

Long-lived <u>ultra-fine ash particles</u> within the Mt Pinatubo volcanic aerosol cloud and their potential impact on its global dispersion and radiative forcing

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Very large magnitude tropical eruption of Mt Pinatubo

- Kerreich and Stein Albert And Stein Albert A
- 14-23 Tg SO₂, ~50 Tg ash emitted at ~21-23km -- well above tropopause
- Largest eruption of 20th C, & observed by several <u>satellite instruments</u>,
 - ground-based lidar at Mauna Loa (19N) & range of mid-latitude sites
 - balloon-borne particle counter soundings at Laramie, USA & Lauder, NZ.





Credit: USG





UK Chemistry and Aerosol project

- Collaboration between UK National Centre for Atmospheric Science (Leeds, Cambridge, Oxford) & UK Met Office since 2005
- Has built aerosol-chemistry sub-model in the UK Met Office
 Unified Model, being applied for a range of applications (climate, air quality, Earth system science, weather)
- Chemistry schemes & aerosol configurations including for stratosphere-troposphere



- Multi-component aerosol microphysics scheme (GLOMAP)
- Global variations in particle size
 WWW . UkCa . aC . Uk
 distribution → sedimentation and SW & LW radiative effects
- UKESM includes UKCA chemistry-aerosol sub-model so that it simulates strat-trop ozone and strat-trop aerosol interactively, each radiatively coupled for composition-dynamics interactions.

Global column sulphur-loadings of SO₂ & aerosol



Stratospheric aerosol optical properties



v7.3 UM-UKCA N48L60 CheS+GLOMAP.

Dhomse et al. (ACP, 2014)



Radiative heating effect on sAOD evolution vs SAGE-II sAOD & AVHRR anomaly

v7.3 N48L60 CheS+GLOMAP with radiative coupling (runs in Dhomse14 had no strat-heating)

v7.3 CheS+GLOMAP N48L60

Stratospheric aerosol sulphur burden in UM-UKCA (thin green) vs HIRS (thick green)



Impact of radiative coupling on lifetime in stratosphere

v7.3 CheS+GLOMAP N48L60

Stratospheric aerosol sulphur burden in UM-UKCA (thin green) vs HIRS (thick green)





Here explore influence from co-emitted ash

- Volcanic ash mostly omitted in global composition-climate model studies with particles assumed to sediment from plume in ~ 1 week.
- However, <u>ash absorption of incoming solar</u> (& outgoing LW) shown to affect the progression of the volcanic aerosol clouds from 1991 Pinatubo (Niemeier et al., 2009), 2014 Kelut (Zhu et al., 2020) and from 2019 Raikoke (Muser et al., 2020).
- Also, <u>impactors on ER-2 & DC-8 aircraft surveys of Pinatubo aerosol cloud</u> during AASE II campaign (Jan 1992 – Mar 1992) collected particles from the Pinatubo plume clearly showing <u>the lower portion of the plume</u> contained ash particles ~0.6-2 µm diameter, even through to February 1992.



Mixed "ash-sulphuric particles" remained present in lowest ~10% of Pinatubo aerosol cloud

(~10km) even 8 months after the eruption

Measurements from impactors on <u>ER-2</u> (~20km) and <u>DC8</u> (~10km), see Pueschel et al., JGR, 1994)

of individual droplets by SEM, as shown in Figure 1. Thirty five of the ER-2 samples and 21 from the DC-8 that were amenable to the size discrimination analysis (examples of which are shown in Figures 1 and 2) are summarized in Tables 1a and 1b, respectively. This represents a sufficiently large sample population to demonstrate the effects of the 1991 Pinatubo volcanic eruption on the physical and elemental characteristics of the stratospheric aerosol.

It follows from Table 1 that on a few occasions at ER-2 altitudes and quite regularly at the lower DC-8 altitudes, a third large-particle mode with geometric mean radius around $0.8 \ \mu m$ existed. SEM imagery identified the shape of these large particles as slightly nonspherical (Figure 3), in contrast to the particles comprising modes 1 and 2 (Figure 1). X ray energy-dispersive analysis (Figure 4b) determined an elemental composition of these particles that is typical of mineral ash. This is different from the X ray energy spectra of the ER-2 samples (Figure 4a), the morphology of which is shown in Figure 1. In these samples the only peak aside from



Figure 3. Plane view of stratospheric H_2SO_4/H_2O droplets and sulfuric-acid-coated volcanic ash particles collected on February 17, 1992, near (53°N; 67°W) at 10.7 km msl.

8 months after the Pinatubo eruption (Feb 92) At 53°N, 67°W, 10.7km

3 rd "ash-sulphuric mode" ~0.6-2 µm mean diam							6-8 months after the Pinatubo eruption							
					Mode 1			Mode 2			Mode 3			
At latitudes 42-87N		\bigcirc		Alt,	N ₀₁	r _{g1} ,		N ₀₂₁	r _{g2} ,		N ₀₃	r _{g3} ,		
Sample	Date	Lat	Long	km	cm_,	μm	σ_{g1}	cm ^{-,}	μm	σ_{g2}	cm ⁻³	μm	σ_{g3}	
1	Jan. 14, 1992	N65.0	W108.0	11.3				0.8	0.29	1.6				
2	Jan. 14, 192	N67.0	W128.5	12.6				3.0	0.17	1.4	0.3	0.77	1.1	
3	Jan. 14, 1992	N67.0	W139.0	12.6	1.7	0.12	1.6	1.3	0.41	1.4				
4	Jan. 16, 1992	N87.0	W93.0	11.3	7.5	0.14	1.9							
5	Jan. 16, 1992	N82.0	W53.0	11.3	9.0	0.13	2.0							
6	Jan. 16, 1992	N66.0	W17.5	12.0	9.1	0.14	1.8				0.3	0.81	1.1	
7	Jan. 19, 1992	N66.0	W51.0	11.3	4.1	0.14	1.9				0.1	1.03	1.2	
8	Jan. 19, 1992	N65.0	W67.0	11.3	5.0	0.10	2.1							
9	Jan. 19, 1992	N59.0	W68.0	11.3	4.7	0.12	2.0				0.3	0.80	1.2	
10	Jan. 22, 1992	N63.0	W100.0	11.3	7.8	0.19	1.6				0.5	0.83	2.2	
11	Jan. 22, 1992	N63.0	W100.0	11.3	2.5	0.04	1.6	1.5	0.20	1.4				
12	Jan. 23, 1992	N68.0	W87.0	11.3	12.5	0.07	1.4	8.0	0.16	1.7	0.4	0.86	1.1	
13	Feb. 12, 1992	N52.0	W122.0	10.7	7.7	0.07	1.3	5.5	0.13	1.3	4.8	0.28	1.6	
14	Feb. 12, 1992	N71.0	W115.0	10.7	8.5	0.10	1.8	3.7	0.31	1.3	0.6	0.62	1.3	
15	Feb. 12, 1992	N73.0	W127.0	12.0	2.2	0.07	1.3	4.8	0.23	1.8				
16	Feb. 17, 1992	N69.5	W13.0	10.7	6.8	0.05	1.3	3.4	0.14	1.8	0.4	0.86	1.1	
17	Feb. 17, 1992	N76.0	W53.0	11.3	4.8	0.06	1.2	9.8	0.17	1.7	0.4	0.77	1.2	
18	Feb. 17, 1992	N51.5	W67.0	11.3	7.7	0.06	1.2	8.3	0.20	1.7	0.7	0.78	1.1	
19	Feb. 20, 1992	N42.0	W72.0	9.5	10.1	0.13	1.2				0.6	0.84	1.1	
20	March 14, 1992	N87.5	W8.0	10.1	24.9	0.08	1.5	3.2	0.28	1.4	0.6	0.71	1.2	
21	March 20, 1992	N44.0	W106.0	11.6	16.9	0.05	1.2	5.3	0.13	1.5	3.2	0.41	1.6	
												$\mathbf{\vee}$		

Table 1b. Lognormal Characteristics of Particle Size Distributions From DC-8 Samples Collected Between 9.5-and 12.6-km

X-ray aerosol composition analysis

Measurements from impactors on ER-2 and DC8 (Pueschel et al., JGR, 1994)



0.000

Figure 3. Plane view of stratospheric H₂SO₄/H₂O droplets and sulfuric-acid-coated volcanic ash particles collected on February 17, 1992, near (53°N; 67°W) at 10.7 km msl. 8 months after Pinatubo eruption (Feb 92) At 53°N, 67°W, 10.7 km





contrast, a nonspherical particle of a DC-8 sample (Figure 3) shows additional peaks (Figure 4b) corresponding to the K_{α} lines of Na at 1.05 keV, of Al at 1.48 keV and of Si at 1.74 keV. Since these elements are typical of minerals, including volcanic ash, we conclude that the large nonspherical particles that were collected aboard the DC-8 are those of ash coated with sulfuric acid. Their predominant location near the bottom of the stratospheric aerosol layer alludes to a higher settling velocity due to both a larger size and greater density of these particles than of the H₂SO₄/H₂O droplets. This finding explains the occasional depolarization of lidar backscattering near the bottom of the Pinatubo layer [*Brow-ell et al.*, 1993].

Q: How widespread are these ash-sulphuric particles?



polarised power) and mean monthly temperature for 6th December 1991 and 21st February 1992 are plotted in Figures 6 and 7 respectively. The depolarisation data have been scaled to a value of 0.014 for the clear air below the tropopause (approx. 10 km). Most noticeable is the very low depolarisation in the region of the main Pinatubo layer (19 - 25 km) and the relatively highly depolarising layer around 16 km. The fact that the layer is still present at the same height on both *δ*>0.01 @10-19km dates and is not falling suggests that it is not the primarily the result of fallout of irregular dust particles. Other possible explanations for the enhanced depolarisation are the agglomeration of partially frozen droplets or a complete phase change (E. Browell, personal communication). Support for this latter suggestion comes from the temperature profiles which show a minimum at the height of peak depolarisation.

Young et al. (1994, ILRC92 extended abstracts)

Young, S. A. et al. (Extended Abstracts of the 1992 International Laser Radar Conference, 1994)

20-24km

(main layer)

6th Dec '91

(base of the layer)

6th Dec '91

Lidar measurements of Mt. Pinatubo aerosols at Aberystwyth from August 1991 through March 1992

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1.3

34

32

30

26

24

22

20

18

16

Ê 28

HEIGHT,

BOVE 7.0 14

3.0 - 5.0 12

5.0 - 7.0

2.0 - 3.0

1.3 - 1.6 1.0 - 1.3

DAY NO. OF 1991 DAY NO. OF 1992 BELOW 1.0 Fig.3: Contour plot of backscatter ratio (in the parallel channel). Vertical bars along the top denote nights when observations were made. Data were averaged over 450 m to produce this plot, thus the maximum values shown differ from those in fig.1. The darkest shading corresponds to R > 7 and the next darkest to R > 5. Vaughan et al. (1994, GRL)

1.6

20

40

60

80

220 240 260 280 300 320 340 360

Lowest few km of Pinatubo cloud clearly depolarizing Aug91 to Feb92 In NH mid-latitude stratosphere.

Can see the steady progression with the depolarizing region slowly descending, consistent with slow sub-µm ash removal



Fig.5: As fig.3 for depolarisation ratio. Contour interval is 1%.

- The presence of the depolarizing ash-sulphuric particles clearly evident in lidar measurements from Aberystwyth.
- -- peak depolarization between 18-20km in Aug 1991
- -- descends steadily to 15-17km Sep-Dec and ~12km Jan-Feb 1992



Stratosphere-enabled GLOMAP and applications

- GLOMAP developed to simulate the global variation in particle size distribution (new particle formation and growth via coagulation, condensation and cloud processing).
- Initially applied only for the tropospheric aerosol layer (Mann et al., 2010; Bellouin et al., 2013; Turnock et al., 2015)
- Then adapted in UM-UKCA to simulate aerosol microphysics across the stratosphere & troposphere (Dhomse et al., 2014)
- Through 2016-7, "strat-enabled GLOMAP" further developed to form stratospheric aerosol particles on meteoric smoke particles, alongside homogeneously nucleated particles
- Strat-enabled GLOMAP within UM-UKCA applied for VolMIP Tambora-ISA expt (Marshall et al., 2018; Clyne et al., 2021).
- And to simulate major volcanic aerosol from 1963 Agung, 1982 El Chichon & 1991 Pinatubo (Dhomse et al., 2020).

CAMS43-Leeds PhD at Leeds on volcanic aerosol (Sarah Shallcross)

Part of co-funded PhD studentship developed GLOMAP v8.3 further to co-emit volcanic ash with volcanic SO₂ \rightarrow understand radiative & microphysical effects on plume dispersion.

UM-UKCA now has GLOMAP simulating uptake of H_2SO_4 onto volcanic ash and radiative effects:



Figure 5. Stratospheric H₂SO₄/H₃O droplets and ash covered in H₂SO₄. From Pueschel *et al.* (1994), collected on February 17th 1992 (53^o N, 67^o W).



Evaluate 21-23km, 18-20km, 18-25km injection height UM-UKCA simulations, comparing to lidar measurements at Mauna Loa, and at NH mid-latitudes.

14 Tg of SO2 emitted at 21-23km altitude

Extinction (550 nm) at Mauna Loa

This is standard UM-UKCA Pinatubo with only SO2 (no co-emitted ash)



Evaluate 21-23km, 18-20km, 18-25km injection height UM-UKCA simulations, comparing to lidar measurements at Mauna Loa, and at NH mid-latitudes.

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14 Tg of SO2 emitted at 18-25km altitude

Extinction (550 nm) at Mauna Loa

This is standard UM-UKCA Pinatubo with only SO2 (no co-emitted ash)



Initial 10-day test <u>Pinatubo ash runs</u> <u>co-emitting SO₂ &</u> <u>different sized ash</u> (1, 3.16, 10 μm) (21-23km emission ht)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0



Figure 6-1 Percentage mass of ash over a one-week period following the eruption for 1, 3.16 and 10 µm ash sizes.

<u>100-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (0.10, 0.316, 1.0 μm)

(21-23km emission ht)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0



Figure 6-2 Percentage mass of ash in the first 3 months following the 1991 Mount Pinatubo eruption, for 3 different ash sizes (0.1, 0.316 and 1 μ m) for 0.05 and 0.5 Tg of ash injected. Straight lines denote 0.05 Tg, dashed denote 0.5 Tg ash.

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

-- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications (21-23, 18-21, 18-25km)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0



Figure 6-4 Monthly average ash (red) and sulfate (blue) burden for the tropics (20°S-20°N) for a 14 Tg SO₂ and 0.05 Tg ash injection mass at 18-20 km (solid lines), 21-23 km (dashed lines) and 18-25 km (dot-dashed line).

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

-- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications (21-23, 18-21, 18-25km)

Assess impacts of the co-emitted ash on Pinatubo sulphate aerosol

(Global SO₂ & SO₄ burden)



n.b. ash particles represented within coarse-insoluble mode (mode 7) with $\sigma = 2.0$

Figure 6-13 Global sulfur burden for (left) SO₂ gas-phase sulfur and (right) sulfate aerosol for 14 Tg injected SO₂ at all injection heights comparing with and without ash.

with $\sigma_g=2.0$ Only a small effect of sub-micron ash when consider the global sulphate burden

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

-- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications (21-23, 18-21, 18-25km)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0

Assess impacts of the co-emitted ash on Pinatubo sulphate aerosol



Figure 6-10 Tropical (Left) SO₂ burden and (Right) SO₄ burden for 14 Tg ash (solid line) and nonash (dashed) simulations.

Peak tropical SO4 burden reduces slightly (~10%) when sub-micron ash included.

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

-- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications (21-23, 18-21, 18-25km)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with $\sigma_g=2.0$



Substantial increase in transport to Southern Hemisphere when UF-ash co-emitted

Comparison to Mauna Loa lidar (19°N)

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

 -- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications
 (21-23, 18-21, 18-25km)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0



Figure 6-17 Extinction plots for MLO with 14 Tg SO₂ and 18-25 km injection height. Plots show observations (top), no ash (middle) and with ash (bottom). Black dots denote where layers are picked out from the layer finding algorithm.

Heating effect from ultra-fine ash <u>causes a modest change to initial progression</u> <u>of Pinatubo plume in tropical reservoir</u> → more concentrated to lower portion

Comparison to Hautes Provences lidar (44°N)

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

 -- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications
 (21-23, 18-21, 18-25km)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0



Figure 6-19 Extinction plots for OHP with 14 Tg SO₂ and 21-23 km injection height. Plots show observations (top), no ash (middle) and with ash (bottom).

Heating effect from ultra-fine ash <u>causes earlier transport to NH mid-latitudes</u> in better agreement with the timing observed from Hautes Provence

<u>180-day Pinatubo</u> <u>volcanic ash & SO₂</u> <u>experiments with</u> <u>sub-μm sized ash</u> (with 0.10μm g.s.d.)

-- <u>3 different injection</u> <u>height scenarios</u> matching ISA-MIP HErSEA experiment specifications (21-23, 18-21, 18-25km)

n.b. ash particles represented within coarse-insoluble mode (mode 7) with σ_g =2.0

Assess vertical distribution of Pinatubo ultra-fine ash in NH mid-latitudes (Aberystwyth lidar backscatter and depolarisation)



Figure 6-20 (Top) Contour plots of backscatter ratio and depolarisation data from Aberystwyth compared to ash ratio model data in 2 scenarios, (bottom) depth of cloud from the layer finding algorithm for observations and the same 2 model scenarios from the top row.

The altitude of the depolarization observed from the Aberystwyth lidar agrees well with the 21-23km injection run --> <u>UF-ash can explain the signal observed</u>

Conclusions

- GA4 "strat-enabled" UM-UKCA simulates aerosol microphysics across the stratosphere & troposphere (Dhomse et al., 2014; Mann et al., 2015)
- Benchmark GA4 "strat-UKCA v3" model (with GLOMAP v8.2) applied VolMIP Tambora-ISA expt (Marshall et al., 2018; Clyne et al., 2021), for volcanic forcing Perturbed Parameter Ensemble expt (Marshall et al., 2019) and to simulate the volc-aerosol clouds from Agung, El Chichon & Pinatubo (Dhomse et al., 2020).
- <u>Further developed for volcanic ash</u> within PhD studentship jointly funded by CAMS43 (IFS global aerosol development) & Leeds Univ (Shallcross et al., 2020): <u>http://homepages.see.leeds.ac.uk/~amtgwm/SarahShallcross_PhD_thesis_Corrected.pdf</u>
- Case study on Pinatubo to co-emit volcanic ash with SO₂ (see chapter 6)
- Ash particles larger than 1 micron sedimented from Pinatubo cloud in ~1 week But "ultra-fine ash" particles remained within the Pinatubo cloud for ~9 months
- Heating effect from UF-ash important, and model confirms hypothesis that observed depolarization of lidar backscatter at Aberystwyth caused by UF-ash.
- Next add SO₂ reactive uptake (het. ox.) removes ~40% for Kelud (Zhu et al. (2020).