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Pinatubo aerosol in  
a CCM

S. S. Dhomse et al.

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# Aerosol microphysics simulations of the Mt. Pinatubo eruption with the UKCA composition-climate model

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## Abstract

We have enhanced the capability of a microphysical aerosol-chemistry module to simulate the atmospheric aerosol and precursor gases for both tropospheric and stratospheric conditions. Using the Mount Pinatubo eruption (June 1991) as a test case, we evaluate simulated aerosol properties in a composition-climate model against a range of satellite and in-situ observations. Simulations are performed assuming an injection of 20 Tg SO<sub>2</sub> at 19–27 km in tropical latitudes, without any radiative feedback from the simulated aerosol. In both quiescent and volcanically perturbed conditions, simulated aerosol properties in the lower stratosphere show reasonable agreement with the observations. The model captures the observed timing of the maximum aerosol optical depth (AOD) and its decay timescale in both tropics and Northern Hemisphere (NH) mid-latitudes. There is also good qualitative agreement with the observations in terms of spatial and temporal variation of the aerosol effective radius ( $R_{\text{eff}}$ ), which peaks 6–8 months after the eruption. However, the model shows significant biases against some observational data sets. Simulated AOD and Surface Area Density (SAD) in the tropics are substantially higher than the gap-filled satellite data products during the first 6 months after the eruption. The model shows consistently weaker enhancement in  $R_{\text{eff}}$  compared to satellite and in-situ measurements.

Simulated aerosol particle size distribution is also compared to NH mid-latitude in-situ balloon sounding measurements of size-resolved number concentrations. Before the eruption, the model captures the observed profiles of lower stratospheric particle number concentrations with radii larger than 5, 150 and 250 nm ( $N_5$ ,  $N_{150}$  and  $N_{250}$ ) very well. However, in the first 6 months after the eruption, the model shows high bias in  $N_5$  concentrations in the lower stratosphere, suggesting too strong nucleation. Following particle growth via condensation and coagulation, this bias in the finest particles propagates into a factor 2 high bias in  $N_{150}$ . Our comparison suggests that new particle formation in the initial phase of large eruptions, and subsequent particle growth to

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optically-active sizes, might be playing an important role in determining the magnitude of the climate impacts from volcanoes like Pinatubo.

## 1 Introduction

Volcanic eruptions can have significant impact on atmospheric composition and climate (e.g. McCormick et al., 1995; Robock, 2000). Powerful explosive eruptions can inject large amounts of SO<sub>2</sub>, ash, water vapour and various other chemical species directly into the stratosphere. Volcanic SO<sub>2</sub> injected into the stratosphere is chemically converted to sulphuric acid vapour over a timescale of days to months, causing substantial new particle formation and aerosol growth by condensation. Such a volcanic enhancement of the stratospheric aerosol substantially increases particle concentrations at optically-active sizes (Deshler et al., 2003) which can persist for several years in the case of tropical eruptions. The optically thicker stratospheric aerosol alters the Earth's radiative balance with increased albedo via enhanced back-scattering of solar radiation, cooling the surface and increased absorption of terrestrial long-wave radiation, warming the stratosphere (Labitzke and McCormick, 1992). The efficiency of these direct radiative effects from the enhanced stratospheric aerosol is strongly influenced by its particle size distribution (Lacis et al., 1992; Hansen et al., 1992).

The long-wave radiative heating induced by the thickened aerosol layer modifies the stratospheric circulation (e.g. Young et al., 1994), leading to indirect radiative effects via dynamical changes in ozone and meridional transport, with important implications for surface climate (Robock and Mao, 1992; Graf et al., 1993). Volcanically increased aerosol surface area density (SAD) can also accelerate heterogeneous chemistry perturbing stratospheric NO<sub>y</sub> species, halogens and ozone (e.g. Solomon et al., 1996). Quantifying the net impact from these direct and indirect radiative effects is very important to better understand volcanic influences within the historical climate records.

There is an increasing recognition that having a good representation of stratospheric processes is important for climate projections (e.g. Scaife et al., 2012). However,

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whereas most coupled atmosphere-ocean climate models (e.g. Jones et al., 2011) that carried out historical integrations for CMIP5 (Taylor et al., 2012) included prognostic treatment of tropospheric aerosol, stratospheric aerosol has always been handled separately. Most of these models used prescribed heating rates from Stenchikov et al. (1998) that are based on time-varying aerosol optical depth from Sato et al. (1993). However, most of these models with prescribed aerosol (and heating rates) are not able to capture the dynamical response that have been associated with volcanic eruptions (Driscoll et al., 2012). An increasing number of chemistry-climate models (CCMs) are designed to simulate the evolution of stratospheric ozone (e.g. SPARC, 2010), but there are still few that include a prognostic treatment of the stratospheric aerosol layer. Many CCMs include prescribed aerosol surface area density. Some also use pre-calculated heating rates to account for stratospheric aerosol induced changes during period of volcanically enhanced aerosol loading. Nevertheless CCMs either overestimate or underestimate the El-Chichon (1982) and Mount Pinatubo (1991) eruption related ozone changes compared with the observations (SPARC, 2010, Fig. 8.20). So, there is great need to have consistent representation of stratospheric aerosols in CCMs.

In this paper, we use the composition-climate model UMUKCA (Unified Model – UK Chemistry and Aerosol) to simulate stratospheric aerosol in both quiescent and volcanically perturbed conditions. The model includes the GLOMAP-mode aerosol microphysics module, enabling the 3-D evolution of the particle size distribution to be quantified. We use the Mount Pinatubo eruption as a test case to evaluate simulated aerosol properties against a range of observational data from the period. Mount Pinatubo erupted in the Philippines (15.1° N, 120.4° E) on 15 June 1991 and was the largest tropical eruption since Krakatoa in 1883. Using daily SO<sub>2</sub> measurements from the TOMS satellite instrument, Bluth et al. (1992) estimated that the eruption injected about 20 Tg of SO<sub>2</sub> into the stratosphere that was chemically converted to sulphuric acid vapour on a timescale of around 35 days. Assuming a 50 % conversion of SO<sub>2</sub> to sulphuric acid by July and using an assumed size distribution and composition to convert the Stratospheric Aerosol and Gas Experiment II (SAGE-II) satellite measurements of aerosol

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extinction, McCormick and Veiga (1992) estimated that the total global aerosol loading was increased between 20 to 30 Tg. Baran and Foot (1994) used infrared satellite measurements from the High resolution Infrared Radiation Sounder (HIRS) instrument to derive a timeseries of the global stratospheric aerosol mass loading, finding a peak of 21 Tg in September 1991 with values in excess of 15 Tg persisting until November 1992 and much earlier and steeper decay in the tropics than Northern Hemispheric mid-latitudes. Monthly balloon soundings of total and size-resolved particle concentrations carried out at Laramie, Wyoming (e.g. Deshler, 1994) showed that although substantially enhanced particle concentrations were detected in the lower-most stratosphere by mid-July, the main part of the volcanic plume was only transported to Northern Hemisphere (NH) mid-latitudes several months later.

There have been many previous global modelling studies to simulate the evolution of stratospheric aerosol following the Pinatubo eruption. However most have use aerosol schemes that simulate only the evolution of aerosol mass, prescribing a fixed particle size distribution for sedimentation and radiative effects (e.g. Timmreck et al., 1999; Oman et al., 2006; Aquila et al., 2012; Arfeuille et al., 2013). However, size-resolved stratospheric aerosol modules which include microphysical processes such as new particle formation, coagulation and condensation have also been developed. The first Pinatubo aerosol microphysics simulations were carried out in two-dimensional models (Bekki et al., 1994, 1996; Weisenstein et al., 1997) with single-moment sectional schemes where mass in numerous size bins is transported. More recently several 3-D general circulation models with aerosol microphysics schemes have also been used, to predict sedimentation and changes in radiative forcing in conjunction with the evolving stratospheric particle size distribution (e.g. Timmreck, 2001; Toohey et al., 2011; English et al., 2012, 2013).

Despite the diversity in model complexities, most of these studies evaluated their simulations against a limited set of observational data sets, primarily AODs derived using Advanced Very High Resolution Radiometer (AVHRR) and Stratospheric Aerosol and Gas Experiment (SAGE II) measurements. Using a mass-based prognostic strato-

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spheric aerosol module in a middle-atmosphere version of the ECHAM4 climate model, Timmreck et al. (1999) showed that the two distinct maxima in AOD apparent in AVHRR and SAGE II based AOD could be simulated, but the model failed to simulate slow AOD decay a few months after the eruption. Similarly, using an offline 3-D chemical transport model, Pitari and Mancini (1999) could simulate SH AOD reasonably well but their model simulations were biased low in the tropical and NH AOD, a few months after the eruption. Aquila et al. (2012), used a mass-based aerosol scheme in a global aerosol transport model, but the model-simulated AOD was slightly lower than AVHRR immediately after the eruption, with better agreement for the later period. Using a sectional aerosol microphysics module, English et al. (2013) achieved good agreement with NH mid-latitude AOD against AVHRR immediately after the eruption but their modelled AOD decayed too rapidly in later months. They compared the aerosol effective radius ( $R_{\text{eff}}$ ) evolution against observations from SAGE II and in-situ measurements (for e.g. Russell et al., 1996; Bauman et al., 2003), finding peak values in the model NH tropical stratosphere occurred earlier than in the observations. Some of these model-observation biases in earlier studies may be linked with the transport related issues (e.g. the lack of a Quasi-Biennial Oscillation, QBO) in the underlying GCM, whereas some may be linked to the simplified treatment of the particle size distribution. Other causes such as interactions with ash, or missing minor eruptions such as Mount Hudson in Chile (September 1991) have also been suggested.

Although they have near-global spatial extent, satellite measurements of AOD and  $R_{\text{eff}}$  constrain only integrated stratospheric aerosol properties over the full particle size range. Balloon-borne measurements (e.g. Deshler, 1994) enable a closer examination of the particle size distribution, but are available at only a small number of sites.

Here, we use both satellite and balloon-borne measurements to evaluate the UMUKCA simulated stratospheric aerosol properties, and seek to better understand the source of model biases. In Sect. 2 we describe the model, including the experimental set-up and the developments to the aerosol and chemistry schemes which extend its applicability to both stratospheric and tropospheric conditions. Section 3 describes

the measurements that are used to evaluate the model. Results and discussion about potential causes of model-observation biases are presented in Sect. 4 and 5, respectively. Summary and major conclusions are presented in Sect. 6.

## 2 Model description

We use an updated version of the stratospheric version of the UMUKCA composition-climate model originally described in Morgenstern et al. (2009) and evaluated in SPARC (2010). It is a high-top low resolution version of the Hadley Centre Global Environmental Model (Hewitt et al., 2011) as used by Braesicke et al. (2013) and Telford et al. (2013). Although UKCA is designed to apply the radiative effects of the simulated aerosol,  $O_3$ ,  $CH_4$ ,  $N_2O$  and other gases to feedback on the model, in present simulations only the simulated  $O_3$  is radiatively coupled. We run UMUKCA in an atmosphere-only configuration whereby sea-surface temperature and sea-ice fields are prescribed from the AMIP time-varying dataset (Hurrell et al., 2008). The simulations are carried out at N48L60 resolution, which is  $2.5^\circ \times 3.75^\circ$  in latitude and longitude with 60 vertical hybrid-height levels from the surface to 84 km. Telford et al. (2009) used a previous version of the model to study effects of the Pinatubo eruption on the stratospheric ozone and found that modelled total ozone changes with specified dynamics showed reasonable agreement with the observations.

To simulate stratospheric aerosol precursor gas phase species, we have extended the existing UKCA stratospheric chemistry scheme to include sulphur chemistry (see Sect. 2.1). The coupling to the GLOMAP-mode aerosol microphysics module (Mann et al., 2010), and its adaptation for stratospheric conditions, is described in Sect. 2.2. Surface emissions of  $NO_x$ , CO and HCHO are from the RCP 4.5 scenario. The lower boundary conditions used to determine surface mixing ratios of  $CH_4$ ,  $N_2O$ , CFC-11 ( $CFCl_3$ ) and CFC-12 ( $CF_2Cl_2$ ) are from WMO (2011). Minor CFCs (other than CFC-11 and CFC-12), that together account for more than 50 % of atmospheric chlorine, are treated implicitly by a lumping approach.  $CH_3Br$  is prescribed at the surface, and

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its abundance is increased to account for halons and very short-lived species (VSLs) which are not treated in the chemistry scheme. This increases  $\text{CH}_3\text{Br}$  concentrations by about a factor of 2 (Morgenstern et al., 2010). Heterogeneous chemical reactions are driven by a prescribed climatology of aerosol surface area density produced for the SPARC Assessment of the Stratospheric Aerosols Report (SPARC, 2006). The calculated aerosols do not influence the model heating rates or ozone chemistry. For the added sulphur species, surface and higher altitude 2-D emissions sources of anthropogenic  $\text{SO}_2$  are included following the IPCC AR5 inventory for the year 2000 (Lamarque et al., 2010). A 3-D source from passively degassing volcanoes from Andres and Kasgnoc (1998) is also included. Marine surface emissions of DMS are determined by wind speed using a seawater concentration climatology of Kettle and Andreae (2000) with the sea–air exchange function of Liss and Merlivat (1986). For OCS, which has tropospheric lifetime of about two years (Montzka et al., 2007), we do not include an emissions source, but instead apply a fixed surface boundary condition of 550 pptm.

### 2.1 Stratospheric chemistry extended for a sulphur cycle

The existing UMUKCA stratospheric chemistry scheme (Morgenstern et al., 2009) covers the oxidation of  $\text{CH}_4$  and  $\text{CO}$ , with chlorine and bromine chemistry and their interaction with  $\text{HO}_x$ ,  $\text{NO}_x$  and  $\text{O}_x$  cycles including heterogeneous reactions on polar stratospheric clouds (PSCs) and liquid sulfate aerosols which is based on Chipperfield and Pyle (1998). Here, we have extended the scheme to also include a stratospheric aerosol precursor chemistry scheme based on that used by Weisenstein et al. (1997) with updates to reaction rates from Sander et al. (2006). The added chemistry includes the steady background source of  $\text{SO}_2$  from OCS, which principally maintains the stratospheric aerosol during volcanically quiescent periods (e.g. Carslaw and Kärcher, 2006). Also included are photolysis reactions for  $\text{H}_2\text{SO}_4$  and  $\text{SO}_3$ , which occur above about 30 km and lead to a reservoir of  $\text{SO}_2$  building up during polar winter, leading to new particle formation in polar spring (Mills et al., 2005). The chemistry is integrated with the ASAD chemical integration package (Carver et al., 1997) with the Newton

Raphson sparse-matrix solver from Wild et al. (2000). Photolysis rates are calculated using the FAST-JX online photolysis (Neu et al., 2007) following the implementation described in Telford et al. (2013). The cross section of  $\text{H}_2\text{SO}_4$  is assumed analogous to the cross section of HCl, at a fraction of 0.016, following the method of Bekki and Pyle (1992). Gas-phase production of  $\text{H}_2\text{SO}_4$  is passed to the GLOMAP aerosol scheme to update the gas phase  $\text{H}_2\text{SO}_4$  tracer on the short “competition timestep” used for nucleation and condensation (see Sect. 2.2). Aqueous sulphate production in (tropospheric) liquid clouds is also passed to the GLOMAP module for growth of accumulation and coarse soluble particles.

## 2.2 The aerosol microphysics module adapted for the stratosphere

The GLOMAP aerosol microphysics module was developed as a component of the TOMCAT 3-D offline Chemical Transport Model (Chipperfield, 2006) with both 2-moment sectional (Spracklen et al., 2005) and 2-moment modal versions (Mann et al., 2010) available. The faster modal scheme (GLOMAP-mode) was specifically designed for longer integrations within UMUKCA and applies similar aerosol microphysics representations as the sectional scheme but with the size distribution parameterised into 7 log-normal modes, being similar in framework to that used in ECHAM-HAM (e.g. Stier et al., 2005). The GLOMAP-mode scheme produces aerosol properties in good agreement with the more sophisticated sectional scheme under most conditions (Mann et al., 2012).

The seven modes used by GLOMAP-mode are a nucleation mode, and Aitken, accumulation and coarse mode aerosol in both soluble and insoluble species. The insoluble modes receive emissions of particles composed of a mixture of black carbon and organic carbon (Aitken-insoluble) or composed entirely of dust (accumulation and coarse insoluble). Since we are focussed on the evolution of the stratospheric aerosol layer after Pinatubo, for this work we use only the four soluble modes and treat only sulphate and sea salt components, the latter included to give reasonable representation of tropospheric aerosol optical properties. Thus far, the GLOMAP aerosol scheme has been

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used to simulate tropospheric aerosol. For this work, the scheme has been updated to apply different methods for the calculation of water uptake, particle density, vapour condensation and new particle formation, for all model levels above the upper troposphere (pressures below 150 hPa).

### 2.2.1 Water uptake

In the standard version of GLOMAP-mode described by Mann et al. (2010), the water uptake to the soluble modes is calculated using the ZSR approach (Zdanovskii, 1948; Stokes and Robinson, 1966) using water activities based on binary electrolyte solutions. In the stratosphere however, aqueous sulphuric acid solution droplets form even at very low relative humidity. We therefore apply the analytical expression of Carslaw et al. (1995) at pressures lower than 150 hPa to calculate water content based on the H<sub>2</sub>SO<sub>4</sub> mole fraction for the binary aqueous sulphuric acid solution droplets. At 225 K and 101 hPa, the composition of the solution is 74.5 % H<sub>2</sub>SO<sub>4</sub> and 25.5 % water, approximating the 75 % weight fraction assumed in some studies (e.g. Stenchikov et al., 1998; Oman et al., 2006).

### 2.2.2 Particle density

As composition of the aqueous sulphuric acid solution droplets also affects their density, we modified GLOMAP-mode for the stratosphere. For pressures lower than 150 hPa, density values for each mode are replaced with the values from a look-up table based on the measurements of Martin et al. (2000) as a function of the sulphuric acid weight-fraction.

### 2.2.3 Condensation and vapour pressure of H<sub>2</sub>SO<sub>4</sub>

In all previous versions of the GLOMAP aerosol module, gas phase H<sub>2</sub>SO<sub>4</sub> partitions into the particle phase assuming zero vapour pressure, i.e. the transfer is represented as a condensation process. Although this approach is entirely appropriate in tropo-

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spheric conditions, above  $\sim 25\text{--}30$  km, the vapour pressure of  $\text{H}_2\text{SO}_4$  can become significant as the temperatures are high enough that sulphuric acid droplets rapidly evaporate (Hamill et al., 1997; Hommel et al., 2011).

The  $\text{H}_2\text{SO}_4$  vapour pressure over a flat surface  $p$  is given by Eq. (1) (Ayers et al., 1980).

$$\ln(-p) = -(10\,156/T) + 16.259 + (\mu - \mu^0)/RT \quad (1)$$

Where  $p$  is in atmospheres,  $R$  is the universal gas constant,  $\mu$  is chemical potential of  $\text{H}_2\text{SO}_4$  in a solution at the given composition and  $\mu^0$  that of pure acid, and  $\mu - \mu^0$  is fitted from experimental data by Giauque et al. (1960). Eq. (1) calculated at 360 K ( $p_{360\text{K}}$ ) is then used to give the vapour pressure for the much lower stratospheric temperatures following Kulmala and Laaksonen (1990):

$$\ln(p_{\text{H}_2\text{SO}_4}) = \ln(p_{360\text{K}}) + \frac{\Delta H_v(360\text{K})}{R} \left[ -\frac{1}{T} + \frac{1}{360} + \frac{0.38}{905 - 360} \cdot \left( 1 + \ln \frac{360}{T} - \frac{905}{T} \right) \right] \quad (2)$$

The reference point 360 K is the base temperature at which Ayers et al. (1980) measured the saturation vapour pressure, 905 K is the critical temperature, and  $\Delta H_v(360\text{K})/R$  is set to 10 156 as in Kulmala and Laaksonen (1990).

In this study we do not allow particle evaporation, but modify the existing GLOMAP approach (e.g. Mann et al., 2010) so that condensation of  $\text{H}_2\text{SO}_4$  proceeds according to the vapour pressure deficit (neglecting the Kelvin effect). This ensures that gas phase  $\text{H}_2\text{SO}_4$  produced in the upper stratosphere only condenses to the aerosol when the vapour is supersaturated.

## 2.2.4 New particle formation

Previous versions of GLOMAP (e.g. Mann et al., 2010) allowed formation of new  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  particles based on the Kulmala et al. (1998) parameterization for binary homogeneous nucleation. This is only applicable at temperatures in the range

233–298 K. Vehkamäki et al. (2002) suggested that conditions for nucleation are also favourable at  $\sim 200$  K in the upper tropical troposphere and they updated the Kulmala et al. (1998) parameterization to be applicable down to lower temperatures and humidities. To allow GLOMAP-mode to be applied in both tropospheric and stratospheric conditions, we have incorporated the Vehkamäki et al. (2002) parameterisation, and used it within the recommended ranges of temperature (190 to 305 K) and  $\text{H}_2\text{SO}_4$  concentration ( $10^4$  to  $10^{11}$   $\text{cm}^{-3}$ ). Note that we also use the expression of Kerminen and Kulmala (2002) to convert the “real” nucleation rate from Vehkamäki et al. (2002) into an “apparent nucleation rate” at 3 nm.

### 2.2.5 Size Distribution

Balloon-borne optical particle counter and condensation nucleus counter measurements in the mid-latitude stratosphere in the 1990s (e.g. Deshler et al., 2003) suggest a bimodal size distribution with the first  $\sigma_g$  mode at about 50–150 nm radius with  $\sigma_g$  between 1.6 and 1.8 and a larger much narrower mode ( $\sigma_g \sim 1.2$ ) at around 300–800 nm radius that is weak in volcanically quiescent conditions but much stronger (in number) following the Pinatubo eruption (e.g. Carslaw and Kärcher, 2006). After such large volcanic eruptions, a significant proportion of stratospheric sulphuric acid aerosol droplets grow to super-micron sizes. For example Deshler et al. (2003) show that in March 1993 (21 months after the Pinatubo eruption), in the NH mid-latitude lower stratosphere there was a 6 km layer (12–18 km) with the number concentration of particles with radii larger than 500 nm greater than  $1 \text{ cm}^{-3}$ . Such coarse particles have grown from their original size of around 1 nm due to coagulation and gas-to-particle transfer of sulphuric acid. Modal microphysics schemes such as GLOMAP-mode represent this condensational and coagulative growth, but must use a technique referred to as “mode-merging” (e.g. Binkowski and Roselle, 2003) to transfer particles to adjacent larger modes following strong growth. In this case of a large volcanic eruption, the mode-merging must transfer particles first from the nucleation mode to the soluble Aitken mode, and following further growth up to the soluble accumulation mode and then to the soluble coarse

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mode. In each case, when particles are received from the adjacent smaller mode, the transferred number and mass is added to that existing in the mode, with the mean size re-formulated according to the prescribed standard deviation of the mode ( $\sigma_g$ ).

Kokkola et al. (2009) compared size distributions simulated by a modal and three sectional schemes in a box model. While the four models agreed well in background stratospheric conditions, in volcanically perturbed conditions, the size distributions were found to be better represented with narrower mode widths. In particular, with the original coarse mode  $\sigma_g$  of 2.0, they found the modal scheme over-predicted the  $R_{\text{eff}}$  compared to a reference sectional scheme with a large number of bins. Niemeier et al. (2009) used an improved version of the same modal microphysics scheme whereby  $\sigma_g$  for the accumulation soluble mode was reduced to 1.2 and the coarse mode was de-activated.

Here we are applying the modal GLOMAP scheme to volcanically-perturbed stratospheric conditions, and also using the same modes to also represent tropospheric aerosol. In the troposphere, the coarse soluble mode in GLOMAP-mode is almost exclusively containing sea-salt, and the scheme has followed Wilson et al. (2001) and Vignati et al. (2004) in using a value of 2.0 for  $\sigma_g$  in this mode, which are based on values given in D'Almeida et al. (1991). However, Heintzenberg et al. (2004) present size distribution observations over several fields campaigns in the marine boundary layer which find a median value of coarse mode  $\sigma_g$  at 1.4.

To best represent coarse particles in both tropospheric and stratospheric conditions, we reduce  $\sigma_g$  for the coarse soluble mode to be 1.4 rather than 2.0. We retain the  $\sigma_g$  value of 1.4 for the soluble accumulation mode in GLOMAP-mode, as reduced by Mann et al. (2012) from the value of 1.59 used in Mann et al. (2010) to better compare with size distributions simulated by the sectional scheme and with observations. We retain the  $\sigma_g = 1.59$  values for the nucleation and Aitken modes. The narrowed coarse soluble mode  $\sigma_g$  of 1.4 matches that for the accumulation soluble mode, which is desirable numerically when the mode-merging algorithm transfers particles between the two modes in strong particle growth conditions.

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## 2.3 Experimental setup

To examine the perturbation to the stratospheric aerosol layer that occurred after the Pinatubo eruption, we performed two simulations with (run Pinatubo) and without (run no-Pinatubo) related SO<sub>2</sub> injection. For run Pinatubo, the model is configured to inject 20 Tg of SO<sub>2</sub> into the tropical stratosphere on 15th June 1991 between 19 and 27 km. Satellite observations of SO<sub>2</sub> after the eruption (Bluth et al., 1992) show that the plume initially dispersed southwards crossing the equator within two weeks. As described in Sect. 2, the simulated aerosol is not radiatively coupled to the atmospheric dynamics, which is believed to be an important limitation in simulating the initial dispersion of the Pinatubo plume correctly (Young et al., 1994). Consequently, to ensure we closely match the initial spatial distribution of the aerosol cloud, as in Timmreck et al. (1999) we apply the SO<sub>2</sub> injection spread across eight model grid boxes between 0–20° N 120.5° E. The aerosol and gas phase sulphur species had 8 yr spin-up from an initialisation with zero aerosol and gas phase species, with other gases from the UMUKCA REF-C1 integration from the SPARC Lifetimes Assessment Report (SPARC, 2013), representative of 1990 conditions.

A key mode of inter-annual variability in the tropical lower stratosphere is the observed QBO of zonal winds between easterly and westerly phases, which occurs with a period of 22 to 34 months with an average of 28 months (Andrews et al., 1987; Baldwin et al., 2001). Using satellite observations, Trepte and Hitchman (1992) showed the important role the QBO phase played in determining the initial dispersion of the Pinatubo plume.

For this experiment, we picked the initialisation year such that lower stratospheric winds in the model were in the easterly phase of the QBO, as at the time of the actual eruption. Figure 1a shows the time evolution of the model monthly and zonal-mean zonal wind in the tropics (15°S–15°N) against those from the ERA interim reanalysis from 1990 until 1995 (Fig. 1b Dee et al., 2011). At the time of the eruption (15 June 1991), the model has an easterly QBO phase with easterly winds stronger

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than  $10 \text{ ms}^{-1}$  above 33 hPa, agreeing well with the ERA-interim tropical zonal wind profile at that time. This model easterly QBO phase continues until around April 1992 (at 30 hPa), the shift to westerly flow occurring slightly earlier than in ERA-interim, which continues until around September 1992. The semi-annual oscillation in the tropical middle and upper stratosphere is also well represented in the model.

How rapidly the Pinatubo-enhanced stratospheric aerosol layer returns to pre-eruption levels is strongly influenced by the strength of the model stratospheric circulation. Sedimentation of particles is also important as it moves particles closer to the tropopause where they can be removed by stratospheric-tropospheric exchange (e.g. Deshler, 2008). A common metric used to assess stratospheric transport in chemistry-climate models (e.g. Strahan et al., 2011) is the mean stratospheric age of air.

Figure 1c shows the latitude and altitude distribution of the model zonal-mean age of air (for 1991–2000) and Fig. 1d compares the model age of air at 50 hPa against that derived from aircraft observations of the long-lived tropospheric source gases  $\text{SF}_6$  and  $\text{CO}_2$  (Hall et al., 1999). The values from other composition-climate models participating in the recent CCMVal lifetime assessment (Chipperfield et al., 2013) are also shown for reference. In the tropics, the model age of air agrees well with the observations, but at mid- and high latitudes there is a low bias compared to the observations; up to 1 yr too young air at high latitudes. The low bias in mid-latitude age-of-air indicates that model may have too rapid meridional poleward transport as well as stronger STE. In turn, such a mixing can cause too fast removal of aerosol from the stratosphere into the mid-high latitude troposphere, and should be considered when drawing inference from the evaluation of the model post-Pinatubo stratospheric aerosol decay.

### 3 Measurements

To evaluate the UKCA simulations, we use measurements from the SAGE II instrument (McCormick and Veiga, 1992), which was launched on the Earth Radiation Budget Satellite (ERBS) in 1984. SAGE II was a seven-channel Sun photometer oper-

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ated in solar occultation mode with a vertical resolution of about 0.5 km. Spectral windows were centred at 385, 448, 453, 525, 600, 940 and 1020 nm. For evaluating the model stratospheric aerosol optical depth, we use the gap-filled SAGE-II (V6.2) product (Hamill et al., 2006) produced for ASAP (Assessment of Stratospheric Aerosol Properties SPARC, 2006). Simulated aerosol extinction is compared against the recently updated version (V7.0) of the SAGE-II data (Damadeo et al., 2013). We also compare to the SAGE-derived SAD product (Thomason et al., 1997) that is obtained from <http://www.sparc-climate.org/data-center/data-access/asap/>. Simulated SAD is also compared against the recently available SAD data Arfeuille et al. (2013) which was created using SAGE II V7.0 data, and is provided for the Chemistry Climate Model Initiative (CCMI) simulations. Further evaluation of the post-Pinatubo simulated aerosol optical depth (AOD) evolution was carried out by comparing to that measured by the Advanced Very High Resolution Radiometer (AVHRR/2), which was onboard on the National Oceanic and Atmospheric Administration (NOAA/11) satellite. For details see [http://www.nsof.class.noaa.gov/release/data\\_available/avhrr/index.htm](http://www.nsof.class.noaa.gov/release/data_available/avhrr/index.htm). The AVHRR instrument measures the reflectance of the Earth in five spectral bands centred around 0.6, 0.9, 3.5, 11 and 12  $\mu\text{m}$ .

To examine the simulated particle size distribution that underpins the model aerosol optical properties, we also compare to profile measurements of size-resolved number concentration made at Laramie, USA (Deshler et al., 2003). The balloon-borne system includes a Condensation Nucleus Counter (CNC) to measure the concentrations of particles larger than 10 nm and an optical particle counter (OPC Deshler et al., 1992) to measure size-resolved particle concentrations in several size ranges in the accumulation and coarse regions of the size spectrum. The OPC is a light counter to derive integrated size distribution from measured aerosol scattering in the forward direction. The standard OPC design gives integral number concentrations larger than 150 nm and 250 nm radius, and has been used in balloon sounding measurements of the stratospheric aerosol since 1963 (Rosen, 1964), also giving important information about the stratospheric aerosol changes induced by the 1980 Mount St. Helen's

erupton (Hofmann and Rosen, 1982). Deshler et al. (1992) present the measurements taken in the first few months after the June 1991 Pinatubo eruption, with most balloon flights using this original 2-channel OPC. An enhanced OPC, using an increased scattering angle, measured concentrations in 8 size channels for radii larger than 150 nm to around 10 microns. The 8-channel OPC had been developed shortly before the eruption, and became the default measuring system a few months after the eruption. The measurement capabilities were later further enhanced to measure up to twelve size ranges (see Deshler et al., 2003).

## 4 Results

Figure 2 shows the January 1991 to December 1994 time-evolution of the total global column mass burden of sulphur in the gas phase (as  $\text{SO}_2$ , red) and in the aerosol particle phase (blue) from our Pinatubo and no-Pinatubo simulations. Separate lines indicating the UTS aerosol sulphur burden (Upper Tropospheric and Stratosphere) and that in the mid- and lower stratosphere are also shown. Without Pinatubo (dotted lines), the global  $\text{SO}_2$  and aerosol sulphur burden timeseries are dominated by anthropogenic emissions which are mostly emitted in NH mid-latitudes. Oxidant concentrations are highest during the summer due to stronger photochemistry, which leads to efficient conversion of  $\text{SO}_2$  to aerosol sulphate. During the NH summer, only 10 % of the total sulphur burden comes from  $\text{SO}_2$ , compared to around 50 % during winter. In our model, 30–40 % of the total aerosol sulphur burden (around 0.5 Tg S) is in the stratosphere, which is considerably higher than the 17 % (15 Tg S) found by Hommel et al. (2011). Tropospheric aerosol burdens are also higher than other models Textor et al. (e.g. 2006) at around 1.25 Tg S on the annual mean. For the Pinatubo simulation, the global  $\text{SO}_2$  burden peaks at 5.75 Tg for the July 1991 monthly-mean. However, note that the June monthly-mean  $\text{SO}_2$  burden of 4.75 Tg included 15 days of simulation before the eruption (which had zero stratospheric  $\text{SO}_2$ ), and can thus be approximated as around 9 Tg of sulphur.

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The July mean  $\text{SO}_2$  burden can be considered as representative of the centre of the month, i.e. about 30 days after the eruption. Subtracting the  $\text{SO}_2$  mass from the no-Pinatubo run (which is all in the troposphere), gives 5.4 Tg S, suggesting 4.6 of the emitted 10 Tg S emitted as  $\text{SO}_2$  has been chemically converted to sulphuric acid over the 30 days. We therefore estimate the e-folding timescale for conversion of  $\text{SO}_2$  into sulphuric acid aerosol as 30 divided by  $\ln(10/5.4)$  which is 49 days. Bluth et al. (1992) derived an e-folding timescale of 35 days from the TOMS satellite  $\text{SO}_2$  measurements, but present this as a tentative estimate. McCormick and Veiga (1992) derived an approximate aerosol sulphur burden assuming a 50 % conversion from  $\text{SO}_2$  to  $\text{H}_2\text{SO}_4$  by the end of July, which corresponds to an e-folding timescale of 43 days. Oman et al. (2006) found an  $\text{SO}_2$  e-folding conversion timescale of 35 days in their model, which used fixed OH concentrations. The Bekki (1995) timescale is 40 days, based on 2-D model simulations with stratospheric sulphur and oxidant chemistry, thus accounting for the depletion of oxidants, as in our study.

The peak in global aerosol sulphur burden occurs 3 months after the eruption in September, in agreement with Baran and Foot (1994). We find a maximum total aerosol burden of 10.2 Tg of sulphur, with 9 Tg of sulphur in the stratosphere, or 36 Tg aerosol mass assuming 75 % sulphuric acid composition, slightly higher than the 21.5 Tg (5.4 Tg of sulphur) estimated by Baran and Foot (1994). The model global aerosol burden reduced to 3.5 Tg of sulphur by June 1992 (12 months after the eruption) suggesting an e-folding time for the model aerosol mass of 15.8 months; slightly longer than the range of 12–14 months estimated by Baran and Foot (1994) and Bluth et al. (1997).

Figure 3 shows vertical profiles of mixing ratios of the three key gas phase sulphur species OCS,  $\text{SO}_2$ ,  $\text{H}_2\text{SO}_4$ , and of sulphuric acid in the particle phase (Aer\_ $\text{H}_2\text{SO}_4$ ). The left and right panels are for mean profiles in the tropics ( $15^\circ\text{S}$ – $15^\circ\text{N}$ ) and mid-latitudes ( $35$ – $60^\circ\text{N}$ ), respectively with the top and bottom rows indicating the means for July 1991 and October 1991, selected as they show the initial period when the  $\text{SO}_2$  is oxidised to  $\text{H}_2\text{SO}_4$  vapour, and the period when the peak global aerosol burden oc-

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curs in the model. The profile of OCS shows the expected shape, being constant in the troposphere and then reducing with increasing altitude in the stratosphere as it is photolysed. In the run without the Pinatubo eruption (dashed line) the  $\text{SO}_2$  mixing ratio reduces sharply with height across the tropopause but then reaches a minimum and begins to increase with height to a local maximum at 30 km corresponding to where the source from OCS photolysis is largest. Below 30 km the sulphuric acid vapour follows a similar shape as  $\text{SO}_2$  (but at lower concentrations) but above that altitude continues to increase as its vapour pressure becomes significant. The vertical profile of the aerosol sulphuric acid concentration is approximately constant in the tropics in these quiescent conditions, but has a slight decrease with altitude in the NH mid-latitudes due to more effective meridional transport at lower altitudes within the stratosphere. Note that, since in these runs we do not include evaporation of sulphuric acid, the profile of aerosol sulphuric acid above 35 km is too high. In reality the rapidly evaporating particles will release their  $\text{H}_2\text{SO}_4$  to the gas phase causing a sharp reduction around 40 km. As well as biasing gas phase sulphur species, our neglect of particle evaporation will overestimate surface area density at the top of the aerosol layer.

When we include the Pinatubo eruption, in the tropics, the July profiles of  $\text{SO}_2$  and aerosol  $\text{H}_2\text{SO}_4$  are very strongly enhanced with concentrations between 20 and 30 km higher by factors  $10^3$ – $10^4$  and factor-100 respectively. The enhanced aerosol- $\text{H}_2\text{SO}_4$  profile indicates that much of the  $\text{SO}_2$  has already been oxidised and condensed into the particle phase. By contrast, the NH mid-latitude July profiles shows that the Pinatubo plume has not yet been transported with  $\text{SO}_2$  and aerosol  $\text{H}_2\text{SO}_4$  still at quiescent concentrations over almost the entire stratosphere, although a small perturbation can be observed in the lowermost stratosphere and uppermost troposphere. It is notable that balloon-borne particle concentration soundings at Laramie ( $41^\circ\text{N}$ ) in July 1991 already show some enhanced layers between 15–18 km (Deshler et al., 1992) which corresponds well with the altitude of the  $\text{SO}_2$  and  $\text{P-H}_2\text{SO}_4$  enhancement seen in the July-mean NH mid-latitude profiles.

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The October mean SO<sub>2</sub> profile is still strongly enhanced (factor 100) in the tropics with the P-H<sub>2</sub>SO<sub>4</sub> enhancement only slightly higher than in July but over a much deeper layer. This tropical enhancement in both SO<sub>2</sub> and Aer-H<sub>2</sub>SO<sub>4</sub> propagates up to above 40 km, although the aerosol enhancement is unrealistic as it will in reality have evaporated at those altitudes. It is interesting that the October 1991 tropical gas phase H<sub>2</sub>SO<sub>4</sub> profile actually shows lower values than in quiescent conditions in the main part of the plume (15–30 km), due to the condensation sink to aerosol being so much stronger. By contrast above 30 km the increase in vapour pressure shuts off the condensation sink leading to the H<sub>2</sub>SO<sub>4</sub> vapour concentrations being higher than quiescent at those altitudes. The October 1991 NH mid-latitude SO<sub>2</sub> and Aer-H<sub>2</sub>SO<sub>4</sub> profiles show only moderate enhancement suggesting the easterly phase of the QBO has prevented transport of Pinatubo-enhanced air masses.

Figure 4a shows the time evolution of the model mid-visible stratospheric AOD, with Fig. 4b and c showing the mid-visible AOD measured from AVHRR and SAGE II. Since AVHRR is a nadir viewing instrument, the derived AODs are from the full column aerosol (covering both troposphere and stratosphere). Hence, in Fig. 4b we have subtracted monthly-mean AODs for the year prior to the eruption (as in Aquila et al., 2012). Note that the SAGE-II derived AOD is lower than AVHRR, due to the measured extinction saturating during the very high loading period after Pinatubo (Hamill et al., 2006). Overall, there is good qualitative agreement between the model and satellite AOD distributions. For example, there is high AOD after the eruption centred around the equator with peak AOD in September 1991 in both the model and the two satellite datasets. However, the model feature is narrower, confined between 10° N and 10° S. Another well-captured feature in the model is that there is no significant enhancement of AOD in NH mid-high latitudes until October 1991. The model does not capture the observed AOD enhancements in the Southern Hemisphere (SH). Extinction profile measurements from SAGE-II between July and September show (e.g. McCormick et al., 1995) that transport to the SH occurred above about 24 km. Aquila et al. (2012) highlighted the importance of resolving the enhanced tropical upwelling which occurred due to the

long-wave absorption by the relatively larger stratospheric aerosol after the Pinatubo eruption. As explained in Sect. 2, in these simulations we do not radiatively couple the simulated aerosol with the model dynamics, which likely explains this discrepancy. Another reason could be the eruption of Cerro Hudson in Chile (46° S, 73° W), which occurred from 12–15 August 1991, and is not included in our simulations.

Figure 5 shows model and SAGE-II aerosol extinctions at 32, 25 and 20 km in the tropics (20° S–20° N). We choose these altitudes to allow comparison with the evaluation presented in Weisenstein et al. (2006, Fig. 6.20) for other stratospheric aerosol models. Here we compare extinction in the mid-visible (panels a to c) as well as the near infrared (panels d to f). We use the updated v7.0 SAGE-II dataset and the profiles shown are averages between 20° S and 20° N. Monthly mean observed values are calculated based on both sunrise and sunset profiles.

At 20 and 25 km, the model captures the general evolution of the tropical mid-visible extinction, with the magnitude and timing of peak values, and the decay timescale, agreeing well with SAGE-II. However, before the eruption, modelled extinctions have a moderate low bias of 30–50 % at these levels. Also, at 25 km, the model extinction reduces sharply after its peak, whereas in the satellite measurements, values plateau for 2–3 months before the decay period begins. At 32 km, simulated extinctions are consistently higher than SAGE, although there are large measurement error bars. The bias may indicate that the model upper injection altitude was too high or be related to the lack of aerosol-induced radiative heating as well as lack of aerosol evaporation in our simulations. As noted above, the SAGE-II instrument saturated in the tropics during this high aerosol loading period and we do not have any observed data points to constrain the simulated mid-visible extinction at 20 km.

At near-infrared wavelengths, the model also agrees fairly well with the observations, although differences are larger than at mid-visible wavelengths. For example, at 20 km, the period of increasing extinction is much longer lasting in the observations than in the model. At 25 km, the model agrees better with the observations, but extinctions are slightly lower than measured by SAGE-II. It is also noticeable that the SAGE-II extinc-

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tion stays approximately constant at close to the known saturation value of  $0.01 \text{ km}^{-1}$  for several months at both altitudes.

Figure 6 shows a similar analysis to Fig. 5, but for NH mid-latitudes ( $35\text{--}60^\circ \text{ N}$ ), again to compare against models shown in Weisenstein et al. (2006). At 20 km, there is good agreement between modelled and SAGE II extinctions, although the model appears slightly higher in the first 6 months after the eruption. At 25 km however, the model is consistently higher than the observations, in common with the models participating in the SPARC (2006). A noticeable seasonal cycle is seen in the SAGE-II extinction at 25 km, which is not captured by the model. This might be due to the absence of aerosol evaporation in our simulations. At 32 km the model has a similar evolution to that simulated at 20 and 25 km, whereas the observations indicate very little volcanic enhancement occurred at this level.

Figure 7 compares the vertical and latitudinal distribution of zonal mean SAD against two versions of the satellite derived SAD dataset, for four selected months between May 1991 and May 1992. Before the eruption (May 1991), the model captures the observed SAD very well with a hemispherically symmetric distribution in the lower stratosphere in the range  $0.5$  to  $2 \text{ m}^2 \text{ cm}^{-3}$ . For September 1991 (three months after the eruption), although the simulated SAD distribution broadly matches the observed shape, it is up to a factor 5 to 6 too high in the tropics. Also, the model Pinatubo-enhanced SAD plume is too strongly confined to the tropical pipe, whereas in the satellite-derived SAD (Fig. 7d) one can see weak meridional transport to NH and SH sub-tropics at about 20–22 km. Young et al. (1994) show that including the aerosol radiative effects on the model dynamics broadens the latitudinal extent of the Pinatubo cloud, in better agreement with the satellite observations, and this is likely the source of the bias seen here. By January 1992, the model high-bias has reduced to a factor 3–4, and the model shows meridional transport to NH mid-latitudes in the lowermost stratosphere, also seen in the observations. However, the satellite derived SAD suggest meridional transport also occurs to the SH, but at slightly higher altitudes. By May 1992, the model SAD high bias is less than a factor of 2 and the general latitudinal and altitudinal distribution

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is still in good qualitative agreement with the observations, aside from the continued low bias in the SH.

When interpreting the model-observation SAD discrepancies, one should consider how the satellite SAD product is derived from the SAGE I, SAGE II, SAM II (Stratospheric Aerosol Instrument II) and SME (Solar Mesosphere Explorer) measurements. As noted earlier, the extinction measured by the SAGE and SAM instruments has an upper limit of  $0.02 \text{ km}^{-1}$ , above which the atmosphere is effectively opaque to the instruments Hamill et al. (2006). During the peak aerosol loading period, when the model SAD is a factor of 5–6 high biased, it is apparent (for example in Fig. 5) that the SAGE II 525 nm and 1020 nm extinctions in the tropical lower stratosphere are saturating at the upper limit value, with actual extinction values likely to have been higher. The late-1991 to 1992 period was flagged as missing data in the original SAGE-II extinction dataset. The data gaps during that period were addressed by Hamill et al. (2006), who used lidar data from two tropical sites (Camaguey, Cuba and Mauna Loa, Hawaii) and two mid-latitude sites (Virginia, USA and Lauder, New Zealand), to fill the missing data.

Another important issue to consider with the SAGE-II derived SAD product is that, even outside the gap-filled part of the dataset, particles smaller than 50 nm are essentially invisible to the satellite and there is little sensitivity to particles smaller than 100 nm. Reeves et al. (2008) derived extinction, SAD and volume concentration from aircraft measurements of the aerosol particle size distribution (in quiescent conditions) and compared to SAGE-II products. They found the aircraft measured SAD was a factor 1.5–3 higher than the SAGE-II derived values, whereas volume concentrations were only 35 % higher. Another product derived from the gap-filled satellite extinction record, that can be used to assess the evolution of the stratospheric aerosol properties following the Pinatubo eruption, is the  $R_{\text{eff}}$ , defined as the ratio of the 3rd and 2nd integral moments in radius and for multimodal distribution can be represented as (Russell et al.,

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1996, Eq. 6):

$$R_{\text{eff}} = \frac{\sum_{i=1}^m N_i r_{gi}^3 \exp \left[ 9/2 (\ln \sigma_i)^2 \right]}{\sum_{i=1}^m N_i r_{gi}^2 \exp \left[ 2 (\ln \sigma_i)^2 \right]} \quad (3)$$

The two gap-filled SAGE/SAM extinction data products thereby provide 3-D time-varying volume concentration and SAD which together give  $R_{\text{eff}}$  throughout the Pinatubo period. This record therefore has the potential to give unprecedented information on how the particle size distribution in the stratosphere was perturbed by the eruption. However, again, when comparing the model to the satellite  $R_{\text{eff}}$ , the limitations associated with the derived product need to be considered. In particular, because of the “blind spot” associated with particles smaller than 50–100 nm, (Hamill et al., 2006) state that since the derived SAD may have an inherent low bias (whereas the derived volume density will be less affected) the derived  $R_{\text{eff}}$  may overestimate the true value.

Figure 8 shows the evolution of the model zonal-mean  $R_{\text{eff}}$  at 20 and 25 km compared to that derived by Bauman et al. (2003) from the SAGE-II and CLAES satellite measurements. The general spatial and temporal evolution of the model  $R_{\text{eff}}$  is in good qualitative agreement with the observations, with values at 20 km larger than at 25 km, likely due to sedimentation. At both altitudes the observations show a gradual decay from peak effective values in around October to December at 20 km and February to March 1992 at 25 km. In the model, the timing of the peak  $R_{\text{eff}}$  is well captured, but at 25 km, the model peaks later than observed, matching the timing at 20 km. Effective radius values are always higher in the tropics than at mid-latitudes, a feature that is consistent between the model and observations. However, the model values are substantially lower than those derived from the satellite, with maximum model  $R_{\text{eff}}$  of around 0.4  $\mu\text{m}$ , compared to around 0.6  $\mu\text{m}$  from the satellites.

At 20 km (Fig. 8b), despite combining the two sets of satellite products, there is no observational constraint on the tropical  $R_{\text{eff}}$  between approximately June 1991 and August 1992, but the overall shape suggests the  $R_{\text{eff}}$  was likely even larger than 0.6  $\mu\text{m}$

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during that period. The model low bias in  $R_{\text{eff}}$  is apparent at about the same extent at all latitudes and altitudes and before the eruption, which suggests it is not associated with sedimentation, since that would be expected to occur mostly during the highest loading period. There appears to be a more persistent bias in simulated particle size distribution, but it is unclear whether the model has too many small particles, or too few large particles.

To give a stronger observational constraint on the simulated size distribution, we compare the model against balloon-borne CNC and OPC measurements made at Laramie, Wyoming, USA (41° N), see Sect. 3. Figures 9 and 10 compare model profiles of size-resolved number concentrations (larger than a given particle diameter) against those measured by the CNC and OPC. In each case we are comparing a monthly-mean size-resolved particle concentration to a single balloon sounding. Note that whereas the number concentration profiles for particles larger than 5 nm, 150 nm and 250 nm are exactly as measured by the OPC, the larger size channels we have interpolated the observations (linearly in  $\log N$  vs.  $\log R$  space) onto regular  $D_p > 550$  nm, 750 nm and 1000 nm size channels from the irregular size thresholds given in the individual sounding data files.

Figure 9 presents the comparison through August to November 1991, in the period after the Pinatubo plume was first detected as Laramie on 16 July (Deshler et al., 1992). In August and September 1991, the model predicts that the eruption enhanced  $N_5$ ,  $N_{150}$  and  $N_{250}$  between 14 and 20 km compared to the no-Pinatubo run, whereas at higher altitudes the profile remains unperturbed. The region with elevated  $N_5$ ,  $N_{150}$  and  $N_{250}$  profiles matches well with the observations, and indicates efficient transport of air from the tropics in the lowermost stratosphere. Intriguingly, the model predicts that the eruption reduced concentrations of particles at sizes larger than 750 nm. One possible explanation could be that there is reduced growth of background nuclei to these coarse sizes when  $N_{150}$  and  $N_{250}$  are so high. However, observations from balloon soundings in the post-Pinatubo quiescent period (Deshler et al., 2003) show much lower concentrations of these coarse particles than in our model. Whereas the observations show

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approximately constant  $N_5$  between 20 and 30 km, the model shows a steady decline. The observations show that concentrations of particles at 150 nm and larger reduce sharply above 32 km, whereas the model profiles show only moderate decline, which is almost certainly due to the particle evaporation not being resolved in the model.

The model predicts that, by October 1991, the region of enhanced particle concentrations larger than 150 nm and larger than 250 nm have deepened to around 24 km and 30 km respectively, whereas  $N_5$  shows enhancement throughout the lower and middle stratosphere. The slight reduction in coarse particle concentrations predicted by the model is still apparent for this month, up to about 28 km. In the lower stratosphere, these perturbed profiles continue to be in good agreement with the observations for most of the size thresholds.

For November 1991, the model shows enhancement for all the particle size thresholds, with the coarse mode higher than the no-Pinatubo run in the lowermost stratosphere. The depth of the enhanced particle concentrations larger than 150 nm has now propagated up to 35 km, suggesting transport of the Pinatubo plume to mid-latitudes throughout the lower and middle stratosphere. There remains a region of suppressed coarse mode particle concentrations, but this is shifted to higher altitudes. The observed  $N_5$  and  $N_{150}$  lines are close together between 19 and 24 km suggesting few particles remain smaller than 150 nm in that part of the stratosphere. By contrast the model size distribution still has high concentrations of these ultrafine particles, perhaps indicating that nucleation rates persists in the model when it should be suppressed by the large surface area, or the particles do not grow fast enough by  $H_2SO_4$  condensation and coagulation. The high bias seen in lower stratospheric  $N_5$  in October has worsened and deepened in November and has propagated through to the  $N_{150}$  profile. By contrast the  $N_{250}$  is in good agreement with the observations, with reasonable agreement also at larger sizes. Again this suggests that the model is generating too many particles at sub-10 nm sizes and their growth is leading to a high bias in  $N_{150}$ . Potential reasons for these biases are discussed in the latter part of this section.

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In order to evaluate the model size distribution profile in quiescent conditions, we compare to the Laramie balloon measurements in March 1991 (Fig. 10a). We then probe the longer-timescale evolution of the size distribution through the Pinatubo period, in March 1992, 1993 and 1994 (Fig. 10b, c and d). Before the eruption, the observations show that  $N_5$  decreases with increasing altitude between 12 and 23 km, whereas  $N_{150}$  and  $N_{250}$  show very little decrease with height. This feature is well captured by the model with  $N_5$  and  $N_{150}$  in excellent agreement with the observations in this altitude range, although  $N_{250}$  has a slight low bias. Between 25 and 30 km, the observed  $N_5$  profile shows a layer of enhanced concentrations, by around a factor of 10. By contrast the model  $N_5$  profile shows a steady decrease with height in this altitude range, at the same rate as found in the lower stratosphere. The altitude of this sudden  $N_5$  increase corresponds approximately to where temperatures become warm enough for sulphuric acid particles to evaporate, which is not included in our simulations. Gas phase sulphuric acid concentrations are known to increase rapidly with height in this region from balloon-borne ion mass spectrometer measurements (e.g. Arnold et al., 1981). These elevated concentrations of gas phase  $H_2SO_4$  have been shown to cause significant nucleation in the middle-stratosphere (Hommel et al., 2011) which may be the cause of this observed feature.

In March 1992 (Fig. 10b), 9 months after the eruption, the observed particle concentration profiles show major enhancements between 17 and 25 km, compared to March 1991 for all size thresholds 150 nm and larger. By contrast,  $N_5$  has increased only slightly, and is only marginally higher than  $N_{150}$  and  $N_{250}$  for this month, suggesting most particles have grown larger than 250 nm in this height range. In the model, the lower stratosphere  $N_5$  and  $N_{150}$  high biases seen for November 1991 (Fig. 9c) have worsened further, by factors of 8 and 2 respectively. However, the model  $N_{250}$ ,  $N_{550}$ ,  $N_{750}$  and  $N_{1000}$  show much better agreement with the observations.

In March 1993 (Fig. 10c), the observations show clear separation between  $N_5$  and  $N_{150}$ , although  $N_{150}$  and  $N_{250}$  are close together. This indicates the formation of a bimodal size distribution consisting of an external mixture of particles which have grown



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tions in the 5–250 nm size range are too high compared to the observations. There are several possible causes for this model size distribution bias. First, nucleation rates at the very low humidity and temperature conditions in the stratosphere are known to be highly uncertain. The Vehkamäki et al. (2002) parameterization used in this paper is the best available for stratospheric conditions, but is essentially an extrapolation from laboratory measurements at much higher temperatures and humidities, based on classical nucleation theory. Second, the nucleation rate expression is a steep non-linear function of the sulphuric acid concentrations, which may vary considerably in the large global model grid-boxes. An approach sometimes used in global models to represent sub-grid-scale nucleation and growth, is to include a small fraction of the SO<sub>2</sub> emitted as a source of “primary sulphate” particles, with a prescribed size distribution. For example, Dentener et al. (2006) recommend including 2.5 % of SO<sub>2</sub> emitted from passively degassing and explosive volcanoes as primary sulphate with an  $R_{\text{eff}}$  of around 60 nm. Including such a sub-grid source of particles would also reduce gas phase H<sub>2</sub>SO<sub>4</sub> concentrations, since immediately after the eruption there would already be substantial particle surface area density. Lower H<sub>2</sub>SO<sub>4</sub> concentrations would hence lead to lower nucleation rates and would also weaken particle growth, which would tend to reduce both  $N_5$  and  $N_{150}$ .

To document the nucleation rate occurring in the no-Pinatubo and Pinatubo simulations, we show in Fig. 11 its global distribution, on the zonal-mean, against latitude and altitude over the period July to October 1991. In volcanically quiescent conditions, the model has nucleation occurring mainly in the tropical upper troposphere with negligible new particle formation in the stratosphere. The observed and simulated lower stratospheric  $N_5$  and  $N_{150}$  profiles at Laramie in March 1991 (Fig. 10a) are in good agreement, and Fig. 11 indicates that these stratospheric particles were actually formed in the tropical upper troposphere, consistent with the stratospheric aerosol lifecycle described by Hamill et al. (1997). The observations at Laramie indicate that only a small proportion of these nucleated particles grow to sizes larger than 150 nm, with most being at smaller sizes.

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Following the eruption of Mount Pinatubo, the balloon observations at Laramie indicate that, by March 1992 (e.g. Figure 10b),  $N_{150}$  is increased by a factor of 8, whereas  $N_5$  has already returned to pre-eruption values. As a consequence, the  $N_5$  and  $N_{150}$  profiles are separated by only a few tens of percent, indicating the majority of particles in the lower stratosphere have grown larger than 150 nm. In the model however, the Pinatubo simulation indicates  $N_5$  larger than  $N_{150}$  by around a factor of two, suggesting the air mass consists of an external mixture combining older particles which have grown to 150 nm and larger, together with other more recently nucleated particles which remain close to their nucleated size, formed after the emitted sulphur has already been oxidised and taken up into the aerosol particle phase.

Figure 11 shows that, for the model Pinatubo simulation, strong nucleation was occurring throughout the injection height range of 19–27 km for at least a month after the eruption. Nucleation rates then reduce in magnitude over August and September as the emitted  $\text{SO}_2$  is converted to sulphuric acid and by October (Fig. 11h) nucleation is similar to the no-Pinatubo simulation. Since the uptake of reactive gases is dependent on particle size, accounting for the shift in size distribution may also be important for better quantification of the influence that volcanically enhanced aerosol has on stratospheric ozone through accelerated heterogeneous chemistry. We therefore investigate the evolution of the SAD distribution across the 4 aerosol modes (Fig. 12) at around 1 month after the eruption (panels a to d) and at around the time of peak aerosol loading (panels e to h). Nucleation mode particles are always smaller than 10 nm, so even during July 1991, when substantial nucleation is occurring; those tiny particles have insignificant SAD. However, although the Aitken mode particles are smaller than 100 nm, during the early part of the eruption they have a very high SAD, with values in excess of  $10 \mu\text{m}^2 \text{cm}^{-3}$  between 22 and 29 km near the equator. The accumulation mode also shows similar very high values, but between 18 and 25 km, and over a wider latitude range. At this stage of the plume evolution, the coarse mode has only low SAD, with values peaking at around  $4 \mu\text{m}^2 \text{cm}^{-3}$ . In the main part of the plume, the SAD is mostly in accumulation mode sizes, whereas in the upper parts, the SAD comes mainly



from Aitken sized particles. We note that in Fig. 7 the model shows highest biases in simulated SAD against the observations during the first few months after the eruption.

## 6 Summary and conclusions

We have extended the UKCA module to incorporate stratospheric sulphur chemistry and stratospheric microphysical processes. Using stratospheric aerosol changes after the Mt. Pinatubo as a test case, we have evaluated simulated aerosol properties against a wide range of observations in both quiescent and volcanically perturbed conditions. The improvements to the model enable a prognostic treatment of stratospheric aerosol with dynamically varying particle size distribution alongside stratospheric transport and chemistry up to a model top of 84 km.

In general, the model captures the observed distribution and evolution of stratospheric aerosol properties well, in both quiescent and volcanically perturbed conditions. For the Pinatubo test case, the peak in global aerosol mass and decay timescale are captured well, with an e-folding time of 15.8 months, slightly longer than the 12–14 months estimated from satellite data (e.g. Baran et al., 1993). Modelled extinction in the tropical and NH mid-latitude lower stratosphere shows reasonably good agreement with SAGE V7 data in both the mid-visible and near infra-red. However, for the first six months after the eruption simulated AOD and SAD are larger ( $\sim 2$ – $4$  times) than the satellite measurements, and the model enhancement in  $R_{\text{eff}}$  is too low (e.g. compared to Bauman et al., 2003). Lack of radiative coupling in these simulations likely explains the underestimate in the transport of the Pinatubo plume to the Southern Hemisphere in the model, and this may also have caused tropical AOD and SAD to be too high in the tropics.

To better understand how the particle size distribution was perturbed during the Pinatubo eruption, we have compared against mid-latitude balloon-borne measurements during that time period, allowing a strong observational constraint on concentrations of particles larger than 5, 150, 250, 550, 750 and 1000 nm. Although there have

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been many model studies covering the Pinatubo period over the 22 yr since the eruption, to our knowledge, this is the first time the full profile of a simulated size distribution in a global model has been compared to these measurements in volcanically perturbed conditions.

5 In volcanically quiescent conditions, the model finds no nucleation occurring in the stratosphere, with particles forming mainly in the tropical upper troposphere. At NH mid-latitudes, the model agrees well with the observations, indicating that only a small proportion of these nucleated particles have grown to 150 nm by coagulation and condensation, with  $N_5$  larger than  $N_{150}$  by around a factor of 10. We have investigated the  
 10 impact on the size distribution of the 20 Tg tropical injection of SO<sub>2</sub> from Pinatubo. Although the spatial and temporal evolution of the  $R_{\text{eff}}$  in the lower stratosphere seen by satellite is well captured by the model, the comparisons to the balloon measurements indicate that, at NH mid-latitudes, the model generates far too many particles in the size range 5–250 nm, although larger particle sizes compare well to the observations. The  
 15 high bias in  $N_{150}$  is consistent with the high biases seen in AOD and SAD in the initial phase of the eruption, and with the general low bias apparent in  $R_{\text{eff}}$ . These findings underline the importance of better constraining nucleation and growth of these finest particles in the first few months after the eruption to improve prediction of volcanic impacts on climate with stratospheric aerosol microphysics models.

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**Table 1.** Additional sulphur chemistry reactions and rates within UKCA, W = Weisenstein et al. (1997), JPL = Sander et al. (2006), K03 = Kreidenweis et al. (2003)

	Rate	Reference
$\text{DMS} + \text{O}(^3\text{P}) \rightarrow \text{SO}_2$	$1.3 \times 10^{-11} \exp(410/T)$	W, JPL
$\text{DMS} + \text{OH} \rightarrow \text{SO}_2$	$1.2 \times 10^{-11} \exp(-260/T)$	W
$\text{DMS} + \text{OH} \rightarrow \text{MSA} + \text{SO}_2$	$3.04 \times 10^{-12} \exp(350/T) \cdot (\gamma/1 + \gamma)$ $\gamma = 5.53 \times 10^{-31} \exp(7460/T) \times [\text{O}_2]$	W
$\text{DMS} + \text{NO}_3 \rightarrow \text{SO}_2$	$1.9 \times 10^{-13} \exp(500/T)$	W
$\text{OCS} + \text{O}(^3\text{P}) \rightarrow \text{CO} + \text{SO}_2$	$2.1 \times 10^{-11} \exp(-2200/T)$	W, JPL
$\text{OCS} + \text{OH} \rightarrow \text{CO}_2 + \text{SO}_2$	$1.1 \times 10^{-13} \exp(-1200/T)$	W, JPL
$\text{SO}_2 + \text{OH} + M \rightarrow \text{SO}_3 + \text{HO}_2$	$k(T) = \frac{A}{1+B} \times 0.6^{(1+(\log B)^2)^{-1}}$ $A = 3.0 \times 10^{-31} \times (300/T)^{3.3}$ $B = A/1.5 \times 10^{-12}$	W
$\text{SO}_2 + \text{O}_3 \rightarrow \text{SO}_3$	$3.0 \times 10^{-12} \exp(-7000/T)$	W, JPL
$\text{SO}_3 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_4$	$8.5 \times 10^{-41} \exp(6540/T) \cdot [\text{H}_2\text{O}]$	JPL
$\text{SO}_2 + \text{H}_2\text{O}_2 \xrightarrow{\text{aqueous}} \text{SO}_4$		K03
$\text{OCS} + h\nu \rightarrow \text{CO} + \text{SO}_2$	Photolysis	W
$\text{H}_2\text{SO}_4 + h\nu \rightarrow \text{SO}_3 + \text{OH}$	Photolysis	W
$\text{SO}_3 + h\nu \rightarrow \text{SO}_2 + \text{O}(^3\text{P})$	Photolysis	W

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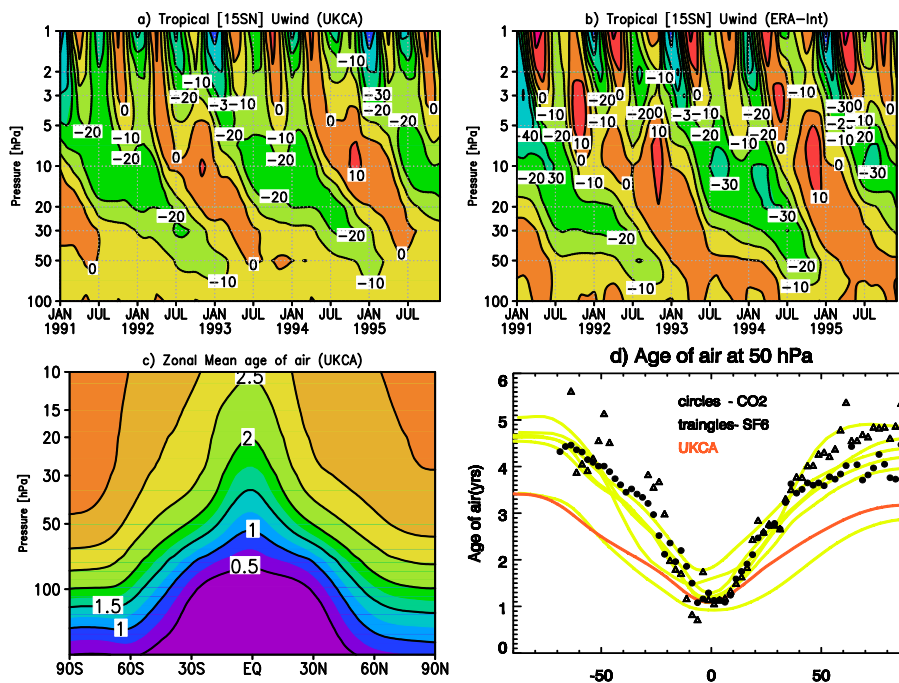
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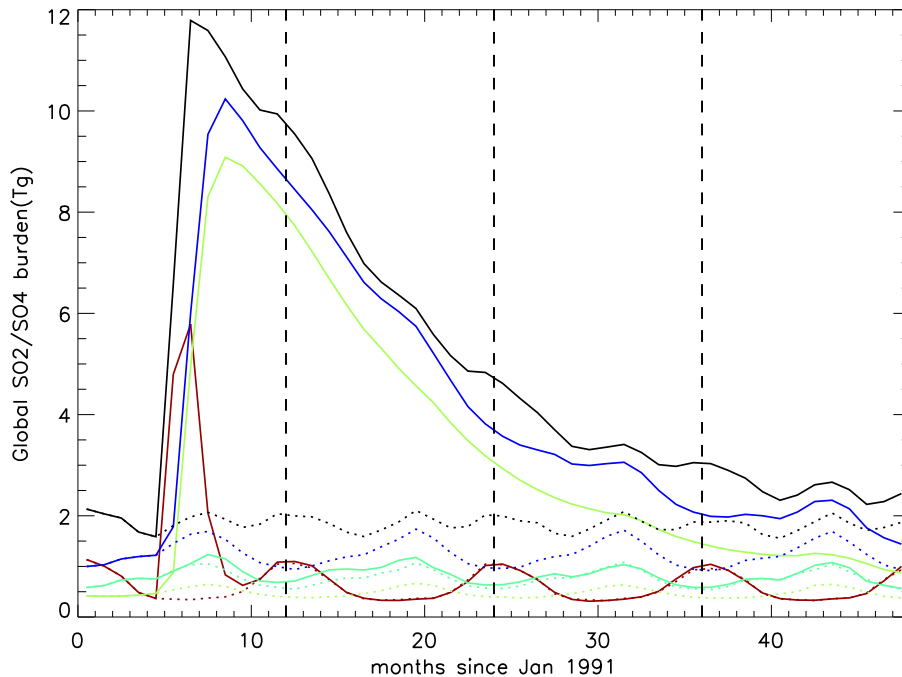
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**Fig. 1.** (a) Model simulated tropical ( $15^{\circ}\text{S}$ – $15^{\circ}\text{N}$ ) mean monthly mean zonal wind ( $\text{m s}^{-1}$ , QBO propagation). (b) Same as (a) but using ERA-interim reanalysis data (Dee et al., 2011). (c) Zonal mean age of air (years, mean 1991–2000), and (d) mean age of air (1991–2000) comparison at 50 hPa. Traingle and filled circles show estimated age of air from  $\text{CO}_2$  and  $\text{SF}_6$  (Hall et al., 1999). Mean age of air from various CCMs which participated in CCMVal lifetime assessment are shown with yellow lines and one from this study is shown with the red line.



**Fig. 2.** Time series of the global burden (in Tg of sulphur) of SO<sub>2</sub> (red), total sulphur (includes both SO<sub>2</sub> and aerosol, black), aerosol sulphur (dark blue) and aerosol sulphur in the upper troposphere and stratosphere (green) and lower and mid-troposphere (aqua), determined by above or below 400 hPa, for the simulations with (solid lines) and without (dashed lines) the Pinatubo eruption.

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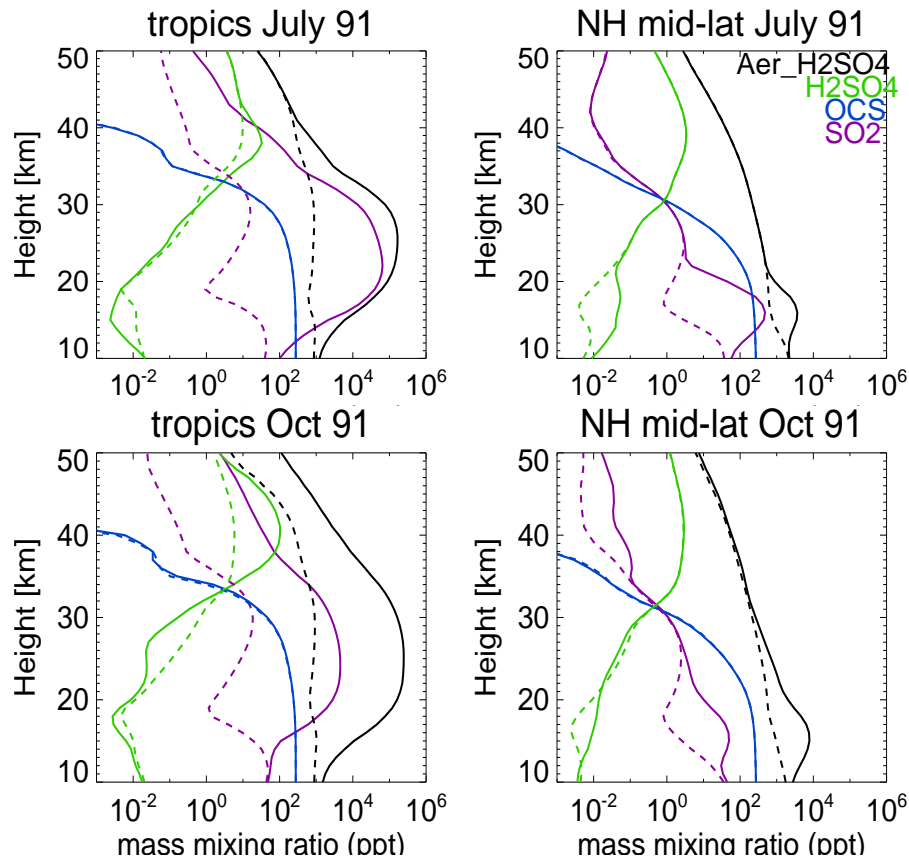
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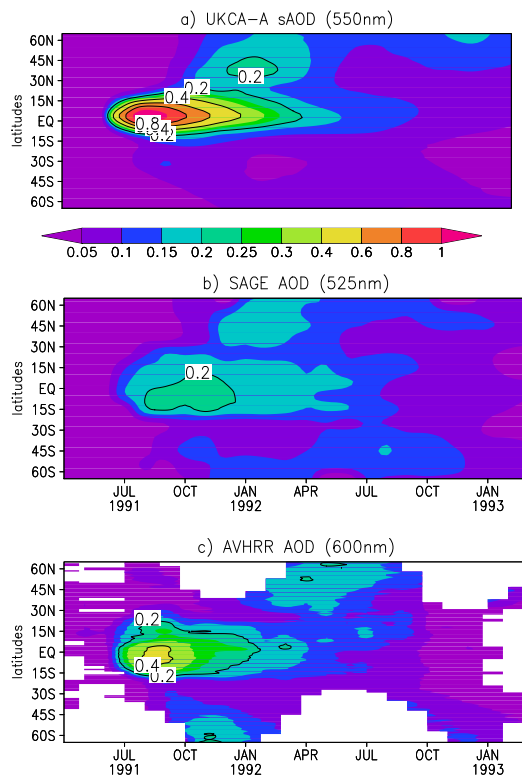




**Fig. 3.** Mixing ratios of various sulphur containing species (pptv) in the tropics ( $15^{\circ} S$ – $15^{\circ} N$ , left) and NH mid-latitudes ( $35$ – $60^{\circ} N$ , right) during July 1991 (top) and October 1991 (bottom). Gas-phase and particle phase  $H_2SO_4$  ratios are shown with black and green lines, respectively. OCS and  $SO_2$  are shown with blue and purple lines, respectively. Mixing ratios from the simulations with and without Pinatubo eruption are shown with solid and dashed lines, respectively.

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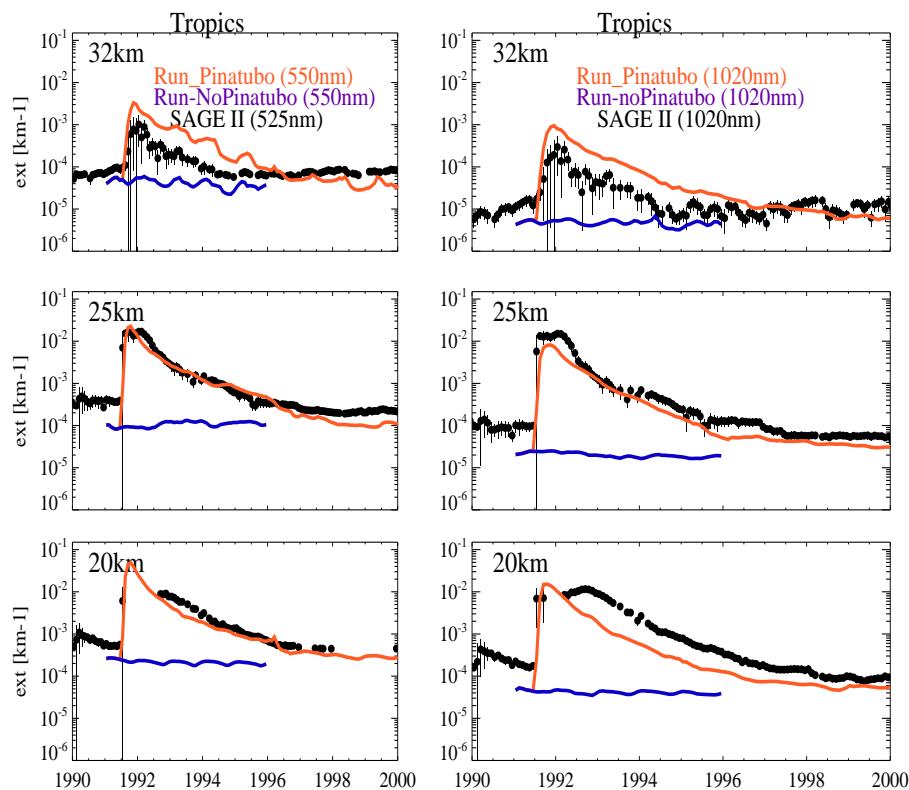
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**Fig. 4.** Time series of model simulated zonal mean **(a)** stratosphere-only (integrated from the tropopause to top of the atmosphere) aerosol optical depth (AOD) derived using 525 nm extinctions. **(b)** and **(c)** are the AOD derived using SAGE II (525 nm) and AVHRR (600 nm) measurements. Due to the different viewing geometry of the AVHRR instrument (nadir viewing) AVHRR AOD shown **(c)** are after removal of background tropospheric AOD (22 months before the eruption).

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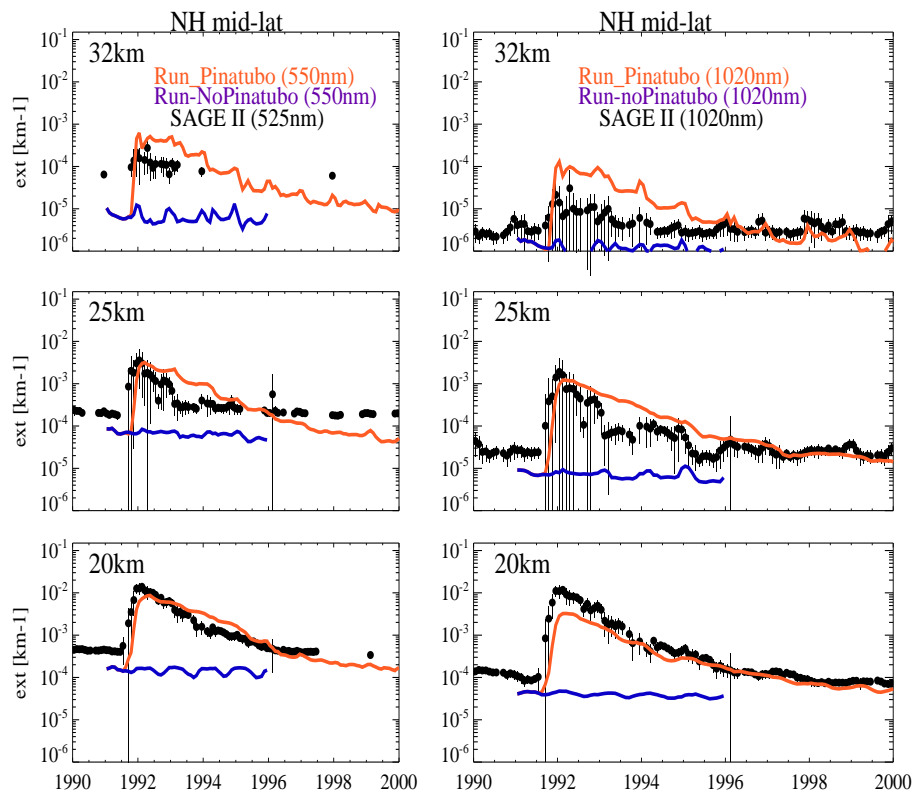


**Fig. 5.** Comparison between modelled and SAGE II (V7.0) retrieved extinction at 525 nm (left) and 1020 nm (right) in tropics (20° S–20° N) for 20 km (bottom), 25 km (middle) and 32 km (top).

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**Fig. 6.** Same as Fig. 5 but for NH mid-latitude (35–65° N).

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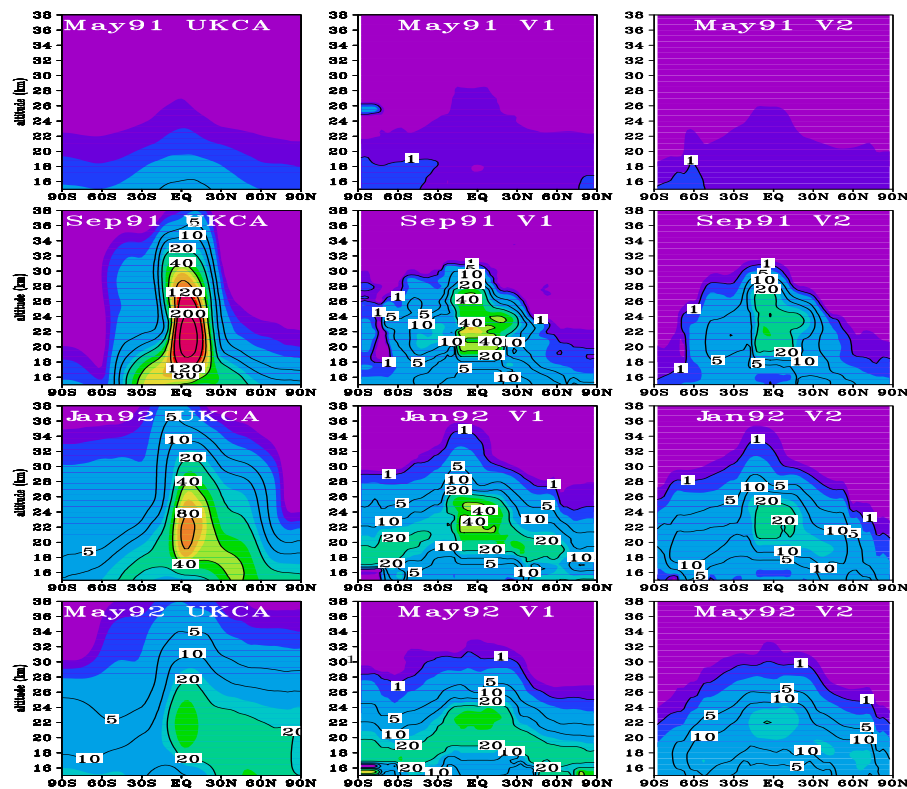
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**Fig. 7.** Comparison between zonal mean modelled and satellite derived V1 and V2 SAD ( $\mu\text{m}^2 \text{cm}^{-3}$ ) from SPARC (2006) and Arfeuille et al. (2013), respectively, for various months before and after the eruption.

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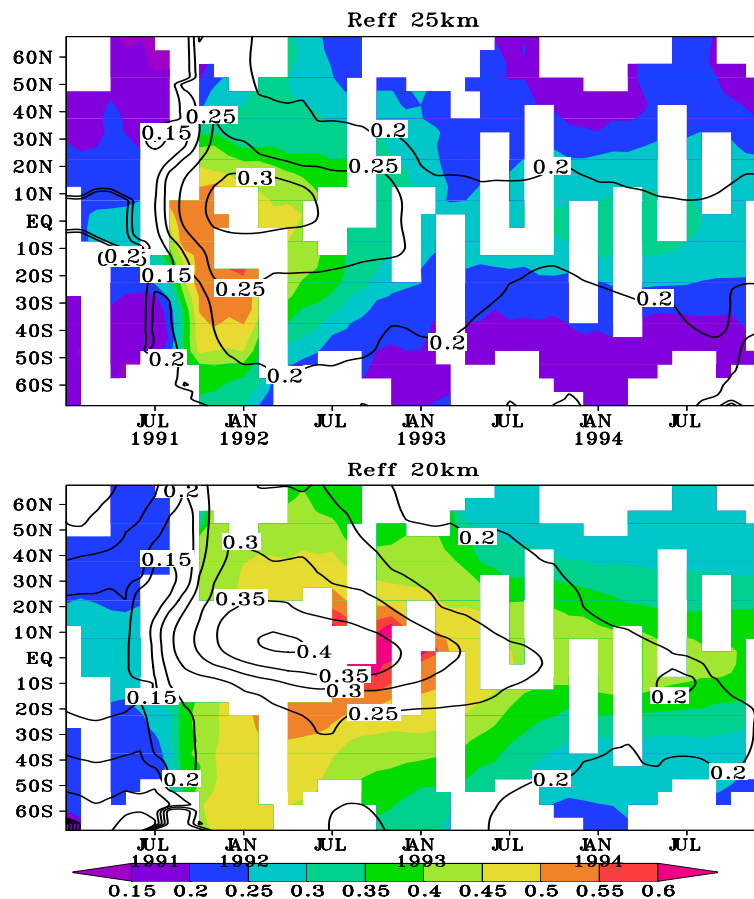
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**Fig. 8.** Satellite derived (shaded, from Bauman et al., 2003) and modelled (contours) effective radius ( $\mu\text{m}$ ) at 25 and 20 km.

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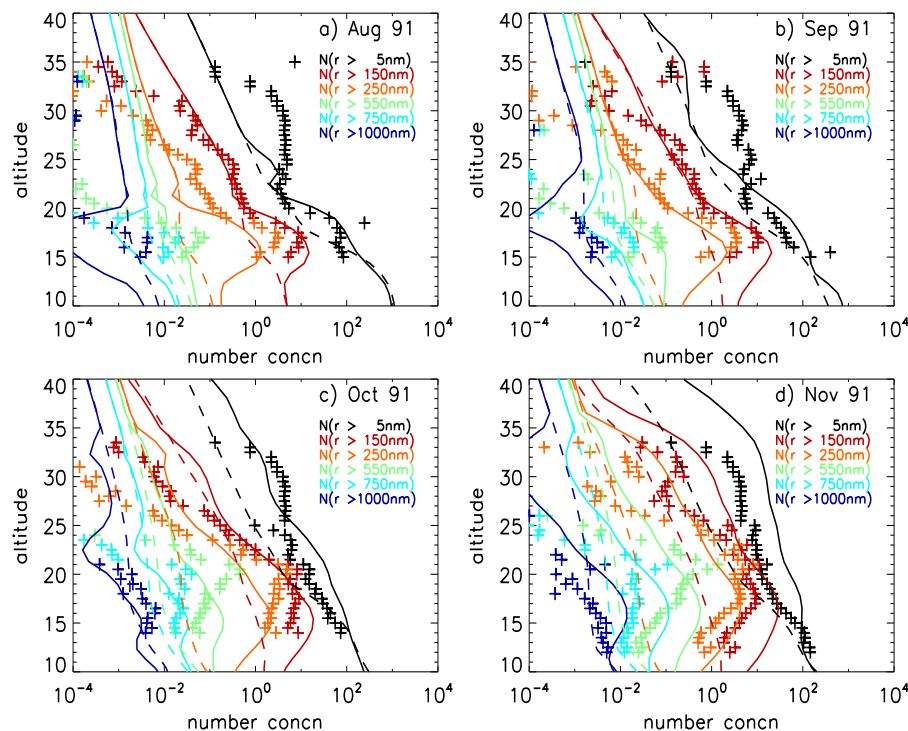
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**Fig. 9.** August, September, October, November profiles of size-resolved number concentrations of particles ( $\text{cm}^{-3}$ ) with radii larger than 5, 150, 250, 550, 750 and 1000 nm from Laramie ( $41.3^\circ \text{N}$ ,  $105.5^\circ \text{W}$ ) are shown with plus (+) symbol. Solid and dashed lines show aerosol profiles from the Pinatubo and no-Pinatubo runs, respectively highlighting the region where model predicts the perturbation in the aerosol profiles.

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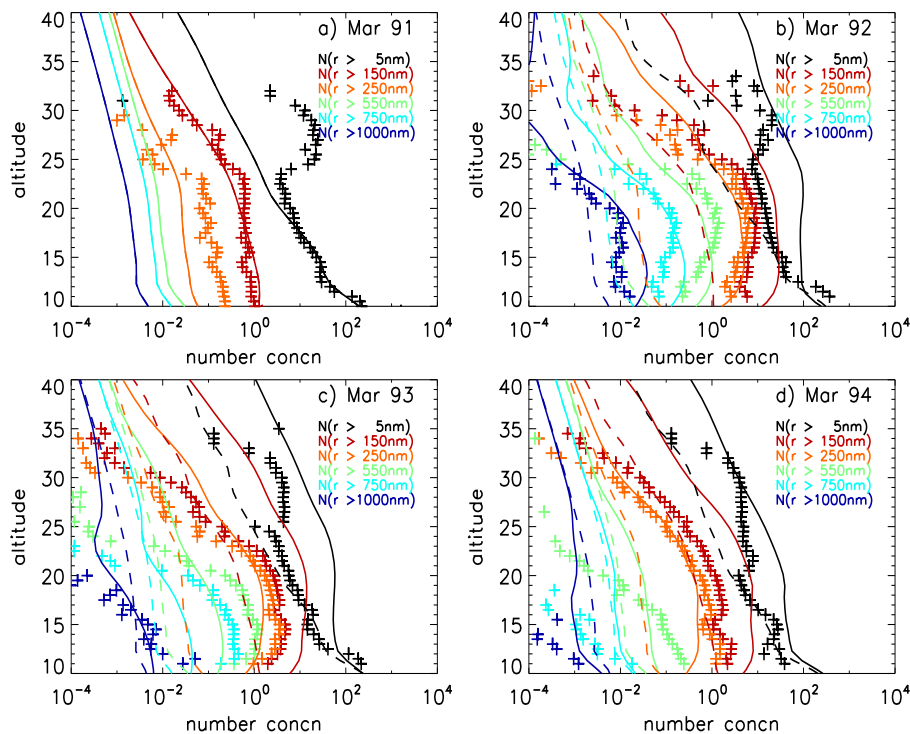


Fig. 10. Same as Fig. 9 but for March 1991, March 1992, March 1993 and March 1994.

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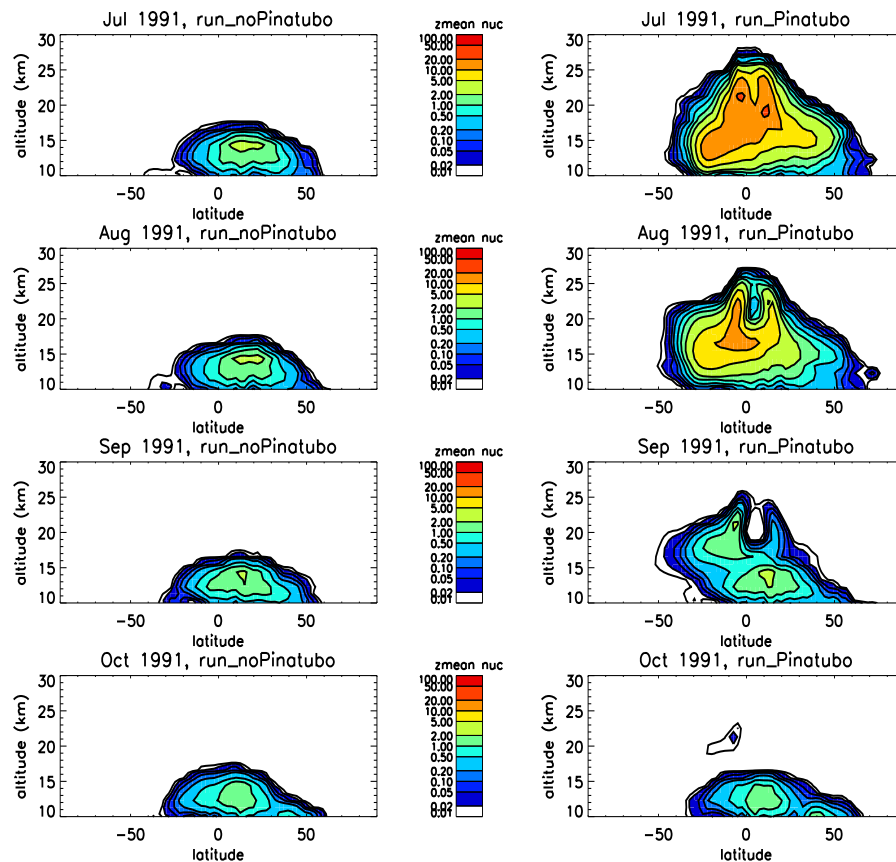
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**Fig. 11.** Modelled nucleation rates ( $\text{cm}^3 \text{s}^{-1}$ ) from non-Pinatubo (left) and Pinatubo (right) runs for (top to bottom) July, August, September and October 1991.

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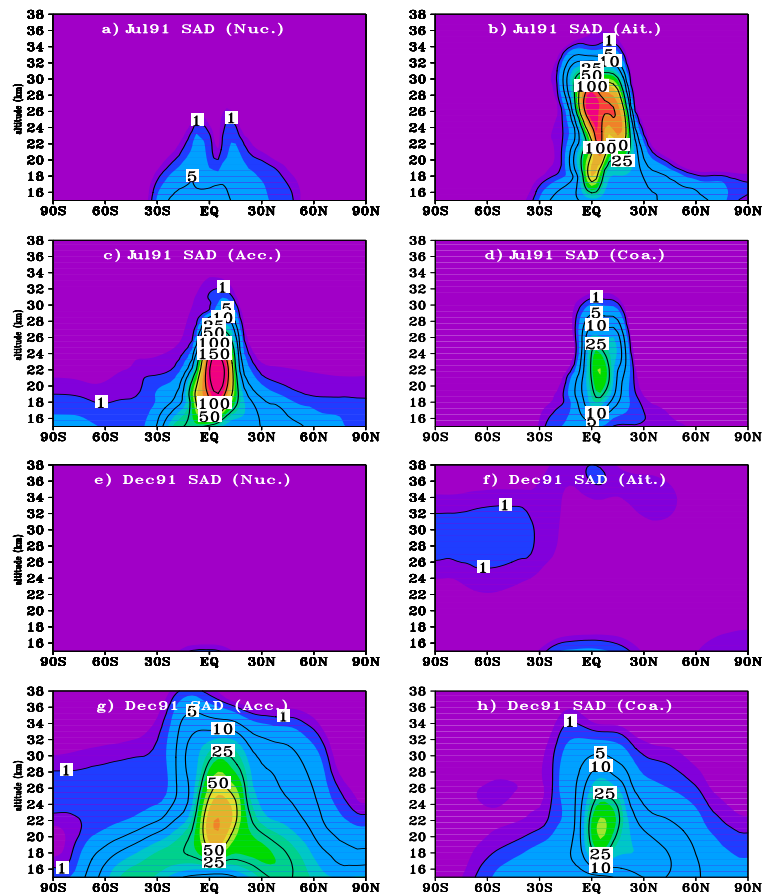
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**Fig. 12.** Zonal mean surface area density ( $\mu\text{m}^2 \text{cm}^{-3}$ ) for each mode against latitude and altitude for July 1991 (a, b, c, d) and December 1991 (e, f, g, h).