Balloon-Borne Measurements of Atmospheric Aerosol Particles at Beijing, China in Summer of 1993: Morphology, Size and Concentration

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Received 21 January 1997
Accepted 10 November 1997

Abstract—Tropospheric and stratospheric aerosol particle size, number density, and morphological feature were observed in August, 1993 at Beijing, China (40°N, 116°E). Irregular shape particles, possibly soil particles, were frequently observed in the free troposphere on the basis of scanning electron microscopic measurements of individual particles collected on the surface of carbon-coated nitrocellulose (collodion) films with a balloon-borne particle counter. In the stratosphere the particles with 'satellite structure' which has been recognized as a typical feature of sulfuric acid droplets on carbon deposited surface were major particles. Ammonium sulfate particles were frequently identified in the mid troposphere. The number density of the particles with their diameter, d, larger than 0.4 μm was about 3 particles/cm³ near the density peak of the stratospheric aerosol layer, suggesting that the disturbance due to the volcanic eruption of Mt. Pinatubo (1991; Philippine) remained in the stratosphere. Less uniformity in number density profiles of the particles with d ≥ 0.4 μm was observed in the troposphere, suggesting the effect of dynamical air motions with various spacial scales over the Asian continent in summer on aerosol density distributions.

Key Words: Stratospheric Aerosol, Tropospheric Aerosol, Asian Dust Particle, Balloon-Borne Measurements.

Introduction

Asian continent has been recognized as an important source area of atmospheric soil particles which are globally diffused over the East Asia-West Pacific region by westerly wind and give large influence on environment and climate in this region through scattering effect of solar radiation and terrestrial radiation, long-range transport of particulate matter on geochemical cycle of various metals and other constituents, and contribution of particles as nuclei of cloud condensation.

Recent estimations show that noticeable radiative forcing of atmospheric aerosols can occur over the East Asia and the West Pacific region. Many investigations suggested that soil particles and other atmospheric constituents were frequently transported from the Asian continent to Japan, Alaska, and the North-West Pacific Ocean in the free troposphere by westerly wind. Electron microscopic measurements of particles collected in the free troposphere over Japan suggested the possibility that the surface of soil
particles played as site of chemical reactions between atmospheric gases such as SO$_2$ and NO$_x$ and particulate matter\textsuperscript{5}).

Additionally, there is a great concern about anthropogenic perturbations of atmospheric particles due to increase sulfur loading with expanding industrial activities and soil particle loading with desertification in the East Asia region (e.g., Scientific Report published by IPCC \textsuperscript{7}).

Although the effect of atmospheric particles on radiation, clouds, precipitation, and overall climate in the East Asia and West Pacific region have been recognized, acquired knowledge pertaining to causes and effects so far is inadequate and information has been based mostly on measurements made in the surface atmosphere but few measurements in the free atmosphere, owing to difficulty of measurements, where long-range transport of atmospheric constituents is expected to be active.

The purpose of this paper is to present the comprehensive measurements on the vertical changes in particle size-number density and morphological feature of individual particles in the Asian continental troposphere and stratosphere on the basis of the balloon-borne measurements done in August and September 1993 at Xianghe, Beijing (40°N, 116°E) (Fig. 1). This is the first balloon-borne measurements done in the Asian region describing behavior of aerosols in the continental atmosphere.

**Observation**

During the balloon-borne measurements made at Xianghe, Beijing (40°N, 116°E) on August 22, 1993, a particle counter and a particle impactor were used to observe aerosol size-number density and individual particle morphology in the troposphere and stratosphere, respectively.

The air pump used in the counter and the impactor, in order to keep the flow rate of the sampled air constant, has the function of feedback making compensation for decrease in flow rate of sampled air under the condition of low atmospheric pressure. The efficiency of the air pump was tested and regulated in the low pressure chamber in order to see that the function of the pumps were effective under the stratospheric conditions. According to the laboratory experiments \textsuperscript{8)}, the efficiency of the pump was 100.0 ± 4.6\% under the condition of atmospheric pressure of 1000 hPa-5 hPa. Additionally efficiency of total system of the balloon-borne optical particle counter and the particle impactor also were tested.

![Fig. 1 Balloon-Borne measurements of atmospheric aerosols (August 22, 1993; Balloon Observation Site at Xianghe, Beijing, China). The particle counter and particle impactor were installed in the gondola containing hygrometer, aneroid barometer, electronics of telemetering, battery, and ballast (left). The gondola was hung 150 m below the balloon to minimize contamination from plastic sheet of the balloon (right). After sampling aerosol particles the gondola was recovered.](image-url)
Rotated by stepping motor

Jet nozzle

TEM Grid

Schematic diagram of particle impactor

**Fig. 2** Schematic diagram of particle impactor. Grids of electron microscopic measurement are set on the surface of rotating disk. The disk is rotated by a stepping motor and each grid is exposed (3 minutes) by turn to the air introduced with an air pump. During the exposure the disk is kept not to rotate in 3 min.

and regulated in the low pressure chamber which simulated the upper tropospheric and lower stratospheric condition before the balloon ascending.

The volume of sampled air was estimated comparing both those experimental data obtained before the balloon-borne measurements and the monitored values during the balloon ascent and descent (rotation number of air pump per unit time, temperature inside of box containing the counter and the impactor, and atmospheric temperature).

Aerosol particles were collected on electron microscopic grids covered with carbon-coated nitrocellulose (collodion) with using a balloon-borne particle impactor (2-mm diameter and the air flow rate of 22 l/min). Those grids were set on the surface of the disk, as shown in Fig. 2. According to Ranz and Wong 9), the particle size for collection efficiency of 50% was 0.25 µm for the density of particulate matter = 2.6 g/cm³. After the recovery of the balloon, the particles collected were installed in the sealed box to minimize the effect of contamination of particulate matter collected. The particles collected on the surface of the grids were coated with carbon alloy to observe the particles with a scanning electron microscope (AKASHI Beam Technology Ltd. ABT-55).

In **Fig. 3**, time-height cross section of flight path of the balloon (August 22, 1993) is shown. The balloon reached a height of 35 km about 1 hour 30 minutes after the balloon launching.

**Fig. 3** Time-height cross section of flight path of the ascending balloon used for the measurement in August 22, 1993.

**Fig. 4** Number concentration of the particles with their diameter larger than 0.4, 0.5, and 2.0 µm measured at Xianghe, Beijing (40° N, 116° E) in August 22, 1993.

Particle size and number density were measured with a balloon-borne optical particle counter. The detailed description about the particle counter used here was already given in other paper 8), and the
outline of the counter only is described here. The counter contains halogen lamp as light source, light scattering chamber, photodiode as detector of the light scattered by the particles, air pump used to introduce aerosols, electronics, and battery used for electric power source.

The vertical changes in concentration of aerosol particles sized at diameter, $d \geq 0.4$, $0.5$ and $0.6$ $\mu$m are shown in Fig. 4. The height of the local tropopause was shown with an arrow (15.6 km). Noticeable density peak of the particles with $d \geq 0.4$ and $0.5$ $\mu$m was found at 22 km and minimum of density at 13 km. The density of the particles with $d \geq 2.0$ $\mu$m decreased above 14 km.

Therefore large difference in aerosol size distribution is present between the regions above and below 14 km, and it suggested that stratospheric aerosols contained only few supermicron size particles.

The peak density of particles with $d \geq 0.4$ $\mu$m in the stratosphere (3 particles/cm$^3$ at 22 km) was apparently larger than the values observed during the quiet stratospheric period without volcanic disturbance $^{11-14}$. According to those previous measurements, the number density of the particles with $d \geq 0.3$ $\mu$m was in the range of 0.5–1.5 particles/cm$^3$ during the calm stratospheric period.

Below the tropopause the size-number density of atmospheric particles showed large irregular variations. The density peak of the aerosols with $d \geq 0.4$ $\mu$m found in about 8 km, 5 km, and others seems to well correlate to the peak of the particles with $d \geq 0.5$ $\mu$m. However, the feature in the profile of density of the particles with $d \geq 2.0$ $\mu$m is apparently different from those profiles. The density profiles of the particles with $d \geq 2.0$ $\mu$m seems to be much uniform than the profiles of the particles with $d \geq 0.4$ and $0.5$ $\mu$m. In the previous measurements made in North American area, $^{12, 15}$ density of the particles with $d \geq 1.9$ $\mu$m appeared less uniform and increased from about $10^{-5}$ to $10^{-1}$ particles/cm$^3$ with decreasing height, which showed large variations in the mid and lower troposphere.

Soil particles with the size of super micron, as described in Discussion, were frequently identified in the free troposphere from the observation of morphological feature of individual particles with the scanning electron microscope. Those electron microscopic measurements seem to be well corresponding to the results of the particle counter.

![Fig. 5](image-url) ELECTRON MICROGRAM OF THE PARTICLES COLLECTED ON THE SURFACE OF CARBON FILM.
(a) : 19.4–20.6 km,  
(b) : 12.1–13.9 km,  
(c) : 4.2–5.7 km.
showing that the particles of $d \geq 2.0 \mu m$ diffused to the free troposphere.

Electron microgram of the particles collected at the height range of 19.4-20.6 km are shown in Fig. 5 (a). Typical feature of particle morphology collected on carbon film in the lower stratosphere in the figure is ‘satellite structure’ which means the structure having many small dots surrounding the relatively large dot (see ‘A’). This feature is known as one of the typical structure which characterizes the particles containing sulfuric acid solution from many laboratory experiments and field measurements e.g. 10, 16).

Electron microgram of the particles collected in the height of 12.1-13.9 km (Fig. 5 (b)) also shows that there are many sulfuric acid particles, but irregular shape particles (see ‘B’ ) are much frequently found in this height range than the region of 19.4-20.6 km. Additionally another kind ‘particles with satellites’ are identified in this region (see ‘C’ ). The ‘C’ type particles, of course, have satellites but number of satellite seems to be smaller than the ‘A’ type particles, and particulate material in center is apparently larger than ‘A’ particles.

The electron microgram of the particles collected in the height of 4.2-5.7 km shows frequently external mixing state of irregular shape particles (‘B’ ) and spherical particles and/or irregular with a few satellite (‘C’ ), and satellite structure like type of A is not typical morphology of particles (Fig. 5 (c)) in those heights.

**Discussion**

The eruption of Mt. Pinatubo (June 1991, Philippine) caused large enhancement of stratospheric aerosols through the injection of lots of volcanic materials such as SO$_2$ gas into the stratosphere. Following the eruption, large enhancement of stratospheric aerosol loading was reported by many investigators who suggested the global diffusion of volcanic aerosols in the stratosphere e.g. 17-21).

Long term trend of the disturbed stratospheric aerosols by the eruption, on the basis of the lidar measurements made in Toyokawa (35°50’N, 137°20’E), showed that the volcanic disturbance of the stratospheric aerosols remained even 2 years after the eruption and did not recover to pre-Pinatubo levels in summer of 1995 (Fig. 6). Therefore it is reasonable to consider that the number concentration of stratospheric aerosols measured at Beijing in summer of 1993 was also in the levels enhanced by the Pinatubo eruption and a little larger than the values observed in the period without volcanic disturbance. The effect of Pinatubo eruption on the stratospheric aerosols was suggested also by Zhou et al. 22) on the basis of the number-size distributions measured with a particle counter in summer and fall of 1993.

Morphological feature of individual Asian dust particles have been discussed by many investigators on the basis of an electron microscopic measurements of the particles collected in the boundary atmosphere e.g. 5, 10, 23). According to those investigations irregular shape particles were frequently found in the air mass transported from the Asian continent and suggested that those particles were soil particles originating from desert areas of the Asian continent.

During the observational periods, large scale cumulus clouds were frequently observed in the east area of the Asian continent suggesting appearance of active air convection over Beijing area. Such active convective motions over the Asian continent in mid-summer seems to be one of possible processes to inject soil particles into the free troposphere from near the ground. From those previous studies on particle morphology, the irregular shape particles like ‘B’ particles shown in Figs. 5 (b) and (c) can

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**Fig. 6** Integrated backscattering coefficient of stratospheric aerosols, $\int \beta (z) dz$: integration is made in the heights from the tropopause to the top of the stratospheric aerosol layer, measured with a lidar at Toyokawa, Japan (35°50’N, 137°20’E). The integrated backscattering coefficient is recognized as the values corresponding to the load of stratospheric aerosols (e.g., Iwasaka et al. 1995).
be suggested to be soil particles. In also Fig. 5 (a), irregular shape 'B' particle is found, but 'B' type particles were very rare in the stratosphere. The large variations in the vertical profiles of aerosol size-number concentration also can be due to such dynamical air motions.

It has been suggested, on the basis of laboratory experiments and field measurements, that sulfuric acid droplets collected on the surface of carbon thin film always show satellite structure, as shown in 'A' particles of Fig. 5 (a). Those measurements showed the possibility that sulfuric acid particles formed through photochemical reactions of SO2 in the atmosphere. Even when sulfuric acid droplets are partially neutralized by atmospheric ammonia through reactions such as H2SO4 (liquid) + NH3 (gas) → HNH4SO4, satellite structure is identified. However, difference in morphological feature of satellite structure is noticeable between sulfuric acid droplets and partially neutralized droplets by ammonia: number of satellite of partially neutralized droplets is apparently smaller than that of sulfuric acid droplets, and size of the material in the center is larger than that of sulfuric acid droplets. Type of 'C' particles has been recognized as morphology of sulfate particles partially neutralized by ammonia.

Comparing the external mixing state of sulfuric acid droplet-ammonium sulfate particles in the mid troposphere (Fig. 5 (c)) and the upper troposphere (Fig. 5 (b)), ammonium sulfate particles were much frequently observed in the mid troposphere than the upper troposphere. Therefore possible interpretation of atmospheric ammonium sulfate particles is that the neutralization of sulfuric acid droplets by anthropogenic ammonia is relatively active in the mid troposphere in comparison with the upper troposphere. Another possibility is that the diffusion of ammonium sulfate particles from the boundary atmosphere to the mid troposphere through convective motion which is very active in summer season.

Conclusion

Aerosol particle size, concentration, and morphological feature were measured in the troposphere and stratosphere in summer of 1993 at Beijing, China, and following facts were revealed,

1) Stratospheric aerosols were mainly composed at sulfuric acid droplets, and their number concentration (in the diameter range \( \geq 0.4 \ \mu m \)) of the peak of the stratospheric aerosol layer was 2-3 times larger than the concentrations in the period without volcanic disturbance. Combining the measurements with the decay trend of the disturbed stratospheric aerosol layer by the eruption of Pinatubo of 1991, it is suggested that the stratospheric aerosol layer measured here was disturbed by the Pinatubo eruption.

2) Irregular shape particles, possibly soil particles, were frequently identified in the free troposphere on the basis of electron microscopic measurements of the individual particles collected with a balloon-borne particle impactor.

3) From the morphological feature of electron microgram of the particles collected on carbon thin film, it is speculated that conversion of sulfuric acid droplets to ammonium sulfate particles (neutralization of sulfuric acid droplet with atmospheric ammonia) actively occurred in the lower troposphere.

4) Less uniformity in vertical distribution of particle concentration in the troposphere is possibly due to active convection in the summer troposphere over the Asian continent. However, the present studies are due to only the measurements in summer of 1993. Further measurements are desired to understand seasonal changes in aerosol size and density and changes in chemical-physical characteristics of atmospheric particulate matter in the continental atmosphere. Measurements of dynamical effect on vertical distribution of the particles originated in the ground also is desired.

Acknowledgement

This research was supported by the fund of Japan Ministry of Education, Science, Sports and Culture (International Science Research Program 06041049). Technical support staff members of Balloon Observation Site, Xianghe, Beijing kindly helped us during the balloon flight operations.

References


