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module
intercomparison

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Global 2-D intercomparison of sectional and modal aerosol modules

D. K. Weisenstein¹, J. E. Penner², M. Herzog^{2,*}, and X. Liu^{2,**}

¹Atmospheric and Environmental Research, Inc., Lexington, Massachusetts, USA

²Department of Atmospheric, Oceanic, and Space Sciences, University of Michigan, Ann Arbor, Michigan, USA

*now at: NOAA Geophysical Fluid Dynamics Laborator, Princeton, New Jersey, USA

**now at: Pacific Northwest National Laboratory, Richland, Washington, USA

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Correspondence to: D. K. Weisenstein (dkweis@aer.com)

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Abstract

We present an intercomparison of two aerosol modules, one sectional, one modal, in a global 2-D model in order to differentiate their behavior for tropospheric and stratospheric applications. We model only binary sulfuric acid-water aerosols in this study.

Two versions of the sectional model and three versions of the modal model are used to test the sensitivity of background aerosol mass and size distribution to the number of bins or modes and to the prescribed width of the largest mode. We find modest sensitivity to the number of bins (40 vs 150) used in the sectional model. Aerosol mass is found to be reduced in a modal model if care is not taken in selecting the width of the largest lognormal mode, reflecting differences in sedimentation in the middle stratosphere. The size distributions calculated by the sectional model can be better matched by a modal model with four modes rather than three modes in most but not all situations. A simulation of aerosol decay following the 1991 eruption of Mt. Pinatubo shows that the representation of the size distribution can have a significant impact on model-calculated aerosol decay rates in the stratosphere. Between 1991 and 1995, aerosol mass and surface area density calculated by two versions of the modal model adequately match results from the sectional model. Calculated effective radius for the same time period shows more intermodel variability.

1 Introduction

Aerosols are important to the radiative balance and chemistry of the atmosphere, and can modify cloud properties. In the stratosphere, aerosol particles provide surfaces for heterogeneous chemistry, modifying the ratio of active NO_x , HO_x , ClO_x , and BrO_x radicals to reservoir species (Fahey et al., 1993; Wennberg et al., 1994) and thus modifying ozone concentrations following volcanic eruptions (Hofmann and Solomon, 1989). In the troposphere, aerosol particles can act as cloud condensation nuclei, influencing cloud droplet number density and size (Penner et al., 2001), and thus cloud

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albedo. They also have direct radiative effects (Haywood and Ramaswamy, 1998) and can modify atmospheric circulation (Labitzke and McCormick, 1992; McCormick, et al., 1995) and temperature (Hansen et al., 2002).

This study was motivated by the requirements of the Global Modeling Initiative (GMI) 3-D chemical-transport model (Rotman et al., 2001). The GMI model was created to be modular and permit intercomparisons between different process modules as a way of studying model sensitivity. It uses a variety of wind fields, from both assimilation systems and GCM simulations (Douglass et al., 1999; Strahan and Douglass, 2004). It can also use a variety of chemical schemes and parameterizations (Considine et al., 2000; Douglass et al., 2004). Sulfur chemistry and aerosol microphysics from the University of Michigan 3-mode model have been added to the tropospheric version of GMI (Liu et al., 2005). Eventually the GMI will operate with aerosol microphysics in a version which will span both troposphere and stratosphere, and can run with either a sectional or modal aerosol representation. This study tests and contrasts these two representations of aerosol size distribution in a 2-D model of the troposphere and stratosphere for both accuracy and computational efficiency.

Tropospheric aerosol models must deal with many types of aerosols, including sulfate, dust, sea salt, organics, and black carbon. Because of the computational requirements of 3-D tropospheric models, the prediction of aerosol mass was often considered adequate and fixed size distributions were assumed to evaluate radiative effects (Penner et al., 2001, 2002). More recent models have added the prediction of number density and size distribution using efficient methods such as modal representations (Whitby et al., 1991) or the method of moments (Wright et al., 2001). Regional tropospheric models have employed more detailed sectional representations (Jacobson, 2001; Zhang et al., 2004) to predict particle size distributions without imposing an a priori shape on the distribution.

Stratospheric aerosol models generally differ from tropospheric aerosol models because resolving the size distribution of aerosol particles becomes more important at altitudes above the tropopause. In the troposphere, only particles larger than $\sim 1 \mu\text{m}$

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settle appreciably, whereas the thinner air in the stratosphere causes sedimentation rates to be a strong function of both particle radius and air density. Even particles of $0.01 \mu\text{m}$ radius have significant sedimentation rates at 30 km. Resolving the size distribution of aerosol particles is crucial to predicting the correct sedimentation rate and therefore the lifetime and vertical distribution of particles in the stratosphere. Thus stratospheric models have generally used the sectional approach to resolve size distributions (Weisenstein et al., 1997; Timmreck, 2001; Pitari et al., 2002). Yet the sectional approach leads to numerical diffusion in size space, which may be excessive for a coarse resolution sectional model. The computational expense of a sectional model is mitigated for stratospheric studies because non-sulfate particles are not important in much of the stratosphere and therefore are generally omitted.

Many of the sulfur source gases are short-lived and have localized emissions, such as industrial sources. Rapid transport in convective cells is believed to play an important role in moving sulfur source gases from the boundary layer to the upper troposphere, where they may interact with clouds. Transport by diabatic ascent, cloud outflow, and horizontal motion moves sulfur from the troposphere into the stratosphere. Thus detailed modeling of tropospheric transport, cloud interactions, and microphysics is important to predicting the sulfur entering the stratosphere. For these reasons, we believe it important to model tropospheric and stratospheric aerosols together in a 3-D model like the GMI.

The University of Michigan aerosol module, referred to as UMaer, is described in Herzog et al. (2004). That paper applied the aerosol module within a zero-dimensional box model and compared results with the Atmospheric and Environmental Research (AER) sectional model using 40 or 150 bins. In that intercomparison, both models were thoroughly tested until the only remaining differences were due to the representation of the size distribution; differences in microphysical parameterizations were removed. Here that intercomparison is extended to two dimensions so that the impact of the different representations of size distribution will be seen in the model transport and sedimentation as well. We have performed simulations of sulfate aerosol under back-

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ground nonvolcanic conditions and a time dependent simulation from 1991 to 2001 including the Mt. Pinatubo eruption.

This paper presents descriptions of the 2-D model framework used as the inter-comparison tool and the two aerosol modules in Sect. 2. In Sect. 3 we describe the intercomparison approach and model versions tested (two sectional, three modal). Section 4 provides results for a background atmosphere calculation in the upper troposphere and stratosphere, showing how differences in aerosol representation affect model results. Section 5 shows results for a volcanic simulation and the differences in aerosol removal rates with the different approaches. A summary and discussion is provided in Sect. 6.

2 Model descriptions

The AER 2-D model is used as the framework for this intercomparison, with transport, sulfur chemistry, and aerosol microphysics performed for the global domain from the surface to 60 km. Grid resolution is 9.5° in latitude and 1.2 km in the vertical. Transport is effected by the residual circulation and by horizontal and vertical diffusion. We use transport parameters from Fleming et al. (1999) which are calculated from observed ozone, water vapor, zonal wind, and temperature for climatological conditions. Wave driving is provided by forcing from six planetary waves and the effects of gravity wave breaking. Diabatic heating rates are computed following Rosenfield et al. (1994), with tropospheric latent heating from Newell et al. (1974). The calculation of horizontal diffusion coefficients follows Randel and Garcia (1994).

Since we are modeling sulfate aerosols for this intercomparison, we model only sulfur chemistry and use necessary radical concentrations from other model simulations. Sulfur source gases include DMS, H_2S , CS_2 , OCS, and SO_2 . Surface fluxes are 25 MT sulfur per year from DMS, 1 MT sulfur from CS_2 , 8.7 MT sulfur from H_2S , and 78 MT sulfur from SO_2 . In addition, we assume a surface mixing ratio for OCS of 500 pptv which provides a stratospheric source of sulfur. Photolysis and reactions with OH, O,

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O₃, and NO₃ convert sulfur source gases to sulfuric acid. Reaction rates are taken from the JPL 2002 compendium (Sander et al., 2003). Homogeneous nucleation of sulfuric acid vapor occurs chiefly in the tropical upper troposphere due to low temperatures and high relative humidity. Subsequently, condensation increases the size of particles, while coagulation limits their number density. Evaporation occurs in the 30–40 km altitude region, yielding H₂SO₄ vapor, and, after photolysis, SO₂.

The University of Michigan aerosol module (UMaer) is capable of treating the nucleation and growth of sulfuric acid-water aerosols, as well as their coagulation with nonsulfate particles (Herzog et al., 2004). Aerosol size distributions, defined by N(r), particle number concentration at radius r, are treated by predicting two moments (mass and number) of the two or more lognormal distributions

$$\frac{dN(r)}{dr} = \frac{N_0}{\sqrt{2\pi}r_g \ln \sigma_g} \exp\left(-\frac{1}{2} \frac{\ln^2(r/r_g)}{\ln^2 \sigma_g}\right) \quad (1)$$

each of which is defined by a mode radius r_g and a distribution width σ_g . The distribution width is specified. As mass is added to each mode and the particles grow, a merging process shifts mass from mode to mode, keeping the mode radius within defined bounds. The module performs dynamic time stepping without operator splitting such that all aerosol processes interact with each other during each time step. It can be applied to both tropospheric and stratospheric conditions, but to date has been used only in the troposphere and lowermost stratosphere.

The AER 2-D sulfate aerosol model was described in Weisenstein et al. (1997, 1998). The aerosol module uses a sectional representation of the particle size distribution and can represent any arbitrary distribution shape. Particle number density in 40 bins between 0.4 nm and 3.2 μm by volume doubling is predicted, though the bin number, smallest radius, and volume ratio between bins are adjustable parameters. The model is intended for stratospheric applications and includes only sulfuric acid-water particles. Aerosol composition, or weight fractions of sulfate and water, is adjusted continuously based on ambient temperature and relative humidity according to Tabazadeh et

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al. (1997). Particle sizes are based on both sulfate and water fractions, so that particles grow or shrink when either temperature or water vapor concentration changes in a grid box. The microphysical solution uses operator splitting with a time step of one hour for transport, chemistry, and microphysics, but 20 substeps for the condensation and nucleation processes to prevent one process from dominating the gas-to-particle exchange rate.

Our goal in this intercomparison, and our previous box model intercomparison (Herzog et al., 2004) was to compare microphysical modules which are identical except in the way that the size distribution is represented. To that end, we have compared each aerosol process carefully and ensured that initial tendencies are identical. We use the Vehkamaeki et al. (2002) nucleation parameterization, which is in agreement with more detailed calculations of hydrated clusters for temperatures greater than 190 K. The parameterization calculates the radius, composition, and production rate of new particles. The UMaer module adds the new particle number and mass to the smallest aerosol mode, exactly preserving the calculated number density. The AER module requires that the nucleated mass be added to a single bin with a fixed radius, so particle number is adjusted to preserve the calculated nucleation mass.

The condensation and evaporation process is treated as described in Herzog et al. (2004). The condensational growth or evaporation in each bin or mode depends of the difference in the gas phase H_2SO_4 concentration, N_{gas} , and the equilibrium concentration of H_2SO_4 above the particle surface, $N_{\text{gas}}^{\text{equ}}$, and is described by

$$\frac{\partial}{\partial t} N_{\text{gas}} = -4\pi\beta D(N_{\text{gas}} - N_{\text{gas}}^{\text{equ}})r_p N_p \quad (2)$$

where β is a term correcting for noncontinuum effects and imperfect surface accommodation, D is the diffusion coefficient for H_2SO_4 molecules in air, and N_p is the number density of particles in the bin or mode. For the sectional model, r_p is the bin radius. For the modal model, the appropriate radius r_p depends on the volume mean wet radius,

$\overline{r_{\text{vol}}}$, and the width of the lognormal distribution, σ_g ,

$$r_p = \overline{r_{\text{vol}}} \exp(-\ln^2 \sigma_g). \quad (3)$$

Wet radius is calculated using the Tabazadeh et al. (1997) parameterization for aerosol composition. The Kelvin effect is included in the calculation of $N_{\text{gas}}^{\text{equ}}$ and depends upon particle radius. The modal model uses volume mean radius in this calculation, and for calculation of the Knudsen number when calculating β . Condensation doesn't change the number of particles, only the mass in each mode for the modal model. In the sectional model, condensational growth shifts particles to larger bin sizes. Evaporation does the opposite, shifting particles to smaller sizes, but net number density is only reduced for evaporation from the smallest bin size or mode.

The coagulation process reduces number concentration and shifts aerosol mass into larger particles. A coagulation kernel defines the collision probability of two particles of different radii, and depends of the radius of each particle and the particle diffusion coefficients. The modal model uses volume mean wet radius for this calculation. In the sectional model, when two particles of radii r_i and r_j collide, where $r_i < r_j$, a particle is removed from bin i and a new particle with size intermediate to bin j and bin $j+1$ is created. We apportion the particle mass between the two bins but this process results in numerical diffusion in size space. If the ratio of adjacent bin volumes is 2.0, then coagulation of two particles in bin j results in a particles of exactly radius r_{j+1} . In the modal model, coagulation within a mode results in a reduction in number in that mode. Coagulation between modes results in a reduction in mass and number of the smaller mode and an increase of mass in the larger mode.

The sedimentation process affects the vertical distribution of aerosol sulfate, particularly in the middle stratosphere, and reduces the residence time of particles. The gravitational settling velocity of a particle with radius r_p is given by

$$v_{\text{grav}} = \frac{2}{9} \frac{g}{\eta_{\text{air}}} r_p^2 \rho_p \left[1 + Kn \left[1.257 + 0.4 \exp(-1.1Kn^{-1}) \right] \right] \quad (4)$$

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with

$$Kn = \frac{\lambda_{\text{air}}}{r_p} \quad (5)$$

according to Stokes law with the Cunningham slip correction factor (Seinfeld and Pandis, 1997). Here λ_{air} is the mean free path of air and η_{air} is the viscosity of air. The sectional model applies the above settling velocity for each bin. The modal model applies one settling velocity per mode, replacing r_p with an effective radius appropriate for sedimentation of aerosol number

$$r_p^{\text{num}} = \overline{r_{\text{vol}}} \exp(-0.5 \ln^2 \sigma_g) \quad (6)$$

or for sedimentation of aerosol mass

$$r_p^{\text{mass}} = \overline{r_{\text{vol}}} \exp(2.5 \ln^2 \sigma_g). \quad (7)$$

Since settling velocity is quite sensitive to particle radius for submicron particles at altitudes above about 25 km, we expect that the difference in a modal and a sectional model may be pronounced above 25 km.

The box model intercomparison between these two aerosol modules (Herzog et al., 2004) showed that the modal model was capable of predicting both aerosol number concentration and surface area to within a factor of 1.2 (4 modes) or 1.3 (2 modes) on average as compared to the sectional model. Prediction of accumulation mode particle number concentration was not as accurate but still generally within a factor of 2.1. This intercomparison is performed within a 2-D model in order to compare differences due to transport and sedimentation over seasonal and decadal time scales. In the sectional model, transport occurs independently for each size bin. In the modal model, transport modifies the number and mass for each mode. Sedimentation in the modal model is a function of the assumed distribution width, since the larger tail of the distribution is much more sensitive to settling, yet the entire modal distribution is given the same settling velocity. Sedimentation not only removes particles from a

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given grid box, it also moves particles from higher to lower altitudes, affecting local size distributions and vertical mass profiles. We expect sedimentation to contribute to much of the difference between model-calculated aerosol distributions, and therefore we perform some calculations with and without sedimentation.

3 Intercomparison approach

Our intercomparison approach is to apply the two different aerosol modules within the same 2-D chemical-transport model. Thus transport and chemistry are treated identically insofar as possible. We use the AER sectional model with 40 bins (our standard treatment) and with 150 bins. The 150-bin version is computationally expensive, but reduces the numerical diffusion inherent in a sectional model. Thus we can also analyze deficiencies in the AER 40-bin model. The model versions and their defining parameters are listed in Table 1, along with the total stratospheric aerosol burden from each background aerosol calculation. The UMaer modal model is run with three modes and with four modes. We present two 3-mode versions differing in the width, σ_g , of the largest mode. For each modal model, we specify the distribution width and size limits of the modes. Each model is run to steady-state for a background atmosphere case. The final state is independent of the initial condition. Some of the selected cases are also run in time-dependent mode for a Pinatubo-like injection of volcanic SO_2 in the tropical stratosphere, so that we can compare the volcanic aerosol decay rates over a 10 year period. These calculations are initialized with the steady-state background condition for the respective model.

Each model version covers the diameter range from sub-nanometer to about $3 \mu\text{m}$. In the sectional model, the bin spacing is described by the parameter V_{rat} which is the ratio of particle volumes between adjacent bins. Typical stratospheric aerosol sectional models (see Bekki and Pyle, 1992; Mills et al., 1999; Timmreck, 2001) employ V_{rat} values of 2.0, as does our 40-bin model. Our 150-bin model uses a V_{rat} of 1.2 to cover the same radius space. Numerical stability demands smaller time steps as the number

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of bins increases. We use a 15 min time step for the 150-bin model and a one hour time step for the 40-bin model, in each case applying 20 substeps for condensation and nucleation. The modal model specifies the width of each mode, σ_g . In addition, merge radii are required which specify when aerosol mass is shifted to the next larger mode as the mean radius of the mode increases. See Herzog et al. (2004) for details of this process. The modal version UMaer-3mA uses mode definitions which were tuned to calculate tropospheric aerosols. UMaer-3mB has an adjustment of the largest mode for better stratospheric performance. UMaer-4m is a 4-mode version which can better represent the details of the size distribution under varying conditions. The computational requirements of the 3-mode model are about half that of the 40-bin model. The 4-mode model requires about 70% of the computational resources of the 40-bin model. The 150-bin model increases the computational cost by a factor of 15 over the 40-bin model and is not practical for global calculations in 2-D, and is prohibitive for 3-D.

4 Nonvolcanic atmosphere intercomparison

We use the AER model with 150 bins (designated AER150) as the best numerical solution for the background atmosphere simulation. We define the background atmosphere for sulfate aerosol as an atmosphere with biogenic and anthropogenic sulfur emissions appropriate to the year 2000, but without volcanic influence. We omit aerosol types other than sulfate for simplicity in our intercomparison. The modeled tropospheric aerosol is not realistic without dust, sea salt, organic carbon, and black carbon and cannot be compared with observations. The stratospheric aerosol is realistic a few kilometers away from the tropopause, and will be compared with global observations from the SAGE II satellite. In Fig. 1 we show calculated annual average aerosol properties from the AER150 model version for the global domain from the surface to 40 km (the top of the aerosol layer). Aerosol mass density in $\mu\text{g}/\text{cm}^3$, including both sulfuric acid and water in particulate form, is shown in Fig. 1a, and surface area density in $\mu\text{m}^2/\text{cm}^3$ in Fig. 1b. These integrated aerosol quantities are important for mass bal-

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ance and heterogeneous chemistry affecting ozone, respectively. Shown in Fig. 1c is the effective radius, defined as

$$R_{\text{eff}} = \frac{\int r^3 \frac{dN}{dr} dr}{\int r^2 \frac{dN}{dr} dr} \quad (8)$$

and in Fig. 1d the number density of particles with radii greater than $0.05 \mu\text{m}$.

We compare the AER sectional module with 40 bins (AER40) with the sectional model using 150 bins (AER150) to assess the accuracy of the AER40 module relative to numerical diffusion in radius. Figure 2 shows calculated aerosol size distributions in April at several latitudes and altitudes for the AER150 model and the AER40 model. The size distributions are very similar, but the AER40 model produces more large particles and more small particles due to the size broadening effect of numerical diffusion. Figure 3 shows percent differences between AER40 and AER150 in aerosol mass, surface area density, effective radius, and number density. Aerosol mass density between AER40 and AER150 is identical in the troposphere, but in the stratosphere AER40 is reduced by 1% at the tropopause to 10–15% at 30 km relative to the AER150 model. The reduction in aerosol mass density with altitude is caused by greater sedimentation in AER40 with its slightly broader size distributions. AER40 also shows less surface area density than AER150, by 4–14% between the surface and 30 km. Reductions in the troposphere are seen due to differences in the size distributions between the two models, even though their mass densities are the same. Effective radius is increased by 0–6% in AER40, consistent with the increase in large particles which leads to increased sedimentation. Number density for particles greater than $0.05 \mu\text{m}$ radius is increased in the middle troposphere and decreased in most of the stratosphere.

We have run the AER40, UMAer-3mA, UMAer-3mB, and UMAer-4m models without sedimentation, since we expect that sedimentation will play a major role in differences between these models. Figure 4 shows the impact of sedimentation on the AER40 model. The difference in aerosol mass with sedimentation vs without is seen in Fig. 4a. Sedimentation reduces aerosol mass by a few percent at the tropopause, up to 40% at

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15 km, and up to 90% at 30 km. In the tropics below 25 km, aerosol mass is increased by sedimentation, since particles sediment from above into this region, and the upwelling circulation causes some particles to stagnate in this region. Figure 4b shows a similar comparison but for total model-calculated sulfur in both gas and particulate forms, plotted up to 50 km. By looking at total sulfur, we avoid model differences in condensation and evaporation rates, which depend on existing particle densities and size distributions. The impact of sedimentation on total sulfur is somewhat less than on aerosol mass, with a maximum effect of only 85% at 30 km.

A comparison of aerosol mass density calculated by the UMaer-3mA model versus the AER40 model without sedimentation is shown in Fig. 5a. In all of our comparisons, we adjust the radii of modal distributions to include condensed water, since the sectional model size distributions include both water and sulfate. Model differences without sedimentation are 0–20% within the stratospheric aerosol layer, and less than 5% in the troposphere. Figure 5b shows the same comparison but for total sulfur (gas and particulate). Here differences are less than 10%, indicating that condensation and evaporation account for some of the differences seen in the aerosol mass density. Total sulfur differences are greater than 5% only above 30 km where aerosols evaporate and at high latitudes. The UMaer-3mB model does not differ substantially from the UMaer-3mA model without sedimentation. The UMaer-4m model differs from the UMaer-3mA model by 2% or less in aerosol mass density when sedimentation is omitted.

When sedimentation is included in the calculations, differences between the modal and sectional models become considerably larger. Figure 6a shows percent differences in aerosol mass density between model UMaer-3mA and AER150. The models don't differ in the troposphere, but the UMaer-3mA model calculates less aerosol mass in most of the middle stratosphere, by as much as 40%. In the tropical lower stratosphere, the UMaer-3mA model calculates up to 20% more aerosol mass, and near the tropopause at high latitudes up to 10% more aerosol mass. In the comparison without sedimentation, the UMaer-3mA model showed an increase of 5–10% in aerosol mass over most of the stratosphere. Sedimentation is removing more aerosol

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mass in the middle stratosphere of the UMaer-3mA model than in the AER150 model, and the excess sedimentation is increasing aerosol mass in parts of the lower stratosphere. Changes in surface area density between Umaer-3mA and AER150 are shown in Fig. 6b. Surface area changes in both the troposphere and stratosphere, with differences within 20% everywhere but high latitudes and high altitudes. Because the models represent the size distribution differently, they have different surface area densities even when the mass density is the same. Figure 6c shows differences in effective radius, which varies by up to 30%, both positively and negatively, below 25 km. Number density, shown in Fig. 6d, varies by up to 20% both positively and negatively, with the largest differences in the troposphere.

The aerosol size distributions generated by the modal models are shown in Fig. 7 for several latitudes and altitudes and compared with size distributions generated by the AER150 model. The red lines represent the UMaer-3mA model and the green lines the UMaer-3mB model. These are both 3-mode models and differ only in the width of the third and largest mode. As seen in the figures, UMaer-3mA always produces more large particles than UMaer-3mB and both produce more large particles than AER150 for the locations shown. The 4-mode UMaer-4m model results are shown with the blue lines. The size distributions that the modal model is capable of reproducing accurately depend on the imposed number of modes and widths of those modes. The distribution shown in Fig. 7b for 20 km at the equator in April is a lognormal distribution of aged aerosol particles centered at about $0.1 \mu\text{m}$ with a secondary peak below $0.001 \mu\text{m}$ due to nucleation. The 3-mode models reproduce this distribution well. The 4-mode model calculates too many particles in the 0.001 to $0.004 \mu\text{m}$ size range. The distribution shown in Fig. 7a for the tropical upper troposphere contains large numbers of particles from $0.0005 \mu\text{m}$ to $0.1 \mu\text{m}$ and results from continual nucleation. The 3-mode models cannot reproduce this distribution over the full range or radii, but capture the distribution well for particles greater than $0.01 \mu\text{m}$. The 4-mode model more accurately captures the particle distributions for radii less than $0.01 \mu\text{m}$. The distributions shown in Figs. 7c and d represent an aerosol population that may have been subjected to evaporation

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or mixing of air masses with different histories. Both the 3-mode and 4-mode models have difficulty reproducing the lower size cutoff of these distributions, with the 3-mode model performing better. However, since integrated aerosol properties depend most strongly on the larger particles in the distribution, this failure may not be significant for many applications.

Figure 8 shows percent changes in integrated aerosol properties for the UMaer-3mB model relative to the AER150 model. This figure can be compared with Fig. 6 to evaluate how the width of the large mode in the 3-mode model affects quantities such as aerosol mass density and effective radius. The narrower width of the large mode in UMaer-3mB does lead to an aerosol mass density above 25 km which is closer to the AER150 mass density than the UMaer-3mA model. Mass density is still 10–30% less than AER150, but not 20–40% less as was UMaer-3mA. In the lower stratosphere and up to 25 km in the tropics, the aerosol mass density is greater than in AER150 and up to 10% greater than with UMaer-3mA. Narrowing the large mode of the 3-mode model improved the simulation of mass density in the middle stratosphere but made the results somewhat worse in the lower stratosphere and tropics. The same is true for surface area density, where in addition the tropospheric values became somewhat worse. Effective radius, however, is improved in the entire stratosphere, but not in the troposphere. Calculated number density is improved in most of the troposphere but becomes worse in most of the stratosphere.

The same comparisons for the 4-mode model are shown in Fig. 9. The maximum difference between UMaer-4m and AER150 is only 20% in aerosol mass density or surface area density below 30 km. Large differences above that altitude are not significant in terms of stratospheric aerosol mass, but reflect differences in how the models simulate evaporation of aerosols at the top of the aerosol layer. Overall, the simulation of effective radius is quite good between 5 and 30 km. Number density simulations are worse than with the 3-mode models, with a 40% overprediction in the tropical upper troposphere and differences up of 60–80% at 30 km in high latitudes.

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Table 2 gives the global aerosol burdens of the five simulations discussed, along with tropospheric and stratospheric burdens. The AER150 model predicts 165 kilotons of aerosol sulfur in the troposphere and 138 kilotons of sulfur in the stratosphere. The AER40 model predicts 1% less in the troposphere and 6% less in the stratosphere. The modal models all predict 162 kilotons of sulfur in the troposphere, but stratospheric burdens range from 130 to 146 kilotons. The UMAer-4m model predicts 141 kilotons of sulfur in the stratosphere, only 2% higher than the AER150 model. The UMAer-3mA model predicts 6% low and the UMAer-3mB model 6% high. Table 2 also shows a breakdown in stratospheric aerosol between that above and below 25 km. The AER150 model produces a stratospheric aerosol mass distribution with 16% above 25 km. The AER40, UMAer-3mB and UMAer-4m models have 15% of the stratospheric sulfur above 25 km, and the UMAer-3mA only 12%. Though the global aerosol mass in the UMAer-4m model is closer to the AER150 model than is AER40, the AER40 model has smaller deviations from AER150 at most latitudes and altitudes (see Figs. 3 and 9) in mass, surface area density, effective radius, and number density. We find that a 4-mode model in general does a better job than a 3-mode model, but in some situations a 3-mode model simulates the size distributions better.

5 Volcanic perturbation intercomparison

We have simulated the Mt. Pinatubo volcanic eruption in the Phillipines in order to compare the different model formulations under very high aerosol loading and to compare the rates of aerosol decay. Our simulations are performed by injecting 20 megatons of SO₂ (Bluth et al., 1992; McCormick et al., 1995) on 15 June 1991 into the tropical stratosphere between 5°S and 15°N at 16–29 km altitude (Read et al., 1993). The simulations cover the 10 year period from the beginning of 1991 until the end of 2000. Simulations are performed with the AER40 model, the UMAer-3mA, UMAer-3mB, and UMAer-4m models. The AER40 model is used here as the benchmark since running the AER150 model for 10 years is not practical.

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Figure 10 show model results of the evolution of aerosol mass density at 20 and 26 km at the equator. All models yield similar peak aerosol mass densities 2–3 months after the eruption. This similarity is not unexpected, as the chemical transformation of SO_2 to H_2SO_4 is independent of the microphysical scheme, and thermodynamics dictate that all gas phase H_2SO_4 in the lower and middle stratosphere will condense into particles, though the condensation rate depends somewhat on the existing particle size distribution. At the equator and 26 km, the UMaer-3mA model calculates a peak mass density 20% less than the AER40 model. The UMaer-3mB model calculated a peak mass density 10% less than AER40. The UMaer-4m model produces the same maximum aerosol density as the AER40 model. The UMaer-3mA model simulation produces aerosol mass decay at a much faster rate than any of the other models. At 26 km, the UMaer-3mB model also decays more rapidly than UMaer-4m or AER40. Figure 11 shows the evolution of surface area density at the same locations. The surface area density shows less variability between models than mass density. The surface area density maximums are very similar between all 4 models. Between 1992 and 1995, the UMaer-3mB, UMaer-4m and AER40 models calculate very similar results, with the UMaer-3mA model noticeably lower. After 1995, the AER40 model calculates lower surface area densities than do any of the UMaer models.

The rate at which the particulate matter is removed from the stratosphere is a function of sedimentation rate, which depends on the amount of aerosol mass in larger particles. Thus the different microphysical schemes calculate somewhat different rates of aerosol decay following this simulated volcanic injection. Models UMaer-3mA and UMaer-3mB differ only in the width of the largest lognormal mode, and show aerosol decay at very different rates, confirming the importance of the large particle distribution to sedimentation rates. The decay rate for UMaer-3mB is only slightly faster than for AER40, but the UMaer-3mA model decays much too fast. The UMaer-4m model gives decay rates close to the AER40 model for the 1992–1995 period.

The time evolution of aerosol effective radius is shown in Fig. 12. This parameter is sensitive to the full particle size spectrum, unlike mass density which depends mostly

on the larger particles which contain the majority of the aerosol mass. The effect of a large increase in gas phase sulfur in the stratosphere is to produce a burst of nucleation, seen here as a drop in effective radius at the time of the eruption, and then to increase particle sizes over several months as the vast majority of sulfur condenses onto existing particles and increases their diameters. As shown in Fig. 13, the AER40 model calculates a narrower size distribution than the modal models in the post-Pinatubo period. The modal models have aerosol mass mainly in the largest mode whose width is specified and wider than that calculated by AER40. The AER40 size distributions are not symmetrical in January 1992 or January 1994, peaking at 0.4–0.5 μm at 20 or 26 km at the equator and dropping faster on the high radius side than on the low radius side. The modal models generate far more particles below 0.1 μm radius, and thus produce a smaller effective radius. Effective radius at the equator and 20 km is up to 40% low in UMAer-3mA relative to AER40 in 1994 and 1995, and falls back to background levels 2 years sooner than AER40. UMAer-3mB does a somewhat better jobs than UMAer-3mA but still calculates much lower effective radii. The UMAer-4m model does a good job of simulating effective radius at 20 km, but not at 26 km.

The SAGE II satellite instrument measured aerosol extinction at several wavelengths during the eruption and subsequent decay of aerosol from Mt. Pinatubo. The SAGE II (version 6.1) 1.02 μm extinction observations are compared with model results in Fig. 14. At the equator and 20 km, the AER40 model and the UMAer-3mB and UMAer-4m models match the observations well. The UMAer-3mA model results are much too low between 1993 and 1996. At the equator and 26 km, the AER40 model calculated higher extinctions than observed between mid 1992 and the end of 1993, but comes close to matching observations after 1993. The discrepancy could be caused by an inaccurate initial representation of the distribution of the SO_2 cloud or by model transport not representative of the particular year. The UMAer-3mA model is again well below observations. The UMAer-3mB model is closest to observations between 1992 and the end of 1993, while the UMAer-4m model lies between the UMAer-3mB and AER40 models. Observations show that the UMAer-3mA model with its wide distribu-

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tion in the largest mode produces too much sedimentation and too rapid removal of volcanic aerosol. The AER40, UMaer-3mB, and UMaer-4m models all appear to adequately represent aerosol decay rates for $1.02\ \mu\text{m}$ extinction when volcanic loading is high (1992–1996). When aerosol loading approaches background levels (1997–2001), the UMaer-3mB and UMaer-4m models produce $1.02\ \mu\text{m}$ extinction levels somewhat above observations. The $1.02\ \mu\text{m}$ extinction is not very sensitive to aerosol particles smaller than about $0.1\ \mu\text{m}$, so provides little information to validate the small end of the size distribution under low aerosol loading conditions.

6 Conclusions

We have performed global 2-D model calculations with two versions of a sectional model and three versions of a modal model. The sectional model with 40 bins has noticeable numerical diffusion compared to the sectional model with 150 bins, resulting in greater sedimentation and 6% less stratospheric aerosol mass, with maximum differences of 15% at 30 km. We tested two three-mode model versions and found a 12% difference in stratospheric aerosol mass as a function of the prescribed width of the largest lognormal mode. Differences in aerosol mass between these models and the 150-bin sectional model are as high as 50% at 30 km. A four-mode version was found to perform better than the three-mode version under some, but not all, conditions. Effective radius was more sensitive to model formulation than mass density or surface area density.

Our 10 year calculations of the Pinatubo eruption period have been compared with SAGE-II observations of aerosol extinction at $1.02\ \mu\text{m}$ and show that the UMaer-3mA version indeed has sedimentation which removes aerosol mass too quickly. The AER40, UMaer-3mB and UMaer-4m versions are all consistent with observations between 1992 and 1996, but the AER40 version better matches observations after 1996 when background aerosol levels are approached. Calculated effective radius shows the clearest distinction between model versions during the 1992–1996 period.

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Based on the model performances documented here and the computational efficiency, we recommend that the AER40 and UMaer-3mB model versions should be incorporated into the GMI stratosphere-troposphere chemistry-transport model. Developing a modal model which can prognostically determine mode width would likely be more efficient computationally and at least as accurate as a 4-mode scheme without this feature.

Acknowledgements. This work was supported by the NASA Atmospheric Chemistry Modeling and Analysis Program and the NASA Modeling Analysis and Prediction program.

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Table 1. Model versions used in 2-D intercomparison study.

Module	Label	Bin Ratios by Volume	Time Step
AER 150 bins	AER150	$V_{\text{rat}}=1.2$	15 min
AER 40 bins	AER40	$V_{\text{rat}}=2.0$	1 h
Module	Label	Distribution Width σ_g	Merge Radii
UMaer 3 modes	UMaer-3mA	1.2/1.514/1.77623	0.005/0.05
	UMaer-3mB	1.2/1.514/1.6	0.005/0.05
UMaer 4 modes	UMaer-4m	1.3/1.6/1.6/1.45	0.001/0.01/0.1

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Table 2. Aerosol burdens in kilotons of sulfur calculated by each model version.

Model Version	Global Burden	Trop Burden	Strat Burden	Strat < 25 km	Strat > 25 km
AER150	303	165	138	116	22
AER40	294	164	130	110	20
UMaer-3mA	292	162	130	114	16
UMaer-3mB	308	162	146	124	22
UMaer-4m	303	162	141	120	21

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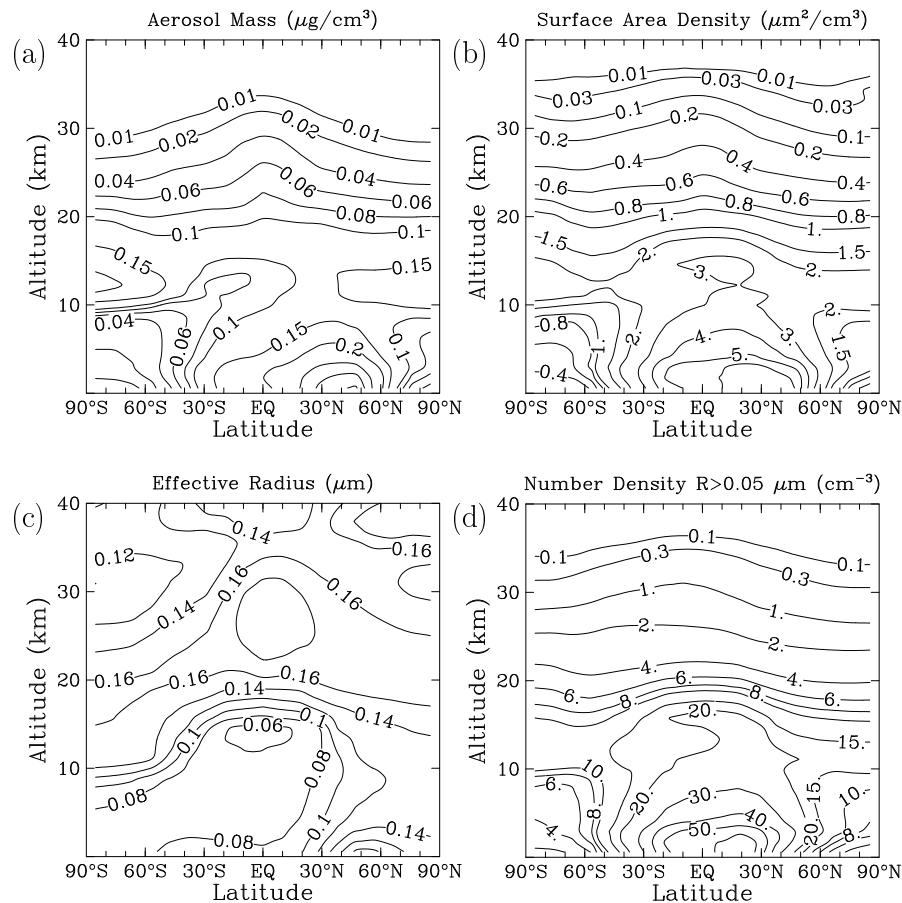


Fig. 1. Model calculated aerosol parameters from the AER 2-D model using the 150-bin sectional aerosol module AER150. Shown are annual average (a) mass density in $\mu\text{g}/\text{cm}^3$, (b) surface area density in $\mu\text{m}^2/\text{cm}^3$, (c) effective radius in μm , and (d) number density of particles with radius greater than $0.05 \mu\text{m}$ in cm^{-3} . 12754

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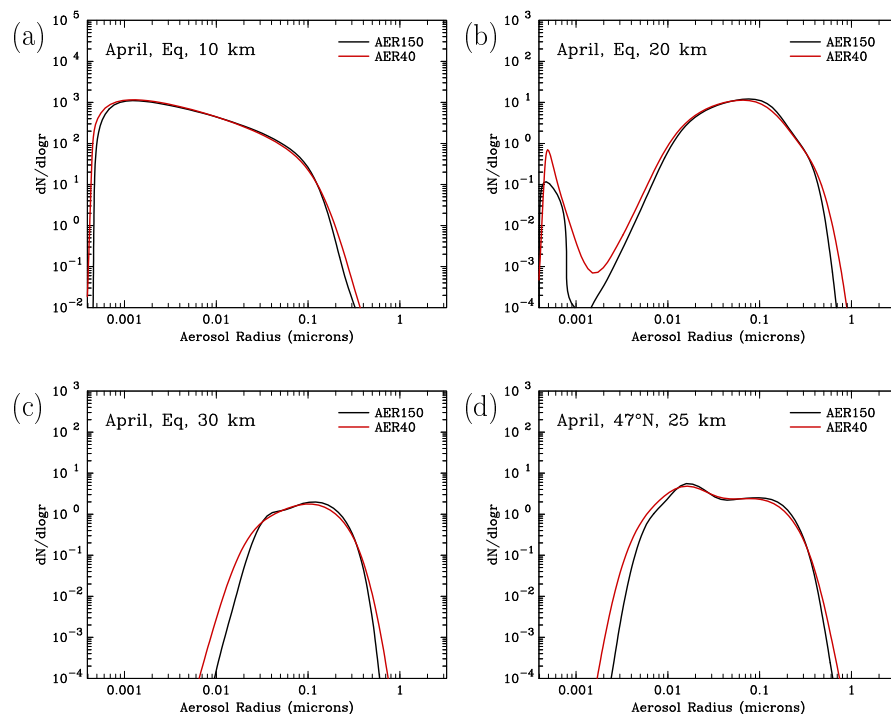


Fig. 2. Calculated size distributions from the AER 2-D model using the 150-bin sectional aerosol module AER150 (black lines) and the 40-bin sectional aerosol module AER40 (red lines) in April at **(a)** the equator and 10 km, **(b)** the equator and 20 km, **(c)** the equator and 30 km, and **(d)** 47° N and 25 km.

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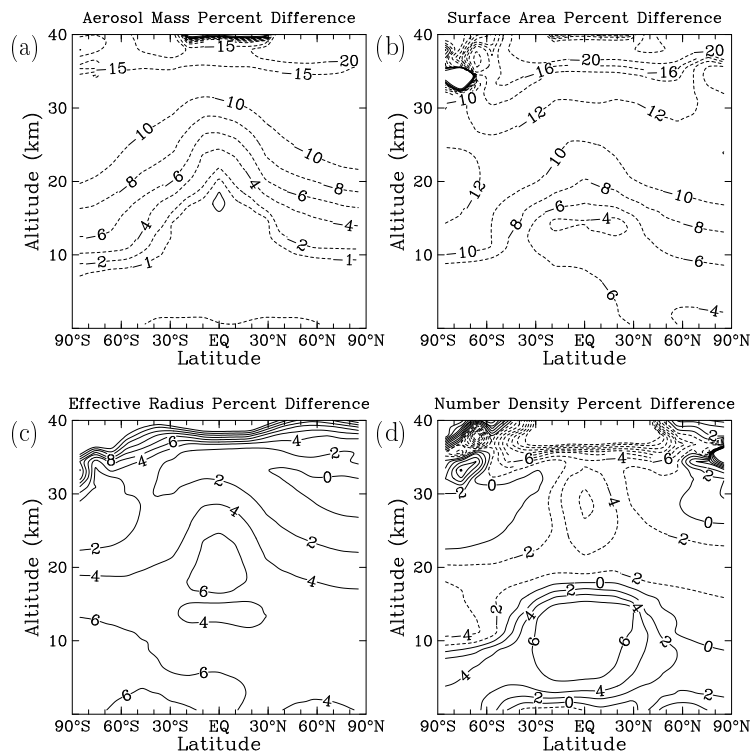


Fig. 3. Percent change in model-calculated aerosol parameters from the AER 2-D model using the 40-bin sectional aerosol module AER40 versus the 150-bin sectional aerosol module AER150. Shown are annual average differences in **(a)** mass density, **(b)** surface area density, **(c)** effective radius, and **(d)** number density of particles with radius greater than $0.05 \mu\text{m}$.

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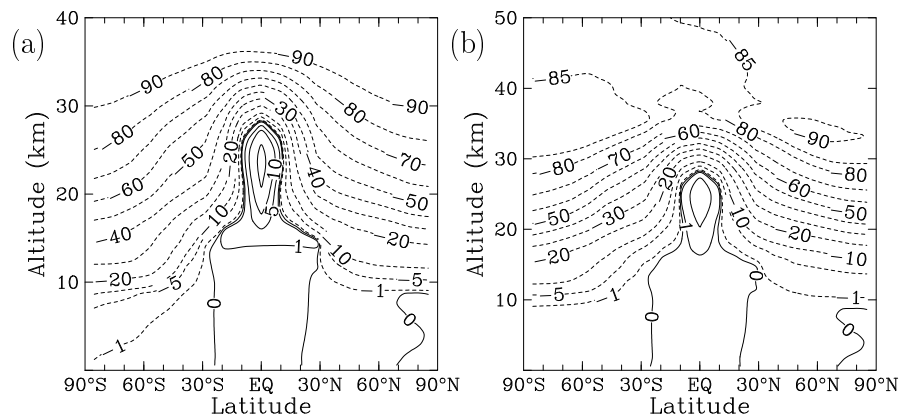


Fig. 4. Percent change in annual average model-calculated **(a)** aerosol mass density and **(b)** total sulfur of the AER40 model with sedimentation versus without sedimentation.

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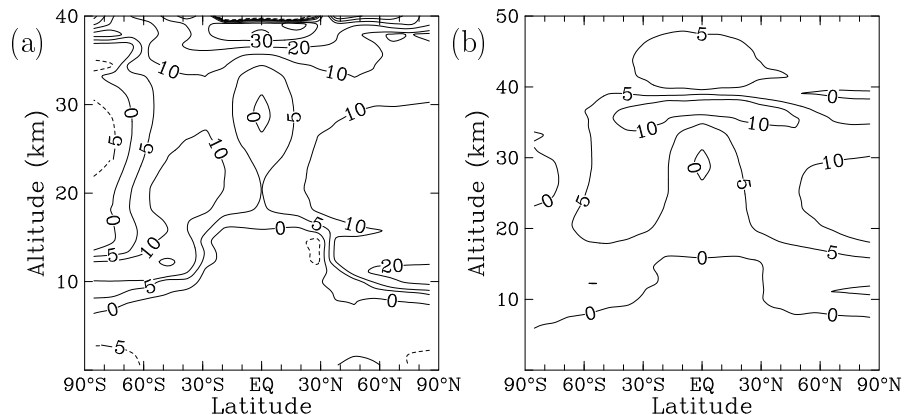


Fig. 5. Percent change in annual average model-calculated **(a)** aerosol mass density and **(b)** total sulfur between the UMAer-3mA model without sedimentation and the AER40 model without sedimentation.

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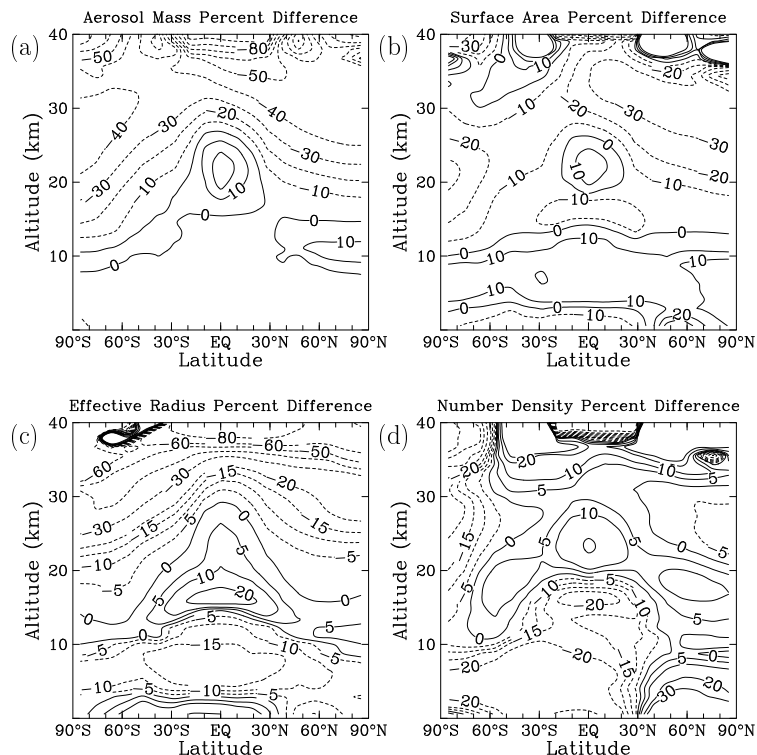


Fig. 6. Percent change in model-calculated annual average aerosol parameters from the AER 2-D model using the UMaer 3-mode aerosol module UMaer-3mA versus the 150-bin sectional aerosol module. Shown are annual average differences in **(a)** mass density, **(b)** surface area density, **(c)** effective radius, and **(d)** number density of particles with radius greater than $0.05 \mu\text{m}$.

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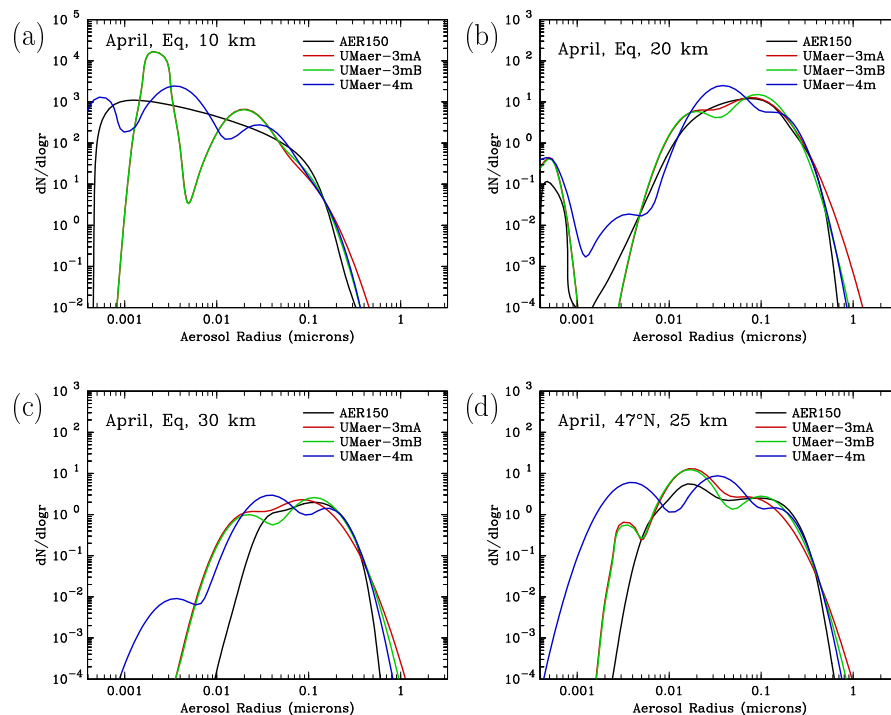


Fig. 7. Calculated size distributions from the AER 2-D model using the 150-bin sectional aerosol module (black lines), the 3-mode UMAer-3mA aerosol module (red lines), the 3-mode UMAer-3mB aerosol module (green lines), and the 4-mode UMAer-4m aerosol module (blue lines) in April at **(a)** the equator and 10 km, **(b)** the equator and 20 km, **(c)** the equator and 30 km, and **(d)** 47° N and 25 km.

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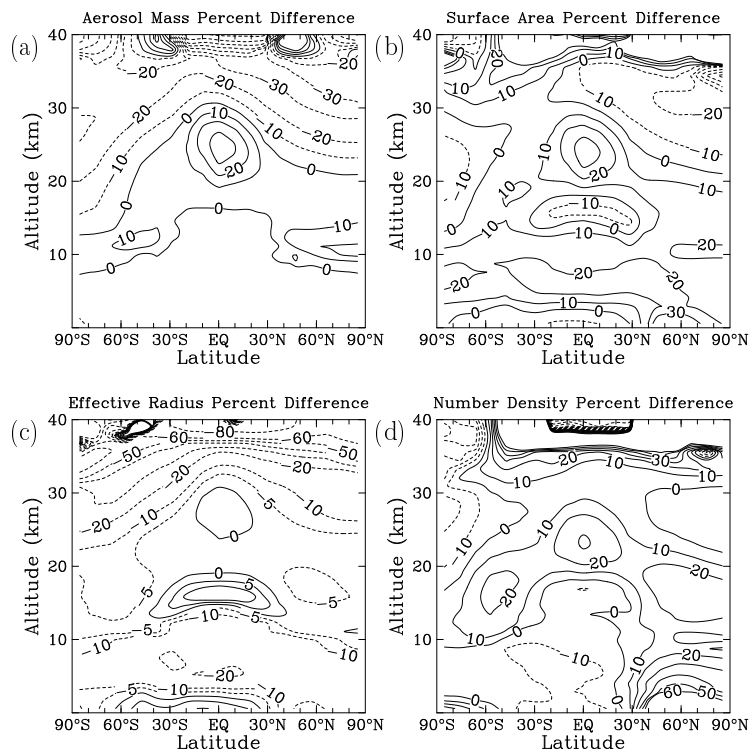


Fig. 8. Percent change in model-calculated annual average aerosol parameters from the AER 2-D model using the UMaer 3-mode aerosol module UMaer-3mB versus the 150-bin sectional aerosol module. Shown are annual average differences in **(a)** mass density, **(b)** surface area density, **(c)** effective radius, and **(d)** number density of particles with radius greater than $0.05 \mu\text{m}$.

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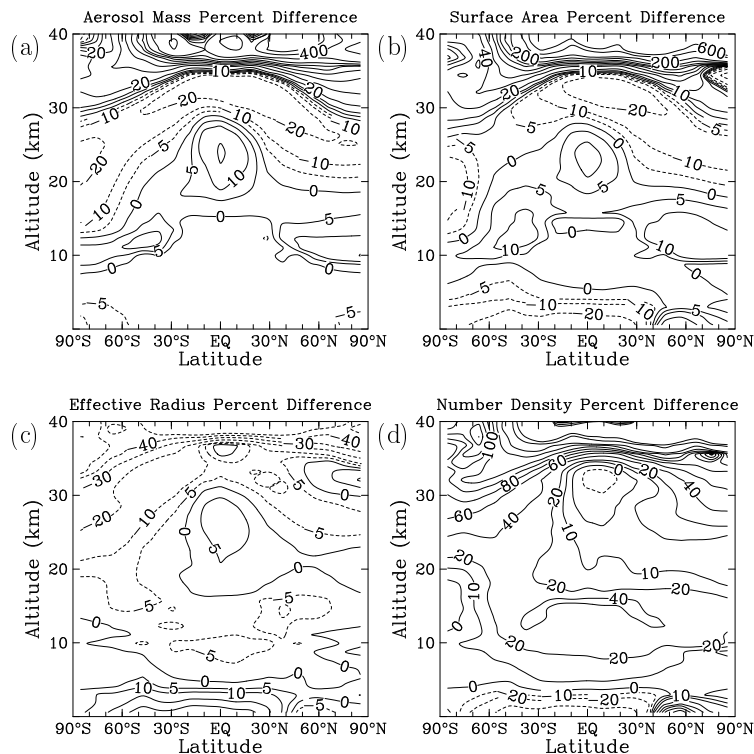


Fig. 9. Percent change in model-calculated annual average aerosol parameters from the AER 2-D model using the UMaer 4-mode aerosol module UMaer-4m versus the 150-bin sectional aerosol module. Shown are annual average differences in **(a)** mass density, **(b)** surface area density, **(c)** effective radius, and **(d)** number density of particles with radius greater than $0.05 \mu\text{m}$.

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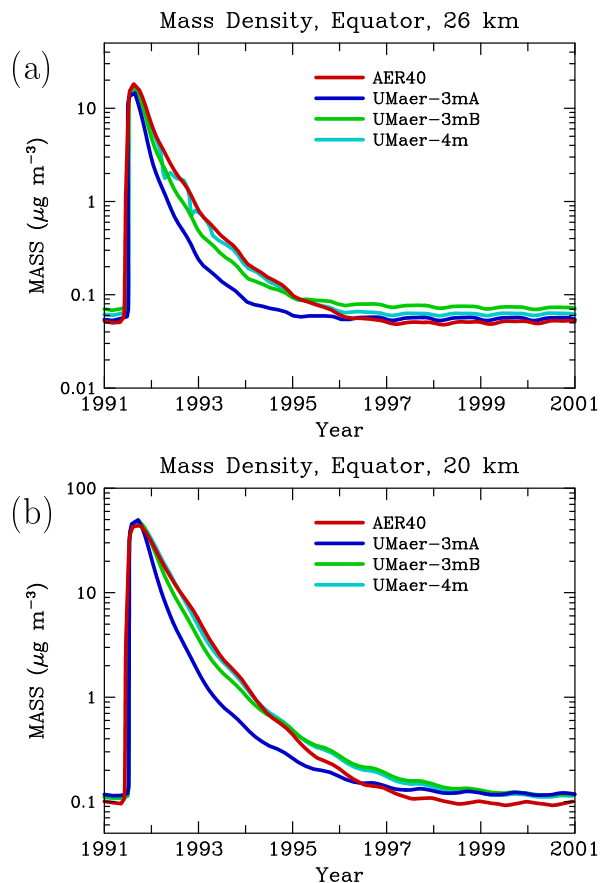


Fig. 10. Aerosol mass density in $\mu\text{g m}^{-3}$ for 1991 to 2002 at the (a) equator and 26 km, and (b) equator and 20 km from simulations with the AER40, UMAer-3mA, UMAer-3mB, and UMAer-4m models.

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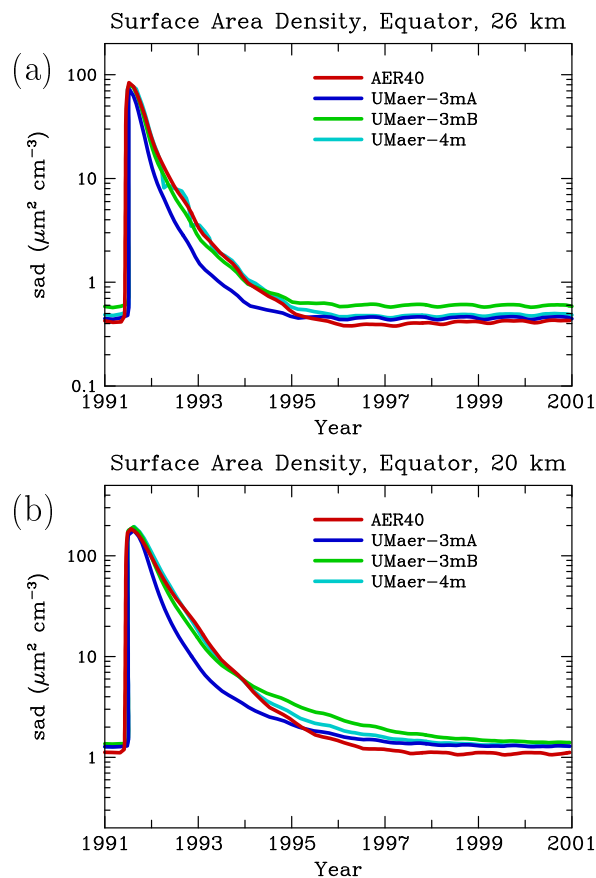


Fig. 11. Aerosol surface area density in $\mu\text{m}^2/\text{cm}^3$ for 1991 to 2002 at the (a) equator and 26 km, and (b) equator and 20 km from simulations with the AER40, UMaer-3mA, UMaer-3mB, and UMaer-4m models.

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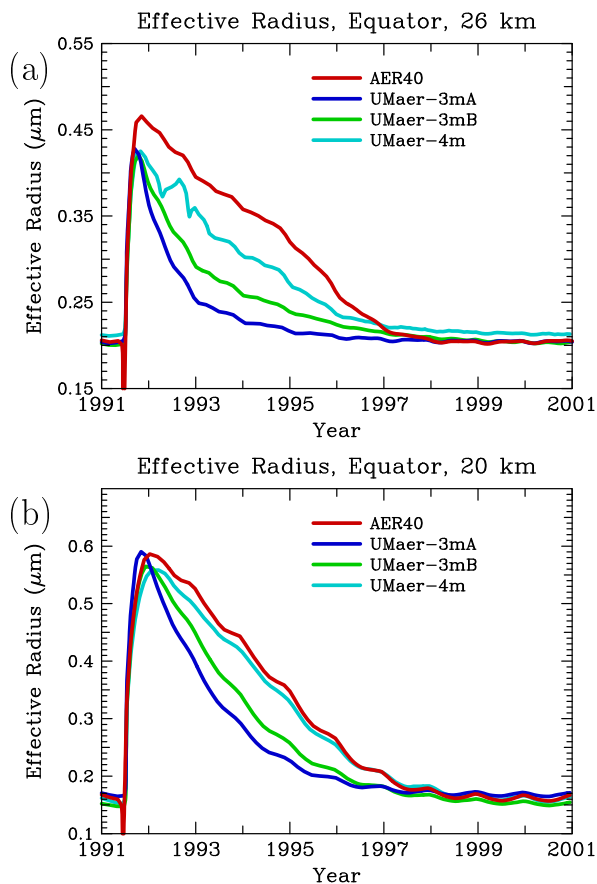


Fig. 12. Aerosol effective radius in μm for 1991 to 2002 at the **(a)** equator and 26 km, and **(b)** equator and 20 km from simulations with the AER40, UMAer-3mA, UMAer-3mB, and UMAer-4m models.

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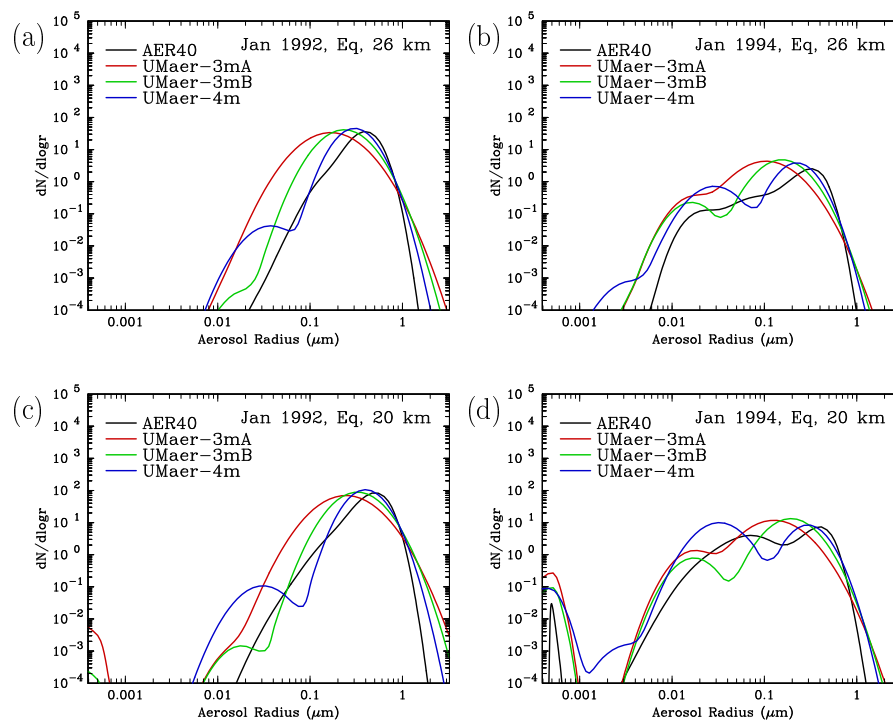


Fig. 13. Calculated size distributions from a simulation of the Mt. Pinatubo eruption using the AER 2-D model using the 40-bin sectional aerosol module AER40 (red lines), the UMAer-3mA module (blue lines), the UMAer-3mB module (green lines) and the UMAer-4m module (cyan lines) at **(a)** the equator and 26 km in January 1992, **(b)** the equator and 26 km in January 1994, **(c)** the equator and 20 km in January 1992, and **(d)** the equator and 20 km in January 1994.

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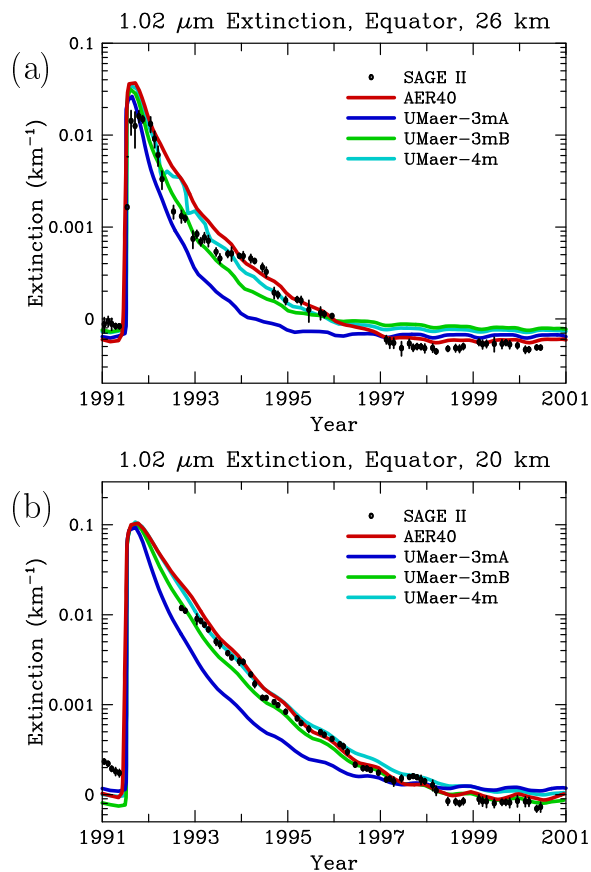


Fig. 14. Aerosol extinction at $1.02 \mu\text{m}$ for 1991 to 2002 at the (a) equator and 26 km, and (b) equator and 20 km. SAGE II data version 6.1 are shown by black symbols with error bars, model results by colored lines.

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