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CHEMICAL AND OPTICAL PROPERTIES OF LOWER TROPOSPHERIC AEROSOLS MEASURED AT Mt.LEMMON IN ARIZONA

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Abstract

Atmospheric concentrations of mass and chemical species of aerosols less than 2 μm in diameter, and gaseous SO_2, NH_3, HNO_3 and HCl were measured at the top of Mt.Lemmon (at a height of 2791 m) near Tucson, Arizona from September 1992 through September 1994. Monthly mean concentration of the aerosols ranged from 0.64 to 3.49 μgm^{-3}, which increased in spring and summer, and decreased late in fall and in winter. Monthly mean concentrations of the aerosol species ranged as follows: elemental carbon (0.07-0.21 μgCm^{-3}), organics (0.15-0.81 μgm^{-3}), SO_4^{2-} (0.29-1.56 μgm^{-3}), Cl^- (0.00-0.03 μgm^{-3}), NO_3^- (0.00-0.02 μgm^{-3}), NH_4^+(0.10-0.54 μgm^{-3}), sea-salt cation (0.005-0.10 μgm^{-3}) and soil (0.02-0.47 μgm^{-3}). The lower tropospheric aerosols in Arizona were made up of above eight species. Monthly mean concentrations of gaseous species in ppbv were SO_2 (0.14-0.51), NH_3 (0.06-0.77), HNO_3 (0.05-0.29) and HCl (0.04-0.27).

Monthly mean single scattering albedo ω and volume extinction coefficient σ_{ext} of the aerosols were calculated based on the measurement results. The values of ω and σ_{ext} for external mixture aerosol ranged from 0.90 to 0.96 and from 3.6×10^{-8} to $1.5 \times 10^{-7} cm^{-1}$, respectively. On the other hand, in case of internal half mixture aerosol which contained mixed nuclei formed with elemental carbon and sulfate, ω decreased to be 0.85-0.93. The albedo effect of aerosols on climate depends strongly on the value of ω as well as the albedo of the underlying earth surface. In our method of estimation of the optical properties, then, it is important to determine concentration of the mixed nuclei in the atmosphere.

KEYWORDS: *atmospheric aerosols, lower troposphere, Arizona, aerosol chemical species, SO_2, NH_3, HNO_3, HCl, single scattering albedo, volume extinction coefficient*

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1. Introduction

Atmospheric aerosols are likely to affect global climate by changing the planetary albedo (the albedo of the earth-atmosphere system) through scattering and absorption of solar radiation. This is called the direct effect or the albedo effect of aerosols on climate (Yamamoto and Tanaka, 1972; Coakley *et al.*, 1983; Charlson *et al.*, 1991).

The aerosols also affect the climate by changing cloud albedo due to increasing cloud droplet concentration, because water soluble aerosols are easy to become cloud condensation nuclei. This is called the indirect effect of aerosols on climate (Twomey 1977; Coakley *et al.*, 1987; Nakajima *et al.*, 1991).

In evaluation of the albedo effect of aerosols, we have to determine the global distribution of optical thickness, size distribution and complex refractive index (or single scattering albedo) of aerosols. We, then, have developed a method to estimate the single scattering albedo based on chemical characterization of aerosols (Ohta and Okita, 1990; Ohta *et al.*, 1990). On the other hand, in evaluation of the indirect effect, we also need to determine the global distribution of aerosol chemical species, in particular, water soluble species such as sulfate and nitrate.

It is, thus, important to obtain the global horizontal and vertical distribution of the chemical species of aerosols. We have some data on the atmospheric concentrations of the aerosol chemical species on the surface in the world. But, in the troposphere above the surface, there are little data available. We, then, have measured the concentrations of chemical species of aerosols and gaseous constituents at the top of Mt.Lemmon, Tucson, Arizona, U.S.A. from September 1992. In this paper we present the measurement results and chemical characterization of the lower tropospheric aerosols in Arizona, and then, estimate the optical properties of aerosols such as the single scattering albedo and volume extinction coefficient based on the chemical characterization.

2. Experimental Procedures

We installed sampling systems in a room of the Cosmic Ray Laboratory, Department of Atmospheric Sciences, the University of Arizona. The laboratory is located at the top of Mt.Lemmon (at a height of 2791 m), 30 km north-east of Tucson, Arizona. Figure 1 shows the sampling system of aerosols (system A), and that of gaseous sulfur dioxide (SO_2), hydrogen chloride (HCl), nitric acid (HNO_3) or ammonia (NH_3) (system B).

We collected atmospheric aerosols simultaneously on a quartz fiber filter(PALLFLEX 2500 QAST-UP) and on a Teflon filter (Sumitomo Fluoropore AF07P) with 47 mm in diameter at a flow rate of $20l$ min^{-1} using cyclone separators whose 50% cut off diameter were 2 μ m in the sampling system A (two lines). By using the cyclone separators, we collected whole fine particles which were the aerosols less than 2 μ m in diameter, but a small portion of coarse particles larger than 2 μ m in diameter.

The atmospheric gaseous constituents were collected simultaneously on reagent soaked paper filters G1 and G2 at a flow rate of $7l$ min^{-1} in the sampling system B (three lines). Removing atmospheric aerosols by using Teflon filter AT, we collected atmospheric SO_2 and HCl on sodium carbonate soaked paper, and HNO_3 and NH_3 on sodium chloride and oxalic acid soaked papers, respectively.

We sampled atmospheric aerosols and gaseous constituents for 30 days every month from September 1992 through September 1994. Before and after the sampling, the Teflon filters used in the sampling system A were placed for 24 hours in a dry box at a relative humidity

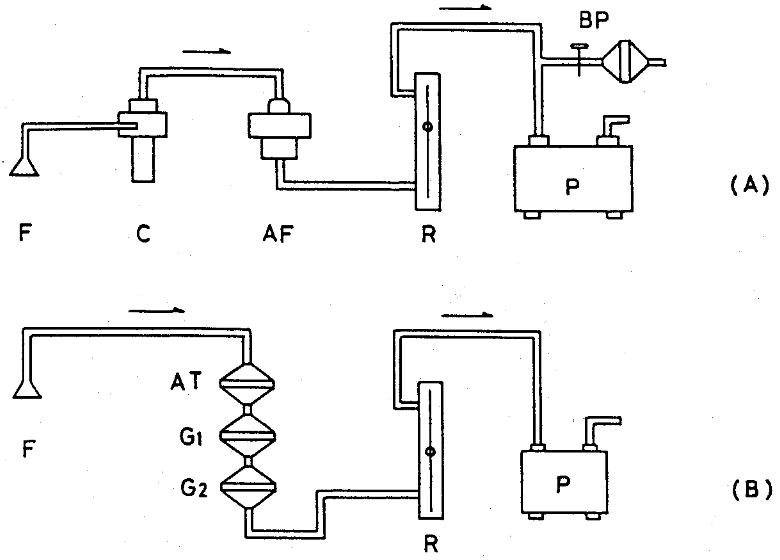

Figure 1. Sampling system of atmospheric aerosol (A) and gaseous constituents (B). F: Funnel, C: Cyclone separator, AF: Aerosol collection filter, R:Rota meter, P: Pump, BP: Bypass, AT:Teflon filter, G1,G2: Gas sampling filter.

less than 30%. We, then, determined the total particulate mass (TPM) by weighing them on an electric balance.

Collected aerosol samples on the Teflon filters were extracted ultrasonically with distilled-deionized water. The concentrations of sulfate (SO_4^{2-}), nitrate (NO_3^-) and chloride (Cl^-) in the extracted solution were determined by using an ion chromatograph (Yokogawa Electric Works Inc., IC-100, column SAM3-125). The concentrations of sodium (Na^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}) were determined with an atomic absorption spectrometer (Hitachi Inc. 170-30). Ammonium (NH_4^+) concentration was measured colorimetrically by using indophenol method (Weatherburn, 1967). The amount of silicon (Si) on the sampled Teflon filter was determined with particle induced X-ray emission (PIXE) method (Kasahara et al., 1993). Carbon contents in the quartz fiber filters collected in the sampling system A were determined by a combustion technique consisting of sample combustion at 850 ℃ in a NC- analyzer and analysis by a gas chromatograph equipped with a nickel catalyst methanizer and a flame ionization detector (Ohta and Okita, 1984). We cut off several pieces of disc-shaped samples with diameter of 1 cm from the quartz fiber filter. One half of the samples served to determine total carbon content(TC). Another half were heated in an electric furnace at 300 ℃ in air for 30 min to remove organic carbon (OC), and then elemental carbon content (EC) was measured by the combustion technique. The difference of the total carbon and elemental carbon gave the amount of organic carbon.

The absorbed SO_2 on the reagent-soaked paper filter was extracted with distilled-deionized water and oxidized to sulfate by dropping H_2O_2. The sulfate concentration was determined with the ion chromatograph. The absorbed HCl, HNO_3 and NH_3 on the reagent soaked paper filters were also extracted with distilled-deionized water. The concentration of HCl in the extracted solution was determined by the ion chromatograph. The concentrations of HNO_3 and NH_3 were measured colorimetrically by using the hydrazine reduction GR reagent method

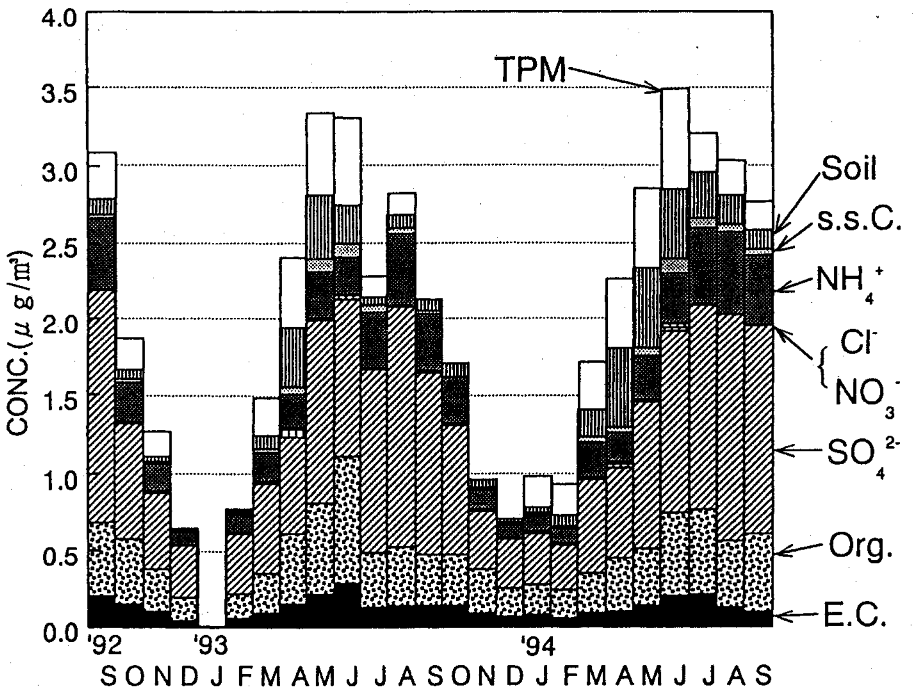

Figure 2. Mass balance of aerosols less than 2 μ m in diameter at Mt.Lemmon, Arizona.

(Ohta et al., 1970) and the indophenol method, respectively.

3. Results of measurements

Figure 2 shows seasonal variation of mass balance of aerosols at the top of Mt.Lemmon. The mark "E.C." denotes amount of elemental carbon, and "Org." denotes amount of organics obtained by multiplying amount of OC by 1.20. The mark "s.s.C." is amount of sea salt cation calculated due to that of Na^+. The amount of soil particles was calculated due to the mass fraction of Si as 31.7% in natural Arizona soil (Air cleaner test dust, Fine, produced by General Motors Corp., Flint, Michigan). Monthly mean concentration of total particulate mass (TPM) ranged from 0.64 to 3.49 μ gm^{-3}. It increased in spring and summer, and decreased late in fall and in winter. According to the radio sonde measurement of temperature profile at Tucson, maximum mixing depth (MMD), which was the height of mixing layer, was generally below 2.8 km altitude of Mt.Lemmon until warm weather arrived in March and surface layer temperature began to surpass 30℃ (Ramsey-Bell, 1987). Thus, the laboratory at the top of Mt.Lemmon stood out above the mixing layer in winter, whereas it was located in the mixing layer in spring and summer. Since in the mixing layer pollutants emitted at the surface are transported upward due to convection, the aerosol concentration at the top of Mt.Lemmon increased in spring and summer.

Monthly mean concentrations of each species of aerosols ranged as follows: elemental carbon (0.07-0.21 μ gCm^{-3}), organics (0.15-0.81 μ gm^{-3}), SO_4^{2-} (0.29-1.56 μ gm^{-3}), Cl^- (0.00-0.03 μ gm^{-3}), NO_3^- (0.00-0.02 μ gm^{-3}), NH_4^+ (0.10-0.54 μ gm^{-3}), s.s.C. (0.005-0.10 μ gm^{-3}) and

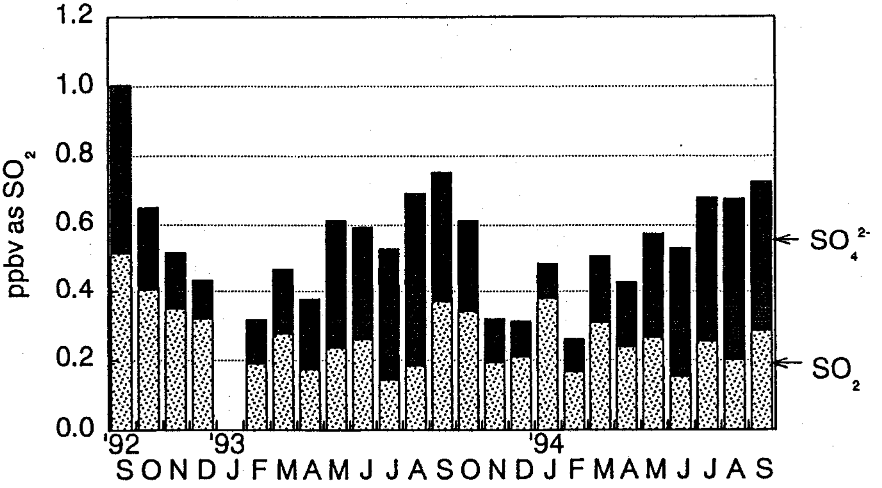

Figure 3. Monthly mean concentration of total sulfur oxides (SO_2 and fine particulate sufate (SO_4^{2-})) as SO_2 at Mt.Lemmon, Arizona.

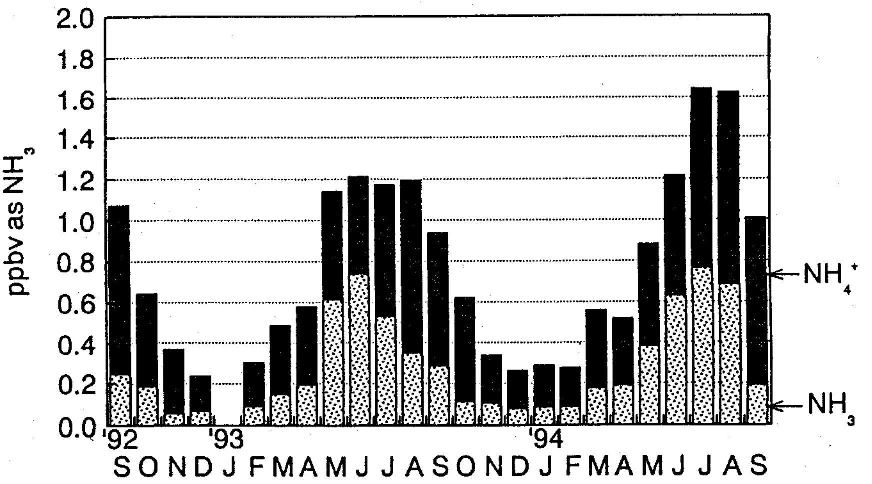

Figure 4. Monthly mean concentration of total ammonia (gaseous ammonia (NH_3) and fine particulate ammonium (NH_4^+)) as NH_3 at Mt.Lemmon, Arizona.

soil (0.02-0.47 μgm^{-3}).

Most of sulfate, ammonium, nitrate and chloride in the fine particles are formed due to conversion from gaseous SO_2, NH_3, HNO_3 and HCl, respectively. We, then, measured concentrations of the gaseous precursors in the atmosphere as well as those of particulate chemical species. Figures 3-6 show monthly mean concentrations of total sulfur oxides (SO_2 gas and particulate sulfate) as SO_2, total ammonia (ammonia gas and particulate ammonium) as NH_3, total nitrate (nitric acid gas and particulate nitrate) as HNO_3 and total chloride (hydrogen chloride gas and particulate chloride) as HCl, respectively. In the figures, mole concentrations of the particulate chemical species were converted to equivalent mole concentrations of

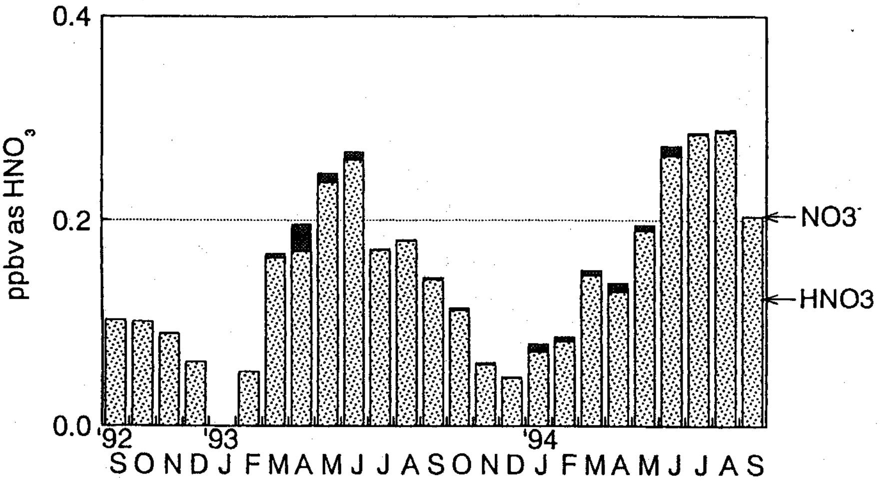

Figure 5. Monthly mean concentration of total nitrate (gaseous nitric acid (HNO_3) and fine particlate nitrate (NO_3^-)) as HNO_3 at Mt.Lemmon, Arizona.

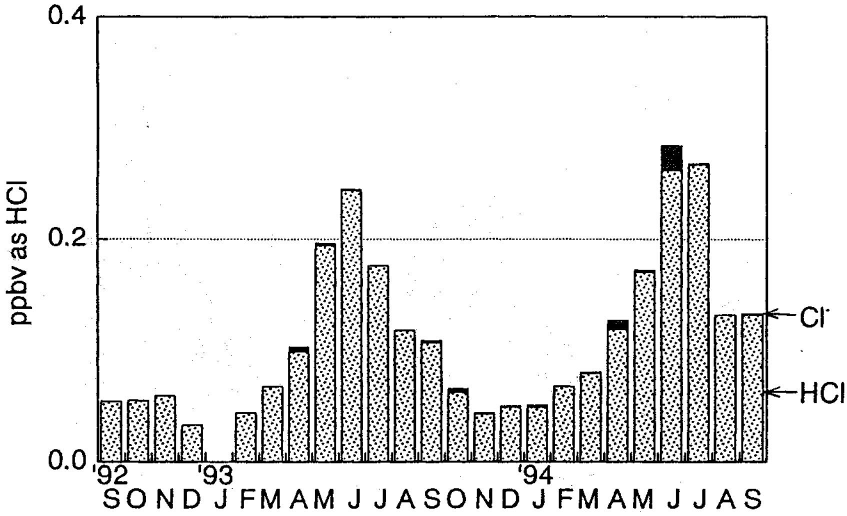

Figure 6. Monthly mean concentration of total chloride (gaseous hydrogen chloride (HCl) and fine particulate chloride (Cl^-)) as HCl at Mt.Lemmon, Arizona.

respective gaseous species, and then expressed in ppbv.

Monthly mean concentration of total sulfur oxides ranged from 0.3 to 1.0 ppbv as SO_2 shown in Figure 3. It had a tendency to increase in summer and decrease in winter, whereas concentration of SO_2 did not have the seasonal variation. It means that the conversion from SO_2 to sulfate occurs in summer in lower troposphere in Arizona.

Monthly mean concentration of total ammonia ranged from 0.2 to 1.6 ppbv as NH_3 shown in Figure 4. Both concentrations of total ammonia and gaseous ammonia increased drastically in summer. In the atmosphere, particulate ammonium is formed through the reaction of gaseous ammonia with sulfuric acid particles, and with gaseous nitric acid and hydrogen chloride as,

$$2NH_3(g) + H_2SO_4(p) \rightarrow (NH_4)_2SO_4(p) \tag{1}$$

$$NH_3(g) + HNO_3(g) \rightarrow NH_4NO_3(p) \tag{2}$$

$$NH_3(g) + HCl(g) \rightarrow \quad NH_4Cl(p) \tag{3}$$

where (g) and (p) denote gaseous and particulate species, respectively. Since the gaseous ammonia was emitted from the surface of Mt.Lemmon in hot season due to activity of soil organisms, atmospheric oncentration of total ammonia increased in summer.

Monthly mean concentrations of total nitrate and total chloride are shown in Figures 5 and 6, respectively. Both concentrations ranged from 0.05 to 0.3 ppbv as HNO_3 or HCl, which increased from May to July.

The gaseous nitric acid is formed through the photochemical reaction in the atmosphere, and the particulate nitrate is formed through the reaction of the nitric acid with ammonia shown in Eq.(2), or the reaction with sodium chloride particles as

$$HNO_3(g) + NaCl(p) \rightarrow NaNO_3(p) + HCl(g). \tag{4}$$

In the troposphere the photochemical reaction is predominant in spring and summer. Thus, the concentration of total nitrate increased in the season.

Since particulate NH_4NO_3 is easy to be decomposed at high temperature in summer and that $NaNO_3$ belongs to the coarse particles larger than 2 μ m in diameter, there were little particulate nitrates collected on the filters in the sampling system A.

In Figure 6, there were little particulate chloride. Particulate chloride derives from NaCl or NH_4Cl particles. But, since NaCl belongs to the coarse particles and that NH_4Cl is easy to be decomposed in the hot season, they were not collected on the filter similar to the particulate nitrate.

4. Estimation of Optical Properties of Fine Particles at Mt.Lemmon, Arizona.

On the basis of the results in Figure 2, we assumed that the atmospheric aerosols at Mt.Lemmon comprised seven species of particles such as elemental carbon (EC), organics, $(NH_4)_2SO_4$, NH_4NO_3, sea-salt, soil particles and mixed nuclei. The mixed nuclei has an inner core of EC and outer shell of sulfate.

EC, organics, $(NH_4)_2SO_4$, NH_4NO_3 and the mixed nuclei belong to the fine particles less than 2 μ m in diameter, and soil and sea-salt particles belong to the coarse particles larger than 2 μ m in diameter. Since we sampled aerosols by the system equipped with the cyclone separator whose 50% cut off diameter was 2 μ m, we measured the whole fine particles, but only small portion of the coarse particles. We, then, estimate the optical properties of only fine particles.

For the fine particles we adopted the particle size distribution of log normal;

$$dN/dlnD = N_0/(\sqrt{2\,\pi}\,ln\,\sigma_G)\cdot exp[-(lnD - lnD_{GN})^2/(2ln^2\,\sigma_G)] \tag{5}$$

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, the standard deviation and the range of particle diameter were assumed to be 0.11 μm, 2.10 and 0.02-4.0 μm, respectively.

We supposed two kinds of mixing conditions for the atmospheric fine particles. External mixture consisted of four species of homogeneous particles such as EC, organics, $(NH_4)_2SO_4$ and NH_4NO_3. Whereas internal half mixture contained mixed nuclei in addition to above four species of homogeneous particles. The mixed nuclei were assumed to be formed with half amount of EC and sulfate in the atmosphere.

By using the complex refractive index of EC, organics, $(NH_4)_2SO_4$ and NH_4NO_3, volume scattering coefficient and volume extinction coefficient for each species of particles were calculated by Mie theory. Those for the mixed nuclei we calculated following Toon and Ackerman (1981). The total volume extinction coefficient σ_{ext} of the atmospheric fine particles was obtained as the sum of the volume extinction coefficients of each species particles σ_{ei},

$$\sigma_{ext} = \sum_i \sigma_{ei}, \tag{6}$$

and the total volume scattering coefficient σ_{sca} of the fine particles was similarly obtained as

$$\sigma_{sca} = \sum_i \sigma_{si}, \tag{7}$$

where σ_{ei} and σ_{si} were the volume extinction and scattering coefficients for i-th species particles, respectively.

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

$$\omega = \sigma_{sca} / \sigma_{ext}. \tag{8}$$

In this study we chose values of the complex refractive index and the density less than 30 % relative humidity (in dry cinditions) for each chemical species as shown in Table 1. We assumed that atmospheric relative humidity was 70% in all season, because we have no data of the mean relative humidity at the top of Mt.Lemmon. According to the experimental studies by Tang *et al.*(1977) and Tang (1980), we calculated the growth of hygroscopic aerosols due to an increase of relative humidity.

Table 1. The density in dry conditions and the complex refractive index of particles of each chemical species at 0.5 μm wavelength.

Particle	Density	Complex refractive index
Elemental C	1.0	1.75-0.60i
Organics	1.4	1.55
$(NH_4)_2SO_4$	1.76	1.55
NH_4NO_3	1.66	1.55

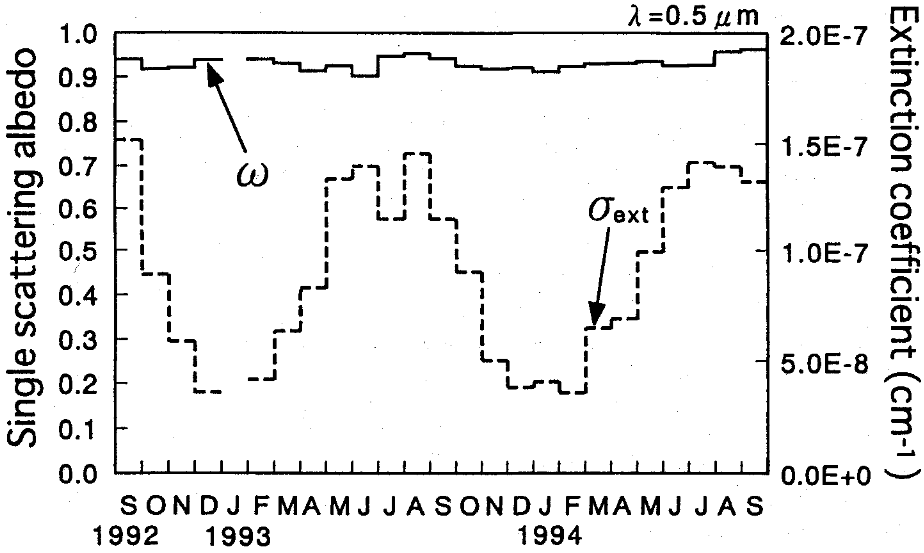

Figure 7. Monthly mean single scattering albedo ω and the volume extinction coefficient σ_{ext} of the fine particles at 0.5 μ m wavelength for the external mixture at the top of Mt.Lemmon.

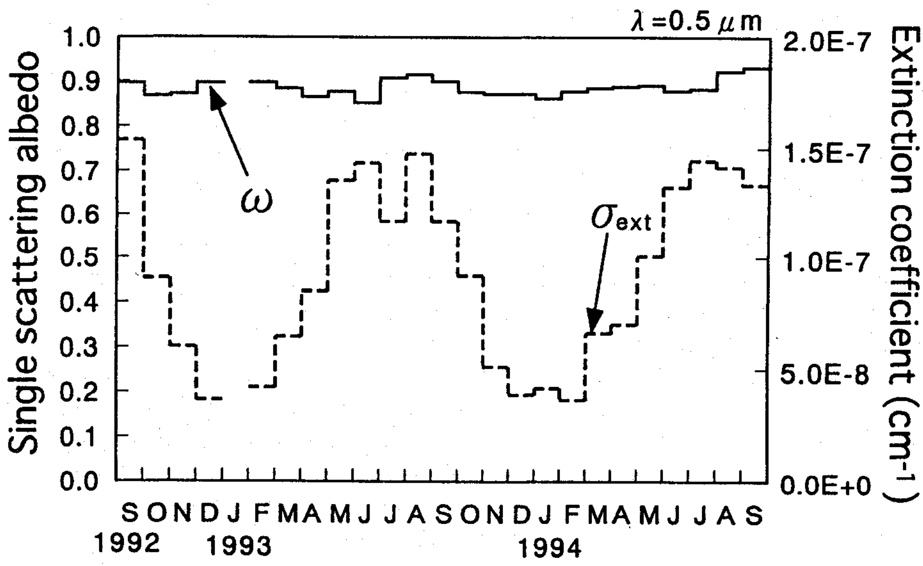

Figure 8. Monthly mean single scattering albedo ω and the volume extinction coefficient σ_{ext} of the fine particles at 0.5 μ m wavelength for the internal half mixture at the top of Mt.Lemmon.

Figures 7 and 8 show the monthly mean single scattering albedo and the volume extinction coefficient of the fine particles at 0.5 μ m wavelength for the external and the internal half mixtures, respectively.

For the external mixture the single scattering albedo and the volume extinction coefficient ranged from 0.90 to 0.96 and from 3.6×10^{-8} to $1.52 \times 10^{-7}\mathrm{cm}^{-1}$, respectively. For the internal half mixture the single scattering albedo decreasd to 0.85-0.93, whereas the volume extinction coefficient increased slightly to $3.7 \times 10^{-8} - 1.55 \times 10^{-7}\mathrm{cm}^{-1}$. There were, thus, differences of 0.03 to 0.05 in the single scattering albedo between the external mixture and the internal half mixture. They derive from the existence of the mixed nuclei.

The albedo effect of atmospheric aerosols on climate depends on the absorptivity of the aerosols (the single scattering albedo ω) and the albedo of the underlying earth surface A_S. According to Ackerman (1988), an increase of weakly absorbing aerosols as $\omega = 0.99$ at 0.5 μ m wavelength always increases the planetary albedo (the reflectivity in the solar wavelenghs at the top of the atmosphere), independent of underlying surface albedo, due to increase of back scatter of solar radiation to space, which causes the global cooling. On the other hand, an increase of strongly absorbing aerosols as $\omega = 0.7$ reduces the planetary albedo for bright earth surface as snow ($A_S = 0.6\sim0.85$) due to aerosol absorption of solar radiation, though it also increases one for dark surface as sea ($A_S = 0.05$). The increase of the strongly absorbing aerosols, thus, causes the heating on the bright surface, but the cooling on the dark surface. Then, for evaluation of the albedo effect of aerosols, we have to determine accurately the absorptivity of the aerosols on the various surface of the earth. In our method of estimation of the optical properties, then, it is important hereafter to determine the concentration of the mixed nuclei in the atmosphere.

5. Summary and Conclusion

We measured atmospheric concentrations of mass and chemical species of aerosols, and gaseous SO_2, NH_3, HNO_3 and HCl at the top of Mt.Lemmon (at a height of 2791m) near Tucson, Arizona from September 1992 to September 1994. Monthly mean concentration of the aerosols ranged from 0.64 to 3.49 $\mu\,\mathrm{gm}^{-3}$, which increased in spring and summer, and decreased late in fall and in winter. The aerosols consisted of elemental carbon, organics, sulfate, nitrate, chloride, ammonium, sea-salt cation and soil components.

Monthly mean concentration of total sulfur oxides (SO_2 gas and fine particulate sulfate) ranged from 0.3 to 1.0 ppbv as SO_2, which had a tendency to increase in summer and decrease in winter. The conversion from SO_2 to sulfate occured predominantly in summer. Monthly mean concentration of total ammonia ranged from 0.2 to 1.6 ppbv as NH_3. It increasd drastically in summer due to an increase of the emission from the surface of Mt.Lemmon by activity of soil organisms. Monthly mean concentrations of total nitrate and total chloride ranged from 0.05 to 0.3 ppbv as HNO_3 and HCl, respectively. Both concentrations increased from May to July due to the photochemical production of gaseous HNO_3 in the troposphere.

We calculated monthly mean single scattering albedo and extinction coefficient of the aerosols at the top of Mt.Lemmon based on the chemical characterization. We assumed two kinds of mixing conditions for the atmospheric aerosols. External mixture consisted of four species of homogeneous particles such as elemental carbon, organics, $(NH_4)_2SO_4$ and NH_4NO_3, whereas internal half mixture contained mixed nuclei in addition to above four species of particles. The mixed nuclei were assumed to be formed with half amount of elemental carbon and sulfate in the atmosphere.

The calculated single scattering albedos were 0.90-0.96 and 0.85-0.93, respectively, in the external and the internal half mixtures. The differences of 0.03 to 0.05 in the single scattering albedo derived from the existence of the mixed nuclei. Since the albedo effect of atmospheric aerosols on climate depends strongly on the value of the single scattering albedo of the aerosols as well as the albedo of the underlying earth surface, it is important hereafter to determine concentration of the mixed nuclei in the atmosphere.

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