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Title	Optical and Chemical Properties of Atmospheric Aerosols at Amami Oshima and Fukue Islands in Japan in Spring, 2001
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Citation	Journal of the meteorological society of japan, 91(1), 63-73 https://doi.org/10.2151/jmsj.2013-104
Issue Date	2013-02
Doc URL	https://hdl.handle.net/2115/53374
Type	journal article
File Information	91_2013-104.pdf

Hokkaido University Collection of Scholarly and Academic Papers : HUSCAP

Journal of the Meteorological Society of Japan, Vol. 91, No. 1, pp. 63–73, 2013
DOI:10.2151/jmsj.2013-104

Optical and Chemical Properties of Atmospheric Aerosols at Amami Oshima and Fukue Islands in Japan in Spring, 2001

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(Manuscript received 6 January 2012, in final form 9 November 2012)

Abstract

The optical and chemical properties of atmospheric aerosols were determined from the ground-based measurements at Amami Oshima in April 2001 during the Asian Atmospheric Particle Environmental Change Studies (APEX) campaign and at Fukue Island in March 2001. At Amami Oshima from April 10 to 16, an aerosol event was observed in which the volume scattering coefficient and sulfate concentration of fine particles increased conspicuously. At the former term of the aerosol event, the single scattering albedo reached 0.98. At the latter term of the event, on the other hand, it was 0.80–0.90 and the concentrations of elemental carbon, aluminum, and zinc increased by a factor of several to ten times compared with the rest of the observation terms. Using chemical and backward trajectory analyses, it was established that the transparent aerosols, rich in sulfate, were converted from sulfur dioxide gas emitted by the Miyake Island volcano at the former term of the aerosol event, while the turbid and absorptive aerosols were anthropogenic aerosols appearing together with Asian yellow dust from continental China at the latter term of the event. The measurements at Fukue Island showed that the volume scattering coefficients as well as the concentrations of sulfate, elemental carbon, aluminum, and zinc were higher than those at Amami Oshima, while the single scattering albedo was relatively low. This study elucidates that in spring, large amounts of anthropogenic particles are frequently transported together with Asian yellow dust from inland continental China to the Pacific Ocean, and that the single scattering albedo and the ratio of organic to elemental carbons are approximately 0.80–0.85 and 1, respectively, over the north-western Pacific Ocean.

Keywords atmospheric aerosols; Amami Oshima; Fukue Island; Miyake Island volcano; anthropogenic aerosols; Asian yellow dust

1. Introduction

In East Asia, large amounts of aerosols are transported to the western Pacific Ocean in spring. This transport has been observed in the Pacific Exploratory Mission in the Western Pacific Ocean (PEM-West A and B) (Arimoto et al. 1996; Carmichael et al. 1997; Chen et al. 1997) during a Kosa (Asian yellow dust) event over East Asia (Uematsu et al. 2002), and in the Asian yellow dust event of April 1998 (Choi et al. 2001; Husar et al. 2001). In April 2001, a heavy dust event was observed by the Asian Atmospheric Particle Environmental Change Studies (APEX; Nakajima et al. 2003) and studied by Chang et al. (2004). In addition, the atmospheric brown cloud (ABC) over East Asia was studied in conjunction with the Atmospheric Brown Cloud East Asian Regional Experiment 2005 (ABC-EAREX 2005) in an overview by Nakajima et al. (2007).

Aerosols may cause cooling or heating in East Asia by a direct effect through modification of the radiation budget because of scattering and absorption of solar radiation (radiative forcing). In estimation of the forcing, it is necessary to determine the optical

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properties of the aerosols, in particular, the single scattering albedo ω, which is the ratio of the volume scattering coefficient to the volume extinction coefficient (Takemura et al. 2002). The value of ω strongly depends on the amounts of absorptive components, such as soot (elemental carbon) and soil in the aerosols. Anderson et al. (1999) measured the optical properties of aerosols at Cheeka Peak, Washington, USA, at a mid-latitude Pacific coastal station. However, there are little data on the optical properties of aerosols in East Asia, in particular, over the north-western Pacific Ocean.

Aerosols also affect the climate by indirect effects arising from increases in the number concentrations of cloud droplets activated from water-soluble aerosols. To better understand these effects, it is necessary to determine the optical and chemical properties of atmospheric aerosols over the north-western Pacific Ocean.

Here, we have measured the optical and chemical properties of aerosols at Kasari, Amami Oshima (28.30°N, 129.40°E) and Kisyuku, Fukue Island (32.44°N, 128.44°E) in the southern part of Japan in the north-western Pacific Ocean, as shown in Fig. 1. At Amami Oshima, the measurements were taken from April 2 to 28, 2001 in cooperation with the APEX campaign, and from March 19 to 26, 2001 at Fukue Island. This paper shows the optical and chemical properties of the atmospheric aerosols measured there.

2. Materials and methods

Atmospheric fine particles (aerosols smaller than 2.0 μm in diameter) were collected on a Teflon filter (Sumitomo Electric Industry, FP-1000, 47 mm in diameter) and on a quartz fibrous filter (Pallflex 2500 QAT-UP, 47 mm in diameter) drawing outdoor air with a hose through a cyclone separator with a 50% cut-off diameter of 2.0 μm at a flow rate of 20 l/min. In particular, the quartz fibrous filter was set behind the Teflon filter (in the tandem QBT method) for correction of positive artificial organic carbon due to adsorption of gaseous organics on the quartz fibrous filter (Kirchstetter et al. 2001; Turpin et al. 1994).

Total particles (all aerosols of any size) in the outdoor air were collected on a Teflon filter (FP-1000, 47 mm in diameter) through a hose without a cyclone separator, at a flow rate of 30 l/min.

Outdoor air was also drawn through a cyclone separator to a manifold. By drawing air from the manifold, the volume scattering coefficient of fine particles at a wavelength of 0.530 μm was measured by an integrating nephelometer (M903, Radiance Research), and the volume absorption coefficient of fine particles at 0.565 μm was measured by an absorption photometer (PSAP, Radiance Research). In the integrating nephelometer measurements, particle-free air was used to establish the instrument zero by passing ambient air through a filter with high collection efficiency, and CO_2 was used as span gas with the scattering coefficient $2.14 \times 10^{-5}\,m^{-1}$ at 20℃ and 1 atm. The measurement principle of the absorption coefficient by PSAP is based on the integrating plate method. Measurements by both instruments were made daily for 23 h, and the data were averaged 5-min values.

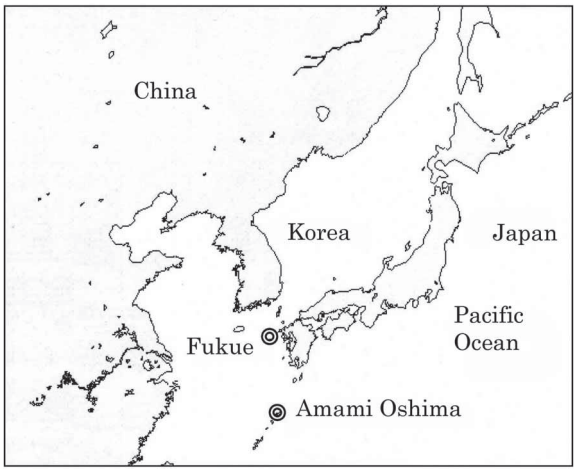

Fig. 1. Map of East Asia. Fukue Island and Amami Oshima are measurement sites in this study.

The aerosol sampling was made daily for 23 h from 11: 00 in the morning until 10: 00 the following morning.

Ionic components of the aerosols collected on the Teflon filter were extracted ultrasonically with distilled de-ionized water and analyzed by an ion chromatograph (Yokogawa Analytical Systems, IC 7000, column Excelpack ICS0-A23 for anion and ICS-C25 for cation). The metal components of the aerosols on the Teflon filter were extracted ultrasonically with a solution of nitric acid and hydrofluoric acid in a polypropylene bottle, left for two days, and analyzed by inductively coupled plasma mass spectrometer (ICP-MS) (Yokogawa Analytical Systems, HP 4500).

The amount of carbonaceous components (elemental carbon and organic carbon) of the aerosols on the quartz fibrous filter was determined by a combustion method with sample combustion in a nitrogen-carbon (NC) analyzer at 850℃ and a flame ionization detector

(FID) with a nickel catalyst methanizer (Ohta and Okita 1984). Several disc-shaped samples with 1-cm diameter were cut off from the filter. One half of the samples was used to determine the total carbon (TC) content, and the other half was heated in an electric furnace at 300℃ in air for 30 min to remove organic carbon (OC) content; then, the elemental carbon (EC) content was measured by the above combustion technique. The difference between the total and elemental carbon content is expressed as [OC] and obtained with the equation:

$$[\mathrm{OC}] = [\mathrm{TC}] - [\mathrm{EC}].$$

To correct the positive artificial organic carbon, the carbon content on the quartz fibrous filter behind the Teflon filter (QBT) was determined. Subtraction of the carbon content of the QBT from the OC value gives a corrected organic carbon amount (Cor. OC).

Fujitani et al. (2004) measured the optical and chemical properties of marine aerosols over the western Pacific Ocean during the Research Vessel (R/V) Mirai cruise in 2002, and Fujitani et al. (2007) measured these properties over the central equatorial Pacific Ocean during the 2003 R/V Mirai cruise. The measurement systems were the same as those in this study. They reported the detection limits for the chemical components of atmospheric aerosols (Fujitani et al. 2004).

They also estimated the detection limit of the volume scattering coefficient σ_{sca} and the volume absorption coefficient σ_{abs} (Fujitani et al. 2004). The detection limit of the 1-h σ_{sca}, determined from signal/noise measurements of particle-free air was 9.9×10^{-8} m^{-1}. Relative humidity was not controlled in that study, and the values in air samples were from 50% to 70%, which is approximately 20% lower than that of the outside air. The detection limit of σ_{abs} for a 1-h interval was 1.0×10^{-7} m^{-1}, estimated from signal/noise measurements of a blank filter.

The single scattering albedo ω was calculated from σ_{abs} and σ_{sca} for the same interval as follows:

$$\omega = \sigma_{sca}/(\sigma_{sca} + \sigma_{abs}).$$

Since the σ_{sca} and σ_{abs} wavelengths were different, ω is considered as the value over a broad band between 0.530 and 0.565 μm.

Bond et al. (1999) estimated a maximum error of 22% in σ_{abs} by overestimation in the absorption because of multiple scattering and scattering misinterpreted as absorption. Fujitani et al. (2004, 2007) further corrected the measured scattering coefficient with the integrating nephelometer for the relative humidity in the manifold to the ambient relative humidity according to Hegg et al. (1996), and estimated systematic errors of σ_{sca} and σ_{abs} to be 7.3% and 22%, respectively; the absolute errors of ω ($|\Delta\omega|$) were then 0.06 (ω = 0.7) and 0.006 (ω = 0.98).

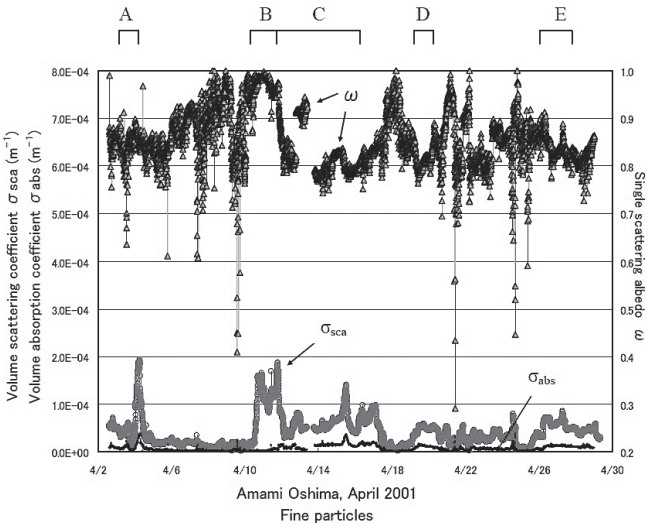

Fig. 2. Volume scattering coefficient σ_{sca}, volume absorption coefficient σ_{abs}, and single scattering albedo ω of atmospheric fine particles at Amami Oshima from April 2 to 28, 2001.

For the long-range transport studies of aerosols, following Murao et al. (1997), a four-day backward isentropic trajectory analysis at 850 hPa was employed, which is considered as top of the planetary boundary layer, every 6 h at Amami Oshima and Fukue Island. This analysis used global objective analysis data from the Japan Meteorological Agency. The trajectories on any given day describe the values at 3: 00, 9: 00, 15: 00, and 21: 00 JST.

3. Measurement results

a. Observation at Amami Oshima from April 2 to 28, 2001

Figure 2 shows the volume scattering coefficient σ_{sca}, the volume absorption coefficient σ_{abs}, and the single scattering albedo ω of atmospheric fine particles at Amami Oshima from April 2 to 28, 2001. The marks A–E on the figure denote the periods of high values of σ_{sca} and high concentrations of sulfate particles shown in Table 1. The maximum of σ_{sca} and σ_{abs} were 190×10^{-6} and 40×10^{-6} m^{-1}, respectively, and the values of ω were 0.70–0.98. From the evening of April 10 to 16, σ_{sca} was high, while the single scattering albedo ω

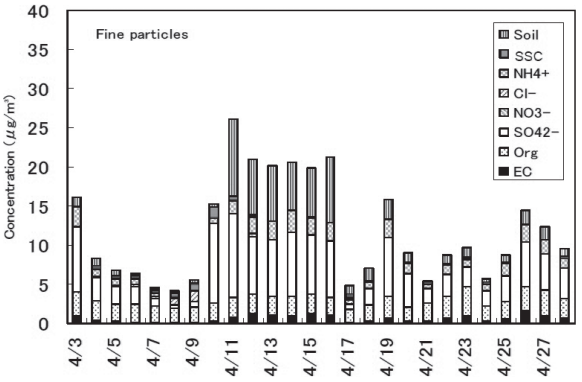

Fig. 3. Concentrations of chemical species in the atmospheric fine particles at Amami Oshima from April 3 to 28, 2001. EC, Org, SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, SSC, and Soil denote the concentrations of elemental carbon, organics, sulfate, nitrate, chloride, ammonium, sea salt cations, and soil particles, respectively.

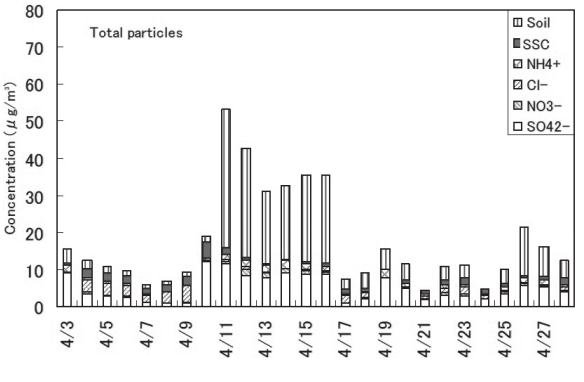

Fig. 4. Concentrations of water soluble and soil components in the atmospheric total particles at Amami Oshima from April 3 to 28, 2001.

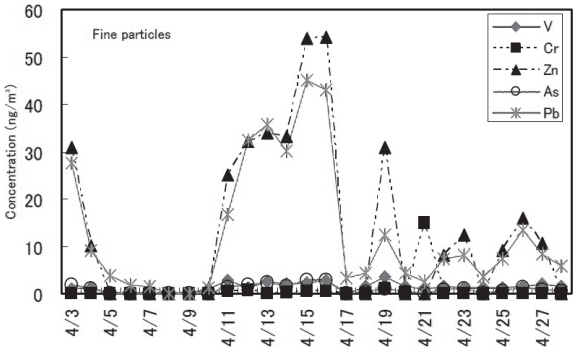

Fig. 5. Concentrations of metal components in the atmospheric fine particles at Amami Oshima from April 3 to 28, 2001. Pb, As, Zn, Cr, and V denote the concentrations of lead, arsenic, zinc, chromium, and vanadium, respectively.

increased to 0.98 from the evening of April 10 to 11 and decreased to 0.80–0.90 from April 12 to 16.

Figure 3 shows the concentrations of chemical species in the fine particles from April 3 to 28, 2001. Figure 4 shows the concentrations of water soluble and soil components in the total particles from April 3 to 28, 2001. Figure 5 shows the concentrations of metal components in the fine particles from April 3 to 28, 2001.

In these figures, EC, Org, SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, SSC, and Soil denote the concentrations of elemental carbon, organics, sulfate, nitrate, chloride, ammonium, sea salt cations, and soil particles, respectively. Pb, As, Zn, Cr, and V denote the concentrations of lead, arsenic, zinc, chromium, and vanadium, respectively.

The amounts of organics [Org], sea salt cations [SSC], and soil particles [Soil] were calculated by Ohta et al. (1998),

$$[\mathrm{Org}] = 1.20 \times [\mathrm{OC}],$$

and by Mason (1970),

$$[\mathrm{SSC}] = 1.194 \times [\mathrm{Na}^+], \text{ and } [\mathrm{Soil}] = [\mathrm{Al}]/0.0813,$$

where $[Na^+]$ is the concentration of water soluble sodium, and [Al] is the concentration of aluminum in the particles.

According to the chemical analysis of atmospheric aerosols in Sapporo, Japan (Ohta et al. 1998), fine particles over Sapporo consist of EC, organics, SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, SSC, soil particles, and water, and the sum of the above nine components represents 90%–116% of the particulate mass. Thus, Fig. 3 represents a chemical characterization of the fine particles at Amami Oshima. In Fig. 3, the concentrations of organics, SO_4^{2-}, and NH_4^+ were high on April 3, April 10–16, April 19, and April 26–27, while those of EC were high on April 3, April 12–16, April 19, April 23, and April 26–27. The concentration of soil was extremely high from April 11 to 16.

In Fig. 4, the concentration of the total particles of SO_4^{2-} was high on April 3, April 10–16, April 19–20, and April 26–27, while that of soil was extremely high from April 11 to 16.

In Fig. 5, the concentrations of Zn and Pb fine particles were high on April 3, April 11–16, April 19, April 22–23, and April 25–27.

Table 1. Optical properties and chemical components in atmospheric aerosols at Amami Oshima in the periods April 3 (period A), from the evening of April 10–11 (B), April 12–16 (C), April 19 (D), and April 26–27 (E), 2001. σ_{sca} and ω indicate the volume scattering coefficient and the single scattering albedo of the fine particles, respectively. The values of SO_4^{2-}, EC, and Zn are the concentrations of sulfate, elemental carbon, and zinc in the fine particles, respectively. The value of Al is the concentration of aluminum in the total particles. In period B, the concentrations of SO_4^{2-}, EC, Al, and Zn in the upper column were measured on April 10 (from the evening of April 10 to the morning of April 11), and those in the lower column were measured on April 11 (from the afternoon to the end of the day).

Period	Date	σ_{sca}	ω	SO_4^{2-}	EC	Al	Zn
	April	$10^{-6}m^{-1}$		$\mu g\ m^{-3}$	$\mu g\ m^{-3}$	$\mu g\ m^{-3}$	$\mu g\ m^{-3}$
A	3	10–190	0.81–0.88	8.5	1.0	0.30	0.030
B	10–11	10–190	0.90–0.98	10.2	0.22	0.10	0.000
				10.6	0.79	3.04	0.024
C	12–16	30–140	0.80–0.90	7.1–8.3	1.0–1.3	1.60–2.37	0.032–0.054
D	19	40–60	0.78–0.88	7.5	0.75	0.45	0.031
E	26–27	30–75	0.82–0.88	5.3	1.3	0.85	0.014

In Figs. 2–5, there are five periods of high values of the volume scattering coefficient σ_{sca} (above 60×10^{-6} m^{-1}) and high concentrations of SO_4^{2-} (above 5.3 μg m^{-3}) in the fine particles. These are denoted as period A (April 3), period B (from the evening of April 10–11), period C (April 12–16), period D (April 19), and period E (April 26–27), as shown by marks A–E on Fig. 2.

Concentrations of EC, soil, Zn, and Pb were high on April 3 (period A), April 11 (period B), April 12–16 (period C), April 19 (period D), and April 26–27 (period E), but not on April 10 in period B. On April 10, only the SO_4^{2-} concentration was high, and the single scattering albedo ω reached 0.98, showing that the aerosols on April 10 were highly transparent because of the low absorption coefficient σ_{abs} with low concentrations of EC (black particles) and the high scattering coefficient σ_{sca} with large amounts of SO_4^{2-} (transparent particles). On April 3, from the afternoon of April 11 to 16, on April 19, and from April 26 to 27, the concentrations of EC and soil particles were high as was SO_4^{2-}, and the single scattering albedo was low, 0.78–0.90, indicating the presence of a large absorption coefficient.

In Fig. 4, the soil particle concentration was high from April 11 to 16. This coincides with the period of the "Kosa event" in Japan and Korea. Fig. 5 shows that in the "Asian yellow dust event", concentrations of Zn and Pb were also high in fine particles. Zn and Pb are anthropogenic, which implies that large volumes of anthropogenic particles were transported from April 11 to 16 together with Asian yellow dust.

Table 1 shows the measurements made at Amami Oshima of the optical properties and concentrations of chemical components in the atmospheric aerosols in the periods April 3 (period A), from the evening of April 10–11 (period B), April 12–16 (period C), April 19 (period D), and April 26–27 (period E), 2001.

In the high turbidity periods A–E, the maximum values of σ_{sca} were 60×10^{-6} m^{-1} (period D) to 190×10^{-6} m^{-1} (periods A and B), and the maximum concentrations of SO_4^{2-} were 5.3 $\mu g\ m^{-3}$ (period E) to 10.6 (period B) $\mu g\ m^{-3}$. Furthermore, in period A, on April 11 in period B, and in periods C, D, and E, the concentrations of EC, Al, and Zn were higher than 0.75, 0.30, and 0.014 $\mu g\ m^{-3}$, respectively; on April 10 in period B, the concentrations of EC, Al, and Zn were much lower than those in other high turbidity periods. In period B, the SO_4^{2-} concentration was also very high, causing the single scattering albedo ω increase to 0.90–0.98. In the other periods, A, C, D, and E, the value of ω was 0.78–0.90 because of the high concentrations of EC.

Figure 6 shows the four-day backward trajectory analysis at Amami Oshima on April 10, 2001 (at 3: 00, 9: 00, 15: 00, and 21: 00 JST on April 10). From the figure, the air mass originated in the area above the north-western Pacific Ocean along the Miyake Island volcano (point M in the figure). The Miyake Island volcano emitted much sulfur dioxide, 20–30 thousand tons per day, in the spring of 2001 (Tomiya 2006).

The peak of the Miyake Island volcano (Mt. Oyama) is at 813 m, and the volcanic plume height above Mt. Oyama was from 800 to 1200 m in May and June 2001, respectively, according to Tomiya (2006). The highest altitude of the emitted sulfur dioxide was 1.6 to 2.0 km at Miyake Island. In Fig. 6, the air mass between 800

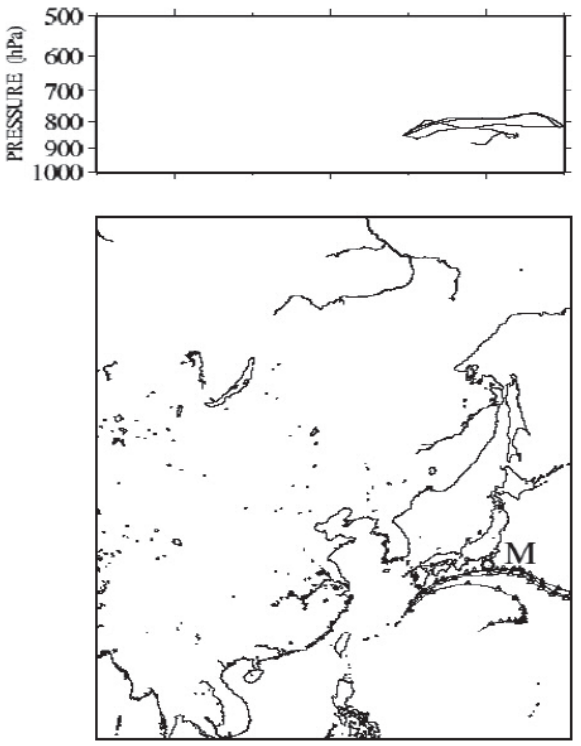

Fig. 6. Four-day backward trajectories reaching Amami Oshima on April 10, 2001 (at 3: 00, 9: 00, 15: 00, and 21: 00 JST on April 10). Point M is the Miyake Island. The air mass came from the area above the north-western Pacific Ocean along a line from the Miyake Island volcano to Amami Oshima.

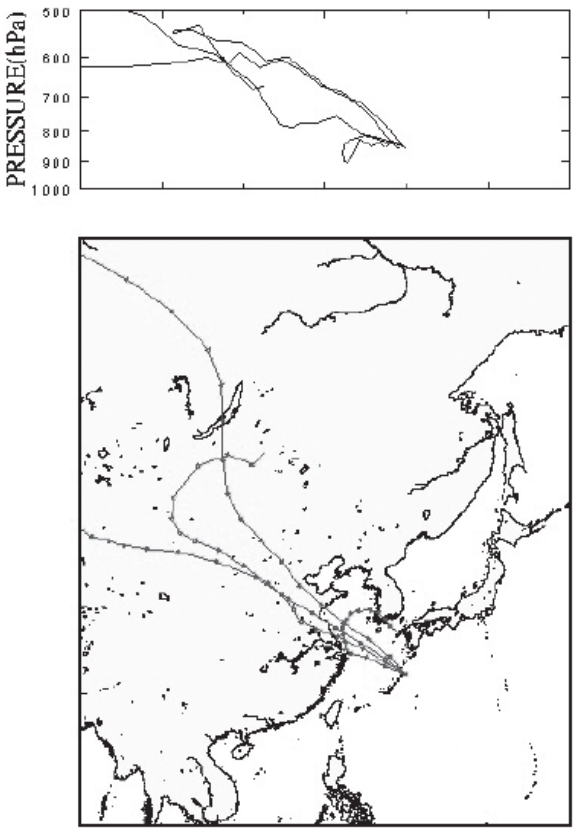

Fig. 7. Four-day backward trajectories reaching Amami Oshima on April 12, 2001 (at 3: 00, 9: 00, 15: 00, and 21: 00 JST on April 12). The air in the morning of April 12 came from the coastal area of Korea and China and in the evening of April 12 from inland continental China.

and 850 hPa (altitude 1.5 and 2.0 km) was transported from Miyake Island to Amami Oshima at 850 hPa (top of the planetary boundary layer) on April 10. It is suggested that the high concentrations of sulfate particles at Amami Oshima in period B were generated by a conversion of sulfur dioxide emitted from the Miyake Island volcano.

Figure 7 shows the four-day backward trajectories reaching Amami Oshima on April 12, 2001 (at 3: 00, 9: 00, 15: 00, and 21: 00 JST on April 12). In the figure, the air mass on the west of Beijing at 600 hPa (altitude 4 km) was transported to the Amami Oshima at 850 hPa (altitude 1.5 km).

Shimizu et al. (2004) made continuous observations of Asian dust and other aerosols by polarization lidars in China (Beijing) and Japan (Nagasaki and Tsukuba) during March to May 2001. They showed time-height sections of the measured backscattering coefficients at Nagasaki by dust and spherical particles with lidars, and of the calculated concentrations of dust and sulfate masses by the Chemical Weather Forecasting System (CFORS). In Fig. 12 of Shimizu et al. (2004), the lidar backscattering coefficients of dust at Nagasaki were high from April 11 to April 17, 2001, from the surface to an altitude of 3 km, while in Fig. 15, the lidar backscattering coefficients of spherical particles were high from April 10 to 16, 2001, from the surface to an altitude of 3 km. In Fig. 12 of Shimizu et al. (2004), the CFORS dust mass concentrations at Nagasaki were high from April 11 to 14, 2001, from the surface to the altitude of 3 km, while in Fig. 15, the CFORS sulfate mass concentration at Nagasaki was high from April 10 to 16, 2001, from the surface to an altitude of 2–4 km. This suggests that in period B, large amounts of transparent sulfate aerosols were generated from the sulfur dioxide transported from the Miyake Island volcano, while in period C, much of the absorptive aerosols were transported from the coastal area of

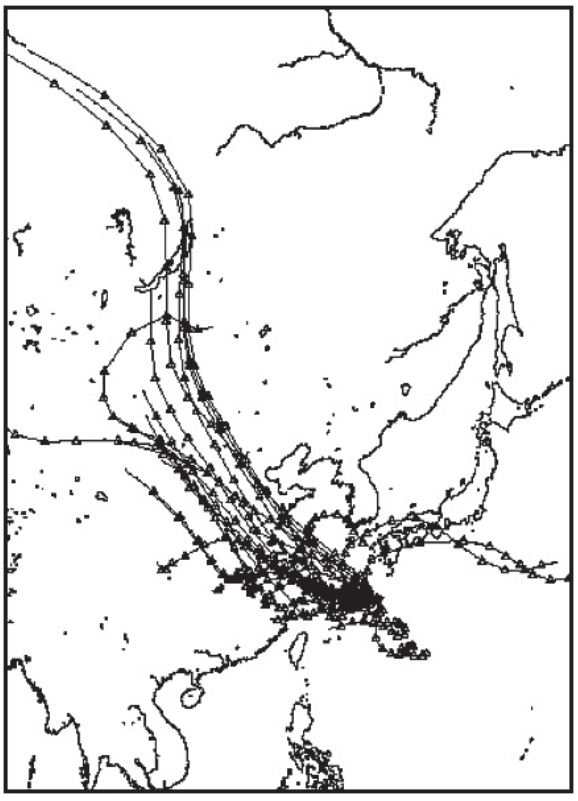

Fig. 8. Four-day backward trajectories reaching Amami Oshima from April 11 to 16, 2001. The air mass came from the western inland desert areas via the coast of central China.

Korea and China and from inland continental China.

Figure 8 in this paper shows the backward trajectories reaching Amami Oshima from April 11 to 16, 2001. Here, the air mass came from the western inland desert areas via the coast of central China. As shown in Fig. 3, at Amami Oshima from April 11 to 16, the SO_4^{2-} concentration was also high.

Thus, the highly concentrated sulfate particles were transported from continental China together with other anthropogenic components including EC, OC, Zn, and Pb as well as soil particles (Asian yellow dust), as shown in Figs. 3, 4, and 5.

Furthermore, the turbid aerosols in period A, on April 11 in period B, and in periods C, D, and E were absorptive, consisting of anthropogenic and soil-derived components, while in period B, they were transparent aerosols rich in SO_4^{2-}, which was converted from sulfur dioxide gas emitted by the Miyake Island volcano. Although the SO_4^{2-} concentrations at Amami Oshima were also high from April 12

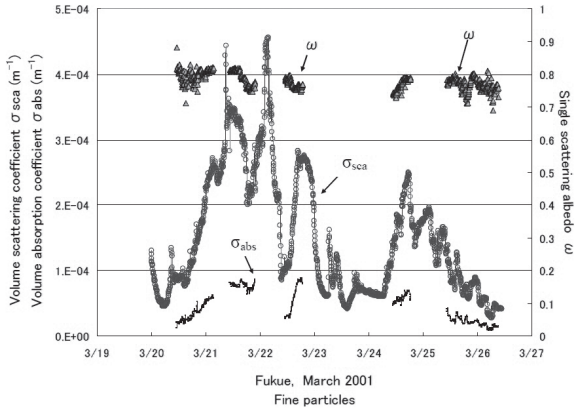

Fig. 9. Volume scattering coefficient σ_{sca}, volume absorption coefficient σ_{abs}, and single scattering albedo ω of atmospheric fine particles at Fukue Island from March 19 to 26, 2001.

to 16 (in period C), here the highly concentrated SO_4^{2-} was transported from the continent of China together with other anthropogenic components such as soot, zinc, and lead, as well as soil particles (Al) (Asian yellow dust).

b. Observations at Fukue Island from March 19 to 26, 2001.

Figure 9 shows the volume scattering coefficient σ_{sca}, the volume absorption coefficient σ_{abs}, and the single scattering albedo ω of fine particles in the atmosphere measured at Fukue Island from March 19 to 26, 2001. The maximum values of σ_{sca} and σ_{abs} were 460×10^{-6} and 90×10^{-6} m^{-1}, respectively, and that of ω was 0.75–0.82. In particular, on March 21 and 22, the scattering coefficient σ_{sca} increased to a maximum of 460×10^{-6} m^{-1}.

Figure 10 shows the concentrations of chemical species in the fine particles. On March 21, concentrations of EC and SO_4^{2-} were high, 6.6 μg m^{-3} and 20.7 μg m^{-3}, respectively. These values are much higher than those in an urban area like Sapporo, Japan (Ohta et al. 1998). Accompanied by the high concentrations of EC, the single scattering albedo for this period in Figure 9 was a low 0.75–0.82.

Figure 11 shows the concentrations of the water soluble fraction and soil components among the total particles for the period March 20 to 25. On March 21, 22, and 24, the soil concentrations were high, in particular, above 57 μg m^{-3} on March 22, and at this time, the SO_4^{2-} concentration was also high.

Figure 12 shows the concentrations of metals in the

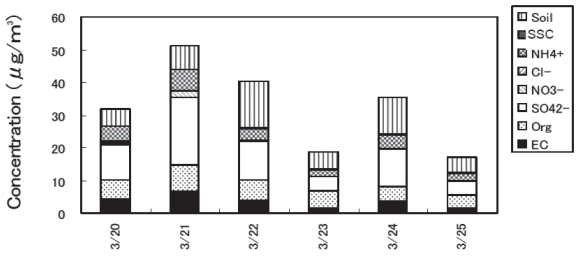

Fig. 10. Concentrations of chemical species in the atmospheric fine particles at Fukue Island from March 20 to 25, 2001.

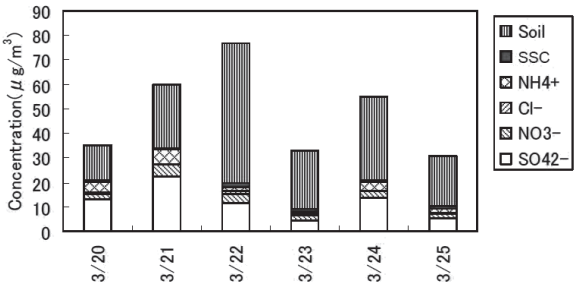

Fig. 11. Concentrations of the water soluble fraction and soil components in the atmospheric total particles at Fukue Island from March 20 to 25, 2001.

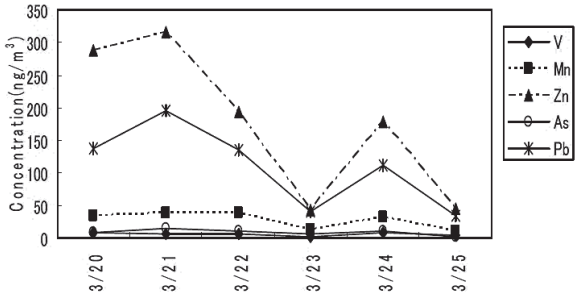

Fig. 12. Concentrations of metals in the atmospheric fine particles at Fukue Island from March 20 to 25, 2001.

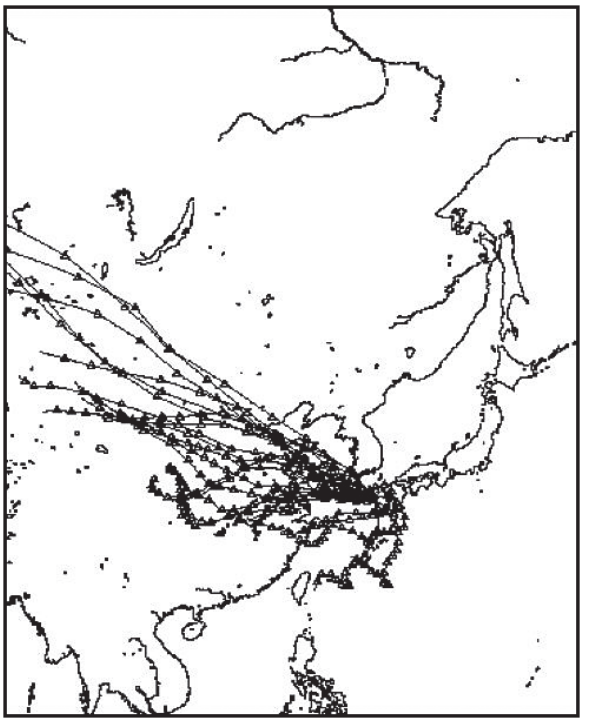

Fig. 13. Four-day backward trajectories reaching Fukue Island from March 20 to 25, 2001. The air mass came from the inland western desert areas via the coast of central China.

fine particles. On March 20, 21, 22, and 24, the concentrations of Zn and Pb were high, particularly on March 21, up to 0.32 and 0.20 μg m^{-3}, respectively.

Figure 13 shows the four-day backward trajectory analysis for the air mass reaching Fukue Island from March 20 to 25, 2001. The air mass came from the western inland desert areas via the coast of central China and carried urban aerosols as well as soil particles (Asian yellow dust). Thus, it may be assumed that the air over Fukue Island in the period had heavy concentrations of anthropogenic particles and Asian yellow dust.

Table 2 shows the measured results at Fukue Island for σ_{sca} and ω in the fine particles, and for the concentrations of SO_4^{2-}, EC, and Zn in the fine particles, with the concentration of Al in the total particles, during March 21 to 22 and on 24, 2001. On these days, σ_{sca} was high, up to 460×10^{-6} m^{-1}, and the concentrations of SO_4^{2-}, EC, Al, and Zn were also high. However, the single scattering albedo ω was low, 0.75–0.82. The high values of σ_{sca} were caused by large amounts of sulfate, and the low values of ω were due to the large amounts of absorptive particles including EC and soil (Al). From the backward trajectory analysis (Fig. 13), the highly concentrated aerosols contained anthropogenic aerosols and Asian yellow dust originating from inland continental China.

Table 2. Optical properties and concentrations of chemical components in atmospheric aerosols at Fukue Island during March 21 to 22 and on March 24, 2001. σ_{sca} and ω are the volume scattering coefficient and the single scattering albedo of the fine particles, respectively. The SO_4^{2-}, EC, and Zn denote the concentrations of sulfate, elemental carbon, and zinc in the fine particles, respectively, and Al is the concentration of aluminum in the total particles.

Date March	σ_{sca} ($10^{-6}m^{-1}$)	ω	SO_4^{2-} (μgm^{-3})	EC (μgm^{-3})	Al (μgm^{-3})	Zn (μgm^{-3})
21–22	200–460	0.75–0.82	11.7–20.7	4.0–6.6	2.11–4.64	0.19–0.32
24	100–250	0.75–0.80	11.6	3.5	2.76	0.18

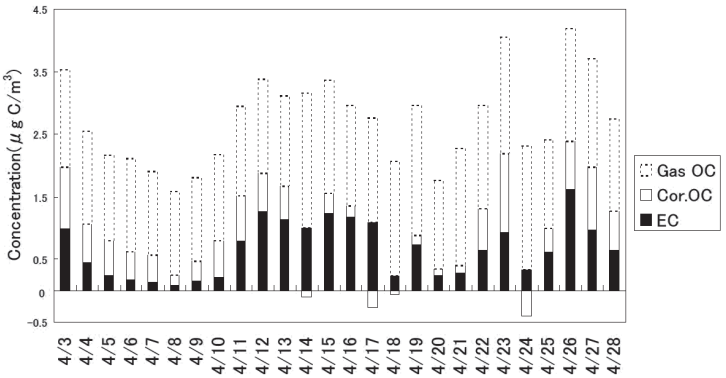

Fig. 14. Concentrations of elemental carbon (EC) and corrected organic carbon (Cor. OC), as well as adsorbed organic carbon (Gas OC) in the fine particles at Amami Oshima from April 3 to 28, 2001.

4. Discussion

Correction of OC concentrations collected by quartz fibrous filters

The measurements here used pre-treated quartz fibrous filters for aerosol sampling, and the quartz fibrous filters were heated at 850℃ in air before the sampling in order to remove carbonaceous contaminants. However, during heating, the quartz filter adsorbs organic gaseous components that may be present (Kirchstetter et al. 2001; Turpin et al. 1994). Figures 14 and 15 show the concentrations of elemental carbon (EC), corrected organic carbon (Cor. OC), and adsorbed gaseous organic carbon (Gas OC) at Amami Oshima and Fukue Island, respectively. At Amami Oshima, in April 2001 (Fig. 14), the ratio of OC (= Cor. OC + Gac OC) to EC (OC/EC) was 3–10, while in the measurements at Fukue Island shown in Fig. 15, the ratio was 1.1–2.3. In the measurements of urban aerosols in Sapporo (Ohta et al. 1998), the OC/EC ratio was approximately 1.0. Thus, in short-sustained sampling for low concentrations of organic particles at Amami Oshima and Fukue Island, the adsorption of organic gas probably showed apparently high concentrations of organic particles. To control this, a correction of OC by the tandem QBT method was made, and Figs. 14 and 15 show the corrected organic carbon concentrations.

In these figures, the Gas OC shows the adsorbed organic gas contribution (difference between OC and Cor. OC, top part of the bars). At Amami Oshima in Fig. 14, the concentrations of Cor. OC are less than half of the apparent OC concentrations, and there are negative values for April 14, 17, 18, and 24. At Fukue Island in Fig. 15, the concentrations of Cor. OC are approximately half the apparent OC concentrations. At Amami Oshima, the concentrations of carbonaceous particles are low, approximately 1.0 μgC m^{-3}, and the correction may not be accurate because of the insufficient analytical technique and the variation in the adsorption capacity of the filters with different lots. From these results, it may be considered that in the north-western Pacific Ocean area in spring, the OC/EC ratio is approximately one, making the concentrations

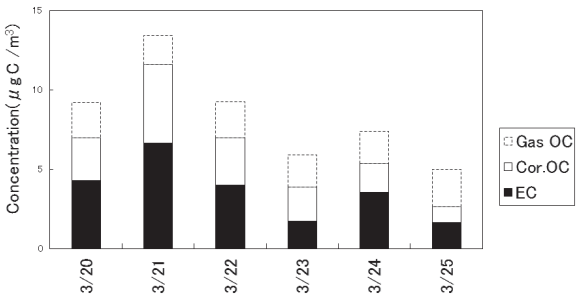

Fig. 15. Same as Fig. 14, but for Fukue Island from March 20 to 25, 2001.

of particulate organic carbon approximately the same as those of EC.

The adsorption is saturated by the large amounts of organic gases passing through the quartz filters. Then, in the weekly and monthly continuous sampling under moderate concentrations of organic particles, and in the sampling under high concentrations of organic particles, it is possible to disregard the amounts of artificial organic particles occurring with the gas adsorption.

5. Summary and conclusions

Measurements of the optical properties and concentrations of chemical components in atmospheric aerosols at Amami Oshima were performed from April 2 to 28 of 2001. There are five periods of high values of the volume scattering coefficient σ_{sca} (above 60×10^{-6} m^{-1}) and high concentrations of SO_4^{2-} (above 5.3 μg m^{-3}) in the fine particles. These are denoted as period A (April 3), period B (from the evening of April 10–11), period C (April 12–16), period D (April 19), and period E (April 26–27). In the high turbidity periods A–E, the maximum value of σ_{sca} was 60×10^{-6} m^{-1} (period D) to 190×10^{-6} m^{-1} (periods A and B), and the maximum concentration of SO_4^{2-} was above 5.3 μg m^{-3} (period E) to 10.6 μg m^{-3} (on April 11, period B). Furthermore, on April 3, 11–16, 19, and 26–27, the concentrations of EC, Al, and Zn were higher than 0.75, 0.30, and 0.014 μg m^{-3}, respectively; on April 10, the concentrations of EC, Al, and Zn were much lower than those in other periods. In period B, the SO_4^{2-} concentration was also high; hence, the single scattering albedo ω increased to 0.90–0.98. In the other periods, the value of ω was 0.78–0.90 because of the high concentration of EC.

The results show that the turbid aerosols on April 3, from the afternoon of April 11 to 16, on April 19, and from April 26 to 27 were absorptive consisting of anthropogenic and soil-derived components, while from the evening of April 10 to 11 (period B), they were transparent aerosols rich in SO_4^{2-}, which was converted from sulfur dioxide gas emitted from the Miyake Island volcano. Though the SO_4^{2-} concentration at Amami Oshima was also high from April 12 to 16 (period C), the highly concentrated SO_4^{2-} was transported from the continent of China together with the other anthropogenic components such as soot, zinc, and lead, as well as soil particles (Al) (Asian yellow dust).

Measurements at Fukue Island of the optical properties and concentrations of chemical components in the atmospheric aerosols were made from March 19 to 26, 2001. On the days, σ_{sca} was high, up to 460×10^{-6} m^{-1}, and the concentrations of SO_4^{2-}, EC, Al, and Zn were also high. However, the single scattering albedo ω was low, 0.75–0.82. The high values of σ_{sca} were caused by large amounts of sulfate, and the low values of ω were due to the large amounts of absorptive particles including EC and soil (Al). From the backward trajectory analysis, the highly concentrated aerosols contained anthropogenic aerosols and Asian yellow dust originating from continental China.

The results suggest that in the spring of 2001, large amounts of anthropogenic particles were frequently transported together with the Asian yellow dust from the inland areas of continental China to the western Pacific Ocean, and the single scattering albedo of the fine particles over the north-western Pacific Ocean was approximately 0.80–0.85 in this period.

The concentrations of corrected organic carbon at Fukue Island were approximately half the apparent OC concentrations. At Amami Oshima, the concentrations of carbonaceous particles were low, approximately 1.0 μgC m^{-3}, and the correction was difficult because of the insufficient accuracy of the analytical technique and variations in the adsorptive capacity of the filters with different lots. From the results, it is concluded that in the north-western Pacific Ocean area in spring, the OC/EC ratio is approximately one, that is, the concentrations of particulate organic carbon are approximately the same as those in EC.

Acknowledgments

We express our sincere thanks to Mr. Ryo Kato for his support and discussion of the matters reported in this paper.

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