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TRANSPORT OF PINATUBO AEROSOLS TO THE ARCTIC REGION: LIDAR MEASUREMENTS IN ALASKA, WINTER 1991/1992

Yasu-nobu IWASAKA[1], Masahiko HAYASHI[1], Takashi SHIBATA[1], Katsuji MATSUNAGA[1], Masahiro NAGATANI[1], Hiroshi NAKADA[1], Ikuko MORI[1], Tetsuro OJIO[1], Motoo FUJIWARA[2], Eiji AKIYOSHI[2], Satosi YASUMATU[2], Koji YAMAZAKI[3], Kouji KONDOH[4] and Hideaki NAKANE[5]

[1]*Solar Terrestrial Environment Laboratory, Nagoya University, Chikusa-ku, Nagoya 464*
[2]*Faculty of Science, Fukuoka University, Jonan-ku, Fukuoka 814-01*
[3]*Meteorological Research Institute, Nagamine, Tsukuba 305*
[4]*Aerological Observatory, Japan Meteorological Agency, Nagamine, Tsukuba 305*
[5]*National Institute of Environmental Studies, Onogawa, Tsukuba 305*

Abstract: The concentration of stratospheric aerosols observed with a lidar in Alaska increased from December 1991 to March 1992. The measurements suggested that particulate matter injected into the stratosphere through the eruption of Pinatubo (Philippines, June 1991) was transported to high latitudes and descended there. The speed of descending motion of the aerosol layer in Alaska was about 50 m/day, larger than the value in mid-latitudes.

1. Introduction

The global dispersion of aerosols injected into the stratosphere through major volcanic eruptions has become a matter of great concern from the viewpoint of volcanic impact on atmospheric chemistry and transfer of solar radiation.

Heterogeneous processes including polar stratospheric clouds (PSCs) are believed to play an important role in ozone destruction in the polar stratosphere since the formation of PSCs denitrifies the stratosphere and active chlorine compounds survive through surface reaction with PSCs (*e.g.*, SOLOMON, 1990). Sulfate particles are believed to act as nucleation sites of PSCs (*e.g.*, WOFSY *et al.*, 1990). Therefore, it is a matter of great concern whether the dispersion of many sulfate particles forming from volcanic SO_2 to the polar region disturbs polar stratospheric chemistry or not.

Recently, it has been suggested that there is a the possibility that sulfate particles can also denitrify the stratosphere and convert inert chlorine to active chlorine which destroys stratospheric ozone through heterogeneous reactions including sulfate droplets (*e.g.*, HOFMANN and SOLOMON, 1989; BRASSEUR *et al.*, 1990; HANSON and RAVISHANKARA, 1991; FAHEY *et al.*, 1993).

The volcano Pinatubo in the Philippines (15°05′N, 120°11′E) underwent a number of eruptions during June 1991. These climaxed in a massive eruption on June 15, 1991, which injected a large cloud of volcanic debris into the strato-

sphere. SAGEII satellite measurements show that the stratospheric volcanic cloud dispersed into about 70°N latitude by late July (McCORMICK and VEIGA, 1992).

Simulation of the Pinatubo aerosol cloud dispersion using a GCM (General Circulation Model)(BOVILLE *et al.*, 1991) suggested that the volcanic cloud initially drifted westward and expanded in longitude and latitude; in the northern hemisphere there was substantial transport into high northern latitudes by August which continued from August to December.

RANDEL *et al.* (1993) presented global maps of nitrous oxide and water mixing ratios obtained by the Upper Atmospheric Research Satellite, and suggested that 'tongues' of tropical stratospheric air extending out from low into middle and high latitudes could be responsible for transporting significant quantities of stratospheric air.

Here lidar measurements of the Pinatubo-enhanced stratospheric aerosols at Poker Flat, Alaska (64°49′N, 147°52′W) in winter 1991/1992 are described, and the geochemical budget of volcanic aerosols in the polar region—accumulation of particulate matter which is due to the inflow of particulate matter from middle and low latitudes through long range transport of stratospheric air and the particle descending from the stratosphere to the troposphere, is discussed.

2. Lidar Measurements

Lidar measurements of stratospheric aerosols were made in mid December 1991 at Poker Flat, Alaska. The lidar system is composed of a Nd-YAG laser (0.5 J/pulse, 532 nm, 10 Hz), a photomultiplier detector, and a 35 cmϕ cassegrain telescope receiver (IWASAKA *et al.*, 1993). The basic lidar signal represents the total scattering containing both molecular and aerosol components. To evaluate the aerosol component, the matching point where an aerosol-free atmosphere was expected was chosen to be above about 30 km from radiosonde data at Poker Flat and weather maps prepared by Japan Meteorological Agency.

Figure 1 shows the vertical profiles of scattering ratio, which is defined by

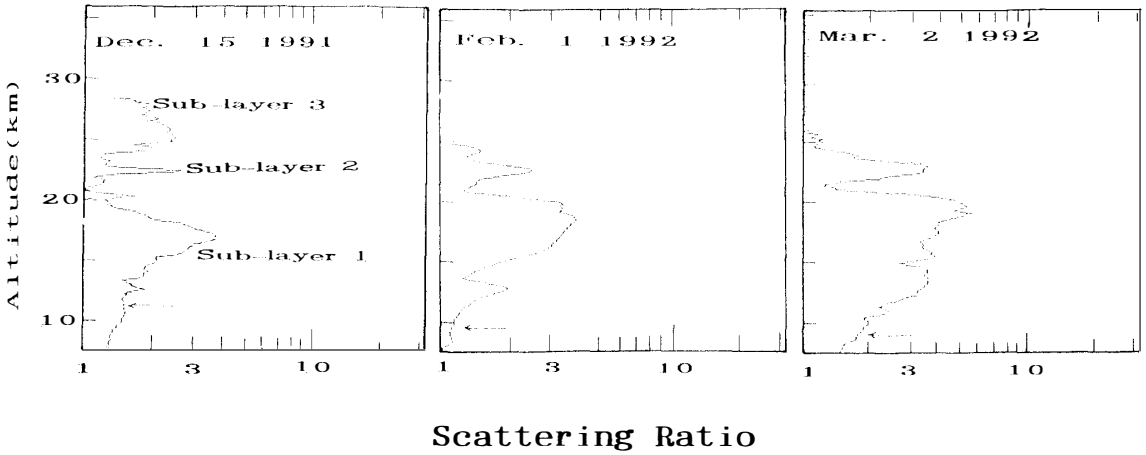


Fig. 1. Scattering ratio measured at Poker Flat, Alaska on December 15, 1991 (left), February 1, 1992 (center), and March 2, 1992 (right). The local tropopause height is shown by arrows '←'.

$[\beta 1+\beta 2]/\beta 1$, where $\beta 1$ and $\beta 2$ are backscattering coefficients of air molecules and aerosols, obtained at Poker Flat, Alaska in winter 1991/1992. The local tropopause height was estimated from both the meteorological sonde measurements at Fairbanks airport and the weather map prepared by the Japan Meteorological Agency.

Enhanced aerosol layers were identified at 15–20 km and 22–23 km during the observational period. The profile on December 15, 1990 showed an additional enhanced layer at 25–27 km.

3. Sporadically Enhanced Aerosol Layer at about 25 km

The profile of December 15, 1991 showed an enhanced layer at 25–27 km (about 16 km above the local tropopause)(layer 3 in Fig. 1).

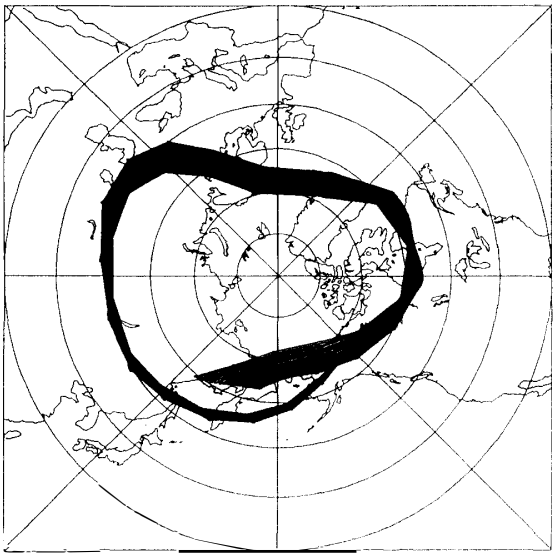
Enhancement of stratospheric aerosols in the cold winter polar stratosphere (Polar Stratospheric Clouds; PSCs) has become a matter of great concern since PSCs control polar ozone destruction (ozone hole) through heterogeneous reactions. According to the measurements by lidar (*e.g.*, IWASAKA, 1986) and satellite (*e.g.*, MCCORMICK *et al.*, 1985), PSCs frequently form in the upper aerosol layer in their early stage.

From the temperature distribution of December 1991 it is suggested that the stratospheric temperature of December 1991 was too warm to form PSCs at about 25 km, and therefore the enhancement of December 15, 1991 was possibly not due to temperature cooling to the frost point temperature but to other processes.

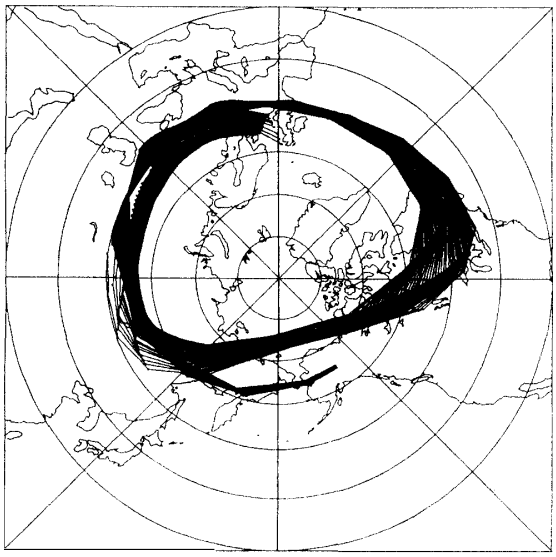
The airmass trajectory on the isentropic surface corresponding to sub-layers 1, 2, and 3 of the measurements of December 15, 1991 was calculated using the 3-dimensional twice-daily JMA data set. The time step for trajectory computations is one hour. The computation procedure has been already described in detail in other papers (YAMAZAKI, 1987; YAMAZAKI *et al.*, 1989). The 10 day backward trajectories shown in Figs. 2a–2c, indicate that the layer of 24–27 km on 15 December is due to an air mass which rapidly moved from low latitude to the polar region.

Therefore, a possible interpretation for the noticeable enhancement of the sub-layer 3 on December 15, 1991 is the rapid and long-distance transportation of air mass containing considerable volcanic material from low to high latitudes. RANDEL *et al.* (1993) describe the erosion of the subtropical barrier, where stratospheric material was pulled off the outer edge, and mixed into middle and high latitudes from UARS data analysis.

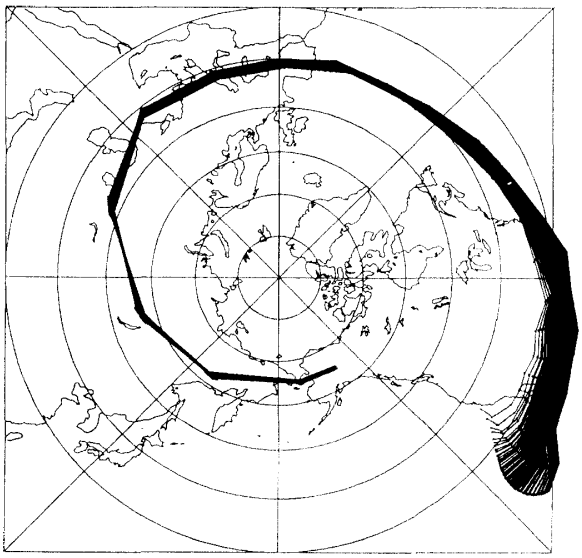
Present sporadic aerosol enhancement associated with the long range transport of stratospheric air from low to high latitudes contributes to global dispersion of volcanic material; this may correspond to the erosion of the subtropical air suggested by RANDEL *et al.* (1993). It is necessary to observe stratospheric aerosols at high latitude more often in order to understand the long-range transport and global dispersion of stratospheric particulate matter.



(a) Initial time, 0800 1991 Dec. 16 (GMT), Initial height, 17 km (sub-layer 1).



(b) Initial time, 0800 1991 Dec. 16 (GMT), Initial height, 22.5 km (sub-layer 2).



(c) Initial time, 0800 1991 Dec. 16 (GMT), Initial height, 25 km (sub-layer 3).

Fig. 2. Trajectory of airmass containing volcanic aerosols (10 days backward trajectory).

4. Accumulation and Descent of Particulate Matter in the Polar Region

During the observational period, noticeable increase in aerosol content was observed in the upper and lower parts of the aerosol layer. However, the mixing ratio of the layer peak observed at about 17 km on December 15 did not increase, although other layer peak showed noticeable increase. The rates of increase of mixing ratio (R-1) were about 42.2, 0.27, and 5.93%/day at 20, 17, and 13 km, respectively.

One possible explanation for the increase observed in the lower part of sub-layer 1 is descending motion of stratospheric aerosols from the upper part of sub-layer 1.

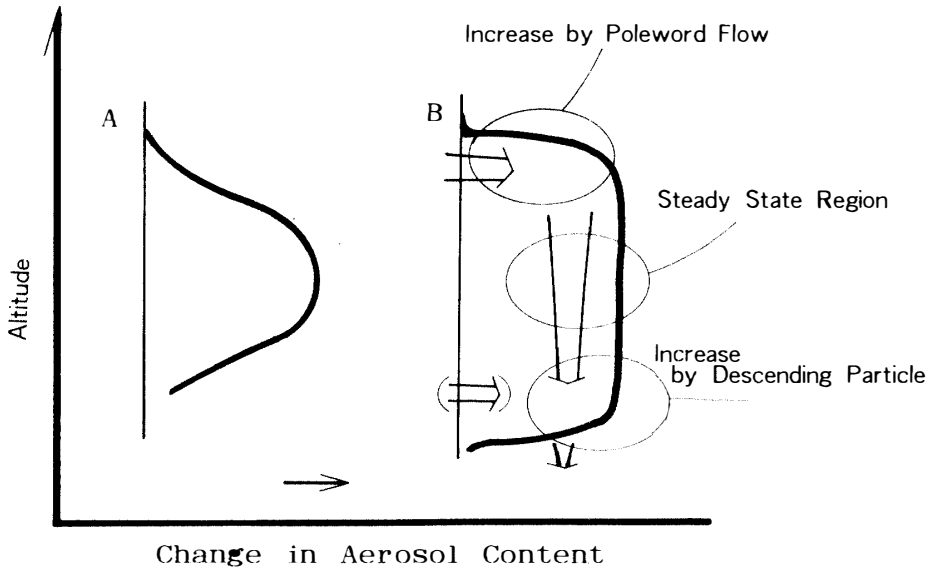


Fig. 3. Horizontal transport and descending motion of particulate matter. In upper aerosol layer, aerosol particle content increases through transport from mid to high latitudes. Near the aerosol layer center, descending motion and horizontal transport produce steady state aerosol load. In the lower layer, aerosol particles may accumulate owing to the decrease in descending speed of particles.

The centroid height of the aerosol layer, H_c, can be defined by

$$H_c = \int z\beta 2\ (z)\ \mathrm{d}z / \int \beta 2\ (z)\ \mathrm{d}z.$$

The value of H_c changed from 17.2 to 14.8 km from December 15, 1991 to March 2, 1992, corresponding to descending motion of about 52 m/day. This value is apparently larger than the results obtained over Japan in the period of the El Chichon disturbance of 1982/1983. According to the lidar measurements at Nagoya, the descending speed was about 20 m/day in the early decay phase (see Fig. 4 in HAYASHIDA and IWASAKA, 1986). When we use this speed for the equation, $F(Z) = V(Z)\cdot\beta 2(Z)$ where F and V are flux and descending speed of H_c, in order to estimate roughly the rate of sedimentation loss, we obtain $F = 4.2\times10^{-5}\mathrm{sr}^{-1}\mathrm{day}^{-1}$ above the tropopause.

More detailed calculations are necessary to estimate accurately loss of volcanic aerosol from the polar stratosphere, since those estimations are based on very limited observations.

The quasi-steady state of aerosol content near 17 km can be explained by balance between inflow from mid-latitudes and sedimentation loss, as shown in Fig. 3.

The polar stratosphere functions as a sink of volcanic material through long range transport from mid latitudes to the polar region and sedimentation of particle matter from the polar stratosphere.

5. Possible Growth of Descending Sulfuric Acid Droplets near the Tropopause

The temperature distribution provided by the Japan Meteorological Agency has a minimum of $-50 \sim -60$°C above the local tropopause during the observation period. Sulfuric acid droplets may grow through dilution during their descending motion from the warmer region to the cooler region. The equilibrium concentration of sulfuric acid in solution changes from about 75%(wt) of H_2SO_4 to 60% corresponding to a temperature drop from about -40 to -60 °C. Those bring an increase of aerosol mass, about 1.2–1.3 times, and the particle sedimentation velocity increases about 1.13–1.19 times. Consequently, downward transport of aerosol material will be activated by dilution growth of droplet in the spring polar stratosphere. It is necessary to know the detailed water vapor concentration in order to discuss the dilution effect on descending particles accurately.

6. Conclusion

Lidar measurements at Poker Flat, Alaska in winter 1991/1992 suggested that the stratospheric aerosol content extremely enhanced. This enhancement is possibly due to the eruption of Mt. Pinatubo (June 1991), considering many lidar measurements and balloon measurements made world-widely are showing the rapid enhancement of stratospheric aerosols after the eruption.

Rapid transport of volcanic materials from low latitudes to Alaska is considered to correspond to the sporadic enhanced layer of aerosols at 25–27 km on December 15, 1991. More observations are necessary to assess the contribution of such transport to global dispersion of stratospheric material.

Descending motion of aerosols from the upper to the lower part of the stratospheric aerosol layer, and from the stratosphere to the troposphere, can be suggested on the basis of the fast descending speed of the centroid height of the aerosol layer, about 52 m/day in the polar region, which seems to be larger than in the early stage of volcanic disturbance in middle latitudes.

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