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The relaxation of volcanic stratospheric aerosols after major eruptions

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Abstract

In this review article, the present state of knowledge about volcanically produced stratospheric aerosols is discussed. These aerosols have gained a growing attention in the last few decades due to the increasing awareness of their impact on the climate, the radiative budget of the Earth-atmosphere system, the atmospheric chemistry and the associated remote sounding measurements. To understand these effects, a discussion on the origin, transport and microphysical evolution of aerosols is presented. Although at present the insight in these processes is advanced, the hypothetical presence of a steady-state background aerosol layer in the stratosphere remains the subject of controversy, and we conclude this article with a brief discussion on this topic.

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1 Importance of volcanic eruptions in stratospheric studies

1.1 Frequency and intensity of major volcanic eruptions

Major volcanic eruptions are known to cause strong perturbations of the atmosphere that can lead to climate changes on many timescales. This influence, suspected since the antiquity, has been studied in pioneering works by [14] and [12] followed by many reviews on climatic effects induced by volcanoes (see e.g. [27],[10],[16]). The most famous modern eruptions (Tambora 1815, Krakatau 1883, Agung 1963, Mount St. Helens 1980, El Chichon 1982, Mount Pinatubo 1991) have produced clear atmospheric impacts such as abnormally cold summers[7], colorful sunrises and sunsets[20] and stratospheric temperature variations [24].

A comprehensive list of known volcanic eruptions can be found in [21], and [12] constructed a "dust veil index" for many eruptions in the period 1500-1968. More quantitative data were reported by [25] who investigated the detailed chronology of major stratospheric dust veils from 1881 to 1960 measured by pyrheliometry. During this period, about ten measurable volcanic dust veils have been clearly identified that produced major optical depth perturbations of the atmosphere. This means that volcanic signals are episodic but they can strongly perturb the atmospheric radiative and chemical properties over periods of 3-5 years after a violent eruption.

1.2 Climatic response

The two strongest eruptions of the century were El Chichon in April 1982 and Mount Pinatubo in June 1991, the latter being the most studied event so far. For several years following these eruptions, significant changes of the global temperature were reported. Volcanic sulfate aerosols reduce the UV solar flux reaching the Earth by pure scattering while absorption becomes dominant in the IR although the total radiative forcing at the top of the atmosphere is negative for submicron particles [24]. This forcing results in tropospheric cooling and a clear positive stratospheric thermal response with temperature increases as large as 4 K in the tropical lower stratosphere[9]. Volcanic aerosols also have the potential to affect the stratospheric chemistry by changing the surface that is available for heterogeneous reactions. Various effects of aerosols on ozone have been described by [22]. A decrease in ozone concentrations reduces UV absorption in the stratosphere which in turn modifies the aerosol heating effect.

1.3 Perturbative effect in remote sounding measurements

Apart from global climatic impact, volcanic aerosols produced by large eruptions can have a strong influence in remote sounding measurements in the UV-visible range. Integrated aerosol backscatter measured by routinely operated ground-based lidars [15] may vary by a factor of 100 and clearly show that the recovery of atmospheric transparency takes several years. Also, the spectral inversion of radiometric measurements where aerosol and other gas components are present is of crucial importance in the assessment of ozone long trends [23],[5]. For the SAGE II experiment, inaccuracies in the aerosol removal have affected the retrieved stratospheric ozone concentrations by interfering with the ozone slant path optical thickness in the Chappuis band ($\lambda \simeq 0.6\mu\text{m}$). This is not only a systematic effect due to the non-orthogonality of the optical thicknesses over the wavelength range but this bias also introduces an

apparent downward trend in lower stratospheric ozone concentrations as the aerosol size distribution evolves during postvolcanic relaxation periods

2 Microphysical evolution of volcanic stratospheric aerosols

2.1 Sources

A pioneering review of observational and theoretical knowledge of the stratospheric aerosols was published by [29] and the Mount Pinatubo eruption in 1991 triggered important modelization work about the origin and the evolution of stratospheric aerosols. [6] studied the processes of maintaining the non-volcanically-perturbed Junge layer by using a global 3-D model describing the SO_2 production from OCS and its latter oxidation to gaseous sulfuric acid. From a dynamical point of view, their model includes the condensation-evaporation equilibrium of H_2SO_4 , condensation growth and particle rainout. The role of OCS as a precursor of the stratospheric background aerosol was reanalyzed by [4] who concluded that the production of stratospheric sulfur aerosol from OCS oxidation was too low to explain the "background" aerosol level. [11] included CS_2 as a precursor gas but also underlined that the main source of stratospheric aerosol was the large amount of SO_2 (about 20 Mt) directly released in the stratosphere by Pinatubo-like eruptions [13].

2.2 Dynamical evolution

The horizontal dispersion of the Pinatubo volcanic cloud has been described in a two dimensional model by [2], assuming an equatorial release of the SO_2 for the sake of realism, and by [32] who only considered climatological means of advective winds. None of both models was able to describe aerosol transport induced by the quasi-biennial oscillation or by other interannual variability. [33] and [11] have described the horizontal dilution of the volcanic cloud by using a cloud area expansion function tuned to reproduce the observations from space-borne experiments. Shortly summarized, the zonal propagation of the cloud encircled the Earth within a month and covered about 1/3 of the surface. The much slower meridional dispersion took about one year and can be considered as independent of altitude. Furthermore, phenomenological vertical velocity and eddy diffusion have to be taken into account for a realistic description of the volcanic aerosol relaxation.

2.3 Microphysical evolution

The time constant for chemical production of H_2SO_4 is equivalent to the SO_2 oxidation time (about 30 days) and much longer than than the time constant for the loss of H_2SO_4 vapour to aerosols (a few hours)[33]. Therefore, although there is continuous conversion of the volcanic emissions into H_2SO_4 , the acid vapour cannot accumulate to high concentrations and is almost immediately converted to particles. Model results show that the homogeneous nucleation of $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ is achieved within the first few weeks after an eruption and the aerosol concentrations are subsequently driven by coagulation. Condensation on the sulfate particles depends on altitude (due to the influence of temperature) and can be considered as finished a few months after the eruption. However, a complete description of the stratospheric aerosol formation requires homogeneous-heteromolecular and heterogeneous-heteromolecular nucleation of water and sulfuric acid vapour[11]. The abundance of homogeneous liquid aerosol

appears a few days after the eruption at about 30 km, then descends to lower altitude and disappears at 20 km approximately 18 months later. Numerical simulations show the presence of 2 maxima in the vertical profile of aerosol concentration: the first one (20 km) coincides with that of homogeneous liquid aerosol; the second one (30 km) is associated with the presence of volcanic solid particles that moderates the evaporation to smaller drops. The formation of new aerosol nuclei is followed by condensation and evaporation of sulfuric acid and water, coagulation, transport and sedimentation.

It is important to realize that the volcanic perturbation on the aerosol population is a long-lasting phenomenon. The concentration of large particles can still increase 5 years after the eruption while smaller particles have already been removed. An important conclusion is that the particle removal is more caused by coagulation than by sedimentation if the latter one is compensated by upward circulation. This was not taken into account by [2] and could explain the discrepancy between the observed and modelled aerosol peak in the stratospheric optical thickness at $1.02 \mu\text{m}$.

Furthermore, it is very important to notice that a correct modelling of the volcanic aerosol requires some accurate estimation of the wind vertical velocities. Indeed, analyzed wind fields show typical velocities of the order of 10 km/year, mostly upward in the tropical zone, that can counterbalance sedimentation. In the past [18], many one-dimensional steady-state stratospheric aerosol models have only taken into account the equilibrium between sedimentation, growth rate, coagulation and the eddy diffusion.

3 The existence of a stratospheric aerosol background layer

3.1 A survey of existing climatologies

The availability of aerosol data measured by the Stratospheric Aerosol and Gas Experiment (SAGE I and II) and Stratospheric Aerosol Measurement (SAM II) instruments has allowed the construction of global climatologies of stratospheric aerosols.

[8] concludes that the aerosol layer is distinctly volcanic in nature and suggest that the decadal average is a more useful estimate of future aerosol loading than a background loading, which is never achieved. They diagnosed a tropical reservoir region with upper and lower transport regimes driven by the quasi-biennial oscillation. These findings were also confirmed by [28] who observed banded structures in stratospheric aerosol optical thickness distributions with minima found between latitudes of approximately 15° and 45° . The structure results from the presence of a tropical aerosol reservoir, advected polewards by non-isentropic transport that has been realistically modelled in the work of [17].

A global climatology of stratospheric aerosol surface area density for the period 1984-1994 has also been published by [26], that confirms the latitudinal "sombbrero" distribution of stratospheric aerosols. The spectral and vertical features of the aerosol extinction profiles have been investigated by [3] and by [30] and were found to be strongly dependent on volcanism. It is also worth mentioning the influence of the tropospheric meridional circulation derived by [31] where the mean circulation features the influence from both the diabatic circulation and the eddy quasi-isentropic transport with some material advection into the upper troposphere from both the lower troposphere and the lower stratosphere.

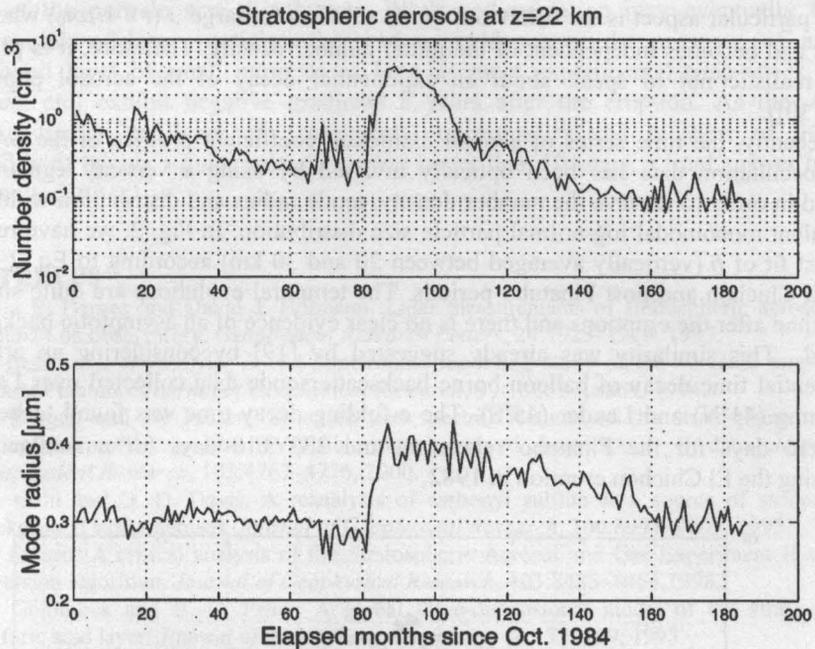


Figure 1. Top: Stratospheric aerosol number density derived from SAGE II data at 22 km. Bottom: Idem for the mode radius of a monomodal lognormal distribution. Notice the delay before the mode radius reaches a maximum

3.2 The quest of the background layer

[1] have investigated the presence of a background layer of stratospheric aerosols. It appears that a somewhat variable background (non-volcanic) stratospheric aerosol level exists at Mauna Loa but its value may be a function of the QBO phase. They concluded that these low aerosol levels are difficult to differentiate clearly.

We perfectly agree with [11] about the pure volcanic nature of the stratospheric aerosol layer. Indeed, it cannot be maintained without injecting SO_2 during volcanic eruptions, which means that the layer never reaches a steady state. This is in fact confirmed by experimental evidence. For a monodisperse particle size distribution, assuming in a first approximation a constant coagulation coefficient α , the aerosol total number density n obeys the Smoluchowski equation:

$$\frac{dn}{dt} = -\alpha n^2 \quad (1)$$

whose solution is simply

$$n(t) = \frac{n_0}{1 + \alpha n_0 t} \quad (2)$$

A particular aspect is the behaviour of the solution for large t ($t \gg 1/\alpha t_0$) where $n(t) \sim 1/\alpha t$ and becomes independent of the initial number density. Therefore, it is probably more realistic not to speak about an exponential decay of the aerosol population ([16],[30]).

Recently, the time series of aerosol extinction coefficient profiles of the SAGE II solar occultation data has been optically inverted by using a vertical regularization method in order to retrieve the number density, mode radius and distribution width of an equivalent monomodal log-normal particle size distribution. In Fig. 2, we have reported the best fit of \bar{n} (vertically averaged between 20 and 30 km) according to Eq. 2 for the post-El Chichon and post-Pinatubo periods. The temporal evolutions are quite similar a long time after the eruptions and there is no clear evidence of an asymptotic background aerosol. This similarity was already suggested by [19] by considering an arbitrary exponential time decay of balloon-borne backscattersonde data collected over Laramie, Wyoming (41°N) and Lauder (45°S). The e-folding decay time was found to be about 343 ± 10 days for the Pinatubo relaxation and 333 ± 10 days for a similar period following the El Chichon eruption in 1982.

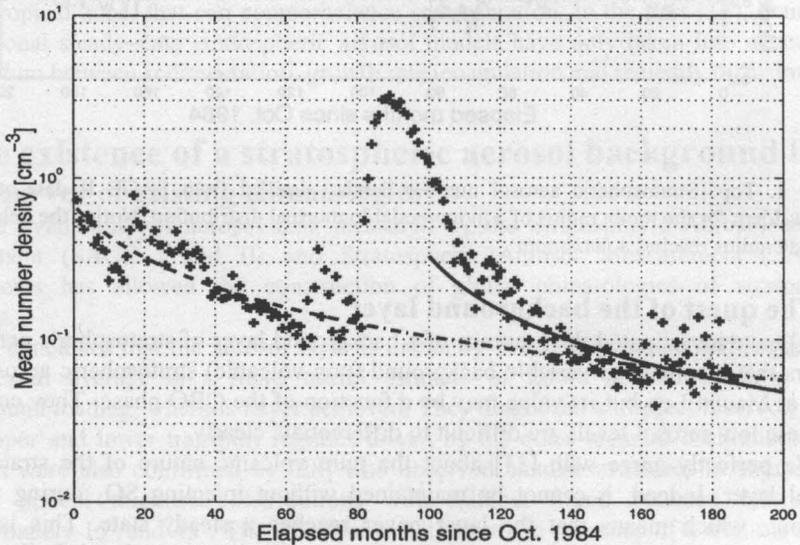


Figure 2. Relaxation of the volcanic aerosol number density. Crosses: Experimental data from SAGE II. Full line: best fit for Eq. 2 (post-Pinatubo). Dashed curve: idem for post-El Chichon (months 0 to 80), extrapolated to post-Pinatubo period (dot-dashed, months 81 to 200).

4 Conclusions

Large volcanic eruptions inject sulfur dioxide into the stratosphere. The normal residual stratospheric meridional circulation lifts the aerosols in the tropics and transports them polewards on one year. There exists a clear experimental evidence that the residence timescale of volcanic aerosols can be as large as 5 years. In particular, the decrease in aerosol number density seems to be associated with an increase in the mode

radius of the particle size distribution. While sedimentation may eventually be the ultimate sink of large particles, there is no evidence of the presence of a clear background level of small particles as long as most experimental time-series (lidar, satellite, ... etc) exhibit negative gradients 8 years after the eruption. As the aerosol decreases under the level of experimental uncertainties and in view of the low probability of having a quiet volcanic period larger than 10 years, a final answer has not yet been given today.

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