current year	
		Allocated	Used	Allocated	Used
High Performance Computing Facility	(units)	3,000,000	109,585 (to Jun16) 109,615 (to Dec16)	3,000,000	0
Data storage capacity	(Gb)	30,000		45,000	

Summary of project objectives

(10 lines max)

A new aerosol module "GLOMAP-mode" has been implemented into the IFS to upgrade on the mass-based scheme developed initially and referred to here as IFS-LMD (Morcrette et al., 2009). The GLOMAP-mode aerosol microphysics scheme (e.g. Mann et al., 2010; Mann et al., 2012) simulates the evolution of the particle size distribution, with explicit sources and sinks of particle number (e.g. via nucleation and coagulation) as well as mass. The scheme tracks the same component masses as the IFS-LMD scheme (sulphate, sea-salt, mineral dust and black and organic carbon) but calculates their composition across the size range resolving internal mixtures and gas to particle transfer. In this 2nd phase of the IFS-GLOMAP special project (SPGBWOOD) the focus is on coupling the model to other aspects such as the sulphur cycle from the TM5 tropospheric chemistry scheme in C-IFS for a sulphate-coupled chemistry-aerosol-microphysics capability.

Summary of problems encountered (if any)

(20 lines max)

The resource request for the spgbwood special project was written in mid-2014 with the expectation that through MACC-III and into the CAMS period (which was intended to immediately follow on from MACC-III), there would be an ongoing PDRA at Leeds working alongside the PI (Mann) and the NCEO PhD student (Tim Keslake), with also the possibility of an additional post (either PDRA or PhD student) to take forward developments to enable an interactive stratospheric aerosol capability.

Over the 1st half of MACC-III (to June 2015), the MACC-III PDRA (Sandip Dhomse) and PhD student Tim Keslake were active users of the spgbwood special project account and also PI Graham Mann and the previous MACC-II PDRA (Will Hewson, now at Leicester University) as occasional users (mainly for referring to their account and assisting with developments & evaluation).

These considerations explain why we requested 3,000,000 SBU's per year when formulating the project – we had the expectation that there would be 2 active users each running C-IFS-GLOMAP experiments which are inherently computationally expensive since they are running atmospheric chemistry (TM5 or strat-trop chemistry scheme) and aerosol microphysics (GLOMAP) interactively.

With Leeds no longer having a PDRA post on CAMS43 aerosol developments, there has been only one user of the spgbwood project in 2017 (Keslake). Keslake is currently writing up his PhD thesis which includes analysis of biomass burning aerosol within C-IFS-GLOMAP integrations through the SAMBBA campaign period. The aerosol microphysics module is now well established in C-IFS, with development and evaluation of C-IFS-GLOMAP being carried out by the CAMS43 PDRA at CNRS (Sam Remy). The two new posts at ECMWF related to aerosol modelling (Zak Kipling, Melanie Ades) will also contribute to enable C-IFS-GLOMAP to transition into the operational CAMS system.

Summary of results of the current year (from July 2016 to June 2017)

(1 to 8 pages; can be replaced by a short summary plus an existing scientific report on the project)

Two alternative tropospheric aerosol configurations of C-IFS-GLOMAP were validated in the first part of SBGBWOOD phase 2. In this current 2nd half of SPGBWOOD phase 2, forecast-cycling experiments with both configuration are applied in experiments at both CNRS and ECMWF.

The first, "stand-alone tropospheric aerosol" configuration of C-IFS-GLOMAP mirrors the simplified "constant sulphate production" approach in the current operational aerosol scheme (Morcrette et al., 2009), e.g. as used in the MACC re-analysis (Innes et al., 2013).

The other "coupled-chemistry tropospheric aerosol" configuration of C-IFS-GLOMAP is identical to the stand-alone configuration, benefits from the 2nd SO₂ tracer and oxidation which is a core part of the TM5 chemistry scheme integrated into C-IFS during MACC (Flemming et al., 2009).

With the achievement of this milestone (validation of tropospheric aerosol capability), the focus of the SPGBWOOD grouping has shifted to analysing experiments run by others and assisting/advising with the two new configurations that are in development in this SPGBWOOD phase 2: the “interactive stratospheric aerosol” and “nitrate-extended” C-IFS-GLOMAP capabilities.

In the June 2016 progress report for SPGBWOOD, we summarized activity in 6 areas, and in this year’s report we continue this approach explaining how activity has continued well in each area as the scheme proceeds towards the operationalisation of the scheme scheduled for 2018.

1) Demonstrating how AOD assimilation improves C-IFS simulation of biomass burning aerosol

The NCEO-funded PhD student at Leeds (Tim Keskake) is currently writing up his PhD thesis, a paper planned to demonstrate how data assimilation of MODIS satellite Aerosol Optical Depth (AOD) improves the skill of C-IFS at simulated biomass burning aerosol, comparing to independent speciated aerosol mass measurements at the surface and from the FAAM aircraft during SAMBBA.

2) Further optimisation of C-IFS-GLOMAP

Progress from the Jun16 report has continued, with Dr. Hook runtime diagnostic code added, and GLOMAP settings evolved to minimise turnaround times for C-IFS-GLOMAP experiments. The specific changes are explained as part of CAMS43 deliverable report D43.1.2 (Apr17), the model now within a factor 2 of the cost of the current operational scheme for “stand-alone” experiments.

3) Paired benchmark C-IFS-GLOMAP experiments to traceably evaluate model developments

The Jun16 report explained how two pairs of benchmark CY40R3 C-IFS-GLOMAP experiments had been submitted to the AeroCom database for independent evaluation. The approach to submit pairs of experiments was to illustrate progress with the scheme, for example the step-change to include DMS emissions (absent in the MACC re-analysis) alleviating systematic biases in the forward model, and the advance to couple to the TM5 sulphur chemistry improving global sulphate prediction, and then more accurately resolving temporal variations in secondary aerosol production.

The approach has continued with CY43R2 simulations compared to CY40R3 runs and submission to AEROCOM planned to enable new developments to be evaluated also to AeroCom benchmark observational datasets. Improved skill compared to AERONET ground-based sun photometer network has resulted from switching to dust emissions (Ginoux et al., 2001) used by the operational scheme and from improved biomass burning emissions (see CAMS43 Deliverable Report D43.1.2).

4) Coupled-chemistry configurations of C-IFS-GLOMAP further tested and port to latest cycle

As explained in the Jun16 report, the coupled chemistry C-IFS-GLOMAP approach remedies an bias in the operational scheme where wintertime sulphate was overestimated over Europe. Porting this coupled-chemistry tropospheric aerosol capability to CY43R2 is complete, in readiness to link with the nitrate-extended GLOMAP (Benduhn et al., 2016) code added to C-IFS during MACC-III. An accurate sulphate seasonal cycle is essential when simulating nitrate aerosol to ensure available ammonia is correct. The surface evaluation is therefore an important step towards readying C-IFS-GLOMAP for simulating inorganic semi-volatile species. The SO₂ emissions datasets used by C-IFS-GLOMAP have also been updated to include the MACC-City inventory (Granier et al., 2011),

5) Development of C-IFS-GLOMAP for interactive stratospheric aerosol capability

In the Jun16 report, the plan to advance the C-IFS-GLOMAP capability for interactive stratospheric aerosol was explained, and two key upgrades were required to enable this. First, the GLOMAP codebase in C-IFS (v7 as used for the tropospheric aerosol hindcast in Turnock et al., 2015) has been updated to that described in Dhomse et al. (2014), thereby enabling simulation of aerosol properties across the troposphere and stratosphere. Secondly, the CAMS42 team (Vincent Huijnen at KNMI and Johannes Flemming at ECMWF) have extended the C-IFS stratosphere-troposphere chemistry (Huijnen et al., 2015) to include carbonyl sulphide, sulphur trioxide and for photolysis of several sulphur species, then matching the representation in UM-UKCA from Dhomse et al. (2014).

When updating GLOMAP, the latest stratospheric-enabled code has been used, taking in further improved capability, as explained in the attached poster by Sandip Dhomse (EGU General Assembly, Apr17) and applied for the UM-UKCA simulations submitted to the VolMIP Tambora interactive stratospheric aerosol intercomparison (see Zanchettin et al., 2016; Marshall et al., 2017).

The changes include the model now resolving heterogeneously nucleated “meteoric-sulphuric particles” (see Brooke et al., 2017), which are now established as a major component of the stratospheric aerosol layer particle community (e.g. Murphy et al., 2014).

With these upgrades, C-IFS-GLOMAP now has world-leading capability to represent the stratospheric aerosol layer and will enable C-IFS to accurately simulate the effects from volcanic injections of sulphur into the stratosphere, a key capability required for the Copernicus atmospheric monitoring system (CAM5).

Further development of the stratospheric GLOMAP module to simulate **volcanic ash** and its interactions with the sulphuric aerosol plume has also begun via the 50%-CAM543-funded PhD studentship analysis (PhD student Sarah Shallcross, supervisors Graham Mann, Anja Schmidt and Ryan Neely. For info please see attached 2-page “thesis-forward-plan excerpt” from her end-of-year-1 Transfer Report and her poster to be presented at the Aug2017 IAVCEI conference.

6) Progress towards nitrate-extended C-IFS-GLOMAP

In the Jun16 report, the description and evaluation paper for the HyDiS dissolution module (also known as “nitrate-extended GLOMAP”) was published in the Discussions forum of ACP (Benduhn et al., 2016a) and, at that time, was currently in peer review. Following the reviewers’ comments, the paper was revised (Benduhn et al., 2016b) to include also the box model testing of two-way gas-particle exchange between particle-phase chloride and gas phase HCl and evaluation of the HyDiS equilibrium algorithm in comparison to the benchmark aerosol thermodynamics code AIM-III (Clegg et al., 1998).

Research at the University of Leeds by PhD student Hana Pearce (supervised by the SPGBWOOD PI Graham Mann) has further tested the HyDiS-GLOMAP system evaluating simulations within the UK’s community composition-climate model UM-UKCA to benchmark global observational datasets of surface sulphate, nitrate and ammonium (to test the seasonal cycle). Other comparisons to aircraft (to test the vertical profile) and ground-based (to test the diurnal cycle in gas-particle exchange) Aerosol Mass Spectrometer measurements (Morgan et al., 2010a; Morgan et al., 2010b) have also been carried out as summarized in the attached poster presentation to the April 2017 EGU General Assembly (presented by Hana Pearce).

Forward-plan for last 6 months of SPGBWOOD phase 2 (July to December 2017)

Whereas the SPGBWOOD project has not yet been used during the first half of 2017, we anticipate considerable use of the account in the final 6 months of SPGBWOOD phase 2. The stratospheric component of C-IFS-GLOMAP will be tested with the Leeds research team (Mann, Dhomse) contributing to run some simulations in collaboration with the CNRS CAM543 PDRA (Samuel Remy). This testing and evaluation of integrations with the new interactive strat-trop aerosol configuration of C-IFS-GLOMAP will likely include runs with alternative 3D initialisation fields from previous UM-UKCA simulations to evolve from different points within both quiescent and volcanically perturbed runs, thereby minimizing the amount of model spin-up required.

Some initial longer integrations to test the interactive stratospheric aerosol configuration of C-IFS-GLOMAP over a multi-year cycle will likely also be carried out by the Leeds team.

List of publications/reports from the project with complete references

- Benduhn, F., G.W. Mann, K. J. Pringle et al., (2016a) “Size-resolved simulations of the aerosol inorganic composition with the new hybrid dissolution solver HyDiS-1.0: Description, evaluation and first global modelling results”, *Geosci. Model Dev. Discuss.*, doi:10.5194/gmd-2015-264, 2016
- Benduhn, F., G.W. Mann, K. J. Pringle et al., (2016b) “Size-resolved simulations of the aerosol inorganic composition with the new hybrid dissolution solver HyDiS-1.0: description, evaluation and first global modelling results”, *Geosci. Model Dev.*, 9, 3875–3906.
- Brooke, J. S. A., Feng, W., Carrillo Sánchez, J. D. et al. (2017), “Meteoric smoke deposition in the polar regions”, submitted (and revised following peer review) for *J. Geophys. Res.*
- Clegg, S. L., Brimblecombe, P. and Wexler, A. (1998): “Thermodynamic Model of the System H^+ - NH_4^+ - Na^+ - SO_4^{2-} - NO_3^- - Cl^- - H_2O at 298.15 K”, *J. Phys. Chem. A*, 102, 2155-2173.
- Flemming, J., Inness, A., Flentje, H. et al., (2009) “Coupling global chemistry transport models to ECMWF’s integrated forecast system”, *Geosci. Model Dev.*, 2, 253–265.
- Ginoux, P., Chin, M., Tegen, I. et al., (2001) “Sources and distributions of dust aerosols with the GOCART model”, *J. Geophys., Res.*, 106 (D17), 20,255-20,273
- Huijnen, V., Flemming, J., Chabrilat, et al. (2016): “C-IFS-CB05-BASCOE: stratospheric chemistry in the ECMWF Integrated Forecasting System”, *Geosci. Mod. Dev.*, 9, 3071–3091.
- Inness, A., Baier, F., Benedetti, A. et al. (2013): “The MACC reanalysis: an 8 yr data set of atmospheric composition”, *Atmos. Chem. Phys.*, 13, 4073–4109.
- Marshall, L., Schmidt, A., Toohey, M. et al. (2017): “Multi-model comparison of the volcanic sulfate deposition from the 1815 eruption of Mt. Tambora”, *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-729>,
- Morcrette, J.-J., Boucher, O., Jones, L. et al. (2009): “Aerosol analysis and forecast in the European Centre for Medium-Range Weather Forecasts Integrated Forecast System: Forward modelling”, *J. Geophys. Res.*, vol. 114, D06206, doi:10.1029/2008JD011235.
- Morgan, W. T., Allan, J. D., Bower, K. N. et al. (2010a): “Airborne measurements of the spatial distribution of aerosol chemical composition across Europe and evolution of the organic fraction”, *Atmos. Chem. Phys.*, 10, 4065–4083.
- Morgan, W. T., Allan, J. D., Bower, K. N. et al. (2010b): “Enhancement of the aerosol direct radiative effect by semi-volatile aerosol components: airborne measurements in North-Western Europe”, *Atmos. Chem. Phys.*, 10, 8151–8171.
- Murphy, D. M., Froyd, K. D. Schwarz, J. P. et al. (2014): “Observations of the chemical composition of stratospheric aerosol particles”, *Q. J. Roy. Meteorol. Soc.*, 140: 1269–1278.
- Turnock, S. T., Spracklen, D. V., Carslaw, K. S. et al. (2015): “Modelled and observed changes in aerosols and surface solar radiation over Europe between 1960 and 2009”, *Atmos. Chem. Phys.*, 15, 9477–9500, 2015.
- Zanchettin, D., Khodri, M., Timmreck, C. et al. (2016): “The Model Intercomparison Project on the climatic response to Volcanic forcing (VolMIP): experimental design and forcing input data for CMIP6”, *Geosci. Model Dev.*, 9, 2701–2719, 2016

Summary of plans for the continuation of the project (10 lines max)

The two phases of the SPGBWOOD special project have successfully incorporated the GLOMAP aerosol microphysics module into C-IFS. Recently appointed ECMWF staff are directly aligned to global aerosol modelling (Zak Kipling) and to completing the link-up of C-IFS-GLOMAP to the AOD data assimilation capability (Melanie Ades). With onward development of C-IFS-GLOMAP being principally carried out by CNRS (Sam Remy) with Leeds advising and assisting, we not anticipate needing to apply for a third phase of the SPGBWOOD project.

Sandip Dhomse, Graham Mann, Lauren Marshall, Ken Carslaw, Martyn Chipperfield (Univ. Leeds, U.K.), Nicolas Bellouin (Univ. Reading, U.K.), Olaf Morgenstern (NIWA, New Zealand), Colin Johnson, Fiona O'Connor (UK Met Office, Exeter, U.K.)

1. Introduction

- Volcanic eruptions can inject large quantity of SO_2 into the stratosphere which oxidises forming source of gas-phase H_2SO_4
- Grows existing particles to larger sizes creating optically thicker stratospheric aerosol layer and exerting direct surface cooling.
- Increased stratospheric aerosol surface area density enhances heterogeneous chemistry causing chemical O_3 loss
- Applied UKCA stratosphere-troposphere composition-climate model to generate microphysically-consistent volcanic forcing datasets for 1991 Pinatubo, 1982 El Chichon and 1963 Agung eruptions
- Includes GLOMAP aerosol microphysics and has been shown to capture global variation in particle size distribution in the stratosphere both in quiescent & volcanically perturbed conditions
- Now also includes important diversity of stratospheric particles (both MSP-sulphuric and pure sulphuric droplets)
- Host UM-UKCA has 85 vertical levels for realistic representation of dynamical loss pathways for the volcanically-enhanced aerosol.

2. Lifecycle of stratospheric aerosol

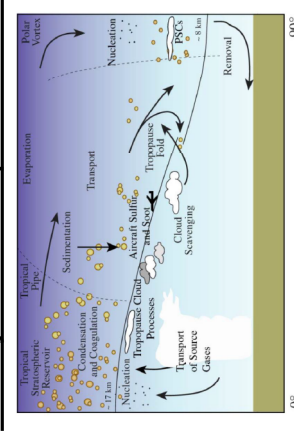


Fig 1: Illustration of the processes maintaining the background stratospheric aerosol
Carslaw & Karcher (SPARC ASAP report, 2006)

5. Stratospheric AOD after Agung, El Chichon and Pinatubo

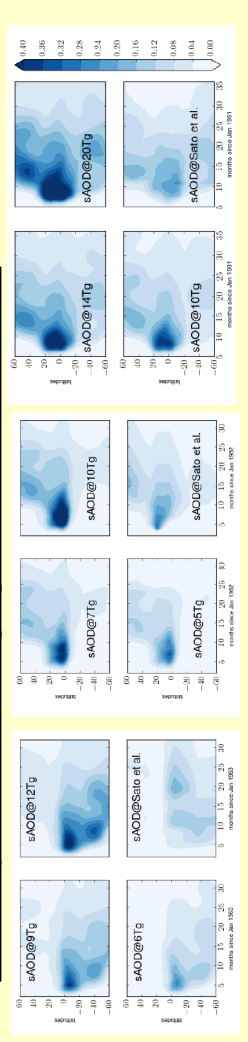


Figure 5: (a,c,d) Mid-visible (550nm) Stratospheric aerosol optical depth comparison. Model has internally-generated QBO and is initialised to capture hemispheric dispersion pattern for each eruption. In each case, lower end of SO_2 injection range compares best to Sato et al. (1993) dataset consistent with findings for Pinatubo in Dhomse et al. (2014)

6. Effective Radius

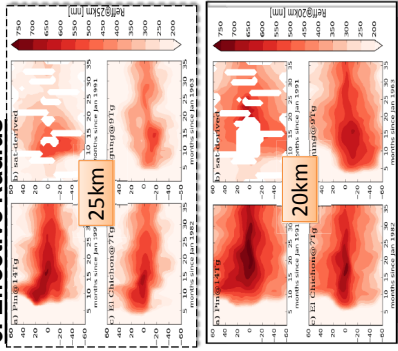


Figure 6: (a,c,d) Simulated effective radius (R_{eff}) at 25km (top) and 20km (bottom) for each eruption. (b) shows multi-sensor satellite R_{eff} for Pinatubo period: SAGE-II and CLAES (Bauman et al., 2003).

7. Extinctions comparison for Pinatubo period

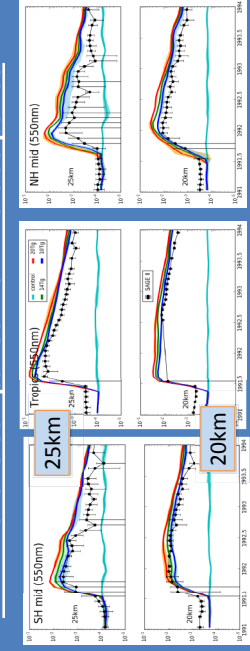


Figure 7: Comparison between modelled extinctions at 550nm with different injection amount and SAGE V7.0 measurements (525nm) for (right) SH mid-latitudes (35S-60S), tropics (20S-20N) and (left) NH mid-latitudes (35N-60N) at 25km (top) and 20km (bottom). Shaded and solid line indicate variance and mean from three-member ensemble. Standard deviation from SAGE II measurements for a given month and latitude bands is indicated with vertical black lines (updated from Dhomse et al., 2014)

3. UKCA sulphur chemistry & aerosol microphysics

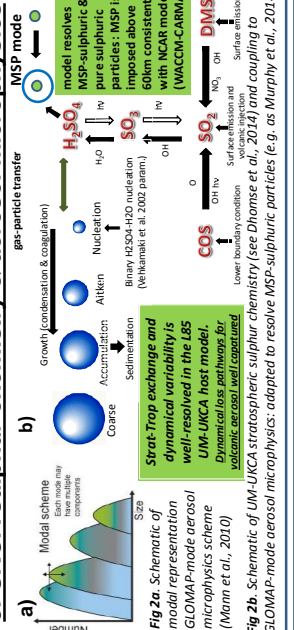


Fig 2a. Schematic of GLOMAP-mode aerosol microphysics scheme (Mann et al., 2010)
Fig 2b. Schematic of UM-UKCA stratospheric sulphur chemistry (see Dhomse et al., 2014) and coupling to GLOMAP-mode aerosol microphysics: adapted to resolve MSP-sulphuric particles (e.g. as Murphy et al., 2014)

8. SAD comparison against CMIP6 3-λ dataset

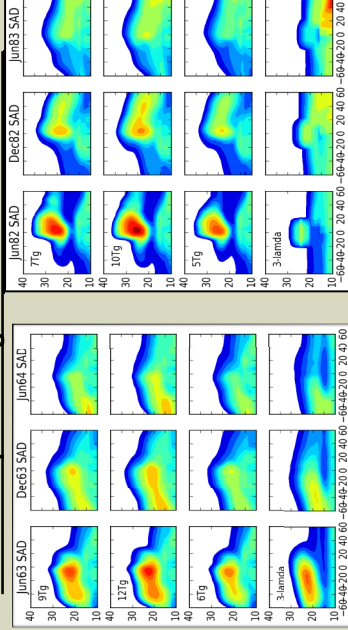


Figure 8: Simulated Surface Area Density (SAD) from mid, high, low "SO₂-injection-realizations", shown at 6-month winter/summer intervals after Agung, El Chichon & Pinatubo. Compares well with CMIP6 dataset and notably has considerably larger SAD in the upper troposphere in all three post-eruption periods. The 3D model for this new dataset resolves dynamical loss pathways for volcanically-enhanced particles more realistically than the 2D model used for the CMIP6 dataset.

4. Model experiments to simulate stratospheric aerosol enhancement caused by major tropical eruptions

- Carried out free-running transient simulations in high-top (80km) version of v8.4 UM-UKCA (GA4) GCM with time-varying prescribed SSTs & sea ice as AMIP2.
- Three sets of 3-member ensemble of simulations for each eruption (mid, high and low SO_2 injection amount) initialised carefully
- Model is internally-generated QBO initialised carefully
- Pinatubo**: at 21-23km (14, 20, 10 Tg) on 15th June 1991, (0-15N spread; easterly QBO through 2nd half of 1991.
- El Chichon**: at 24-28km (7, 10, 51g) on 4th April 1982 (5-18N spread; QBO transitioning easterly-into-westerly)
- Agung**: at 20-22km (9, 12 and 6 Tg) on 17th March 1963 (12S-2N spread; QBO transitioning easterly-into-westerly)
- Compare to observation-only-based volcanic forcing dataset (Sato et al., 1993) & identify objectively which SO_2 injection compares best to available observations for each eruption.

9. Model Pinatubo forcing time-signatures vs ERBE anomaly timeseries (10Tg eQBO runs)

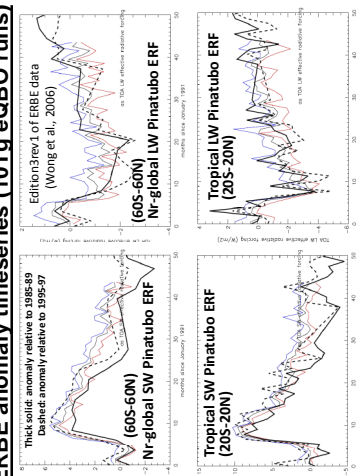


Fig 9 Time-signature of 10Tg Pinatubo SW & LW effective forcing (cooling/warming signal) consistent with ERBE in tropics & near-global.

10. Summary

- New volcanic forcing datasets produced from 3D composition-climate model with well-resolved stratospheric dynamics and aerosol microphysics.
- High, low and mid SO_2 injection runs for Agung, El Chichon & Pinatubo compared to Sato et al (1993) dataset which has many observational constraints.
- For all three eruptions, the lower SO_2 injection amount compares best to multiple observations.
- Microphysically-consistent 3D volcanic aerosol datasets: SAD for chemistry-climate models and aerosol optical properties for historical climate runs.
- Future work will quantify how much of the model volcanic forcing time-signature (Fig. 9) is caused by direct aerosol ERF & how much from indirect effects.

11. References

- Bauman et al. (2003), *J. Geophys. Res.*, 108, D13, doi:10.1029/2002JD002992.
Carslaw & Karcher (2006), chapter 1, *SPARC-ASAP report*.
Dhomse et al. (2014) *Atmos. Chem. Phys.*, 14, 2799-2855. S.S.Dhomse@leeds.ac.uk
Mann et al. (2010), *Geosci. Model. Dev.*, 3, 519-551. G.W.Mann@leeds.ac.uk
Morgenstern et al. (2009), *Geosci. Model. Dev.*, 2, 43-57.
Murphy et al. (2014), *Q. J. Roy. Meteor. Soc.*, 140, 1269-1278.
Sato et al. (1993), *J. Geophys. Res.*, vol. 98, D12, 22,987-22,994.
Wong et al. (2006), *J. Climate*, 19, 4028-4040.

2 AIMS AND OBJECTIVES

Project aim: To identify how significantly ash influenced the global dispersion and radiative forcing from explosive volcanic eruptions.

This thesis will investigate radiative and microphysical interactions of volcanic ash, focussing on how these impact the global dispersion of sulphuric aerosol plumes from major tropical eruptions. This will be achieved by initially comparing interactive stratospheric aerosol simulations of the 1991 Pinatubo plume to lidar observations from near-tropical, mid-latitude and high-latitude in the first year following the Mount Pinatubo 1991 eruption. The analysis links with in situ balloon measurement observations, that show fine ash particles ($\sim 0.1\text{-}0.2\ \mu\text{m}$) were present both individually and within sulphuric particles in the stratosphere for several months following the eruption (Deshler, 2016).

In order to accomplish this aim, this study will involve introducing an ultra-fine ash element into UM-UKCA and running experiments with and without the ash-sulphuric interactions. Extinction profiles will then be compared both to the original lidar analysis and particle count profiles from balloon-borne measurements.

Model experiments will be designed to explore the two mechanisms by which volcanic ash influences global dispersion.

- 1) **Radiative interaction** – The absorption of short wave (solar) and long wave (terrestrial) radiation by ash particles heats the air surrounding the ash, lofting the plume.
- 2) **Microphysical ash-sulphuric interaction** – After Pinatubo, a combination of “pure” sulphate particles and sulphuric acid coated ash particles existed, with these “ash sulphuric” particles being removed much more effectively than those “pure” sulphate particles, due to their larger sizes.

These mechanisms act in opposite ways. Mechanism 1) acts to likely enhance the plume lifetime by lofting particles and particle rich air masses to higher altitudes (further from the tropopause). Mechanism 2) however, will likely shorten the lifetime of particles, as ash-included particles are larger than they would otherwise be and will likely sediment out faster.

The UM-UKCA interactive stratospheric aerosol simulations will explore each effect individually and then assess what the net effect of the two processes are in the changing of the efficacy of the radiative effects of the emitted sulphur.

In particular, the modelling will explore how process 2) accelerates the removal of volcanic sulphur in the first few months after the eruption. It is hoped this will help explain why, for the

1991 Pinatubo eruption, the observed peak in global sulphur mass in the sulphuric particles was only half that of the observed SO₂ in the first few days after the eruption (see discussion in Dhomse *et al.*, 2014).

Hypothesis 1 – Radiative interaction:

The absorption of short-wave radiation and longwave radiation by ash particles heats the air surrounding the ash, lofting the plume. Consequently, it is hypothesised that the inclusion of ash within the model will affect the dispersion of the volcanic plume. It is believed this lofting will drive particles to higher altitudes in the early stages after the eruption.

Hypothesis 2: Microphysical ash-sulphuric interaction:

A combination of “pure” sulphate particles and sulphuric acid coated ash particles existed after Pinatubo. These ash-sulphuric particles are believed to be removed more effectively than those “pure” sulphate particles. Therefore, it is hypothesised that more sulphate will be removed as a result of the ash being included within the model, as speculated by balloon-borne measurements in Deshler (2016).

Below there is an initial thesis plan. If thesis by publication is possible, potential papers are outlined in section 5.2.

Preliminary thesis chapter outline:

Chapter 1 Introduction and motivation

Chapter 2 Literature review (stratospheric aerosol, observations and models)

Chapter 3 UM-UKCA model overview: representing stratospheric aerosol and ultra-fine ash

Chapter 4 Lidar analysis of first phase of global plume dispersion

Chapter 5 Baseline and ash lofting (ash-radiation interaction) vs. lidar and SAGE II?

Chapter 6 Influence of ash-sulphuric interaction – compare to lidar, SAGE II and balloon measurements

Chapter 7 Overall effects of fine-ash on volcanic forcing from major tropical eruptions

Chapter 8 Summary and conclusions

1) Introduction

- Explosive volcanic eruptions are capable of injecting huge quantities (10s of teragrams) of sulphur dioxide (SO₂) and ash into the atmosphere.
- These cause significant enhancement to the stratospheric aerosol layer and have complex effects on Earth's climate (Fig.1').
- Volcanic enhancement to the "background levels" of stratospheric sulphuric acid aerosols occur chemically as the emitted SO₂ is oxidised in the atmosphere².
- This volcanic perturbation acts to increase both scattering of solar radiation and also absorption of longwave radiation in the stratosphere, resulting in a net cooling effect.

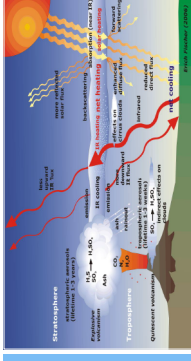


Figure 1. Updated from Robock (2000) a schematic diagram of subsequent and explosive volcanic effects on the radiative balance of the Earth (Bischof et al., 2007).

4) Initial analysis

- Initial analysis of lidar data from Mauna Loa (19.4°N) (Fig.4) show layers within the early stages of the transport of the volcanic cloud.
- Some initial UM-UKCA (Unified Model – United Kingdom Chemistry and Aerosols) model simulations also indicate these layers (Fig.5).

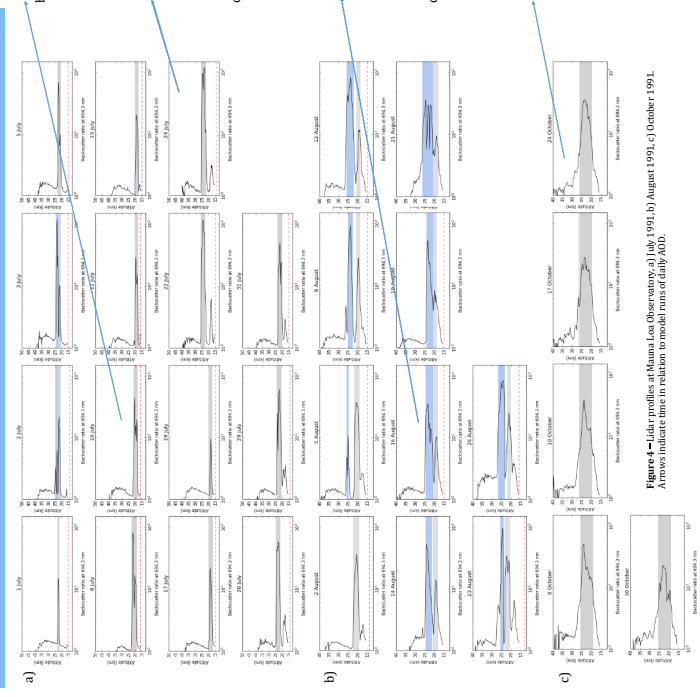


Figure 4 – Lidar profiles at Mauna Loa Observatory. (a) July 1991, (b) August 1991, (c) October 1991. Arrows indicate time in relation to model runs of daily ash.

2) Ash I - Radiative effect and residence time

- The radiative effects caused by particles is strongly influenced by how large particles in the stratosphere become after volcanic perturbation. For example, radiative calculations indicate that if the effective radius of the particle (area weighted mean radius of the aerosol particles) exceeds 2 µm then the eruption would have a warming effect on the surface^{3,6}.
- Particles > 1 µm have sedimentation rates of between a few hours and 5 days⁴ (Fig.2a). This restricts their abundance in the stratosphere and, therefore, their warming effect⁵, however, localised short-term heating can occur which can affect aerosol distribution within volcanic plumes⁷.
- Ash is largely disregarded in many atmospheric studies due to these short residence times. However, fine-ash particles (< 1 µm) can persist for years (Fig.2b)^{7,8}.

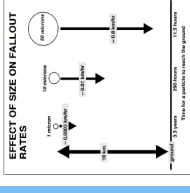


Figure 2b - From the literature, it is estimated that 50 µm particles will fall out within the first day of an eruption, whereas < 1 µm may persist for years in the atmosphere.

4b) Lidar – model comparison

- Figures 6 and 7 show observational lidar against UM-UKCA (Fig.6) plots over the same time period, both showing these progressive layers in the early months following the eruption, becoming more homogenous in later months.
- There are discrepancies between the heights and timings of these layers, potentially due to this lack of ash within the model.

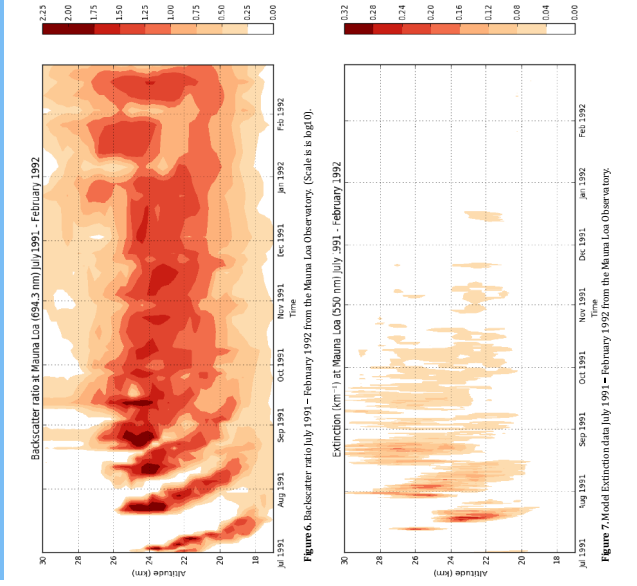


Figure 6. Backscatter ratio July 1991 – February 1992 from the Mauna Loa Observatory. (Scale is log 10).

Figure 7. Model Extinction data July 1991 – February 1992 from the Mauna Loa Observatory.

3) Ash II – self lofting and scavenging effects

- H₂SO₄ and H₂O coated ash particles have been found in the stratosphere up to 8 months after the Mount Pinatubo eruption^{9,10} (Fig.X) which may indicate SO₂ scavenging both in the early and later stages of aerosol cloud development.
- Ash-core particles occur when the co-existence of ash and SO₂ causes some ash particles to become coated in sulphuric material, consequently becoming a sulphate particle with a fine-ash inclusion¹⁰.
- These particles can persist for up to 3 months, as found with the Kaula 2014 eruption, potentially longer for explosive eruptions such as Mount Pinatubo 1991^{9,10}.

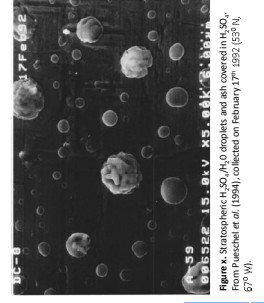


Figure X. Scanning electron microscope (SEM) images of ash particles, ash coated in H₂SO₄ from Pinatubo et al. (1994), collected on February 17th 1992 (135°N, 67°W).

5) UKCA sulphur chemistry & aerosol microphysics

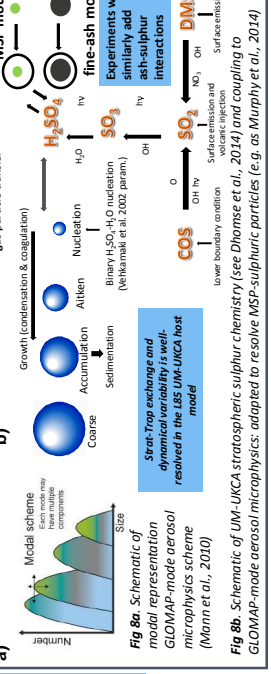


Fig 8a. Schematic of the UKCA stratospheric sulphur chemistry (see Dhomse et al., 2014) and coupling to GLOMAP-mode aerosol microphysics scheme (Mann et al., 2010)

6) Future work

- Adding initial dust-scheme to simulate ash in UM-UKCA (using soluble and insoluble modes from GLOMAP)
- Control simulations without ash – SO₂ dispersion.
- Simulations with ash
- How are self-lofting and ash-sulphate particle removal modelled?
- How do these compare to observations?

References

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1. Background

Semi-volatile inorganic aerosol species, such as ammonium nitrate, form a substantial component of the sub-micron particulate composition in polluted regions of Europe.

Furthermore, nitrate aerosol concentrations exhibit strong diurnal variability and the relative contribution of nitrate to $PM_{2.5}$ is substantially increased in high mass loading events (e.g. 1.2¹).

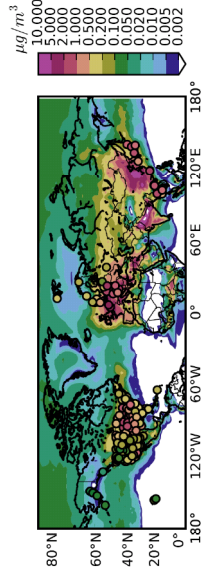


Figure 1. Annual-mean simulated nitrate aerosol concentration with filterpack observations from the EMEP, CASTNET, IMPROVE and EANET monitoring networks.

The UM-UKCA model with the GLOMAP hybrid solver for inorganic aerosol dissolution⁽³⁾ performs well against observed inorganic aerosol concentrations on annual and monthly-mean timescales⁽⁴⁾ (e.g. Figure 1).

Here, we present an evaluation of hourly UM-UKCA simulated nitrate aerosol concentrations at Cabauw, the Netherlands, in May 2008 and investigate the importance of considering the diurnal variability of nitrate in calculations of regional aerosol direct radiative effects.

This time period in May 2008 is well observed and has been the subject of several previous model evaluation studies of simulated inorganic gas-to-aerosol partitioning and regional radiative effects (e.g. 5,6,7⁵).

2. Model Setup

- HadGEM3-UKCA composition-climate model (v7.3 HG3-A+r2 CheT + GLOMAP).
- 1.25° latitude x 1.874° longitude with 63 vertical levels.
- GLOMAP-mode + HyDis hybrid dissolution solver of inorganic gas-to-particle partitioning.
- Tropospheric heterogeneous chemistry included.
- Year 2008 MACCity anthropogenic emissions and ERA-Interim nudged meteorology.
- Hourly output frequency at grid boxes containing EMEP and EUCAARI observation sites.

3. Results: Near-Surface Diurnal Variability at Cabauw

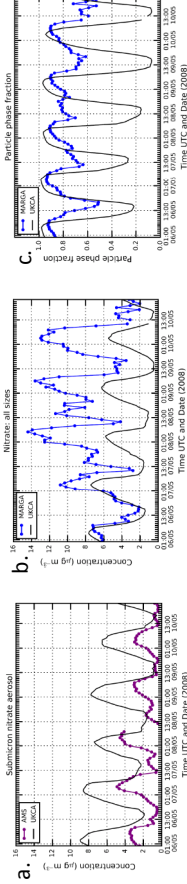


Figure 2. Nitrate near-surface diurnal variability from 6 – 11th May 2008. a) UKCA-simulated and AMS-observed submicron nitrate aerosol concentrations at 60m height. b) UKCA-simulated and MARGA-observed nitrate aerosol concentrations at 4m height. c) UKCA-simulated and MARGA-observed particulate phase fraction i.e. particulate nitrate / (gaseous nitric acid + particulate nitrate).

4. Results: Vertical Diurnal Variability

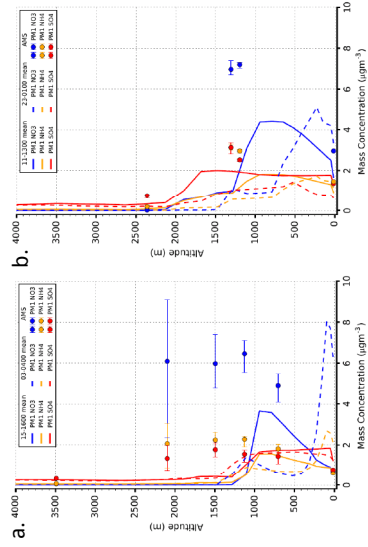


Figure 3. AMS-observed (circles) and UKCA-simulated (solid lines) vertical distribution of nitrate, ammonium and sulphate aerosol in the vicinity of Cabauw (4°-6°E, 51°-53°N) at the times indicated on a) 6th May flight B379 B366 and b) 21st May flight B379 B368. Simulated night-time concentrations are also presented (dashed lines) to indicate diurnal variability.

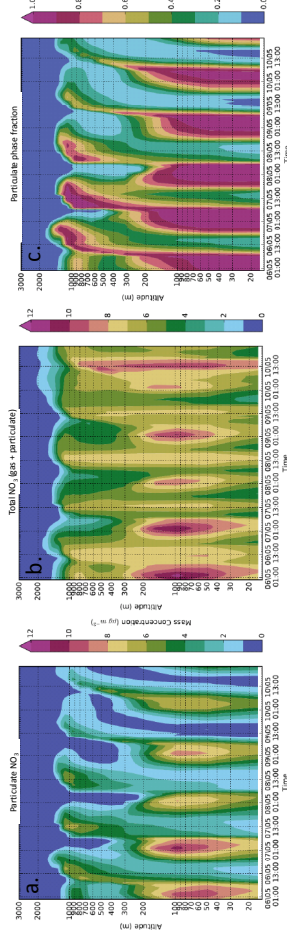


Figure 4. Diurnal variability in simulated vertical distribution of a) particulate nitrate b) total nitrate (nitric acid + particulate nitrate) and c) particulate phase fraction.

5. Key Points

- UKCA-simulated ammonium nitrate concentrations are consistent with ground-based and airborne observations during high pollution conditions at Cabauw, the Netherlands, in May 2008.
- Key features of the nitrate aerosol concentration include:
 - Strong diurnal variability near the surface (Fig. 2).
 - Peak concentrations in the upper boundary layer during the daytime, although the UKCA underestimates the magnitude and altitude of the peak concentration (Fig. 3).
 - Diurnal variability in the altitude of maximum nitrate concentration. We note that simulated partitioning toward the particle phase is favoured at the surface during night-time and in the upper boundary layer during day-time (Fig. 4). Other processes (e.g. dynamical, chemical) also play a role.

We conclude that gas-to-particle partitioning of inorganic aerosol on diurnal timescales should be included in models because it is may have implications for regional radiative forcing calculations and evaluation of model skill against observed aerosol optical depth (AOD).

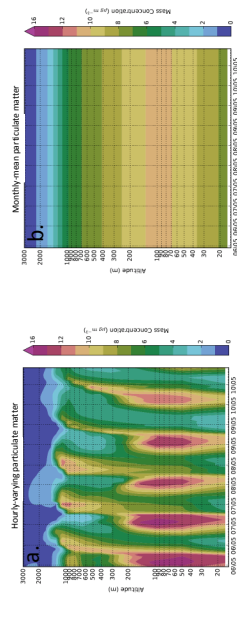


Figure 5. Simulated a) hourly-mean and b) monthly-mean total particulate matter over Cabauw from 6th to 11th May 2008.

Future Work:

- To compare the clear sky direct radiative effects in the column above Cabauw with hourly-varying and monthly mean aerosol fields e.g. Fig. 5.
- To identify regions where this strong diurnal variability in nitrate aerosol concentration occurs, with the aim of applying a flag or reduced weight to these sites when evaluating models against AOD measurements.

6. References

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7. Acknowledgements

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