A model study on the decay of volcanic aerosol layer and verification with Pinatubo and El Chichon data

S RAMACHANDRAN, A JAYARAMAN and B R SITARAM

Physical Research Laboratory, Ahmedabad 380 009, India

The time evolution of stratospheric aerosol layer formed after a volcanic eruption is studied taking into account the aerosol microphysical processes of growth, coagulation and sedimentation. Using a simple model we could explain the observed evolution of the Pinatubo volcanic layer which decayed in about 3 years. The experimental data obtained by Nd:YAG backscatter lidar over Ahmedabad further supports this finding. The data obtained after the El Chichon volcanic eruption also showed that the El Chichon aerosol layer decayed in about 3 years time. Thus, though the amount of SO_2 injected has been higher, in the case of Pinatubo, about two to three times more than El Chichon, it has resulted in the production of larger aerosol particles due to faster growth and coagulation processes, and subsequently a faster removal rate, to give more or less a similar background aerosol amount at the stratosphere in about 3 years time.

1. Introduction

Volcanic eruptions give an opportunity to study the various physico-chemical processes involved in the formation and evolution of stratospheric aerosol layer and predict the impending climatic effects. The latest major eruption is of the Mount Pinatubo in Philippines (15.14°N, 120.35°E) in June 1991. It had put as much as two to three times more material (about 15 to 30 megatons of SO_2 (McPeters 1993; Bluth *et al* 1992)) into the stratosphere compared to the El Chichon eruption in Mexico in 1982, which resulted in a global stratospheric aerosol mass load of about 20–30 Mt (McCormick and Veiga 1992). Also for the few months after the eruption the bulk of the material was found confined over the tropics (Stowe *et al* 1992; Trepte *et al* 1993) from the tropopause level to about 28 km (DeFoor *et al* 1992).

Two balloon experiments were conducted from Hyderabad (17.5°N, 78.6°E), India one in October 1991 and the other in April 1992, 4 and 10 months after the eruption, respectively. The balloons carried identical payloads of multiwavelength filter photometers to obtain the vertical profiles of aerosol extinction as well as the aerosol number density and size distribution parameter from the direct and scattered radiation intensity measurements. The Pinatubo volcanic aerosol layer was also continuously monitored by a Nd:YAG backscatter lidar operating at 532 nm over Ahmedabad (23°N, 72.5°E), India since April 1992.

Earlier results using balloon-borne Sun-tracking photometers for the El Chichon layer over Hyderabad, showed that the layer produced by the eruption decayed in about 3 years (Jayaraman 1991). Since Mt. Pinatubo had injected as much as two to three times more material compared to El Chichon, it was not clear whether the lifetime of these aerosols will be longer because of the larger number density of aerosols, or, perhaps, the aerosol microphysical processes such as growth, coagulation and sedimentation will take place at a much faster rate to give more or less the same decay time and whether the residual aerosol masses, would be similar, as shown through model calculations (Pinto *et al* 1989). To understand better, the relative roles of these processes in influencing the formation and decay of the stratospheric aerosol layer after major volcanic eruptions, a simple model was developed to study the time evolution of a volcanic aerosol layer (17–30 km) and aerosol size distribution using the microphysical processes of growth, coagulation and sedimentation.

Keywords. Pinatubo; stratospheric aerosols; volcanic aerosol layer; optical depth; evolution.

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Though this paper is aimed at explaining the model developed and the results obtained, for the sake of completion a brief summary of the results obtained from the balloon and lidar experiments, is given in the next two subsections.

1.1 *Balloon-borne optical studies of the Pinatubo aerosol layer*

Two balloon experiments, which carried identical payloads of multiwavelength Sun-tracking and Sun-scanning filter photometers were conducted on 26th October 1991 and 20th April 1992. The instrument essentially consists of a sensor assembly containing filter photometers, a Sun-tracking mechanism, and a motor assembly for scanning the sky along the solar almucantar, ±90° with respect to the Sun for scattered radiation measurements. The measurements were made in eight spectral bands centered around 280, 310, 440, 500, 750, 850, 950, and 1050 nm, each having a typical bandwidth of about 10 nm. The data analysis mainly involves the estimation of the attenuation of the incoming solar radiation to determine the aerosol extinction coefficients and deriving the angular distribution obtained from the scattered radiation intensity measurements to determine the aerosol size distribution parameter ν (slope of the Junge power law) and the aerosol number density (Ramachandran *et al* 1994a). The important results obtained from these two balloon measurements are summarized here: The aerosol extinction coefficients measured during October 1991 and April 1992 are found to be the highest ever obtained, over a decade of stratospheric aerosol measurements over Hyderabad (Jayaraman 1991), with a distinct aerosol layer between 16 and 30 km (figure 1a). The aerosol extinction coefficients obtained in 1991 and 1992 at all wavelengths are about 2 orders higher in magnitude at the peak altitude when compared with that of 1985 (background, volcanically quiescent) values over Hyderabad, but only the aerosol extinctions obtained at 500 nm are given, as an example. The recent results showing the highest values ever obtained over the same site indicate the magnitude of Pinatubo eruption which is also the strongest eruption in this century. The aerosol layer peak occurs in October 1991 at 23 km with about 40 particles per cm^3 (figure 1b). In April 1992, 10 months after the eruption, the aerosol number densities have shown a decline, with a peak aerosol number density of about 20 particles per cm^3 at an altitude of about 20 km. When compared to the volcanically quiescent 1985 values, the aerosol number densities obtained during 1991 and 1992, are about an order of magnitude higher, at the peak altitudes. More details on the instrument and results can be obtained from Ramachandran *et al* (1994 a and b).

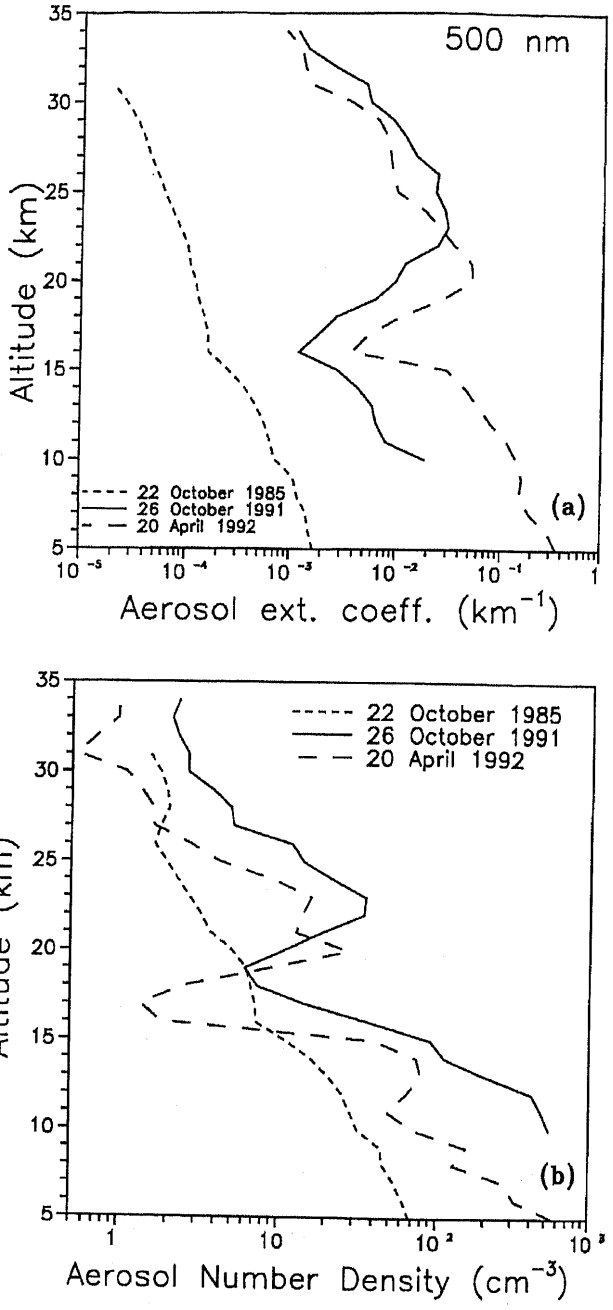

Figure 1(a and b). Profiles of aerosol extinction at 500 nm and number density obtained over Hyderabad (17.5°N, 78.6°E) during the background (nonvolcanic) conditions (22nd October 1985) and the post-Pinatubo phase (26th October 1991 and 20th April 1992).

1.2 *Nd: YAG backscatter lidar measurements*

A newly set up Nd:YAG backscatter lidar system (Jayaraman *et al* 1995a) was employed to study the Pinatubo volcanic aerosol layer over Ahmedabad (23°N), from April 1992 to May 1994. The system employs a transmitter consisting of a pulsed Nd:YAG laser, operating at its second harmonic of 532 nm, a 40 cm Cassegrain telescope which acts as the receiver, a thermoelectrically cooled photomultiplier operating in the photon counting mode, signal processing unit

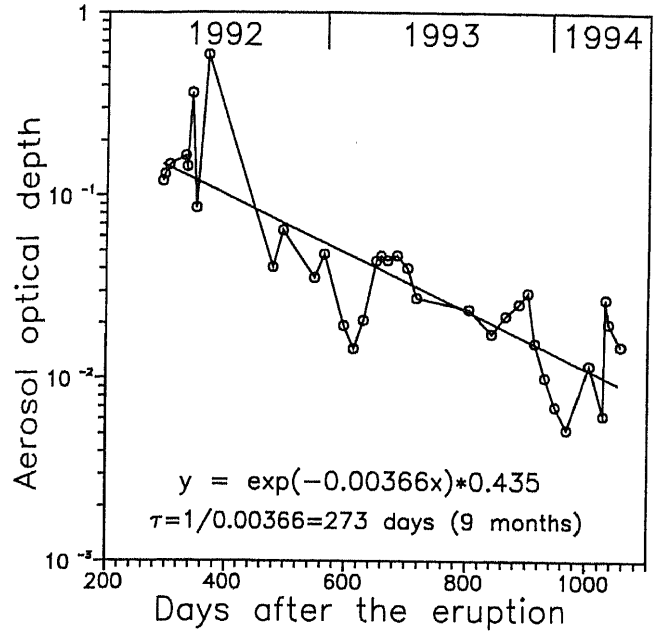

Figure 2. Time evolution of Pinatubo aerosol optical depth (integrated from 17 to 30 km) from April 1992 to May 1994 as obtained from the lidar measurements made over Ahmedabad (23°N).

and data processor. An interference filter having a central bandwidth at 532 nm with a bandwidth of 1 nm is used to reduce the background light noise in the collected backscattered signal. The backscattered signals are then analysed using the top to bottom inversion algorithm proposed by Klett (1985) to determine the aerosol backscattering coefficients and scattering ratios (Jayaraman *et al* 1995b). The obtained aerosol backscatter values are then multiplied by an altitude independent value of 50 sr (Jäger *et al* 1988) to determine the aerosol extinction coefficients which are then integrated to determine the optical depth of the Pinatubo aerosol layer. Lidar measurements are made during clear nights. Each lidar profile is obtained by averaging 5000 individual laser firings made within 500 seconds. In a night about 4 to 5 such observations are made and the results of which are used in the present study. The backscattering ratio profiles obtained from April 1992 till May 1994, a period corresponding to about 10 months after the eruption to 3 years, indicate a dense Pinatubo volcanic aerosol layer between 17 and 30 km, in the initial period. The layer peaks initially at 23 km during April 1992 and by May 1994 the peak has come down to about 20 km. Also the peak scattering ratio which is about 8 in April 1992, increases to about 22 in May 1992 and then subsequently decreases to 3 in October 1992. During the rest of the observation period though the Pinatubo layer is seen, the broad layer which existed between 17 and 30 km in the initial stages, has in May 1994 reduced to a narrow region of 17 and 24 km, with a subsequent decline in the scattering ratio to 1.5, at peak altitudes. An exponential fit to the aerosol optical depths obtained between 17 and 30 km, gives a 1/e-folding time of 9 months, for the Pinatubo layer to decay (figure 2).

2. A time dependent stratospheric aerosol layer model

2.1 *Aerosol microphysical processes responsible for the formation and decay of stratospheric aerosol layer*

The aerosol continuity equation can be written as

$$\frac{\partial n}{\partial t} = \frac{\partial n}{\partial t}\Big)_{\text{nuc}} + \frac{\partial n}{\partial t}\Big)_{\text{growth, evap.}} + \frac{\partial n}{\partial t}\Big)_{\text{coa.}} + \frac{\partial n}{\partial t}\Big)_{\text{sed.}} + \frac{\partial n}{\partial t}\Big)_{\text{diff.}} + \frac{\partial n}{\partial t}\Big)_{\text{washout}} \quad (1)$$

where the various terms refer to the rate of change of aerosol concentration due to processes such as nucleation, growth or evaporation, coagulation, sedimentation, diffusion and washout.

Nucleation refers to the formation of a new stable solid or liquid particle from a gas phase consisting of either a gaseous species only (homogeneous, homomolecular nucleation) or involving two or more gaseous species, one of which most commonly is water (homogeneous, heteromolecular nucleation) or when two or more gaseous species condense onto pre-existing particles (heterogeneous, heteromolecular nucleation) (Hamill *et al* 1977a). The nucleation lifetime is of the order of a few days (Turco *et al* 1979a). Immediately after the El Chichon as well as Mt. Pinatubo eruptions, high concentrations of aerosol droplets were observed suggesting that homogeneous nucleation is the most likely production mechanism of aerosols at the stratospheric altitudes (Hofmann and Rosen 1983; Deshler *et al* 1992).

After nucleation the sulphuric acid-water solution droplets can grow by heteromolecular condensation of H_2SO_4 and H_2O vapours. At a given temperature, the water vapour equilibrium uniquely determines the weight percentage of H_2SO_4 in the solution and consequently the H_2SO_4 vapour pressure over the droplet. Accordingly, on the basis of H_2SO_4 vapour pressure, some of the sulphuric acid molecules incident on the droplet are absorbed or could be evaporated from the existing droplet (Turco *et al* 1979a).

Under stratospheric conditions, Brownian coagulation, which occurs when particles impinge during random thermal motions is important (Turco *et al* 1982). The rate of change of concentration of particles of radius r due to coagulation can be expressed as the difference between two integrals over the particle size distribution: one integral represents the production rate of particles of size r by the coagulation of two smaller particles and the second integral represents the loss rate of particles of size r by coagulation with all other particles. The coagulation integrals are

calculated from Turco *et al* (1979b). The coagulation kernels for each pair of particles, is calculated based on Fuchs (1964) and Hamill *et al* (1977b). In our model, we have adopted a discrete formulation of the particle size distribution. In this formulation, coagulation is considered as an interaction process between pairs of particles whose sizes are limited to a finite set of discrete values. The continuous particle size distribution also yields the same results for the average rates of particle coagulation, but the discrete formulation is much simpler to use (Turco *et al* 1979b).

The removal mechanism involves horizontal dilution due to diffusion of aerosols to other latitudes, vertical diffusion and gravitational sedimentation in which particles fall with respect to the air surrounding them and are returned to the troposphere. When the aerosols grow to a few micron size, gravitational sedimentation becomes important. The settling velocity v_s, for particle of radius r is defined as,

$$v_s = \frac{2\rho r^2 g}{9\eta}[1 + (\lambda/r)(A + B\exp\{-Cr/\lambda\})], \qquad (2)$$

where ρ is the mass density ($1.65\,\mathrm{g\,cm^{-3}}$ for 75% H_2SO_4 droplets), g is the gravitational acceleration, λ is the mean free path, η is the viscosity of air and A, B and C are constants whose values are 1.249, 0.42 and 0.87, respectively (Kasten 1968). Using the above equation it is estimated that a particle of radius 1 μm will fall from 20 to 10 km in less than a year but a 0.1 μm particle will take about 10 years to fall the same distance. Therefore gravitational sedimentation is a significant removal mechanism for particles with radius $\geq 1\,\mu$m and this accounts for the fact that the fraction of particles $\geq 1\,\mu$m is smaller in the stratosphere than in the troposphere. Gravitational sedimentation is not a significant removal process for particles in the size range $\leq 0.1\,\mu$m (Hamill *et al* 1977b).

2.2 *Model specifications*

As homogeneous-heteromolecular nucleation and heterogeneous-heteromolecular nucleation are the most important production mechanisms of aerosols only in the initial stages of volcanic eruption and also as the nucleation life time is of the order of a day (the process is almost instantaneous), the process of nucleation is not included in the model. Hamill *et al* (1977a) showed through model calculations that the heterogeneous-heteromolecular nucleation rate is generally many orders of magnitude higher than the nucleation rate for any competing process. An analysis of the volcanic particles suggested that large numbers of sulphate particles have been formed by homogeneous nucleation in the early stages of Pinatubo volcanic eruption (Sheridan *et al* 1992; Deshler *et al* 1992). Recent model studies of Pinatubo aerosol layer evolution by Zhao *et al* (1995) showed that the new particle formation by homogeneous nucleation almost ceases in about a month after eruption. The enormous amount of SO_2 injected gets rapidly converted into H_2SO_4 in about 30 days, which is the chemical conversion time and the H_2SO_4 vapour thus formed build up very quickly to levels sufficient for homogeneous nucleation to occur. This chemical oxidation of SO_2 into H_2SO_4 is crucial for aerosol formation and evolution and the conversion rate is controlled by OH concentration. The H_2SO_4 vapour thus formed results in the nucleation of new particles followed by co-condensation with water to form the observed aerosol. Winker and Osborn (1992) estimate that one month after the eruption of Mount Pinatubo, about half the SO_2 had been converted into sulphate aerosols. Both experimental (TOMS (Bluth *et al* 1992) and MLS (Read *et al* 1993)) observations and model calculations (Bekki and Pyle 1994) show that more than 90% of the conversion of SO_2 to sulphate aerosols was complete within 3 months after the Pinatubo eruption. In the case of El Chichon, the characteristic aerosol growth time was about 45 days (Hofmann 1987).

Hence, in these calculations an aerosol size distribution is chosen which starts from $t + 45$ days and not at $t = 0$, where t is the day of eruption. Also, the process of aerosol evaporation due to the negative growth rate is not included. Average growth rates of aerosols for the altitude region of 20–30 km are taken from Hofmann and Rosen (1984) for a sulphuric acid concentration of $10^7\,\mathrm{cm^{-3}}$ (estimated after 40 days of El Chichon eruption) and a water vapour mixing ratio of 3 ppmv. In the absence of growth rate data as a function of time, an exponential decay curve of H_2SO_4 vapour was drawn assuming an initial concentration of $10^7\,\mathrm{cm^{-3}}$ (45 days) and a background concentration of $10^4\,\mathrm{cm^{-3}}$ at about 3 years, in the 20–30 km altitude region (Turco *et al* 1982). Growth rates are then calculated at every 100 day time interval and then are given as inputs in the calculation.

In general, the sedimentation flux is calculated as

$$\left(\frac{\partial n}{\partial t}\right)_{\text{sed.}} = -\frac{\partial}{\partial z}(-v_s n), \qquad (3)$$

where v_s is the settling velocity ($\mathrm{cm\,s^{-1}}$) and n is the aerosol number density ($\mathrm{cm^{-3}}$).

Assuming v_s is uniform within the aerosol layer considered (17 to 30 km) and particles are uniformly distributed in z, then, in time Δt, all particles lying in the height region 17 km to 17 km $+v_s\Delta t$ will sediment, which is given by $v_s\Delta t\, n/13$, where 13 is the vertical extent of the layer. Thus the sedimentation flux can be expressed as,

$$\left(\frac{\partial n}{\partial t}\right)_{\text{sed.}} = \frac{(-v_s n)}{13}. \qquad (4)$$

In the model, the coagulation kernels are calculated for 23.5 km (middle of the layer) with a temperature 218°K. It is assumed in the model that all the aerosols

settle down from 17 km. However, as only an average number density distribution curve was taken to be a representative of 17–30 km altitude region, we calculated sedimentation velocities at 3 altitudes namely 17 km (bottom), 23.5 km (middle) and 30 km (top of the layer) respectively and we have studied the effects of varying sedimentation velocities on the aerosol optical depths, which are discussed later. The typical settling velocities at 17, 23.5 and 30 km altitudes for a particle of radius 0.5μm are 0.0015, 0.0035, 0.009 cm s^{-1}, respectively and the settling velocities increase with increase in radius and altitude.

A bimodal lognormal aerosol size distribution signifying an eruption of magnitude of El Chichon or Pinatubo with $r_1 = 0.02\,\mu\text{m}$, $\sigma_1 = 2.2$, $N_1 = 5000\,\text{cm}^{-3}$ and $r_2 = 0.7\,\mu\text{m}$, $\sigma_2 = 1.6$, $N_2 = 50\,\text{cm}^{-3}$, after 45 days of eruption, is taken as the initial boundary case (Model 2). Model 1 has a higher aerosol concentration by a factor of 10 when compared to Model 2 and Model 3 is lower by a factor of 10, while the mode radii and sigma are unchanged. This size distribution is taken, based on the optical particle counter data of stratospheric aerosols following El Chichon volcanic eruption by Hofmann (1988) and Mt. Pinatubo eruption by Deshler *et al* (1993). 15 size bins which cover the aerosols in the radius range of 0.01 μm to 15 μm are considered. The initial size distribution ($t + 45$ days) is made to undergo the physical processes of growth, coagulation and sedimentation and the evolution of the final size distribution after $t + 100$ days, $t + 200$ days, $t + 300$ days, $t + 400$ days, $t + 500$ days, $t + 600$ days, $t + 700$ days, $t + 800$ days, $t + 900$ days, $t + 1000$ days and $t + 3$ years is considered for the present study. In the current model no inflow of aerosols from the lower and upper altitudes i.e., from below 17 km and above 30 km is considered and hence an average aerosol size distribution representing the volcanically perturbed case in the stratosphere is taken. The initial size distribution ($t + 45$ days) has apart from the volcanically produced bimodal size distribution, embedded in it the background aerosol size distribution. The aerosol continuity equation in addition to the terms for growth, coagulation and sedimentation, has a contribution of aerosols from the background conditions, which is referred to as C here. Hence the equation becomes

$$\frac{\partial n}{\partial t} = \left.\frac{\partial n}{\partial t}\right)_{\text{growth}} + \left.\frac{\partial n}{\partial t}\right)_{\text{coa.}} + \left.\frac{\partial n}{\partial t}\right)_{\text{sed.}} + C. \tag{5}$$

If the number density n becomes equal to the background aerosol size distribution which is in an equilibrium condition i.e., $n = n_{\text{eq}}$ then

$$\frac{\partial n}{\partial t} = \frac{\partial n_{\text{eq}}}{\partial t}. \tag{6}$$

Therefore the contribution from the background, equilibrium condition C becomes

$$C = \frac{\partial n_{\text{eq}}}{\partial t} - \left.\frac{\partial n_{\text{eq}}}{\partial t}\right)_{\text{growth}} - \left.\frac{\partial n_{\text{eq}}}{\partial t}\right)_{\text{coa.}} - \left.\frac{\partial n_{\text{eq}}}{\partial t}\right)_{\text{sed.}}. \tag{7}$$

This contribution C is subtracted from the volcanic aerosol size distribution at each time step and hence, the volcanic stratospheric aerosol size distribution is corrected for the background, equilibrium aerosol size distribution.

The sulphur dioxide put into the stratosphere by a volcano is converted into submicron size sulphuric acid droplets within a month. Typically in the same period the particles circumnavigate the whole globe once. The particles are removed only by gravitational settling, which occurs when the particle size grows beyond 1 μm, through multiple collisions and coagulation. In the present work, sedimentation, which is the major removal mechanism of aerosols from the stratosphere is considered in detail.

The dynamical processes such as vertical diffusion and horizontal dilution due to advection responsible for the removal of particles from the stratosphere are shown to be negligible compared to sedimentation considered in the present study.

The rate of vertical diffusion of particles by atmospheric mixing is usually derived from the relation

$$\phi_D = -D n_M \frac{\partial}{\partial z}\left(\frac{n}{n_M}\right), \tag{8}$$

where ϕ_D is the diffusive flux (particles cm^{-2} sec^{-1}), D is the eddy diffusion coefficient (cm^2 sec^{-1}), n_M is the air density (molecules cm^{-3}) and n is the diffusing species (number cm^{-3}). For the tropics the eddy diffusion coefficient varies from about 0.75 to 1×10^5 cm^2 sec^{-1} in the altitude region of 30 to 17 km. It is estimated that $\partial n/\partial t)_{\text{diff.}}$ is of the order of 6.2×10^{-8} which is about two orders of magnitude less than the sedimentation which is of the order of 2.8×10^{-6} for particles of size around 0.5 μm.

In a similar work, the horizontal dispersion of volcanic cloud is treated by Zhao *et al* (1995) by a parameter called 'cloud area expansion function'. Bluth *et al* (1992) and McCormick and Veiga (1992) have shown that the Pinatubo volcanic plume initially propagated zonally and encircled the Earth within a month. However, the meridional dispersion was relatively slow and it took almost a year for the plume to cover the whole globe. Taking the initial area of the plume as 10^7 km^2 Zhao *et al* (1995) have shown that in about 45 days the plume area has become about 15×10^7 km^2 and after a year it became 40×10^7 km^2. In other words, from a period of 45 days to about one year after the eruption the areas have increased by a factor of about 2.7, resulting in a decrease of particle amount inside the plume by the same factor. However, as shown in figure 5(a) the aerosol amount (shown by the equivalent optical depth) decreases by more than two orders of magnitude from 45 days after the eruption to one year.

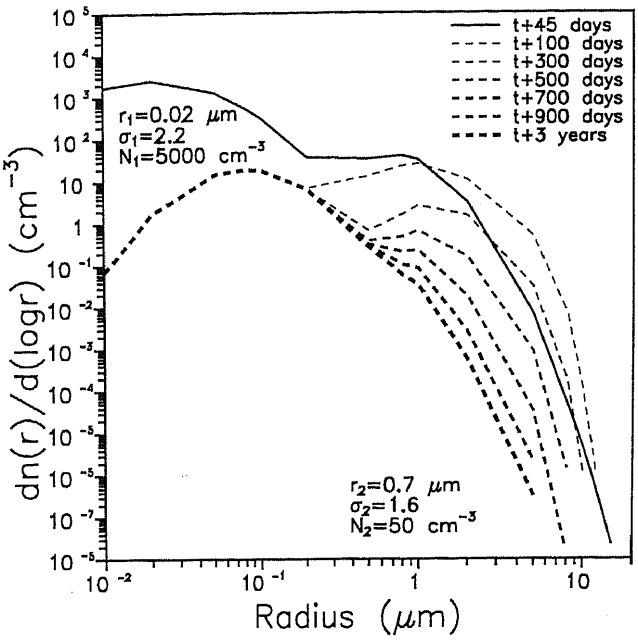

Figure 3. The evolution of aerosol size distributions undergoing the microphysical processes of growth, coagulation and sedimentation and after applying background corrections. The parameters of the initial guess size distribution are shown in the figure.

Rain washout, another removal mechanism of atmospheric aerosols, is effective only below tropopause and is not considered in the model.

Figure 3 shows the evolution of volcanic aerosol size distribution after background corrections. Results corresponding to Model 2 aerosol size distribution are shown as an example and also the evolution of the aerosol size distribution at selective time periods only are shown. About 2 months after the eruption, the primary mode in the bimodal lognormal distribution grows to an average size of 0.08 μm and also the secondary mode is not prominent. About 3 months after the eruption the total number of particles starts to decay as a result of growth, coagulation and sedimentation. Aerosols in the size range 0.2–1 μm continue to grow through coagulation process, even for about a year or so, making the secondary mode to shift to a higher radius of about 1–2 μm, with a subsequent reduction in the total number density. It is clearly seen, that as gravitational sedimentation is a very effective removal mechanism for larger aerosols in the range of a few microns, their removal rate is faster and in about 2–3 years, the size distribution returns to the background aerosol size distribution.

3. Results and discussion

The size distributions obtained at various time intervals are used to calculate the aerosol extinction coefficient at 500 nm (75% H_2SO_4 droplets, $m = 1.431 - i1 \times 10^{-8}$) and then integrated for the whole layer, to study the time evolution of the optical depth of the Pinatubo layer.

3.1 *Contribution of physical processes in the evolution of the stratospheric aerosol layer*

The percentage contribution of the different physical processes considered in the model namely growth, coagulation and sedimentation to the evolution of the aerosol size distribution shows that growth and coagulation dominate immediately after the eruption to about 150 days, whereafter sedimentation becomes dominant (figure 4). The contribution due to growth increases from about 4% at 45 days to about 40% in 160–180 days and then decreases gradually. The contribution due to coagulation which dominates the aerosol formation in the initial stages decreases from about 90% at 45 days to about 10% in about 6 months and the decrease is very rapid. While sedimentation increases from about 0% at 45 days to about 95% at about 3 years and the increase is gradual. While condensational growth and

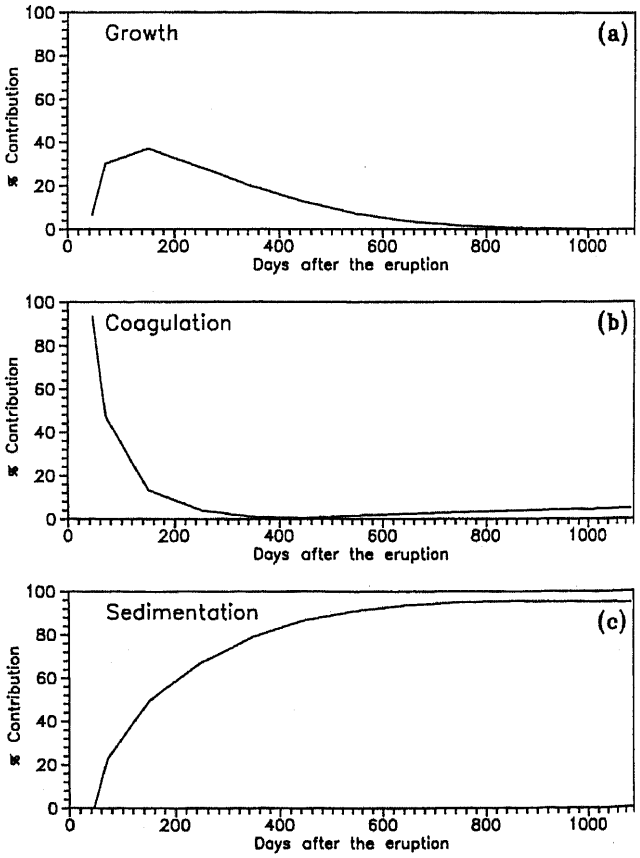

Figure 4(a, b and c). Percentage contribution of the different physical processes to the evolution of aerosol size distribution (figure 3) at different times after a volcanic eruption. Growth (**a**) and coagulation (**b**) are responsible for the increase in aerosol size while sedimentation (**c**) is responsible for the removal of particles from the stratosphere.

coagulation aid in producing larger particles, gravitational sedimentation becomes an effective removal mechanism for larger particles. The steep fall in the percentage contribution due to coagulation in the initial period, when the number of aerosols are abundant, is due to the fact that while the coagulation term exhibits a nonlinear dependence on the number density n i.e., it is proportional to n^2, growth and sedimentation terms are proportional to n. It is important to note that the coagulation process essentially brings down the number of particles very rapidly, whatever be the initial number of particles taken. Even if the sedimentation velocities at different altitudes are considered different, a feature which is present in the other aerosol models, our assumption of considering them as uniform throughout the layer as explained above, will not likely affect the end result as the process is proportional to n. Though the results shown are representative of Model 2 size distribution, they are found to be more or less similar for other size distributions also.

Sensitivity studies conducted by changing the growth rates and coagulation kernels by ±10% indicate that the percentage change in the calculated aerosol optical depth steadily increases from about ±2% to about ±20% in the case of growth rate, while due to coagulation, the change increases from about ±0.5% to about ±8%, for a period corresponding from $t+45$ days to $t+400$ days. Afterwards, the percentage change in optical depth decreases and becomes less than ±1% in about 2 years, indicating that the processes of growth and coagulation are important, in the first year of the eruption in maintaining the aerosol layer, whereafter when the particles grow sufficiently larger, of the order of a few microns, sedimentation becomes important. These results obtained with a change in the growth rates and coagulation kernels are in agreement with the results obtained from the percentage contribution calculations.

3.2 *Aerosol optical depth: Model results*

Figure 5(a) shows the aerosol optical depth obtained for the three model size distributions. The values decrease by about 5 to 9%, when calculated for 532 nm. Though in the initial stages of the eruption, eruptions of widely varying magnitudes, corresponding to the three model size distributions, used here (marked 1, 2 and 3 in the figure), exhibit large differences in the optical depths, but after about 2 years, they are almost the same. In figure 5(b) the effect of varying sedimentation velocities on the aerosol optical depth for model 3 is plotted, as an example. As the sedimentation velocity increases from about 17 km to 30 km and also very rapidly with the size of the aerosol particle, the removal of aerosols from the bin (17–30 km) is much faster in the

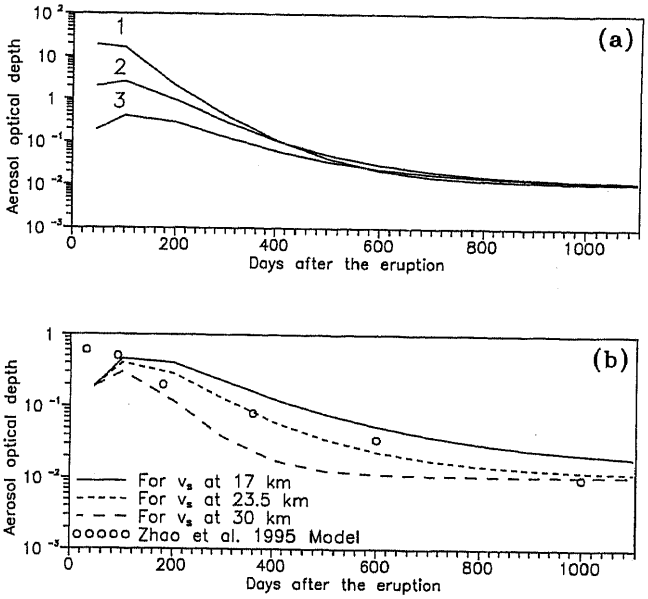

Figure 5(a and b). Variation in the integrated (17 to 30 km) aerosol optical depths at 500 nm as a function of days after a volcanic eruption. (a) For the three initial aerosol masses considered in the present study but for a constant settling velocity v_s at 17 km. (b) For curve 3 in figure 5(a) but for the three settling velocities at 17, 23.5 and 30 km respectively. For comparison Zhao *et al* (1995) model results have also been plotted.

higher altitude region of 30 km than in the lower altitude region of 17 km, hence, while the aerosol optical depth decreases gradually at 17 km, it is faster at 30 km. The aerosol optical depths plotted for the 3 sedimentation velocities, may be considered as the range of aerosol optical depth values obtained for the entire layer. But for the comparison of results with the experimental data, the aerosol optical depths obtained with sedimentation velocities at 17 and 23.5 km seem to be more appropriate, as will be seen later.

For comparison, the results obtained by Zhao *et al* (1995) from a one-dimensional, time-dependent model in simulating the Pinatubo volcanic aerosols in the stratosphere are also plotted in figure 5(b). As can be seen, since the process of nucleation, which is important in the initial stages after the eruption in producing aerosols and their evolution, is not included in the present work, there are discrepancies in the results obtained till about 100 days after the eruption. Bekki and Pyle (1994) using a global two-dimensional chemical-radiative-dynamical model, simulated the formation and temporal evolution of Pinatubo layer. Due to the absence of homogeneous nucleation of aerosols in their model, there were similar discrepancies in the timing of the peak in aerosol loading and the magnitude of surface area density, which made them to suggest that homogeneous nucleation plays an important role in the early stages of the volcanic eruption in determining the average size of the

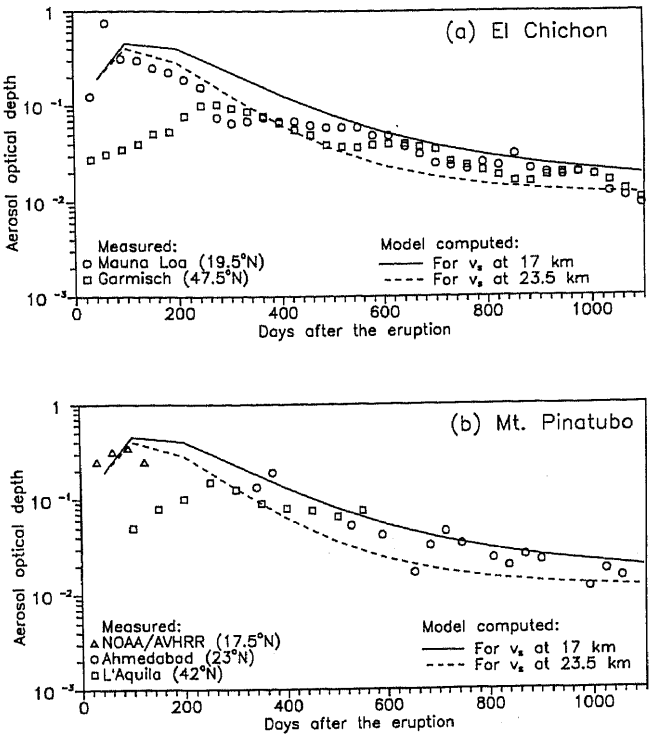

Figure 6(a and b). Comparison of model results with the experimental data (**a**) for the case of El Chichon eruption and the experimental data are from Robinson and DeFoor (1988) at Mauna Loa, Hawaii and Jäger *et al* (1988) at Garmisch-Partenkirchen, Germany, (**b**) for the case of Pinatubo eruption and the experimental data are from Minnis *et al* (1993), Jayaraman *et al* (1995b), Ahmedabad, India and D'Altorio *et al* (1993), L'Aquila, Italy.

volcanic sulphate particles and their residence times in the atmosphere.

3.3 *Comparison of model results with experimental data*

The comparison of model results with the experimental observations is done in figure 6(a) for El Chichon and in figure 6(b) for Mt. Pinatubo volcanic eruptions. The stratospheric aerosol optical depth data for the case of El Chichon is taken from Jäger *et al* (1988) and Robinson and DeFoor (1988) obtained using lidars. In both the cases, the data obtained at 694.3 nm are wavelength corrected, for 532 nm using the values given by Jäger (1992). In the case of Mauna Loa data, the NonRayleigh Back-Scatter obtained for the stratospheric aerosol layer is multiplied by 50, to get the optical depth and then wavelength corrected. It is seen that the computed results for model 3 reproduces the data obtained in both the cases. As the model calculations are made for the tropical region, the computed results compare well with the Mauna Loa (19.5°N) data (Robinson and DeFoor 1988) than with the data obtained over Garmisch-Partenkirchen (47.5°N) data (Jäger *et al* 1988) within one year after the eruption. However, after a year the aerosols are well-mixed globally and hence better agreement is seen between the two experimental data as well as with the model computed results.

For the case of Mt. Pinatubo eruption (figure 6b) the experimental data are taken from the Nd:YAG lidar measurements at 532 nm made at Ahmedabad (23°N) (Jayaraman *et al* 1995b), the Differential Absorption Lidar (DIAL) measurements at 589 nm made at L'Aquila (42°N), Italy (D'Altorio *et al* 1993) and the NOAA/AVHRR satellite measurements at 500 nm at 17.5°N (Minnis *et al* 1993). The satellite data however could be obtained only for the first four months after the Pinatubo eruption. In all the above results only the monthly mean values are shown.

It is interesting to note that the same feature of latitudinal dependence present in the El Chichon case is repeated in the case of the Pinatubo eruption. The stratospheric aerosol optical depth (15–30 km) at L'Aquila (42°N), Italy reproduces the same feature of the latitudinal dependence in the initial stages of the eruption. This feature was observed by using balloon-borne Sun-tracking photometers over Hyderabad during October 1991 and April 1992, about 4 and 10 months after the Pinatubo eruption, where a systematic increase in the aerosol extinction coefficients was seen above 24 km with decreasing latitude, indicating that the upper edge of the layer extends to higher altitudes in the equatorial region (Ramachandran *et al* 1994a). This feature is expected, as both El Chichon and Pinatubo, are in tropical latitudes and also for the first few months after the eruption, bulk of the aerosol cloud lay in the tropics (Hofmann 1987; Trepte *et al* 1993) which were then carried to the higher latitudes. The NOAA/AVHRR satellite data match closely with model 3.

The decay of both the El Chichon and Pinatubo aerosol layers in about 3 years to values very close to background level, shows that, though the mass injected by Pinatubo was more than 2 times as compared to El Chichon, faster growth rate and coagulation resulted in larger particles and hence a faster removal rate, due to sedimentation. Table 1 shows a comparison between the mass of the aerosol layer as well as the IBC (Integrated aerosol back-scattering coefficient) values obtained by Jäger and Hofmann (1991) for the case of El Chichon at 694.3 nm but corrected for 532 nm wavelength (Jäger 1992) and the values obtained in the case of Mt. Pinatubo eruption. In both cases the results are integrated from tropopause +1 km to 30 km. In spite of the large latitudinal difference between the two measurement sites, Garmisch-Partenkirchen (47.5°N) and Ahmedabad (23°N) and a large difference in the quantity of mass injected by the two eruptions, the mass of the aerosol layer and the integrated

Table 1. *Comparison of the mass of the volcanic aerosol layer and the integrated aerosol backscattering coefficient (IBC) at 532 nm in the case of El Chichon eruption as reported by Jäger and Hofmann (1991) and in the case of Pinatubo eruption (Jayaraman et al 1995b).*

	El Chichon		Mt. Pinatubo	
Time after eruption	Mass (gm^{-2})	IBC (sr^{-1})	Mass (gm^{-2})	IBC (sr^{-1})
1 year	3.75×10^{-2}	2.25×10^{-3}	2.39×10^{-1}	1.17×10^{-2}
2 years	1.25×10^{-2}	6.88×10^{-4}	1.12×10^{-2}	5.50×10^{-4}
1056 days	6.00×10^{-3}	3.13×10^{-4}	6.11×10^{-3}	3.00×10^{-4}

backscattering coefficients become comparable, about 2 years after the eruption. This feature was also seen by Pinto *et al* (1989) who through model calculations showed that when the mass of SO_2 injected increases beyond 10 Mt, the aerosol microphysical processes of condensation and coagulation produce larger particles, rather than a larger number of particles of the same size. Consequently the size of sulphate aerosol increases and the removal rate accelerates. The residual sulphate masses could be quite similar after about 2 years for eruptions of a wide range of magnitudes, suggesting that volcanic effects may be self-limiting. Recently Rosen *et al* (1994) from conjugate *in situ* observations of the stratospheric aerosol following Pinatubo eruption over Laramie, Wyoming (41°N) and Lauder (45°S), New Zealand, using balloon-borne backscattersondes, showed that similar aerosol loading and decay rates occurred over both midlatitude stations. A comparison of the results obtained on El Chichon over Laramie indicated higher mixing ratios in the case of Pinatubo but very similar decay times for both the eruptions.

When the models used were fitted with an exponential curve, the $1/e$-folding times, were found to be 4.6 months (Model 1), 6.1 months (Model 2) and 9.5 months (Model 3), showing that eruption of higher magnitude produces larger particles to give a faster decay time. This was experimentally verified by fitting an exponential fit to the Mauna Loa data, which gave a $1/e$-folding time of 11 months, for the El Chichon aerosol layer. For Pinatubo, an eruption which ejected 2 to 3 times more material into the stratosphere compared to El Chichon, a $1/e$-folding time of about 9 months, was found.

Some differences between the model and experimental results, could have been due to the assumption of average conditions, inside the layer, whereas it has been found using lidar data that the decay pattern can be different in different altitude regions (Jayaraman *et al* 1995b). The integrated aerosol extinction coefficient obtained from lidar for three altitude regimes, namely, 15–20 km, 20–25 km and 25–30 km give $1/e$-folding times of 16.4, 8.8 and 5.4 months, respectively. It should be noted that the decrease in aerosol extinction coefficient at a particular altitude layer is both due to removal of particles downward due to gravitational settling and horizontal dispersion to other latitudes. Also, the lower layers have a slower decay pattern compared to the top layer, because they not only lose particles due to gravitational settling but also gain particles from above. This has been also shown by recent model studies of Zhao *et al* (1995) that the aerosol optical depth at 500 nm at different altitudes in the stratosphere can be different.

4. Conclusions

A simple model developed to study the time evolution of a volcanic aerosol layer (17–30 km) and aerosol size distribution using the microphysical processes of growth, coagulation and sedimentation shows that the Pinatubo layer has decayed to the background aerosol level in about 3 years, over the tropics. The experimental lidar data obtained continuously on the Pinatubo layer also gives this result. The experimental data obtained for El Chichon (available in literature) also is well reproduced by the model. The latitudinal dependence of the aerosol layer is important only in the first few months after the eruption, as later on, the low latitudes become the source of aerosols to higher latitudes. The data obtained in the case of El Chichon show that the layer decayed in about 3 years. The latitudinal dependence in the case of El Chichon also is clearly seen in the first few months after the eruption. The model study also indicates that coagulation, which is the only process proportional to the square of the aerosol number density, brings down the number of particles very rapidly in the initial stages, whatever be the number of particles put in. The decay of the volcanic aerosol layers produced due to El Chichon and Mt. Pinatubo in about 3 years, to values very close to background levels, shows that though the amount of SO_2 injected by Pinatubo is more than two times that of El Chichon, the aerosol microphysical processes of growth, coagulation, have taken place faster giving rise to larger particles, and hence the removal rates of these large aerosols have been faster.

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