

SIZE DISTRIBUTION OF AEROSOLS AT BARROW IN ALASKA —A CASE STUDY IN SPRING

Yuji ZAIZEN¹, Kikuo OKADA¹, Miwako IKEGAMI¹, Teruo AOKI¹, Yousuke SAWA¹,
Fumihiko NISHIO² and Yoshihiro TACHIBANA³

¹*Meteorological Research Institute, Nagamine, Tsukuba 305-0052*

²*Hokkaido University of Education, Shiroyama 1-chome, Kushiro 085-8580*

³*Research Institute of Civilization, Tokai University, 1117, Kitakaname,
Hiratsuka 259-1292*

Abstract: Concentration, size distribution and morphological features of aerosol particles were observed at Barrow in Alaska during the period 13–28 April 1997 together with concentrations of gaseous species (SO₂, NO and NO₂). The concentrations of gaseous species were less than 50 pptv except for locally polluted air. Aerosol size distributions suggested the occurrence of new particle formation from the gas phase in spite of very low concentrations of gaseous species, especially in the air mass from lower latitudes during the latter part of the observational period. In the air mass from higher latitudes, new particle formation was assumed to be not active. The largest maximum was found at 0.1–0.2 μm radius in most of the observation periods, suggesting that the size distributions have been influenced by well-aged particles. The dominant component of aerosol particles in this size range is considered to be sulfate-containing particles. Another maximum was found occasionally at 0.02–0.05 μm radius. This kind of air mass was possibly influenced by air from lower latitudes.

1. Introduction

Atmospheric aerosols are one of the major components influencing climate change through direct radiative forcing and indirect forcing by acting as cloud condensation nuclei (CCN). The most effective particles in these processes are thought to be accumulation mode particles. Accumulation mode particles originate from the growth of nuclei mode particles which are formed by homogeneous nucleation from the gas phase. This growth process is thought to be coagulation, heterogeneous condensation and interaction with clouds.

In the Arctic region, aerosols have been studied with a special interest in Arctic haze phenomena (*e.g.*, BARRIE, 1986). In Arctic haze, particles in the accumulation mode are dominant (PACYNA *et al.*, 1984; SHAW, 1985; LEWIS, 1985). The great majority of Arctic aerosols are in or near the accumulation mode (HEINTZENBERG, 1980; SHAW, 1984). Besides, the particles also exist in smaller size ranges. For example, particle formation by homogeneous nucleation occurs through the oxidation of precursor gases from dimethylsulfide (DMS) originated from the sea surface in summertime (HEINTZENBERG and COVERT, 1987). The possibility of nucleation depending on low temperature in an Arctic haze event in winter has also been suggested (SHAW, 1983).

The aim of this paper is to show the variation of aerosol size distribution, especially in the smaller size range, in April 1997 at Barrow, together with the concentrations of gaseous species concentrations.

2. Observation Site and Instruments

This aerosol observation was conducted during the period from 13 to 27 April 1997 at the building of NOAA/CMDL (National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostic Laboratory) located northeast of Barrow, Alaska (Fig. 1).

Aerosol size distribution in the smaller size range ($0.004 \leq r < 0.13 \mu\text{m}$) was measured with a DMA (differential mobility analyzer; TSI, model 3071) and a CNC (condensation nucleus counter; TSI, model 3025). The size distribution in the larger size range ($0.15 \leq r < 5 \mu\text{m}$) was measured with an OPC (optical particle counter) (Dan Industry Co. Ltd., model PM-730-NS15P). Concentrations of gaseous species (SO_2 , NO and NO_2) were measured with a SO_2 analyzer (Thermo Electron, model 43S) and a NO_x analyzer (Thermo Electron, model 42S). The principles of these two instruments are chemiluminescence. The precisions of these instruments, which mainly depend on the S/N ratios, are approximately ± 20 pptv. Individual aerosol particles of $0.05\text{--}0.5 \mu\text{m}$ radius were collected on a carbon-covered nitrocellulose (collodion) film with a cascade impactor and examined with an electron microscope. Before examination by electron microscopy, the collected samples were coated with Pt/Pd alloy at a shadowing angle of $\arctan 0.5$.

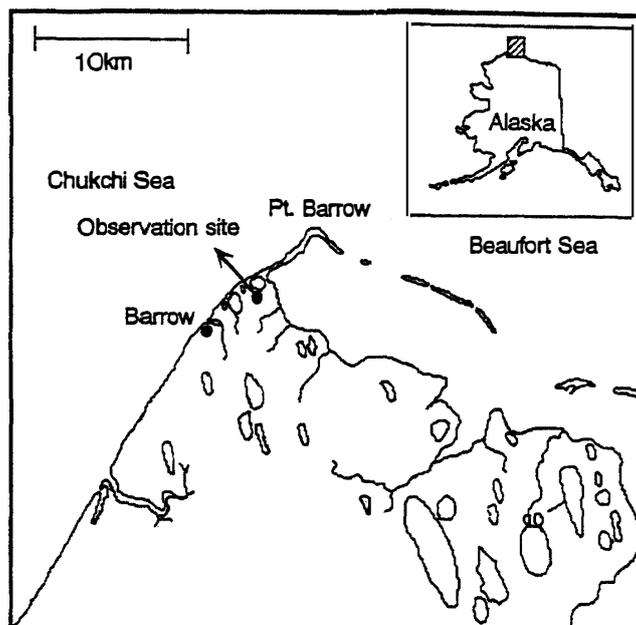


Fig. 1. Location of the observation site ($71^{\circ}49'23''$ N, $156^{\circ}36'32''$ W).

3. Results and Discussion

3.1. Meteorological condition and backward trajectory of air

During the former part of the observational period (approximately 14–18 April), northern Alaska was covered by a polar air mass and the temperature was $-20\sim-30^{\circ}\text{C}$. In the latter part of the observational period (approximately 19–28 April), air from relatively low latitude was dominant and the temperature was $-5\sim-25^{\circ}\text{C}$. The weather was cloudy on 14 April, clear on 16–18 April, cloudy on 19–21 April and clear on 22–28 April. The isentropic backward-trajectories calculated for the previous 5 days from the altitude of 1000 m are shown in Fig. 2. The surface air was considered to be from the same origin, because the height of the mixed layer was approximately 1500 m. These trajectories were calculated with the objective analysis data of the JMA (Japan Meteorological Agency).

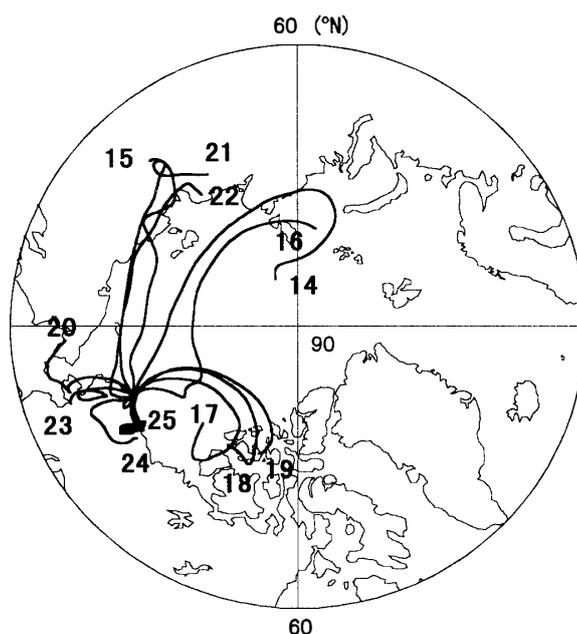


Fig. 2. Isentropic-backward trajectories of air parcels from the 1000 m altitude over the observation site (Barrow) for 5 days. Number indicated at the end of the trajectory denotes the day when the air arrived over the observation site.

3.2. Aerosol concentration

Figure 3 shows the particle concentration measured with the CNC equipped with the DMA in the radius range of $0.004\text{--}0.13\ \mu\text{m}$. The concentration varies between 10^0 and $10^4\ \text{cm}^{-3}$. Many short spikes were found, especially during westerly wind from the direction of Barrow town. These spikes are due to contamination of local origin. During 13–18 April, the daily minimum concentrations were approximately 2–4

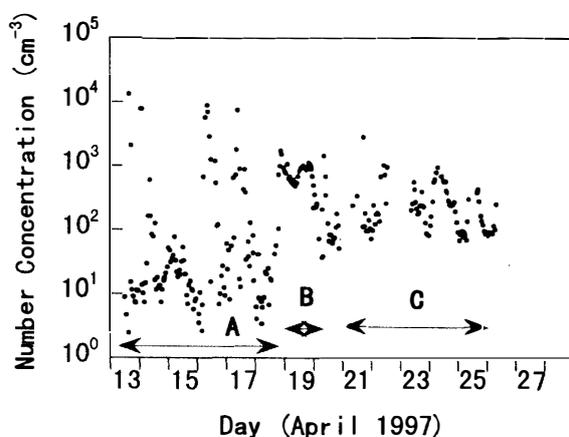


Fig. 3. Aerosol concentration in the small size range ($0.004 \leq r < 0.13 \mu\text{m}$). A, B, and C show the periods. Each dot shows an average concentration over 1 h.

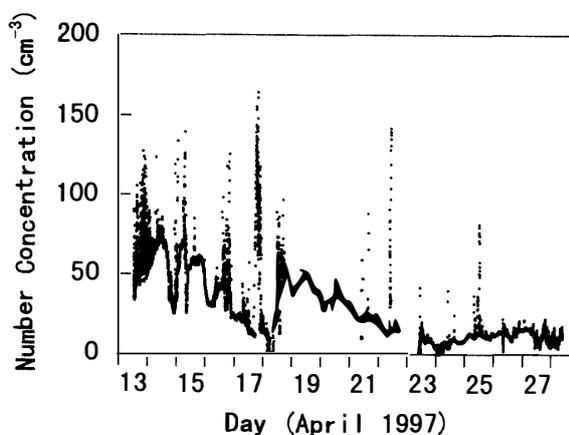


Fig. 4. Number concentration of aerosol particles larger than $0.15 \mu\text{m}$ radius. Each dot shows an average concentration over 1 min.

cm^{-3} . These low concentrations were considered to be associated with the polar air mass. These low values suddenly increased to $400\text{--}1000 \text{ cm}^{-3}$ on 19 April, and decreased slightly to $70\text{--}100 \text{ cm}^{-3}$ during 21–25 April. The high concentrations during these periods were thought to be associated with the air mass from relatively low latitudes. On 19 April, the concentration was very high without wide fluctuation. This uniformity suggests that the high concentration in this period was not due to local contamination. Considering these observations, three periods (A, B and C) were defined as shown in this figure and will be used in following discussion.

Figure 4 shows the number concentrations of aerosol particles larger than $0.15 \mu\text{m}$ radius measured with the OPC. The baseline is relatively clear in spite of large variations as measured for smaller particles in Fig. 3. The baseline value in period A shows a wide fluctuation from 10 to 70 cm^{-3} . However, the values are relatively larger than in period C ($\leq 20 \text{ cm}^{-3}$). The value is constantly high ($40\text{--}50 \text{ cm}^{-3}$) in period B.

The variation of larger particle concentration is different from that of smaller particles (Fig. 3). In period A, the concentrations of smaller particles are low, however, the concentrations of larger particles are relatively high. In period C, the concentrations of smaller particles are high, however, the concentrations of larger particles are low. In period B, particles in both size ranges show high concentration.

3.3. Characteristics of aerosol size distribution

The average size distribution for each period was calculated (Fig. 5). Data with concentrations in the smaller size range ($0.004\text{--}0.13 \mu\text{m}$) higher than 200 cm^{-3} were excluded from the average size distributions A and C to avoid the influence of local contamination.

In each distribution, a maximum was found in the radius range of $0.1\text{--}0.2 \mu\text{m}$. The existence of this peak was reported by former investigators in Arctic areas (*e.g.*,

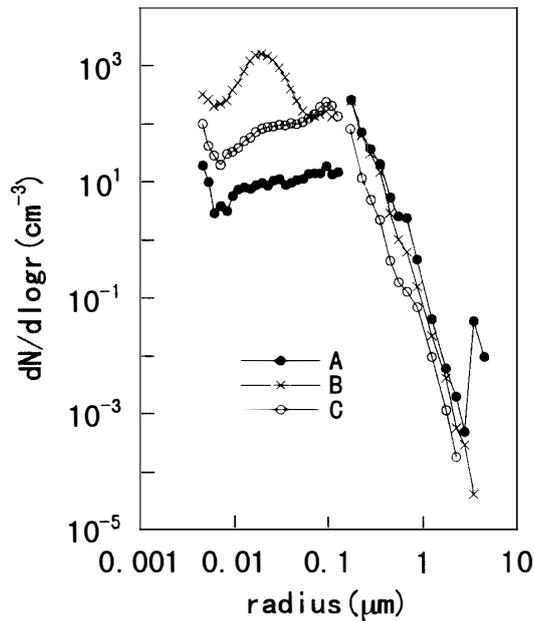


Fig. 5. Averaged size distributions for 3 periods.

COVERT, 1993). This maximum possibly exists constantly over the Arctic air in spring and is assumed to consist of well-aged aerosols. On the other hand, the concentrations of particles smaller than $0.005 \mu\text{m}$ radius are elevated in each size distribution, suggesting the occurrence of nucleation from gas phase material. In period A, concentrations in the smaller size range ($r < 0.15 \mu\text{m}$) are relatively lower, suggesting that new particle formation is passive in the polar air mass. Higher concentrations of smaller particles in period C might suggest relatively active nucleation in the lower latitudinal air mass. The concentrations in the larger size range ($r \geq 0.15 \mu\text{m}$) are higher in period A than in period C. The higher concentration of large particles in period A might be due to a long residence time of aerosols connected with small air mass exchange rates and small amounts of precipitation in the polar region. This high concentration of larger particles tends to restrict new particle formation by reducing the vapor pressure of sulfuric acid by heterogeneous condensation on the surface of pre-existing particles. In period B, the concentration of particles in the nuclei mode is highest and another maximum around $0.02 \mu\text{m}$ radius is found, suggesting that these aerosols were relatively newly formed. Besides, the concentration and size distribution was relatively stable, implying that this air was not influenced by local pollution. The maximum around $0.02 \mu\text{m}$ radius is typically found in background aerosols in middle and low latitudes (ZAIZEN *et al.*, 1996). Therefore, the size distribution of aerosols is considered to be a consequence of relatively active particle formation.

3.4. Concentrations of gaseous species

Figures 6a, b and c show the concentrations of SO_2 , NO and NO_2 , respectively. Large short spikes are considered to be due to local contamination. Especially, the air was contaminated by local pollution on 16 and 17 April. The concentrations of NO_2

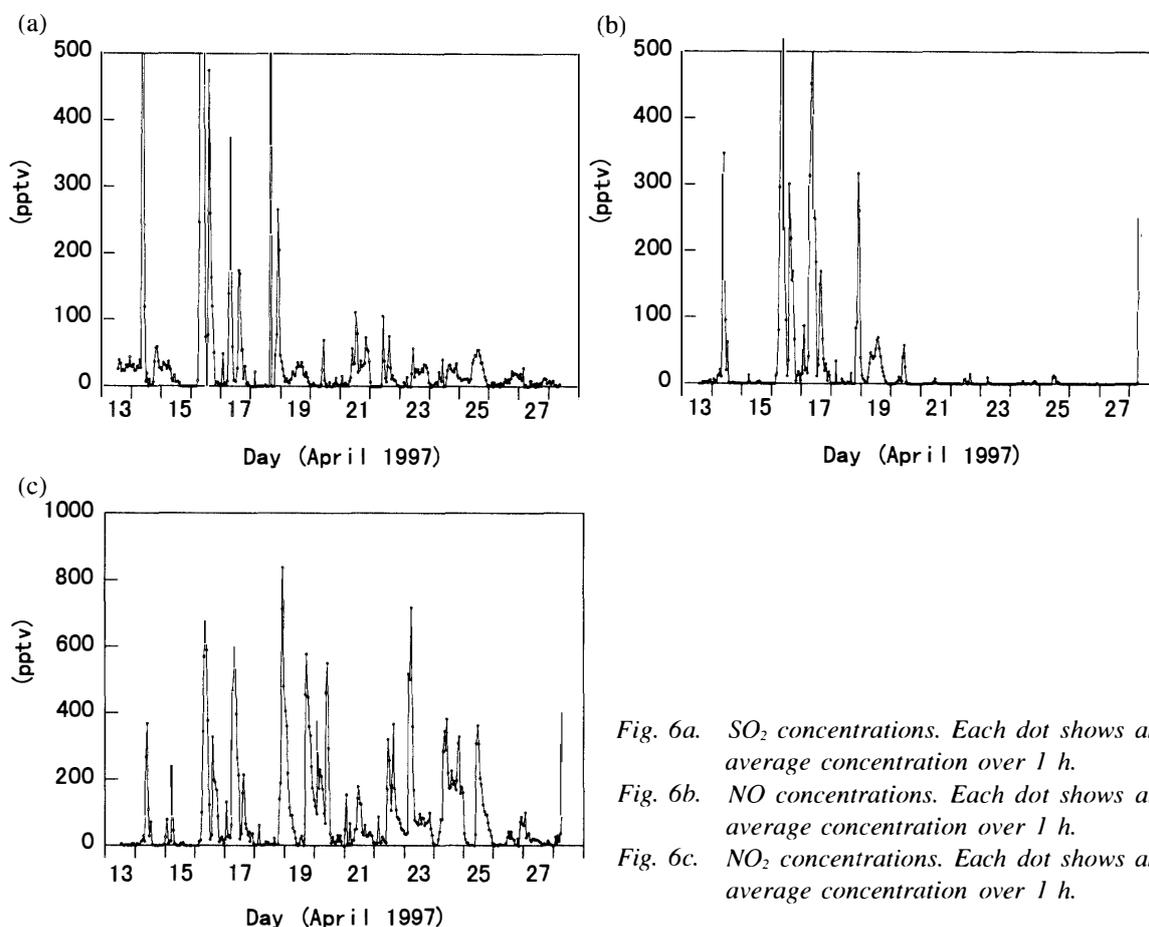


Fig. 6a. SO₂ concentrations. Each dot shows an average concentration over 1 h.
 Fig. 6b. NO concentrations. Each dot shows an average concentration over 1 h.
 Fig. 6c. NO₂ concentrations. Each dot shows an average concentration over 1 h.

in the first several hours after large spikes are not correct because traces of the spikes remain. NO₂ data show many spikes and large values than SO₂ and NO concentrations because of local contamination such as exhaust gas from automobiles.

By excluding the large values from local pollution, the concentrations of these gaseous species were considered to be basically less than 50 pptv. The low concentrations of SO₂ at Barrow in this season may be explained by relatively short residence time (BARRIE *et al.*, 1984) and small amount of the supply.

In period A, there were several interruptions of local pollution and systematic diurnal variation was not found. However, during latter periods, diurnal variations with maxima in the daytime were found on several days. This type of diurnal variation is inconsistent with the variation of mixing layer thickness. Therefore, some production mechanisms of SO₂ should exist in the daytime. One of the possibilities is oxidation of DMS originating from the sea surface. In southern wind (18–22 April), the cracks of sea ice tended to be open. It is suggested that nucleation from SO₂ possibly produced the relatively higher concentration of small particles during the latter periods (periods B and C).

Small maxima of SO₂ and NO concentrations were measured on 19 April, at the time of the maximum of aerosol concentration shown in Fig. 2. The concentrations of the gases in this period were low without large fluctuation. This suggests that the air

in period B was possibly affected by polluted air. The large concentrations of smaller particles in the nuclei mode might be explained by nucleation from injected gaseous species. On the afternoon of 15 April, the concentrations of the three gases were near zero. This would lead to the very low concentrations of particles in the nuclei mode as shown in Fig. 2. On the other hand, the concentration of particles in the larger size range was relatively high on 15 April (Fig. 3).

3.5. Morphological features of individual aerosol particles

During the period 21–24 April (period C), aerosol particles were collected by the cascade impactor. Figure 7 shows the electron micrographs of these samples. Sample A was collected during the period 1058–1104 LST on 21 April and sample B was collected during the period 1007–1013 on 22 April. The aerosol particles shown in these pictures were in the accumulation mode. Most particles in sample A show the appearance of satellite droplet rings on the collecting surface, suggesting that these particles contain sulfuric acid (*e.g.*, FRANK and LODGE, 1967). In contrast, the particles in sample B do not show satellite structure. On the basis of the morphology, the particles are estimated to be ammonium sulfate. From this fact, sulfate is considered to be one of the dominant components of accumulation mode particles in the Arctic region.

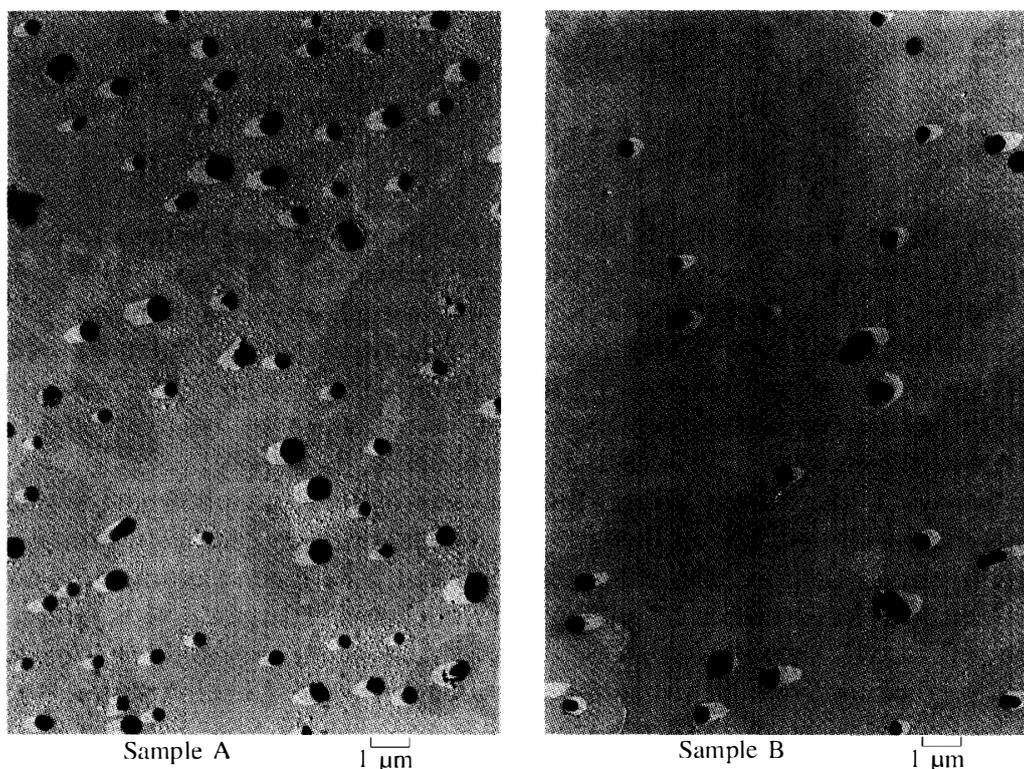


Fig. 7. Electron micrographs of particles collected in the period C. The sample A was collected during the period of 1058–1104 LST on 21 April 1997. The particles show satellite droplet ring on the collecting surface, suggesting the presence of sulfuric acid. The sample B was collected during the period of 1007–1013 LST on 22 April. These particles are considered to be composed of neutralized sulfate.

4. Conclusions

Concentration, size distribution, features of aerosol particles and concentrations of gaseous species were observed at Barrow in Alaska during 13–28 April 1997. From backward-trajectory analyses, the polar airmass was dominant during the former part of the observational period and air from lower latitudes was dominant in the latter part. The concentrations of SO₂, NO and NO₂ were less than 50 pptv in most of the observation period except for locally polluted air. In the latter period, diurnal variation of SO₂, which has a maximum in the daytime, was found, implying SO₂ production by DMS oxidation.

In number-size distributions of aerosol particles, the largest maximum was found in the radius range of 0.1–0.2 μm in most of the observation periods, suggesting that the dominant particles were well-aged. Particle concentrations in the size range smaller than 0.005 μm radius were elevated constantly, suggesting the occurrence of new particle formation from the gas phase in spite of very low concentrations of gaseous species. Another maximum in the middle size range (around 0.02 μm) was found on some occasions. This kind of air mass was possibly influenced by air from lower latitudes.

Concentrations of aerosols in the smaller size range ($r < 0.1 \mu\text{m}$) suggested that new particle formation was passive in the polar air mass and relatively active in air from lower latitudes. In polar air, low amounts of precursor gases and large surface areas of pre-existing particles would reduce the vapor pressure of sulfuric acid in the atmosphere, and suppress new particle formation in the polar air.

On the basis of the morphological features of individual aerosol particles, sulfate is the dominant component of Arctic aerosols.

Acknowledgments

We thank Dr. D. ENDRES and Dr. M. GAYLORD of NOAA/CMDL for offering us an observational site and supporting these observations. We also thank Mr. Y. NAKAJIMA and Ms. Y. TSURUGA of RESTEC (Remote Sensing Technology Center) and Mr. Y. SARUYA for their logistic support. This work was carried out as a part of the ADEOS Field Campaign supported by the NASDA (National Space Development Agency of Japan).

References

- BARRIE, L. A. (1986): Arctic air pollution: An overview of current knowledge. *Atmos. Environ.*, **20**, 643–663.
- BARRIE, L. A. and HOFF, R. M. (1984): The oxidation rate and residence time of sulphur dioxide in the Arctic atmosphere. *Atmos. Environ.*, **18**, 2711–2722.
- COVERT, D. S. (1993): Size distributions and chemical properties of aerosol at Ny-Ålesund, Svalbard. *Atmos. Environ.*, **27A**, 2989–2997.
- LEWIS, N. F. (1985): Particle-size distributions of the Arctic aerosol. M. S. thesis, University of Rhode Island.

- FRANK, E. R. and LODGE, J. P., Jr. (1967): Morphological identification of airborne particles with the electron microscopy. *J. Microscopie*, **6**, 449–456.
- HEINTZENBERG, J. (1980): Particle size distribution and optical properties of Arctic haze. *Tellus* **32**, 251–260.
- HEINTZENBERG, J. and COVERT, D. S. (1987): Chemically resolved submicronic size, distribution and external mixing of the Arctic haze aerosol. *Tellus*, **39B**, 374–382.
- PACZYNA, J. M., VITOLS, V. and HANSEN, J. E. (1984): Size-differentiated composition of the Arctic aerosol at Ny-Ålesund, Spitsbergen. *Atmos. Environ.*, **18**, 2447–2459.
- SHAW, G. E. (1983): On the aerosol particle size distribution spectrum in Alaskan air mass systems: Arctic haze and non-haze episodes. *J. Atmos. Sci.*, **40**, 1313–1320.
- SHAW, G. E. (1984): Microparticle size spectrum of Arctic haze. *Geophys. Res. Lett.*, **11**, 409–412.
- SHAW, G. E. (1985): Aerosol measurements in central Alaska, 1982–1984. *Atmos. Environ.*, **19**, 2025–2031.
- ZAZEN, Y., ISEGAMI, M., TSUTSUMI, Y., MAKINO, Y., OKADA, K., JENSEN, J. and GRAS, J. L. (1996): Number concentration and size distribution of aerosol particles in the middle troposphere over the western Pacific Ocean. *Atmos. Environ.*, **30**, 1755–1762.

(Received January 21, 1998; Revised manuscript accepted April 23, 1998)