

EVALUATION OF OPTICAL PROPERTIES OF ATMOSPHERIC AEROSOLS BASED ON CHEMICAL CHARACTERIZATION

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ABSTRACT

Atmospheric fine particles, aerosols less than $2\mu\text{m}$ in diameter, were collected on filters and chemically analyzed in Sapporo, Okinawa island in Japan and Ester-Dome, Alaska in U.S.A. They were made up of nine components such as elemental carbon, organics, sulfate, nitrate, ammonium, sea-salt cations, soil and water.

Based on the chemical characterization, it was assumed that atmospheric aerosols comprise seven species of particles such as elemental carbon, organics, ammonium sulfate, ammonium nitrate, sea-salt, soil particles and mixed nuclei which have inner core of elemental carbon and outer shell of sulfate. Using log-normal size distribution, complex index of refraction and atmospheric concentrations of each chemical species particles, single scattering albedo and volume extinction coefficient of aerosols were evaluated by calculation with Mie scattering theory.

Keywords; chemical characterization of aerosols, Mie scattering theory, single scattering albedo, volume extinction coefficient, mixed nuclei

1. INTRODUCTION

In remote sensing of ocean color, vegetation or surface reflectivity of land surface, it is necessary to determine optical thickness, size distribution and single scattering albedo of atmospheric aerosols. But, it is difficult, in particular, to measure the single scattering albedo by a handy measuring instrument. We have developed a method to estimate the single scattering albedo based on chemical characterization of the atmospheric aerosols. In this paper, we present the chemical characterization of atmospheric aerosols, and evaluation of the single scattering albedo based on the characterization in Sapporo and Okinawa island in Japan, and Ester-Dome near Fairbanks, Alaska in U.S.A.

2. CHEMICAL CHARACTERIZATION OF ATMOSPHERIC AEROSOLS IN SAPPORO

2.1. Experimental

Atmospheric fine particles, aerosols less than $2\text{ }\mu\text{m}$ in diameter, were collected on two types of filters such as Teflon and quartz fiber filters simultaneously, equipped with a cyclone separator whose 50% cut off diameter was $2\text{ }\mu\text{m}$ at a flow rate of 20 l min^{-1} . The samplings were carried out every half month, continuously for 14 days, from November 1991 to October 1992 on a roof top of our office building at a height of 26m above the ground in the University Campus in Sapporo.

We analyzed chemical species of the fine particles collected on both Teflon and quartz fiber filters as in Ohta and Okita (1990) except for total particulate mass (TPM), water content and aluminum (Al). TPM was determined by weighing Teflon filters on an electric balance in a dry box with silica gel. Before and after the sampling, the Teflon filters were left in the dry box for more than 24 hours at a relative humidity less than 30%, and then weighed them by the electric balance.

The amount of water associated with aerosols on the Teflon filter was determined by Karl-Fischer method. The water was vaporized in a water evaporator, introduced in an aqua-counter, and titrated coulometrically in the Karl-Fischer reagent solution.

We used Al as a tracer material of soil. The amount of Al on the sampled Teflon filter was determined with particle induced X-ray emission (PIXE) method (Kasahara et al., 1993).

2.2. RESULTS

Atmospheric concentrations of chemical components of the aerosols were determined in elemental carbon (EC), organic carbon (OC), sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), chloride (Cl^-), sodium (Na^+), calcium (Ca^{2+}), aluminum (Al) and water.

By using above results, we calculated the following chemical species concentrations in the atmosphere.

The amount of organics was determined by multiplying the amount of organic carbon (OC) by 1.20 according to Countess et al. (1980),

$$\text{Organics} = 1.20 \times \text{OC}.$$

We denoted SS.Ci as the sum of sea-salt cations (Na^+ , Mg^{2+} , Ca^{2+} , K^+ and Sr^{2+}). Since Na^+ consists of 83.7% of the sum of the sea-salt cations, we obtained SS.Ci as follows,

$$\text{SS.Ci} = (1/0.837) \times \text{Na}^+.$$

The mass fraction of Al in Sapporo soil was 8.6% (Ohta and Okita, 1990). The amount of soil particles was, then, calculated by

$$\text{Soil particles} = (1/0.086) \times \text{Al}.$$

Figure 1 shows semimonthly mass balance of atmospheric fine particles in Sapporo.

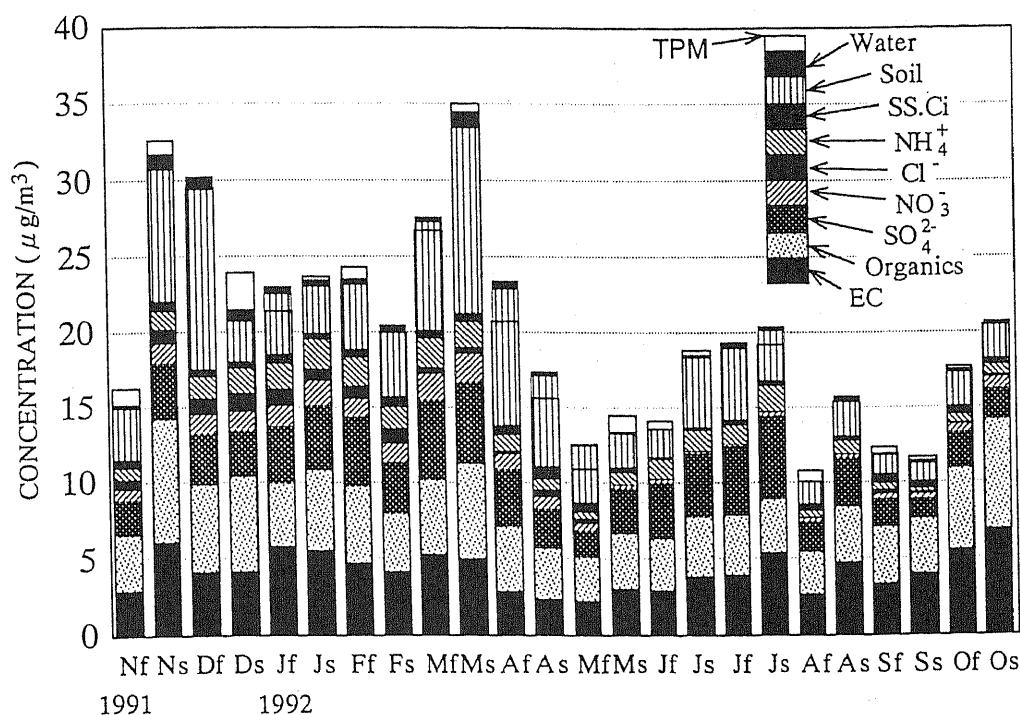


Fig.1 Semimonthly mass balance of atmospheric fine particles in Sapporo from November 1991 to October 1992. TPM, ss.Ci and EC are semimonthly concentrations of totla particulate mass, the sum of sea-salt cations and elemental carbon, respectively. Sampling periods, e.g. Nf and Ns represent the first and second half of November, respectively.

We defined the sum total of chemical components (TCC) as

$$\text{TCC} = \text{EC (elemental carbon)} + \text{OC} + \text{SO}_4^{2-} + \text{NO}_3^- + \text{Cl}^- + \text{NH}_4^+ + \text{SS.Ci} + \text{soil particle} + \text{water}$$

The ratio of the sum total of chemical components (TCC) to the total particulate mass (TPM) ranged from 90 to 116 %. We, thus, concluded that atmospheric fine particles in Sapporo almost consisted of nine components such as elemental carbon, organics, SO_4^{2-} , NO_3^- , Cl^- , NH_4^+ , sea-salt cations, soil particles and water. Under the condition of relative humidity less than 30%, in particular, the water content was negligible.

3. CHEMICAL CHARACTERIZATION OF ATMOSPHERIC AEROSOLS IN OKINAWA ISLAND AND ESTER-DOME, ALASKA.

We measured atmospheric concentrations of chemical species of fine particles, aerosols less than $2\mu\text{m}$ in diameter, weekly from May 1995 to September 1996 at Okinawa island in Japan, and monthly from August 1991 to August 1992 at Ester-Dome near Fairbanks, Alaska, U.S.A. The chemical characterization of the fine particles at Okinawa island and Ester-Dome are shown in Figure 2 and 3, respectively.

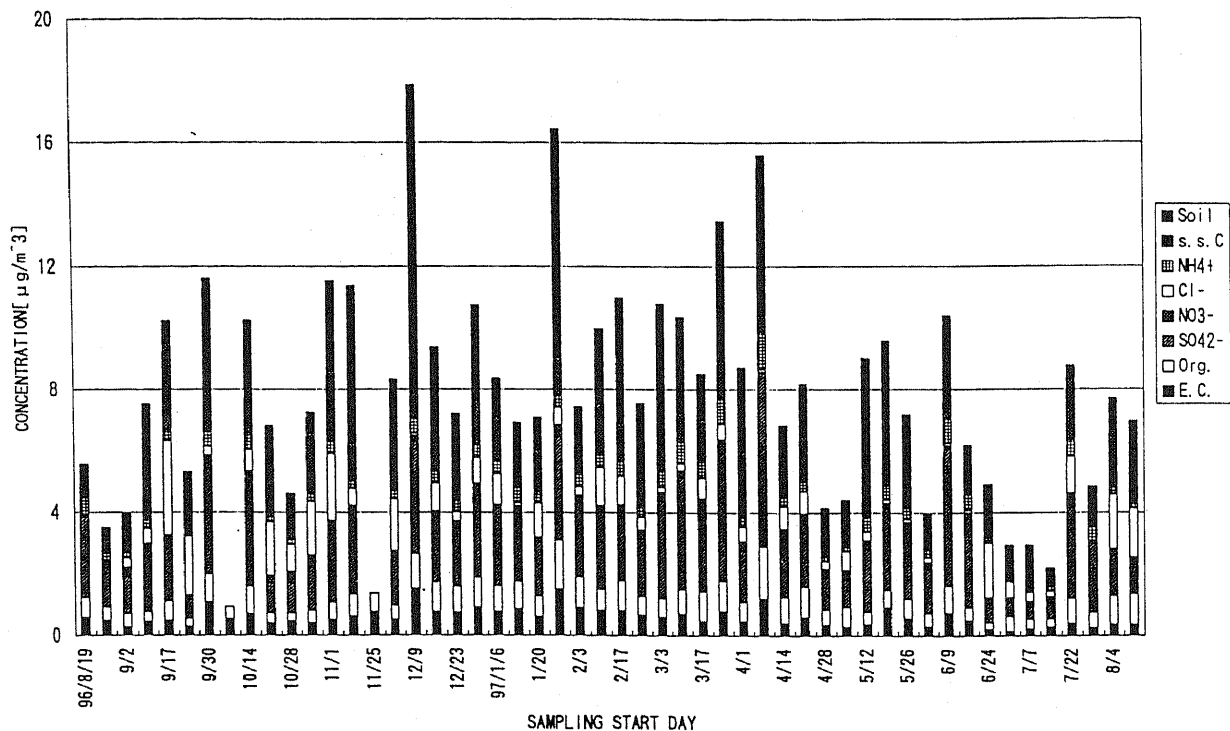


Fig.2 Chemical characterization of atmospheric fine particles in Okinawa island.

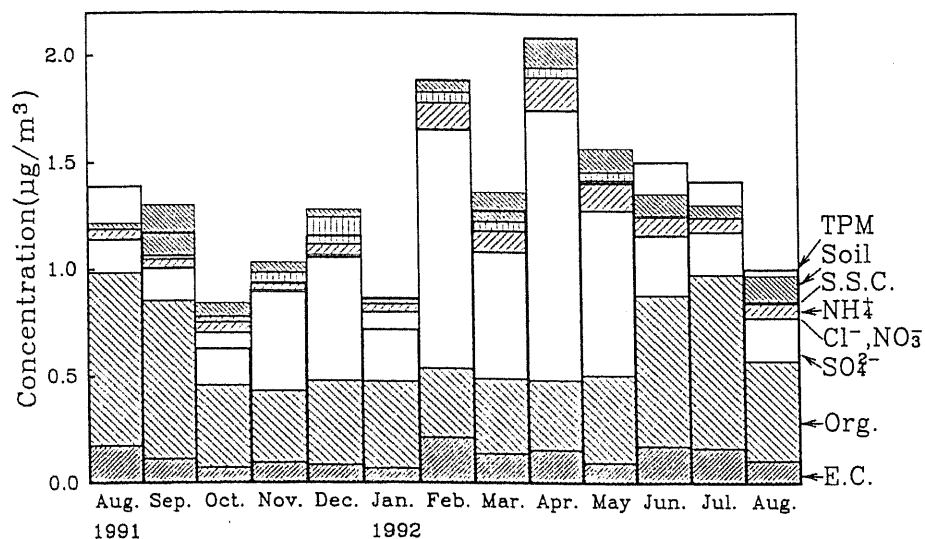


Fig.3 Chemical characterization of atmospheric fine particles at Ester-Dome near Fairbanks in Alaska.

4. EVALUATION OF OPTICAL PROPERTIES OF ATMOSPHERIC AEROSOLS BASED ON CHEMICAL CHARACTERIZATION

On the basis of the chemical characterization, we assumed that atmospheric aerosols had two kinds of mixing condition such as external mixture and internal half mixture. The external mixture comprises six species of

particles such as elemental carbon (EC), organics, $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , sea-salt and soil particles. Whereas internal half mixture contains mixed nuclei in addition to above six species of homogeneous particles. The mixed nuclei are assumed to have an inner core of elemental carbon and outer shell of sulfate, formed with half amount of EC and half amount of sulfate in the atmosphere.

The particles of elemental carbon, organics, $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and the mixed nuclei belong to the fine particles less than $2\mu\text{m}$ in diameter and sea-salt and soil particles belong to the coarse particles which had a size distribution in volume with a peak in the $2\text{--}20\mu\text{m}$ region.

We adopted the size distribution of each species of particles as log-normal in number distribution,

$$dN/d\ln D = N_0 / (\sqrt{2\pi} \ln \sigma_g) \cdot \exp[-(\ln D - \ln D_{gN})^2 / (2 \ln^2 \sigma_g)]$$

where dN is the number density of particles of diameter between D and $D+dD$,

N_0 is the total number density, D_{gN} is the geometric number mean diameter and σ_g is the geometric standard deviation of the distribution.

The geometric number mean diameter and the standard deviation were assumed to be $0.11\mu\text{m}$ and 2.10 for the fine particles, and $0.98\mu\text{m}$ and 2.15 for the coarse particles, respectively. We took account of the condensational growth of hygroscopic particles such as $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and sea-salt for monthly mean relative humidity. The growth of each species of particles was calculated according to the experimental studies by Tang et al. (1977).

Since we sampled aerosols by the system equipped with the cyclone separator whose 50% cut off diameter was $2\mu\text{m}$, we measured the whole fine particles, but only small fraction of the coarse particles.

The complex index of refraction at $0.50\mu\text{m}$ wavelength was assumed to be $1.50\text{--}0.55i$ for EC, $1.55\text{--}0.00i$ for organics, $1.55\text{--}0.00i$ for $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and sea-salt, and $1.55\text{--}0.008i$ for soil particles.

Using above size distribution and complex index of refraction for each species of particles i , the volume extinction coefficients σ_{Ei} and the volume scattering coefficient σ_{Si} were calculated by the Mie scattering theory. Those for the mixed nuclei we calculated following Toon and Ackerman (1981).

Thus, the total volume extinction coefficient σ_E of the atmospheric aerosols was obtained as the sum of volume extinction coefficients of each species of particles σ_{Ei} ,

$$\sigma_E = \sum \sigma_{Ei},$$

and the total volume scattering coefficient σ_s was similarly obtained as

$$\sigma_s = \sum \sigma_{Si}.$$

The single scattering albedo ω of the atmospheric aerosols was defined as the ratio of the total volume scattering coefficient to the total volume extinction coefficient,

$$\omega = \sigma_s / \sigma_E.$$

5. OPTICAL PROPERTIES OF ATMOSPHERIC AEROSOLS

Figures 4 and 5 show the semimonthly mean single scattering albedo and volume extinction coefficient of the atmospheric aerosols in external mixture and internal half mixture, respectively, at the wavelength of $0.5 \mu\text{m}$ in Sapporo from November 1991 to October 1992. In Figure 4 the single scattering albedo and the volume extinction coefficient in external mixture ranged from 0.73 to 0.85 and from 5.8×10^{-7} to $1.47 \times 10^{-6} \text{ cm}^{-1}$, respectively. In the calculation there were no mixed nuclei, that is, EC and sulfate suspended in isolation in the atmosphere. Whereas, in the internal half mixture shown in Figure 5 which had mixed nuclei formed with half amount of EC and half amount of sulfate in the atmosphere, the single scattering albedo and the volume extinction coefficient ranged from 0.68 to 0.78 and from 5.8×10^{-7} to $1.46 \times 10^{-6} \text{ cm}^{-1}$, respectively. Thus, the calculated single scattering albedo decreased by 0.05 to 0.07 compared with that in the external mixture.

Figur 6 shows weekly single scattering albedo and volume extinction coefficient of atmospheric aerosols in internal half mixture in Okinawa island. The single scattering albedo and volume extinction coefficient ranged from 0.80 to 0.95 and 1.5×10^{-7} to $8.0 \times 10^{-7} \text{ cm}^{-1}$, respectively. Whereas, in external mixture condition, the single scattering albedo increased in the range of 0.87 to 0.97.

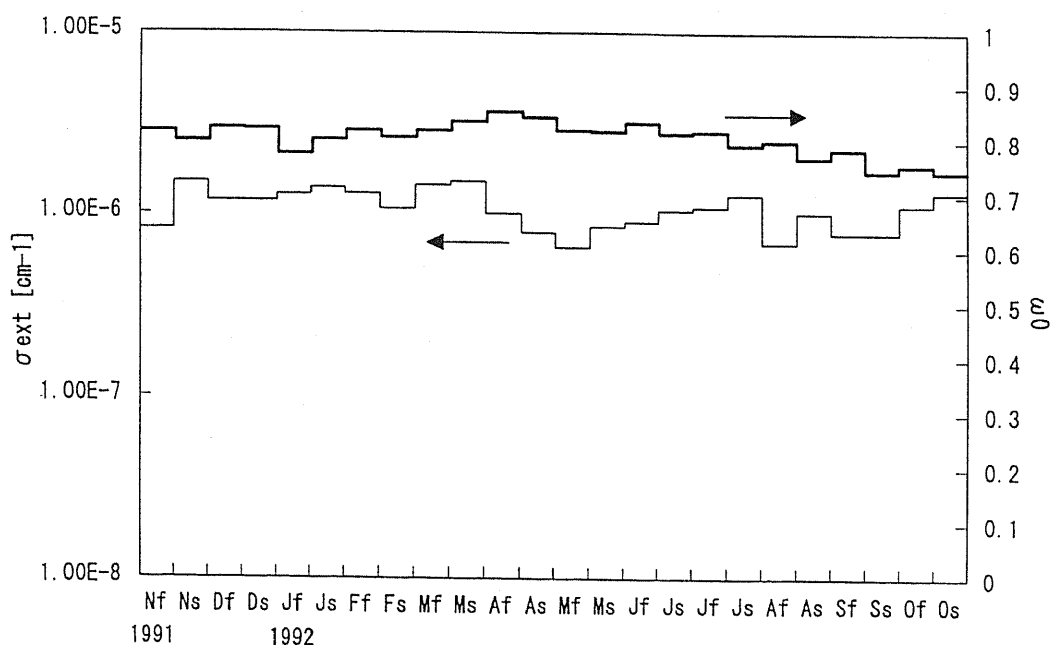


Fig.4 Semimonthly mean single scattering albedo and volume extinction coefficient of atmospheric aerosols in external mixture in Sapporo

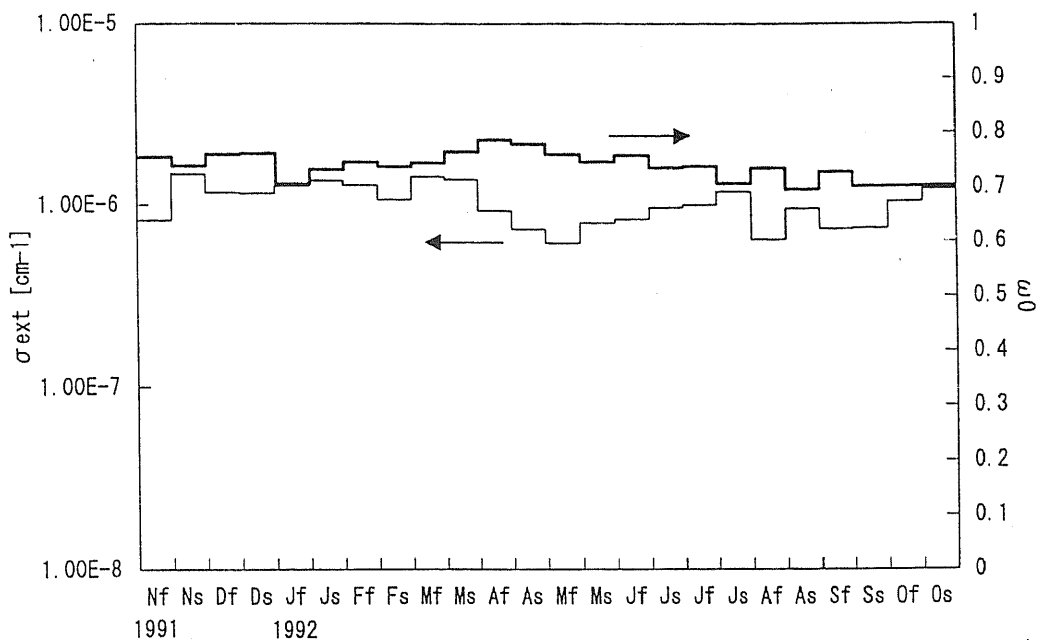


Fig.5 Semimonthly mean single scattering albedo and volume extinction coefficient of atmospheric aerosols in internal half mixture in Sapporo.

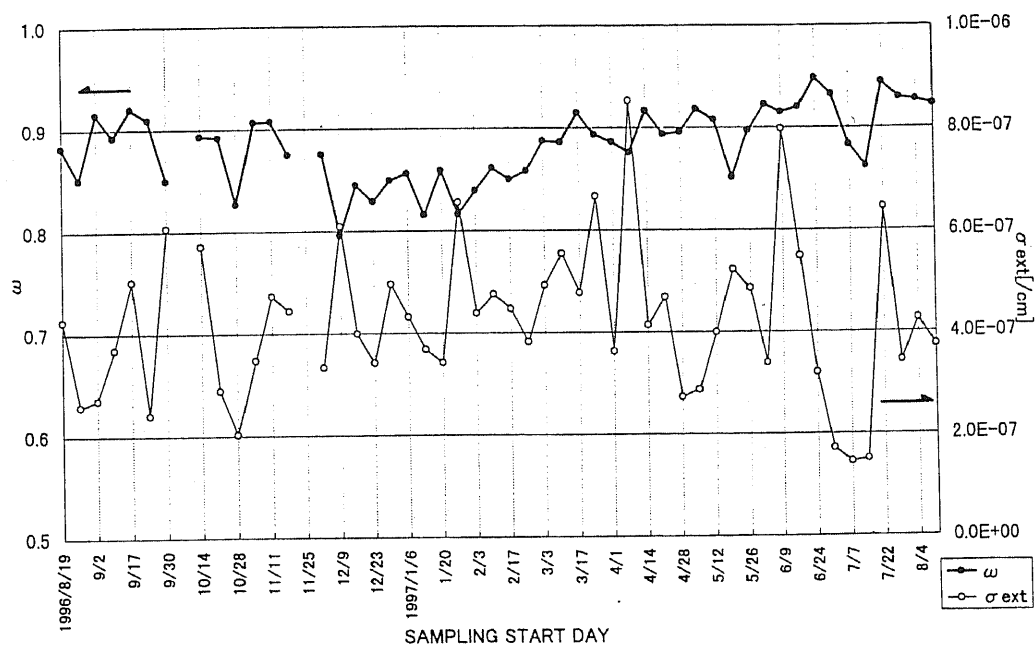


Fig.6 Weekly mean single scattering albedo and volume extinction coefficient of atmospheric aerosols in internal half mixture in Okinawa island.

Figure 7 shows the monthly mean single scattering albedo and volume extinction coefficient of atmospheric aerosols in internal half mixture at Ester-Dome, Alaska. The single scattering albedo ranged from 0.83 to

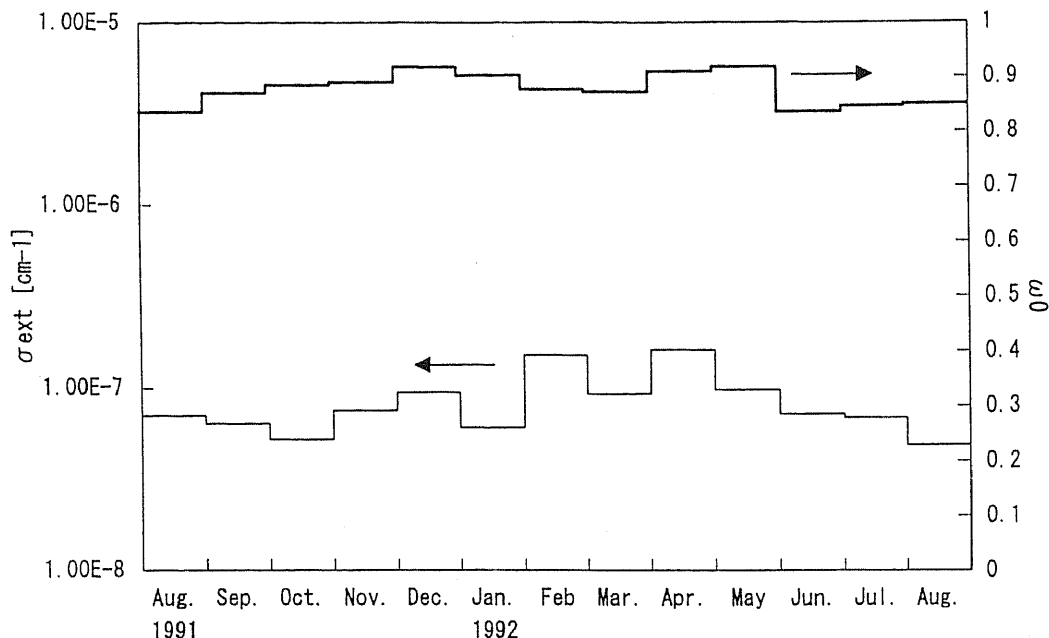


Fig.7 Monthly mean single scattering albedo and volume extinction coefficient of atmospheric aerosols in internal half mixture at Ester-Dome near Fairbanks, Alaska.

0.92 and from 5.2×10^{-8} to $1.7 \times 10^{-7} \text{ cm}^{-1}$, respectively. In the external mixture condition, the single scattering albedo ranged from 0.88 to 0.96.

We found that there were differences of 0.04 to 0.07 in the single scattering albedo between in the external mixture and the internal half mixture aerosols. Thus it is important, hereafter, to determine the concentration of the mixed nuclei in the atmosphere.

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