

## STRATOSPHERIC AEROSOL INCREASE AFTER ERUPTION OF PINATUBO OBSERVED WITH LIDAR AND AUREOLEMETER

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### ABSTRACT

An increase in the amount of stratospheric aerosol due to the Pinatubo eruption (June 12 -15, 1991, 15.14 °N, 120.35 °E) was observed from the end of June, 1991 by a lidar in NIES (National Institute for Environmental Studies), Tsukuba (36.0 °N, 140.1° E). After large fluctuations in summer of 1991, the amount of the aerosols increased in mid-September as a result of enhanced transportation from the sub-tropical region. In autumn and winter of 1991, dense aerosol layers were continuously observed. Aureolemeter (scanning spectral radiometer) measurements were also carried out with lidar measurements and columnar size distribution of stratospheric aerosols was estimated for some cases. Collaborative measurements with the lidar and aureolemeter provided some information on height distribution of the surface area of aerosols in late 1991.

### 1. INTRODUCTION

The Mt. Pinatubo eruption in the Philippines on June 15, 1991 was reported as the most violent of this century. The total amount of SO<sub>2</sub> injected into the stratosphere was estimated to be about 20 megatons from Satellite data (Bluth et al, 1992; McCormick and Veiga, 1992). These results indicate the amount of volcanic material transported into the stratosphere by the Pinatubo eruption was over twice that of the El Chichon (Mexico) eruption on March 29 and April 4, 1982.

Stratospheric ozone decreased about 10% (Mantis et al., 1986), after the eruption of El Chichon in 1982. Hofmann and Solomon (1989) suggested that heterogeneous reactions of chlorine-related substances on the surface of the volcanic aerosols would cause a significant decrease in the ozone amount. Therefore, it is speculated that Pinatubo aerosols would cause ozone depletion of more than 10%.

As the heterogeneous reaction rates are proportional to the surface area of aerosol particles, it is important to estimate the surface area of aerosols to examine the effect of heterogeneous reaction on ozone depletion.

At NIES, Tsukuba Japan, lidar measurements of the stratospheric aerosols after the Mt. Pinatubo eruption have been carried out. Aureolemeter measurements have been also carried out at NIES since November 1991. The aureolemeter provides information on the columnar size distribution of aerosols although it cannot include information on its height distribution.

In this paper, we report some results of the lidar measurements and show the time variation and height distribution of volcanic aerosols. We also report on some examples of the estimation of surface area of stratospheric aerosols derived from collaborative measurements with the lidar and aureolemeter at NIES.

### 2. MEASUREMENTS AND DATA PROCESSING

Specifications of the NIES lidar system for aerosol measurements were described in Shimizu et al. (1985). An Nd-YAG laser emits laser pulses of about 400 mJ for a 532 nm wavelength with a repetition rate of 25 Hz. The diameter of the telescope is 1.5 m. Data processing was described in Hayashida-Amano et al., (1991) in detail. The algorithm of Fernald (1984) was applied to correct for attenuation of the laser signal in the atmosphere. The ratio of extinction to backscatter of aerosols is denoted as S<sub>1</sub> which depends on the size distribution of the particles. S<sub>1</sub> is essentially important for determining aerosol profiles after large volcanic eruptions because the attenuation of signal is strong in the aerosol layer.

The integrated backscattering coefficient (IBC) is defined as

$$IBC = \int_{z_1}^{z_2} \beta_a(z) dz \quad (1)$$

where  $\beta_a$  denotes the backscattering coefficient for aerosols and  $z$  is altitude. IBC times S<sub>1</sub> signifies the optical thickness of the aerosol layer for the altitude region between  $z_1$  and  $z_2$ . Here  $z_1$  and  $z_2$  were taken as 15 km and 30 km, respectively.

The aureolemeter was set in November 1991 on the top of the same building of the NIES lidar. Specifications of the instrument and data processing are described in Nakajima et al. (1986). The aureolemeter was developed to measure not only direct solar irradiance but sky radiance in the solar almucantar. The aureolemeter is equipped with a silicon photodiode and seven interference filters at wavelengths of 332, 368, 500, 675, 777, 862 and 937 nm.

An algorithm developed by Nakajima et al., (1986) was used for retrieving the size distribution of aerosols from the measurements. The solar aureole including the effect of multiple scattering was calculated theoretically and compared with the observed data to derive scattering phase function  $P$ .

Scattering phase function  $P$  at scattering angle  $\theta$  is given by

$$P(\theta) = \int K(\theta, \ln r) \frac{dV}{d \ln r} d \ln r \quad (2)$$

where  $dV/d \ln r$  is the volume size distribution of aerosols,  $r$  is the radius of an aerosol particle, and  $K$  is the kernel function calculated from Mie's amplitude function. Equation (2) was solved as an inverse problem to obtain volume spectrum  $dV/d \ln r$ .

### 3. RESULTS OF LIDAR MEASUREMENTS

Stratospheric measurements were routinely carried out every clear night. The time series of IBC for the altitude region from 15 to 30 km are shown in Fig. 1. The IBC shown here includes an extinction correction for  $S_1 = 50$ .

First arrival of the volcanic aerosol layer was observed near 16 km on June 28, 1991: only two weeks after the eruption. This is due to advection associated with an Asian anticyclone (Hayashida and Sasano, 1992, McCormick and Veiga, 1992). In summer of 1991, aerosol layers appeared sporadically at an altitude of over 20 km. A thick aerosol layer with a peak at about 22.5 km was first observed on September 25. Although there were only limited measurements in September due to bad weather conditions, we could confirm that there was no distinct layer on September 12. It was difficult to estimate IBC quantitatively on September 12 and 25, because the measurements were carried out in daytime with only an insufficient S/N ratio. Therefore, IBCs obtained only on September 5 (before the increase of IBC) and on October 3 (after the increase of IBC) were plotted in Fig. 1.

The increase in the aerosol amount corresponded to the change in wind system. Transportation mechanism was discussed in Hayashida and Sasano (1992) with analysis of meteorological data.

During the autumn and winter of 1991, a thick and dense aerosol layer was continuously observed from about an altitude of 17 to 30 km. Although there were still large fluctuations in the amount of aerosols, IBC increased up to about  $6 \times 10^{-3} \text{ sr}^{-1}$ .

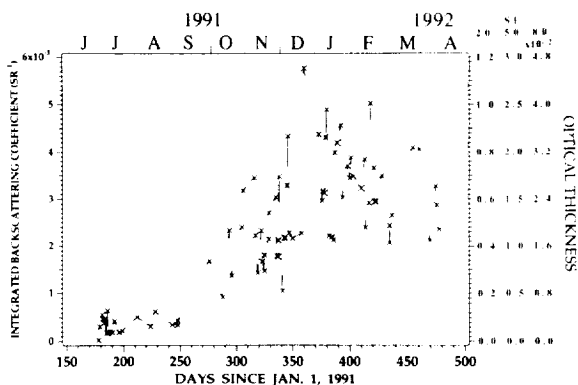


Fig.1. The time series of the integrated backscattering coefficient (IBC) for the range from an altitude of 15 to 30 km after the Pinatubo eruption.

### 4. SURFACE AREA ESTIMATION

In autumn and winter, tropospheric optical thickness is usually small and the weather condition is good in Tsukuba. We were able to make many measurements with the lidar and aureolemeter in autumn and winter of 1991. As the optical thickness of tropospheric aerosols was superimposed on to the stratospheric optical thickness in aureolemeter data, tropospheric lidar measurements were used to select some cases for clear troposphere. November 25, November 29 and December 13 were selected as clear tropospheric cases.

The columnar size distribution was derived from the aureolemeter measurements. Here, we fitted the derived size distribution to a composite of Junge and log-normal distribution functions using the data in the range from 0.2 to 2.0  $\mu\text{m}$ . Tropospheric aerosols usually have a bi-modal distribution with a mode radius of about 0.2  $\mu\text{m}$  and about 3 - 4  $\mu\text{m}$  (Shiobara et al., 1991). Therefore, the size distribution function adjusted for the range from 0.2 to 2.0  $\mu\text{m}$  represents mainly contribution from the stratospheric aerosol. After the El Chichon eruption, stratospheric aerosols had a mode radius of about 0.5  $\mu\text{m}$ , and therefore, it was possible to distinguish stratospheric size distribution from a tropospheric one. The size distribution observed in late 1991 was similar to that observed after the El Chichon eruption.

The distribution function adjusted is expressed as

$$dV/d \ln r = C_0 + C_1 \exp \left[ -1/2 \left( (\ln r - \ln r_0) / \ln \sigma \right)^2 \right] \quad (3)$$

where  $C_0$  represents concentration of the Junge distribution, and  $C_1$ ,  $r_0$  and  $\sigma$  represent concentration, mode radius and dispersion of log-normal distribution, respectively.

Table 1 shows the parameters  $C_0$ ,  $C_1$ ,  $r_0$  and  $\sigma$  for the three days. Junge mode is usually observed in the troposphere (Shiobara et al., 1991). Here we assume the log-normal distribution mode can be regarded as a representative mode of stratospheric aerosol.

Table 2 shows some parameters such as IBC, optical thickness and  $S_1$  for 532 nm which were calculated from Mie

TABLE 1. LOG-NORMAL MODE DERIVED FROM AUREOLEMETER MEASUREMENTS

Date	$C_0(\text{cm}^3/\text{cm}^2)$ ( $\times 10^{-6}$ )	$C_1(\text{cm}^3/\text{cm}^2)$ ( $\times 10^{-6}$ )	$r_0(\mu\text{m})$	$\sigma$
Nov. 25	0.84	1.2	0.47	1.56
Nov. 29	0.48	2.2	0.58	1.67
Dec. 13	0.34	2.7	0.51	1.62

TABLE 2. OPTICAL PARAMETERS CALCULATED FOR THE LOG-NORMAL MODE DERIVED FROM AUREOLEMETER\*

Date	Optical thickness**	IBC**	S1**	$\zeta$ ***
Nov. 25	0.073	0.0017	43.9	56.2
Nov. 29	0.13	0.0034	36.5	48.6
Dec.13	0.16	0.0040	40.9	53.7

\* refractive index was assumed for 1.44 - 0.00i

\*\* calculated for 532 nm on the basis of log-normal mode shown in Table 1.

\*\*\* the ratio of surface area to backscattering coefficient ( $\text{sr}^{-1}$ )

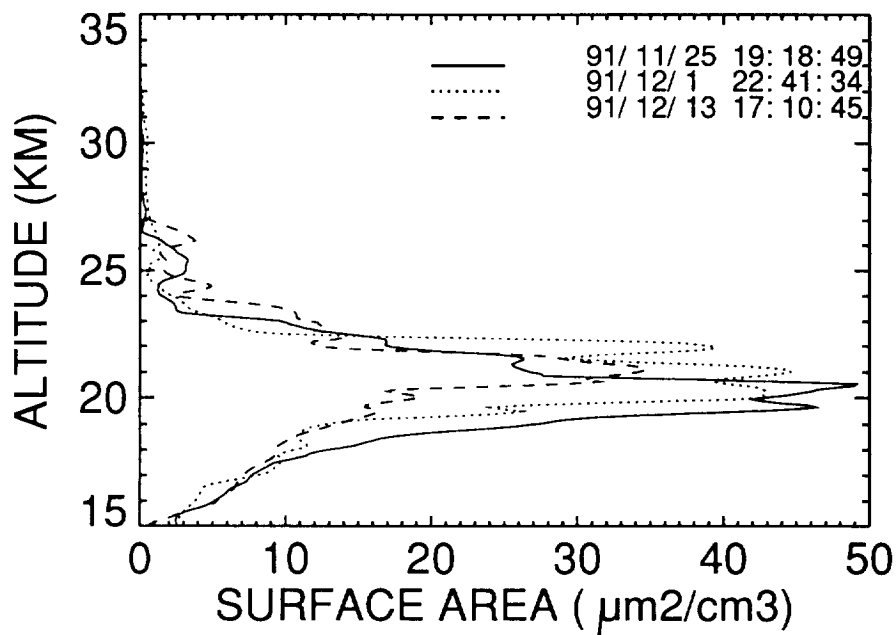


Fig. 2. The surface area of stratospheric aerosols on November 25, December 1 and December 13, 1991 estimated from the lidar and aureolemeter measurements.

theory based on the log-normal distribution shown in Table 1. The refractive index was assumed to be  $1.44 - 0.00i$  on the assumption of 75 % sulfuric acid. The ratio  $\zeta$  of surface area (A) to backscattering coefficients ( $= A/\beta$ ) is also shown. If size distribution of aerosols does not depend on altitude so much, the ratio  $\zeta$  times local value of  $\beta(z)$  means the surface area at altitude  $z$ . Figure 2 shows the surface area profiles ( $\zeta \times \beta(z)$ ) for those three days. These profiles were obtained by assuming  $S_1 = 43.9, 36.5, 40.9$ , respectively according to the calculated values for the log-normal mode (see, Table 2). As there was no stratospheric lidar measurement on November 29, the lidar data on December 1 was compared with the aureolemeter data on November 29.

Even if the size distribution is dependent on altitude,  $\zeta$  in Table 2 should be taken as a vertically averaged value through the stratosphere. The value of  $\zeta = 53$  averaged for the three days should be taken as a representative value for the period during late November to early December 1991.

The calculated values of IBC in Table 2 are not always consistent with those obtained from the lidar data. One of the reasons for this discrepancy is because the lidar and aureolemeter measurements are not carried out simultaneously. Another possible reason is the refractive index used for the calculation. The calculation for refractive index  $= 1.44$  would mislead the results when aerosols include volcanic ash.

In this paper, the Junge mode was assumed for tropospheric aerosols and removed from the aureolemeter data. Tropospheric lidar data should be analyzed in more detail to estimate tropospheric aerosol contribution to the optical thickness observed by aureolemeter in more detail in the future study. Other information on size distribution (multiple wavelength lidar measurements, for example) should be referred for further studies.

## 5. CONCLUDING REMARKS

Lidar measurements over Tsukuba after the Pinatubo eruption on June 12-15 revealed an increase of stratospheric aerosol amount since the end of June. After mid-September, a thick aerosol layer was observed continuously.

Collaborative measurements with the lidar and aureolemeter were carried out and succeeded in estimating the profile of the surface area of aerosols. Ratio  $\zeta$  showing the surface area to backscattering coefficient is about 53 sr<sup>-1</sup> in late 1991. The surface area of Pinatubo stratospheric aerosol was estimated to be about 40  $\mu\text{m}^2/\text{cm}^3$  at the peak of the aerosol layer (about 21 km).

## ACKNOWLEDGMENTS.

We wish to acknowledge our indebtedness to H. Satoh and F. Iida from FIT Inc. for their help in the lidar and aureolemeter measurements.

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