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# Transport of Asian dust (KOSA) particles; importance of weak KOSA events on the geochemical cycle of soil particles

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ABSTRACT

Lidar measurements and airborne-particle collections were made during KOSARP 87 ('KOSA' Research Program of Nagoya University; April–May 1987) at Nagoya (35°N, 137°E) to investigate the vertical distribution of Asian dust (KOSA) particles and the transport of these particles. According to the lidar measurements, the highly concentrated particle layers with large depolarization ratio were frequently in the range from about 2 km to about 6 km.

Electron microscope observations on the morphology of individual particles in the height range from near the ground to about 4400 m suggested that the particle layers contained many soil particles. It is reasonable to consider that KOSA particles were very frequently transported from Asian desert areas to the islands of Japan in the middle troposphere, even when the effect of the KOSA was not detected near the ground. This 'background KOSA' has concentrations of about $1.9 \sim 25\ \mu g/m^3$ at the layer peaks and one order of magnitude smaller than the values of severe KOSA. However, the contribution of the 'weak KOSA' to the global budget of soil particles is not negligible since the frequency of occurrence of 'weak KOSA' is high. The reaction of soil particles sampled on the vapour-deposited Ca thin-film suggested that some of the particles were coated by water or solution containing $SO_4^{2-}$. Such particles can absorb various atmospheric gases, and therefore the KOSA particles can play an important rôle in the geochemical cycle of many chemical constituents, as chemical reaction sites in the atmosphere and as carriers of the chemical products.

## 1. Introduction

The transport of Asian dust storm particles in the atmosphere has attracted the attention of many investigators. Braaten and Cahill (1986), and Darzi and Winchester (1982) observed significant increases in atmospheric particulate load at Mauna Loa Observatory which was due to the long-range transport of soil particles from eastern and central Asia. Many investigators suggested the important contribution of Asian dust particles on the sedimentary geochemistry of the Northern Pacific Ocean (e.g., Duce et al., 1980; Prospero, 1981; Tsunogai and Kondo, 1982; Uematus et al., 1983). The Asian dust events well-known as 'KOSA' in Japan, are frequently observed in spring.

When a severe KOSA attacks the islands of Japan, noticeable atmospheric disturbances such as an appearance of yellow sky, a decrease in visibility, etc., are observed. Extensive chemical analysis of the particles sampled near the ground surface and sea surface have been extensively to study the deposition rate of chemical elements contained in soil particles through the KOSA events (e.g., Duce et al., 1983; Tsunogai et al., 1985). However, studies on the soil particles sampled in the free atmosphere are very limited.

Satellite photography was useful to study the spatial scale of the KOSA events and their temporal changes on a global scale (Murayama 1980, Iwasaka et al., 1983). Severe KOSA events have been treated directly as a subject of investigation on the long-range transport of soil particles from the Asian continent to the Pacific Ocean. The KOSA episode of mid-Apil, 1979 was a good example. Lidar measurements showed that many KOSA particles travelled at about 5 km over Japan during this KOSA event (Iwasaka et al., 1983). Merrill et al. (1985) made isentropic trajectory analyses for the KOSA event of April 1979, and suggested that the main transport path which caused a noticeable increase in atmospheric soil particles over the northern Pacific Ocean was at 305–315°K in spring.

However, few observations have been made for weak KOSA events. To understand the global cycle of soil particles, measurements on only severe KOSA events are not sufficient and information on weak KOSA events is necessary. Satellite measurements were very useful for studies on the global dispersion process of KOSA events, but not for the weak KOSA, since the soil particle content is too low to be detected by satellite.

Sensitivity of lidar is apparently higher than satellite, and thus a lidar measurement should be effective for weak KOSA events. The KOSA particle layers were frequently observed in the height range of approximately 2 km–6 km in spring even when the atmospheric phenomena related to the KOSA layers were hardly observed near the ground surface (of course, meteorological observatories gave no report on KOSA for such atmospheric conditions). Here we call such KOSA events 'background KOSA' or 'weak KOSA'.

The background KOSA can give an important effect on a global geochemical cycle of soil, since its appearance is extremely frequent in comparison with severe KOSA. We present here lidar measurements on particulate matter in the mid and low troposphere during KOSARP 87 ('KOSA' Research Program of Nagoya University; April–May, 1987) at Nagoya (35°N, 137°E), and electron micrographs of individual particles collected on the surface of vapor-deposited films of Ca or C during the aircraft measurements (300 m–4400 m).

## 2. Observations

The mixing ratio and the depolarization ratio of particulate matter were observed by a lidar from about 700 m to 7500 m at Nagoya during KOSARP 87 periods. Particle collection was also made by an aircraft from near the ground to about 4400 m height to observe the particle shape and to test the chemical composition of the particles using an electron microscope.

### *2.1. Lidar measurement*

The main characteristics of the lidar used here were described by Iwasaka et al. (1985). A scattering ratio is defined by

$$R = [B_1 + B_2]/B_1,$$

where $B_1$ and $B_2$ are back-scattering coefficients of air molecules and aerosol particles, respectively. The value of $[R-1] = B_2/B_1$ can be recognized as the mixing ratio of particulate matter. To obtain the value of $B_2$, the "matching method" (e.g., Russell et al., 1976) was used, and the correction of $B_2$ was made taking into account attenuation of laser beam intensity due to extinction. The depolarization ratio of back-scattered light is a good parameter for identifying the non-spherical particles (e.g., Iwasaka, 1986). The depolarization ratio is given by

$$D = B_{2\perp}/B_{2\parallel},$$

where $B_{2\parallel}$ and $B_{2\perp}$ are back-scattering coefficients of particles measured at the polarization plane parallel to the polarization plane of the emitting laser beam and the coefficient measured at the cross polarization plane, respectively. If the particles are spherical, as sulfuric acid droplets, $D = 0$ is expected (e.g., Iwasaka, 1986).

Figs. 1a, b show the vertical profiles of the scattering ratio and the depolarization ratio measured by the lidar during the KOSARP 87, respectively. The heights of potential temperature of 290°K, 300°K, and 310°K in Fig. 1a are based on the radiosonde measurements at Hamamatsu (about 100 km northeast of Nagoya).

### *2.2. Chemical composition and morphology of particles*

Aerosol particles were collected in the range of approximately 300 m–4400 m by an impactor with cutoff of 0.5 $\mu$m aerodynamic diameter

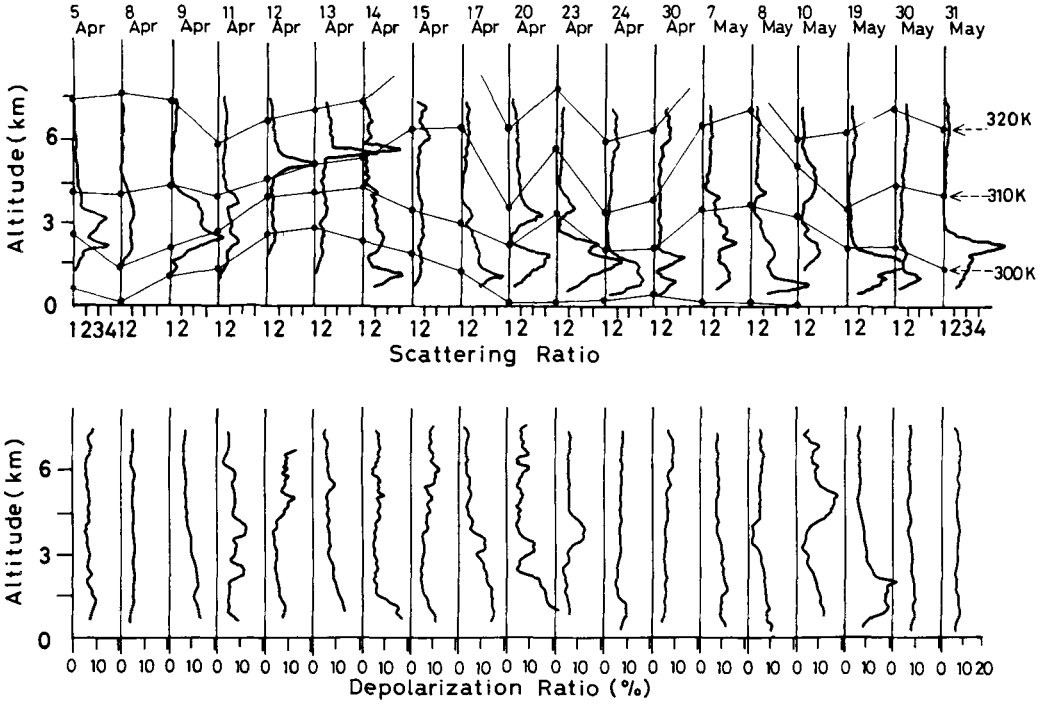



*Fig. 1.* Scattering ratio (upper figure) and depolarization ratio (lower figure) measured by a lidar at Nagoya (35°N, 137°E) during the KOSARP 87 period. The heights of potential temperature of 290°K, 300°K, 310°K and 320°K are estimated using the radiosonde measurements at Hamamatsu (about 100 km northeast of Nagoya).

which was mounted on the aircraft (Cessna 402B). The aircraft measurements and the flight path are shown in Fig. 2. The distance between Nagoya (A) and Wakasa Bay (B) is about 130 km. Sample air was introduced into a cabin of the aircraft through the isokinetic decelerator. Particles were collected on a carbon-covered nitrocellulose film and a vapor-deposited film of Calcium. Before the particle collection, the particles passed through a diffusion drier (relative humidity $\leqq$ about 10%). As described by Ono et al. (1983), sulfuric acid solution can be identified by $CaSO_4$ crystals produced on the film surface through chemical reaction between $H_2SO_4$ and Ca.

Fig. 3a, b show transmitting electron micrograms (TEM) of the particles collected on carbon film during level flights of 2250 m and 4350 m at 9 April 1987. Many irregular shape and electron dense particles were collected at these heights. Comparing these with the TEM imagery of KOSA particles sampled near the ground during the KOSA event of mid-April 1979 (Okada et al., 1987), many similar points were found concerning the morphology of particles. The existence of the electron-dense particles with irregular shape corresponds reasonably to the lidar measurements showing an aerosol layer with high depolarization ratio at this height.

Fig. 4 is the TEM imagery of the particles collected on the calcium films during the flight at 4350 m of 23 April. It is worth noting that TEM imagery shows a trace indicating the existence of water surrounding the electron-dense particles. A calcium thin film is very sensitive to moisture, and therefore the trace easily forms on the calcium film surface. Measurements from meteorological sondes at Hamamatsu showed that the relative humidity was about 75% in the vicinity of the aerosol layer peak at 23 April.

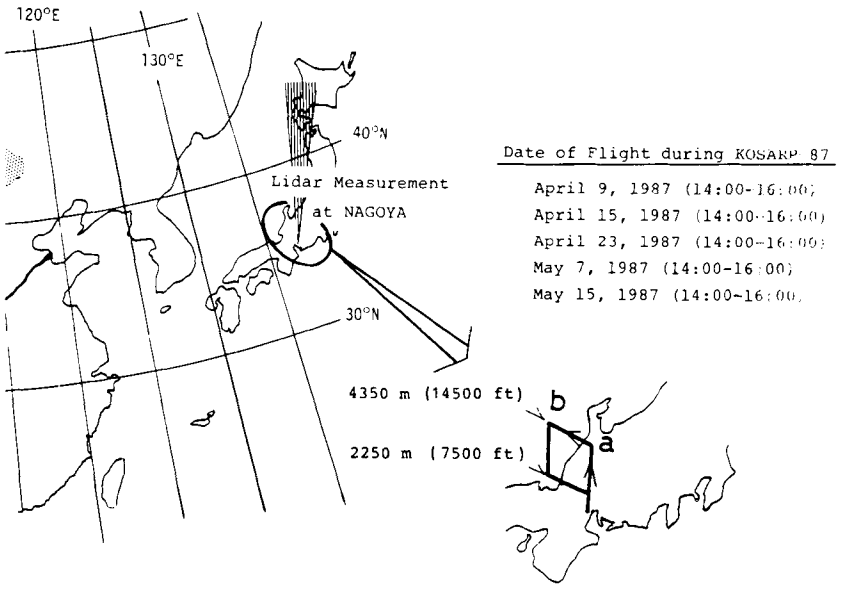



*Fig. 2.* Flight path used for aerosol sampling. The distance between Nagoya (a) and Wakasa Bay (b) is about 130 km. Particles were collected on carbon and calcium thin surface by an impacter.

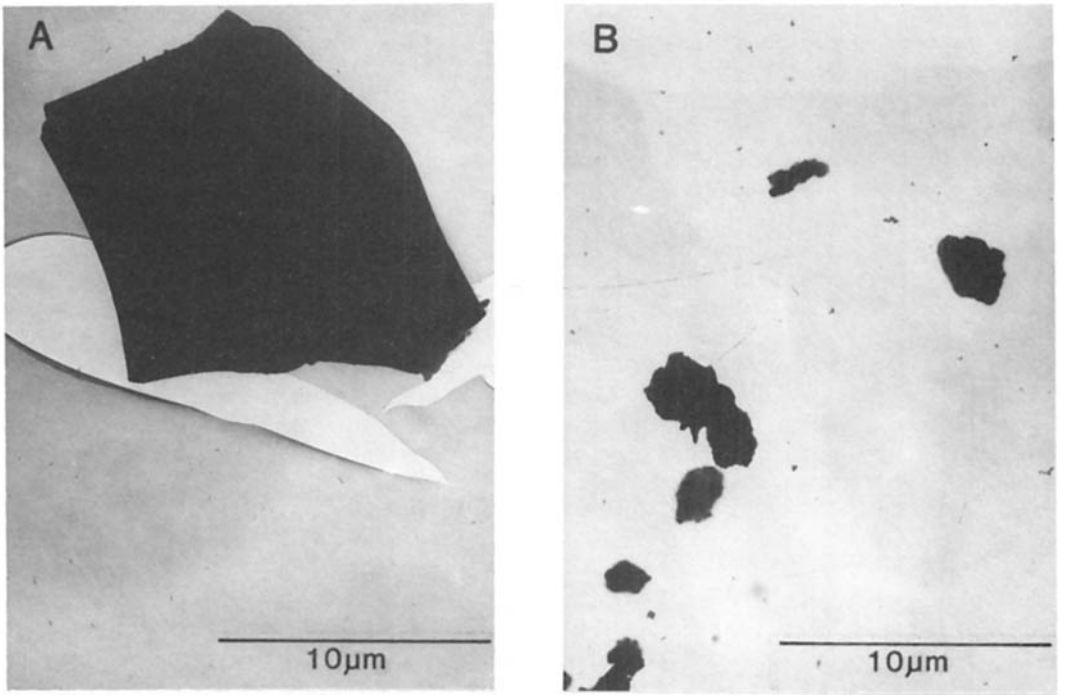



*Fig. 3.* Electron micrograph of particles collected on carbon film. The particle collection was made at 4350 m (A) and 2250 m (B) heights during the flight of 9 April (14:00–16:00 LT).

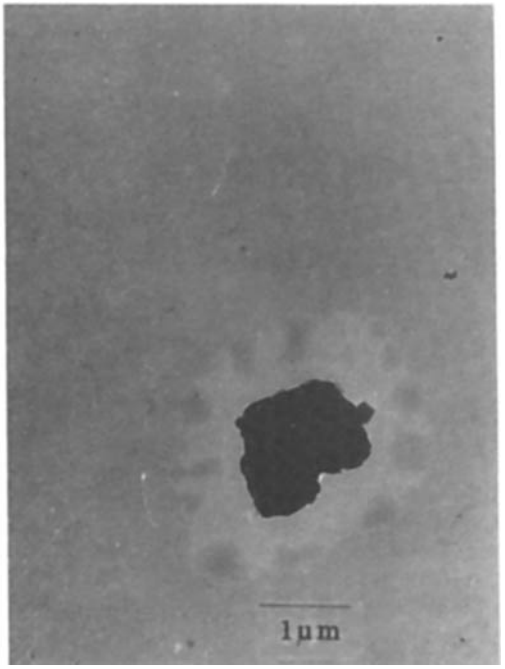



*Fig. 4.* Electron micrograph of particles collected on calcium thin film. Particle collection was made from 14:00 to 16:00 LT on 23 April at the height of 4350 m. The trace of water is observed around KOSA particles.

Figs 5a–d shows significant differences between a polluted urban atmosphere and clean air containing the KOSA particles. All particles in the figure were collected during the level flight at 2250 m height from Wakasa Bay to Nagoya on 15 May. The particles collected near Nagoya are shown in Figs. 5a, b, and the particles at Wakasa Bay in Figs. 5c, d. The morphological features of particles in Figs. 5a, b are common to the morphology of $(NH_4)_2SO_4$ particles produced through the reaction of ammonia with sulfuric acid droplets in the urban atmosphere (see e.g., Okada, 1985; Yamato and Ono, 1985). Figs. 5c d showed that KOSA particles and the particles having Liezegang's ring produced through the chemical reaction between Ca and $H_2SO_4$ were externally mixed in the unpolluted air mass. It is reasonable to expect that the aircraft crossed two different air-masses during the flight, one polluted and the other with relatively clean air containing KOSA particles.

## 3. Discussion

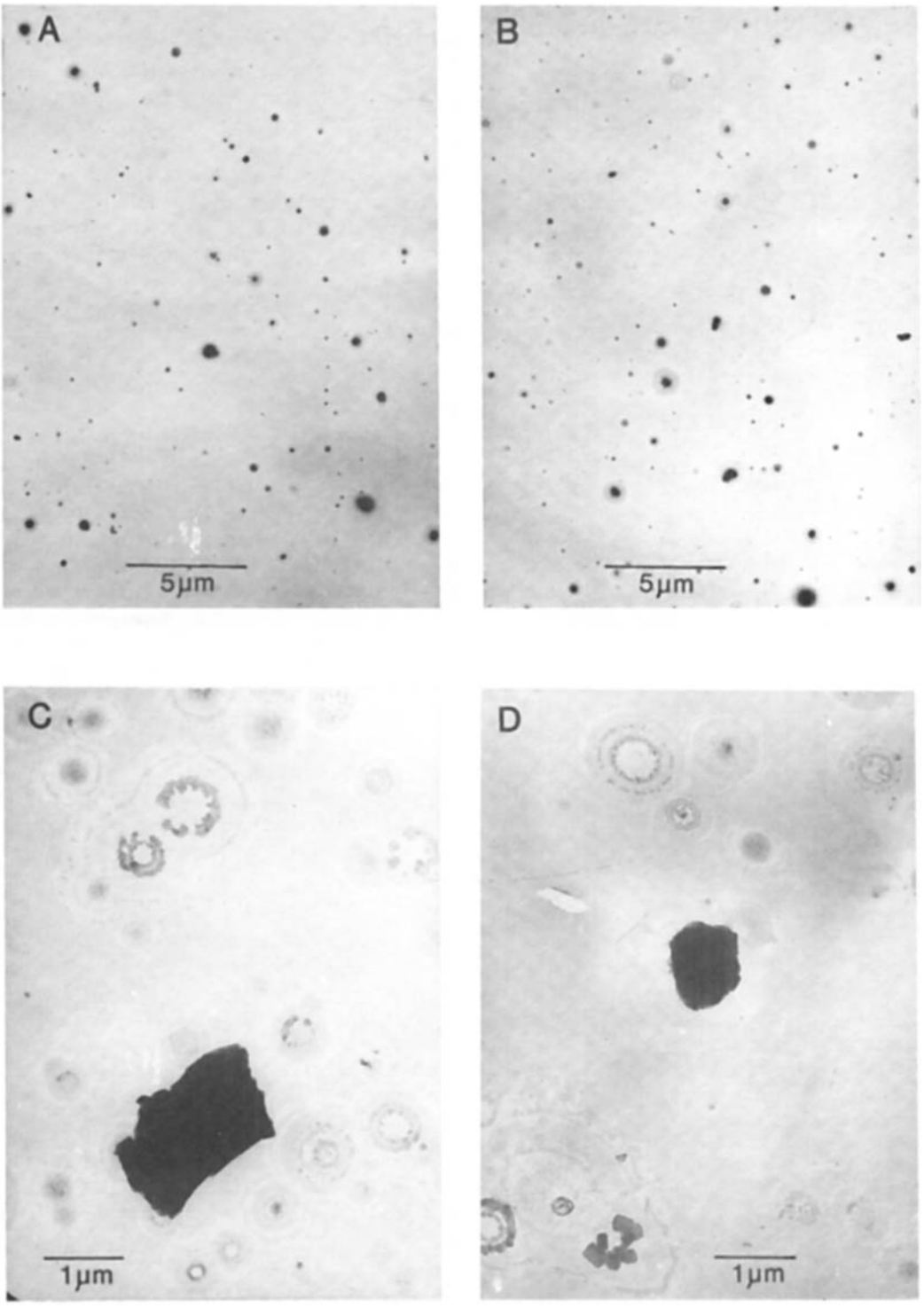
### *3.1. Aerosol layer in the spring lower troposphere*

The highly concentrated aerosol layer was frequently observed in the region of approximately 2 km–6 km by the lidar during the KOSARP 87 period. However, an aerosol layer is rarely observed in those height regions in other seasons at Nagoya (e.g. Kobayashi et al., 1985). It is worth pointing out that most of the layers were observed at heights of potential temperature of about 300°K. According to the trajectory analysis of air masses which approached the subtropical region in the Pacific Ocean in 1979, air parcels containing KOSA particles frequently travelled on the potential-temperature surface of about 300°K from Asian desert areas to the subtropical region in spring (Merrill et al., 1985). Present lidar measurements showed good agreement with the theoretical analysis concerning the heights and season of travel of KOSA particles.

The lidar measurements showed that the depolarization ratio of the lower troposphere was relatively high in comparison with the measurements in other seasons (e.g., Kobayashi et al., 1985). Additionally, layers having values larger than 0.2 were frequently observed. One possible interpretation of the measurements is that irregular particles, possibly KOSA particles, were distributed from the lower troposphere to the mid-troposphere, sometimes with a highly concentrated particle layer at about 300°K height. Considering the wind system of the Asian-Pacific area in the spring troposphere, there is a large possibility that most of the aerosol layers measured at the potential temperature height of about 300°K contained many KOSA particles. The electron microscopic observations on the particles sampled in the free atmosphere confirmed that these layers contained large and irregular shape particles, possibly soil particles.

### *3.2. Particle shape*

Fig. 3a, b are the TEM imagery of the particles sampled at 4350 m and 2250 m on April 9, and corresponding to the large depolarization ratio of the particle layer of April 9 in Fig. 1. Air mass trajectory analysis suggested that most air parcels which were at a height corresponding to the aerosol layer with large depolarization ratio were from Asian desert areas. As one example of these


A
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B
5μm
C
1μm
D
1μm

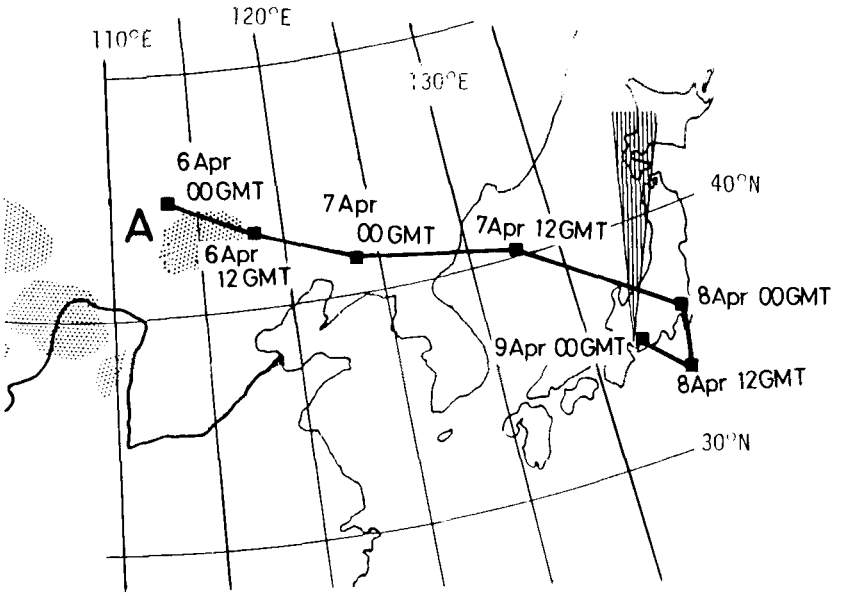


*Fig. 6.* Trajectory of the air mass which was over Nagoya on 9 April. The trace-back analysis was made on the surface of the potential temperature × 305°K, which corresponded to the peak height of the aerosol layer. A small dust storm was reported from 5 to 7 April near the desert area (point of A).

analyses, we show the trajectory of an air mass which produced the aerosol layer of 9 April in Fig. 6. The peak height of the aerosol layer of 9 April was the potential temperature of 305°K. The air parcel of the potential temperature of 305°K was traced back to the desert area of east Asia on 6 April. A dust storm event, possibly a small-scale one, was reported at the meteorological observatory (the point of A in the figure) near the desert area during the period from 5–7 April. There was no KOSA report from meteorological observatories in Japan in April. It was possible that this KOSA episode was too small to affect the atmosphere near the ground and meteorological observatories could not detect the decrease in visibility. Present measurements suggested that KOSA episodes frequently appear in the free atmosphere but that the KOSA phenomenon does not always correspond to the appearance of KOSA in the free atmosphere.

Summarizing the lidar data, TEM imagery, and the trajectory analysis of air masses, we can speculate that most of the aerosol layers with large mixing ratios and depolarization ratios of particles at the height of the potential temperature of approximately 300°K measured during KOSARP 87, were KOSA particle layers.

### *3.3. Soil particles coated by solution*

The TEM imagery of the particles collected at 4400 m on 23 April was very suggestive of the rôle of KOSA particles as chemical reaction sites of atmospheric gases. Many KOSA particles were sampled in the aerosol layer having a large depolarization ratio in 23 April. Some irregular particles were coated by solution. Considering the traces of moisture surrounding KOSA particle shown in Fig. 5, the solution did not contaon $SO_4^{2-}$ ions (or the concentration of the ion was lower than detectable limit). The existence of the par-

*Fig. 5.* Electron micrograph of particles collected on calcium thin film. Particle collection was made at 2250 m height on 15 May (14:00–16:00 LT). The particles sampled over Nagoya ((A) and (B)) did not show the Liezegang's ring and their features were frequently observed in the urban atmospheric particles (e.g., Okada, 1985; Yamato and Ono, 1985). The collection at Wakasa Bay ((C) and (D)) showed that large, irregular and electron-dense particles externally mixed with the particles producing Liezegang's ring (certainly sulfuric acid droplets).

ticles coated by solution may correspond to the high relative humidity (about 75%) in the region where the aerosol layer was observed. When a KOSA particle coated by water travels in the atmosphere, various atmospheric gases, e.g., $NH_2SO_4$, $SO_x$, and $NO_x$, may be absorbed and complex heterogeneous reactions can follow during the transport. If so, KOSA particles seem to play an important role as a chemical reaction site in the atmosphere and as a carrier of products of these chemical reactions.

In Fig. 7, the particle coated by solution containing $SO_4^{2-}$ is shown, but the concentration of $SO_4^{2-}$ may not be high (see A). Fine crystals of $CaSO_4$ were observed surrounding the particle 'A' and suggested that particle 'A' was coated with a solution containing $SO_4^{2-}$ ion. The TEM imagery obtained under the weak electron beam conditions (left) and that under strong electron beam radiation (right) are compared in Fig. 7. Small $CaSO_4$ dots surround the particle 'A', and a hard core remained after strong-beam radiation; materials, which were not identified on the surface of the particle 'A' were vaporized. A similar change was observed for particles 'B', 'C', and 'D'. These indicate possible complex chemical-physical processes on the particle surface during the particle travel in the atmosphere. At present, the origin of $SO_4^{2-}$ in solution is not clarified directly, but the following two processes are considered possible. It is well-known that the soil of Asian desert areas originally contains

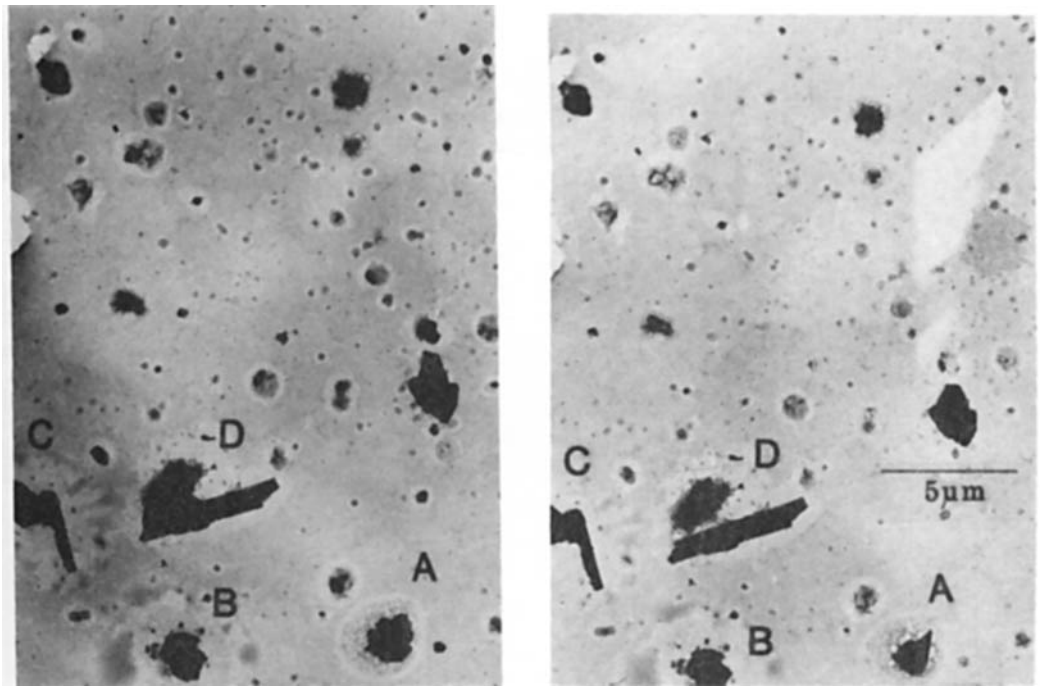


*Fig. 7.* KOSA particle coated solution containing $SO_4^{2-}$ (A, B, C and D). These particles were sampled at 4350 m during the flight of 23 April, 14:00–16:00 LT. Electron micrograph under the weak electron beam (left) and that under the strong beam (right). Faint $CaSO_4$ dots produced from Ca and $SO_4^{2-}$ in solution on the particle surface are observed around particle A. After the radiation of the strong electron beam, unknown volatile material evaporated from the particle surface. Particles B and C were also coated with solution. However, the trace of the solution does not clearly show the existence of $SO_4$. The electron micrograph taken under the strong electron beam showed that the particle D was composed of two different particles. This suggests that there is a possibility of coagulation between KOSA particles and other types of particle in the atmosphere.

sulfur in the form of $CaSO_4$. Therefore, after the particle is coated with a water film, the $SO_4^{2-}$ ion can dissolve into the water film. Another possible process is the gas-to-particle conversion of $SO_x$ on the particle surface during transport in the atmosphere. Detailed chemical identification for the solution is desired to clarify the formation processes of the solution coating KOSA particles.

According to Wu (private communication, 1987), KOSA particles coated by solution containing $NO_3^-$ were observed in the atmosphere (10 m above the ground) at Nagoya during the KOSRAP 87.

### *3.4. Concentration of KOSA particles*

The lidar measurement and the particle collection suggested that a KOSA particle layer frequently appeared in the free atmosphere during the observational period, but there was no report on KOSA for meterological observatories in Japan, since the atmospheric disturbance due to the KOSA was not observed near the ground. This shows the importance of knowing the vertical distribution of KOSA particles for the assessment of KOSA-particle content in the atmosphere and the estimation of KOSA particle flux from the atmosphere to the ground. The situation that the content of KOSA particles in the free atmosphere was higher than that near the ground should be an important factor for clarifying the spatial variations of the particle sedimentation rate on the global scale. According to the present measurements, horizontal mass flux of KOSA particles seems to be large, but vertical flux due to a particle sedimentation was very small at Nagoya.

Concentration of KOSA particle in the atmosphere can be estimated according to the method presented by Iwasaka et al. (1983) from the lidar data. The typical density is in the range from 1.9–25 $\mu g/m^2$, with an average value of 18 $\mu g/m^3$, assuming the particle-size distribution function given by Ishizaka and Ono (1982), density of soil = 2.6 $g/cm^3$ (Iwasaka et al., 1983), and refractive index of soil = 1.5 + Oi at the wavelength of lidar (0.6943 $\mu m$). This concentration is apparently lower than the value of severe KOSA (Iwasaka et al., 1983). However, it does not mean that the weak KOSA contributes much less to soil particle transport on a global scale, since the frequency of appearance of weak KOSA is larger than that of severe KOSA. Thereby, the total contribution of weak KOSA cannot be negligible for the global budget of the atmospheric soil particles. At present, there is little information on the horizontal scale of 'background KOSA', since KOSA particle concentrations are too low for detection by satellite, and lidar stations are too few to cover the scale of the KOSA. Even so, it is of interest to estimate roughly the weak KOSA effect.

The lidar measurements suggest the duration of a few days for individual weak KOSA events. If a typical wind speed is assumed to be 10 m/s at the 300°K surface (meteorological sonde measurement at Hamamatsu observatory), the typical scale, $L$, is from about 1700 km (2 days duration) to about 2600 km (3 days duration). The amount of soil particles transported by the weak KOSA is approximately given by,

$$M = \rho H L^2,$$

where $\rho$ is density of KOSA ($g/cm^2$), $H$ the typical thickness of the particle layer, and $L$ the typical length of weak KOSA events. When we use the values $\rho = 18$ $\mu g/m^3$, $H = 2$ km, and $L = 2600$ km, the value of $M$ is about $2.4 \times 10^5$ metric tons. This value is one order smaller than the measurement reported by Iwasaka et al. (1983). However, if we take the higher frequency of weak KOSA events into consideration for the estimate, the total mass per year transported by weak KOSA will be more than that. Concerning the lidar measurements during KOSARP 87, three weak KOSA episodes were identified. Therefore, the total mass transported by these weak KOSA is about $7.2 \times 10^5$ metric tons at least, and this load is comparable to a severe KOSA.

The present estimate suggests the importance of weak KOSA events for the global geochemical cycle of soil particles. More detailed observations are required, especially on the horizontal scale of weak KOSA events.

## 4. Conclusions

Lidar and aircraft measurements on Asian dust (KOSA) particles suggest the following.

(1) There were KOSA particle layers in the mid- and lower troposphere during spring, even

when atmospheric phenomena related to KOSA events were not observed near the ground surface.

(2) KOSA layers frequently appeared at the heights of potential temperature of about 300°K. The height of the weak KOSA is important for global diffusion of KOSA particles according to the trajectory analysis of air masses from Asian desert areas to the subtropical regions of the Pacific (Merrill et al., 1985).

(3) The weak KOSA can contribute to the global cycle of soil particles, since the frequency of appearance is larger than the severe KOSA events.

(4) A portion of the KOSA particles were coated by solution containing $SO_4^{2-}$ or water. This indicates the possibility that KOSA particles act as chemical reaction sites in the atmosphere and as carriers of chemical products.

Weak KOSA events hardly draw the attention of investigators and therefore little information has been obtained. More information is necessary to clarify the geochemical effects of weak KOSAs. A knowledge of the heterogenous reactions involving KOSA particles is required in order to understand the effect of KOSA on global geochemical cycles.

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