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Anti-correlation between stratospheric aerosol extinction and the Ångström parameter from multiple wavelength measurements with SAGE II – a characteristic of the decay period following major volcanic eruptions

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Abstract. Stratospheric Aerosol and Gas Experiment (SAGE) II data at multiple wavelengths were analyzed to show how aerosol extinction decays with time following major volcanic eruptions. Comparisons were made between the lowest background level in 1999 and the past data record of the stratospheric aerosol layer. The time variation of the extinction coefficients was compared with the Ångström parameter, which is a good indicator of particle size. A clear anti-correlation was found between the extinction and the Ångström parameter after the eruption of Mt. Pinatubo. Comparison of the extinction coefficients and Ångström parameters in 1989 and 1999 made it clear that the aerosol layer was affected by volcanic eruptions in 1989. The distinguishing negative correlation is a characteristic feature of decay periods following volcanic eruptions.

1. Introduction

Hofmann [1990] noted that the amount of stratospheric sulfate aerosol measured by balloon-born OPCs at 41°N increased by 30 - 50% between 1979 and 1989. Hofmann [1991] suggested that this increase in background aerosols could be attributed to increased jet fuel emission in the lower stratosphere.

Thomason et al. [1997] compared global stratospheric aerosol levels from the Stratospheric Aerosol and Gas Experiment (SAGE) in 1979 with those from SAGE II in 1989-1991, and concluded that 1989-1991 was characterized by an ongoing global recovery from the eruptions of El Chichon (1982), Nevado del Ruiz (1985), and Kelut (1990). The major eruption by Pinatubo in June 1991 made it impossible to follow these measurements through the 1990s to confirm the changes in background levels. With the decay of the Pinatubo perturbation, the investigation of the background can now be continued.

As part of the 1998 WMO/UNEP ozone assessment, long-term records of the integrated stratospheric aerosol column observed with some lidars were compared with an OPC data record at Laramie, showing their consistency and the levels in late 1997 being well below pre-Pinatubo values. Therefore, the estimate of the anthropogenic effect based on the 1979-1989 comparison is now considered an overestimate [WMO, 1998]. At the time of the WMO report in late 1997, the stratospheric background aerosol level was still decreasing, and we continued to track this trend from 1997 to the end of 1999.

In this study, we analyzed SAGE II aerosol extinction data at multiple wavelengths from January 1985 through December 1999. This includes a later period than that analyzed by Thomason et al. [1997] and WMO [1998]. We present the time variation of the extinction coefficient, and the changes in particle size indicated by changes in the Ångström parameter. This study demonstrates that the decay terms after volcanic disturbances are characterized not only by a decrease in the mass of aerosols, but also by a change in particle size.

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2. Analysis method

SAGE II measures aerosol extinction at four wavelengths, (1.019, 0.525, 0.452, and 0.386 μm), at altitudes from 10 to 40 km, using solar occultation technique. We used version 6.0 of the aerosol data in this study. The extinction coefficients were averaged for each five-degree latitude band from 80°N to 80°S, at each altitude, and for each month, where the number of measurements exceeded ten per grid, after screening out the data with more than 80% relative error. The wavelength dependence of the extinction was approximated as

$$\sigma = \sigma_o \lambda^{-\alpha} \tag{1}$$

where σ is an extinction coefficient (km^{-1}), λ is wavelength (microns), and α is the Ångström parameter. The Ångström parameters were derived from all events, and are averaged for the five-degree latitude bands. Because of concerns as to quality, the extinction data at the shortest wavelength, 0.386 μm, were not used.

3. Results and Discussion

3.1. Long-term variation of the 1.0-micron extinction coefficient and Ångström parameter

Figure 1 shows the time series of the 1.0-μm extinction coefficient ($\sigma_{1.0}$) at 20 km, from January 1985 through December 1999, as observed by SAGE II. An enormous increase in $\sigma_{1.0}$ occurred after the eruption of Mt. Pinatubo (June 1991, Philippines), although there was a short-lived data void immediately following the eruption, because of data truncation [McCormick and Veiga, 1992]. The subsequent decay phase lasted for more than seven years, as indicated in the figure.

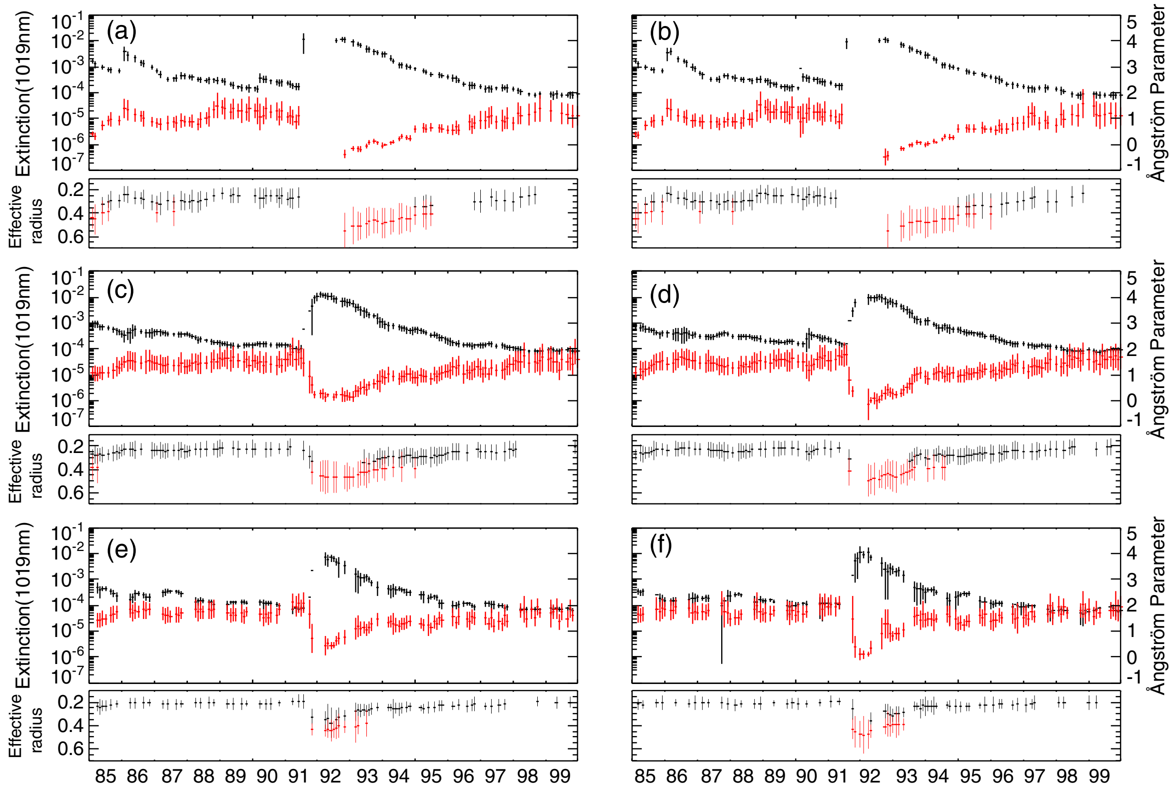

Figure 1. (upper panels) Time series of the 1.02-μm extinction coefficient at 20-km altitude, observed with SAGE II (black). The horizontal bars indicate the range of the average over one month. Ångström parameters are also plotted in the same panel (red). The panels are for (a) 0-5°N, (b) 0-5°S, (c) 30-35°N, (d) 30-35°S, (e) 55-60°N, and (f) 55-60°S. (lower panels) Time series of the effective radius. Black shows the unimodal aerosol type and red corresponds to the bimodal aerosol type. All data are plotted with one-sigma error bars. The effective radius is inversely plotted to compare with the Ångström parameters.

The time series of the Ångström parameter (α) in Figure 1 (red crosses) shows a significant negative correlation with $\sigma_{1.0}$, suggesting that particle growth was enhanced by the sulfur compounds emitted from the volcanic eruptions. In particular, α dropped remarkably (down to zero) after the eruption of Mt. Pinatubo. Although the figures at other altitudes are not shown here, there was a similar tendency at altitudes from 17 to 26 km.

Figure 1 shows that the extinction coefficient continued to decrease after 1997, when the WMO [1998] comparison was made. The unusually prolonged period of disturbance following the Pinatubo eruption was caused by the large amount of sulfate injection [McCormick et al., 1995]. By 1999, the stratospheric aerosol extinction was as low as $\sim 10^{-4}$ /km at 20 km, the lowest value recorded in the entire period of SAGE II observations.

3.2. Relationship between the Ångström parameter and particle size

What is the relationship between particle size and α? Yue [1999] proposed a practical method that derives particle size distribution from multiple wavelength extinction coefficients from SAGE II; he showed that the number density can be expressed as a linear combination of the extinction coefficients observed with SAGE II. For this study, we used coefficients C(k,i,t,p) that were offered by Glenn K. Yue [personal communication, 2000], deriving the size distribution. The derived size distribution yields other parameters, such as the effective radius or sonde ratio. The correlation between α and effective radius is very compact; however, the relationship depends on aerosol type: non-volcanic (unimodal) or volcanic (bimodal).

The time series of the effective radius (r_{eff}) derived by Yue's method is plotted in the lower panels of Figure 1. Results for both unimodal (black crosses) and bimodal (red crosses) models are shown. In some period after the Pinatubo eruption, the data for only the bimodal type (red) are shown, because unimodal type does not give reasonable size distribution. The behaviors of r_{eff} and α are very similar.

We also simulated the extinction coefficient in SAGE II channels based on Mie theory for various values of the parameters r_m and s for the lognormal size distribution function: where r_m is mode radius and s is standard deviation. The refractive index of 1.44 was taken from Russell et al. [1996] (at 223K, 5 ppm of water vapor). We made a diagram showing the correlation between α and $\sigma_{1.0}$ for various size distribution parameters r_m and s (not shown here). The correlation between $\sigma_{1.0}$ and the total mass of aerosols was the better than the other wavelengths, as expected, so $\sigma_{1.0}$ can be used as a parameter representing aerosol mass. On the diagram of α and $\sigma_{1.0}$ obtained from Mie calculations, we plotted the observed $\sigma_{1.0}$ and α data with data uncertainties. All candidates for r_m and s that are expected

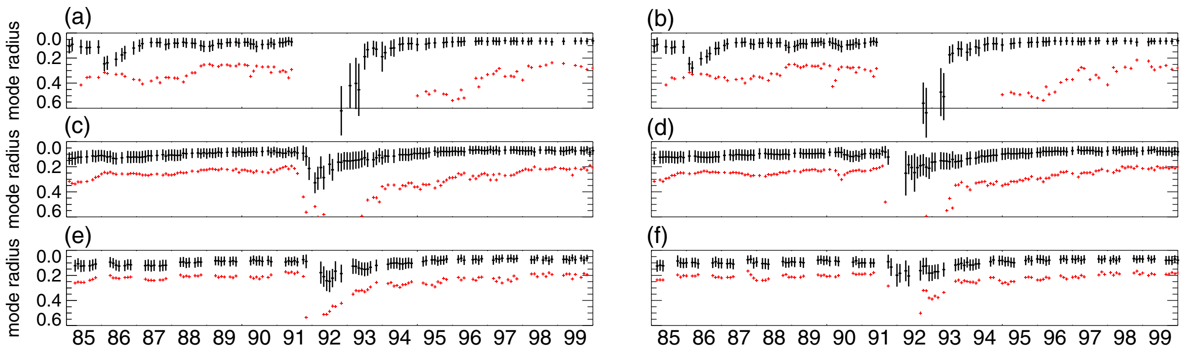

Figure 2. (The time variation of the estimated mode radius, $\bar{r}_m$ in Equation (2) (black crosses) and the estimated effective radius (red) at an altitude of 20 km corresponding to the variation of the extinction and the Ångström parameter shown in Figure 1. The time variation of the effective raius is comparable to that determined by Yue's method, as shown in Figure 1.

theoretically in the range of data uncertainties were averaged to determine the best-estimated size parameters, $\bar{r}_m$ and $\bar{s}$. Figure 2 depicts the time variation of $\bar{r}_m$ and the corresponding effective radius at an altitude of 20 km, determined in this way. The trend in the time variation of r_{eff} is comparable to that for a unimodal size distribution derived by Yue's method, as shown in Figure 1.

When the size distribution function is expressed by a Jünge distribution function, the relationship between α and the power law exponent p is expressed as α = p - 3. In the background period, the size distribution function is similar to the Jünge distribution, as r_m is small enough, compared to the wavelength region of the SAGE II measurements. After large volcanic events, a bimodal size distribution with a large mode radius is often observed, so that the wavelength dependence of extinction is sometimes not linearly related to the logarithm of the wavelength, resulting in non-negligible residuals in Eq. (1). Therefore, during periods of disturbance α is not a straightforward indicator of particle size. However, the clear negative correlation between σ and α, shown in Figure 1, suggests that there is a strong connection between aerosol mass loading and particle growth.

3.3. Anti-correlation of the extinction and the Ångström parameter during the decay period

Figure 3 is a scatterplot of α vs. $\sigma_{1.0}$ on an isentropic surface during the decay period after Pinatubo. The different colored dots represent data for different years. Linear regression lines for each year are shown in the same color as the dots for that year, and their lengths show the range of values. For example, the dark blue line in each panel depicts the data observed in 1993 for all latitudes, with a larger $\sigma_{1.0}$ and smaller α at lower latitudes, and a smaller $\sigma_{1.0}$ and larger α at higher latitudes. The spatial anti-correlation between $\sigma_{1.0}$ and α is now apparent.

As already discussed, σ was large and α was small during the period of disturbance after the Pinatubo eruption, the opposite of the conditions during the background period. In 1999, all the extinction data were at the background level, and scattered over the range reflecting seasonal variation. The range of the seasonal variation of α is relatively large depending on latitudes, resulting in an unclear negative correlation.

The negative correlation is clearer at higher altitudes (compare Fig. 3(a) and (c)). Hamill et al. [1999] discussed changes in the size distribution due to differences in the rate of descent, which depends on particle size, during the decay period. The more rapid descent of larger particles causes extinction to decay faster at longer wavelengths, increasing α. In other words, the changes in α with time after volcanic disturbances provide an indicator of the decay phase. The

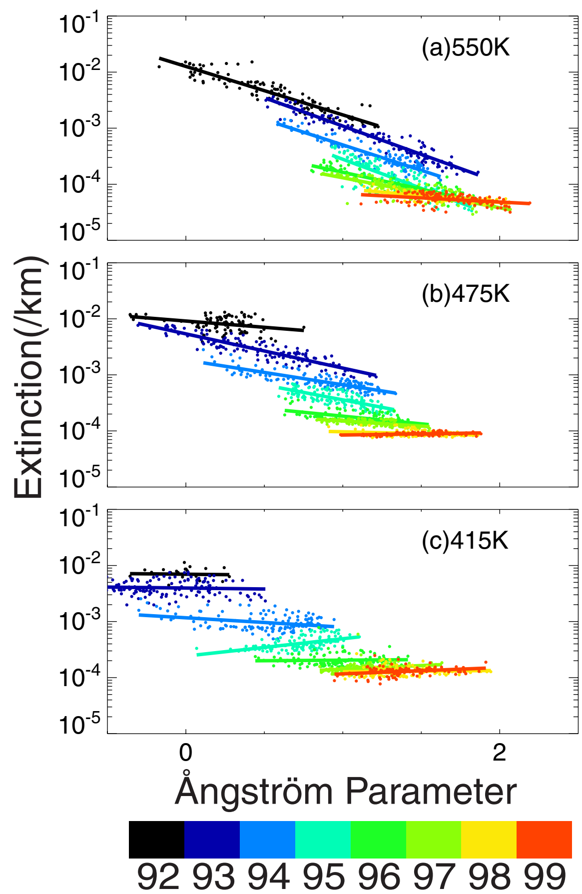

Figure 3. ((a)- (c) Scatter plot of the extinction coefficient and Ångström parameter on the same isentropic surfaces at (a) 550 K (~23 km), (b) 475 K (~20 km), and (c) 415 K (~17 km). Different colors are used to represent data (dots) and linear regression lines for different years. The lengths of the lines indicate the extent of the values.

negative correlation is less clear at lower altitudes, implying settling of larger particles from higher altitudes.

Jäger et al. [1995] traced the temporal evolution of the height of the maximum backscattering coefficient over Garmish-Partenkirchen (47.5°N), Tsukuba (36.1°N), and Naha (26.2°N) using lidar (see Fig. 2 of Jäger et al., [1995]). At all three latitudes, the height of the maximum backscattering coefficient descended from around 23 km in mid-1991 to about 16 km in mid-1993. Our analysis of SAGE II data showed similar results. The heights of the maximum of both $\sigma_{1.0}$ and α descended for about 2 years after the eruption, after which time they rose. This suggests that the larger particles generated from sulfate matter from volcanic injection gradually fall to the tropopause; i.e., large particles are removed by mixing into the troposphere. Similarly, the negative correlation at upper altitudes in the early stage of decay after the Pinatubo eruption can be interpreted as the removal of large particles due to their gravitational settling. At lower altitudes, the negative correlation is less clear, as the influx of particles from upper altitudes and the downward outflow cancel each other out, complicating the situation.

The scatterplot for 1988-1989 (not shown here), before the Pinatubo eruption, shows a negative correlation that is similar to that observed during the decay period after Pinatubo. This is further evidence that the period before Pinatubo was a decay phase and not background.

When examined more closely, in 1999 α shows a latitudinal gradient even in the background period. Although the classic theory of aerosol microphysics depicts growth to 'mature particles' in a poleward meridional circulation, this study suggests that particles diminish in size toward poles. Future studies should consider other microphysical processes, such as evaporation, as well as meridional transport.

4. Concluding Remarks

SAGE II version 6.0 extinction data from 1985 through 1999 were analyzed, focusing on the Ångström parameter. The Ångström parameter decreases after major volcanic eruptions, apparently corresponding to enhancement of extinction. A clear anti-correlation between extinction and the Ångström parameter was observed during the decay after the Pinatubo eruption. The level of extinction in 1999 was the lowest in the history of systematic measurement of the stratospheric aerosol layer, while the Ångström parameter was the highest. Comparison of the extinction coefficients and Ångström parameters in 1989 and 1999 made it clear that even in 1989 the aerosol layer was still affected by previous eruptions, and that a negative correlation is a distinguishing feature of a decay period after volcanic eruptions.

Comparisons of the Ångström parameter with the effective radii obtained by the method of Yue [1999] and derived from simple Mie-calculations proved that the Ångström parameter is an effective indicator of particle size. More extensive analysis, including microphysical considerations, would shed light on the particle formation processes and the mechanism of their loss from the stratosphere.

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