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Sea-salt particles in the upper tropical troposphere

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ABSTRACT

Aircraft observation of aerosol particles was carried out at an altitude of 11.2 km over the tropical Pacific Ocean. Individual particles were collected on electron microscopic grids using an impactor, together with measurement of aerosol number concentrations using an optical particle counter. High concentrations of aerosols were observed intermittently with space, synchronized with the presence of deep convective clouds. In this region, sea-salt particles were collected in large number fractions of submicron particles; that is, the fractions were evaluated to be 0.42 for particles of 0.05–0.1 μm radius and 0.64 for particles of 0.1–1 μm radius. Elemental composition of the particles was examined with an energy-dispersive X-ray analyzer (EDX). The EDX analysis showed that the weight ratios of Cl/Na in sea-salt particles tended to decrease with increasing excess sulfur. Also, the Cl deficiency (loss) was large in smaller sea-salt particles. Moreover, the present research also suggests that sea-salt particles play an important role in the heterogeneous formation of sulfuric acid particles in the tropical free troposphere.

1. Introduction

Knowledge on features and behavior of aerosol particles in the middle and upper troposphere is important in assessing the effect of aerosol particles on global climate through cloud formation. Aircraft measurements in the middle and upper troposphere have been carried out (Blifford and Ringer, 1969; Dinger et al., 1970; Cadle et al., 1970; Gillette and Blifford, 1971; Huebert and Lazrus, 1980; Patterson et al., 1980; Iwasaka et al., 1988; Lechner et al., 1989; Yamato and Ono, 1989; Ikegami et al., 1993). Blifford and Ringer (1969) measured number-size distributions of aerosols at altitudes to 10 km over the United States of America and suggested the importance of cloud modification processes in the ageing of tropospheric aerosols. Patterson et al. (1980) found that marine aerosols were present in the middle troposphere over the equatorial Pacific, probably transported through convective clouds. Moreover, Huebert and Lazrus (1980) stated that high chloride concentrations were measured in the free troposphere over the ITCZ (intertropical convergence zone). By electron microscopy, Ikegami et al. (1993) found high proportions of submicrometer sea-salt particles in the middle troposphere over the tropical Pacific associated with the vertical transport through clouds. Also, presence of mineral particles was observed in the middle (Patterson et al., 1980; Iwasaka et al., 1988; Ikegami et al., 1993) and upper troposphere (Gillette and Blifford, 1971).

Variation of aerosol amounts in the upper troposphere has been measured by lidar (Takeda et al., 1979; Parameswaran et al., 1991) and balloon soundings (Hofmann et al., 1975). Although chemical composition of upper tropospheric particles has been measured by Cadle et al. (1970) and Gillette and Blifford (1971), information on physicochemical properties of aerosol particles in the upper troposphere is meager at the present.

During the INSTAC (International Strato/Tropospheric Air Chemistry) flight campaign in February–March 1990, observations of aerosol particles were carried out at an altitude of approximately 12 km from 65°N to 65°S. Sea-salt

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particles were found to be dominant in the upper tropical troposphere. The aim of this paper is to study the composition of individual sea-salt particles and to show the spatial variation of aerosols associated with the presence of deep convective clouds.

2. Methods

Aircraft observation was carried out on board aircraft (Gulfstream-2) at an altitude of approximately 12 km from 65°N to 65°S during the INSTAC-2 campaign (Fig. 1). The results obtained over the tropical Pacific Ocean are reported in this paper. The instruments for measuring and collecting aerosol particles were mounted inboard the aircraft. The sample air was introduced through an air intake projecting into the airstream from the aircraft fuselage. The sampling at the inlet tip was carried out in the isokinetic situation. The diameter of the sharp-edged inlet is 4 mm. The inner diameter of the tube increases from 4 mm to 7.75 mm at the distance of 1 cm–1.5 cm from the inlet tip. The sample air was then introduced through the tube (inner diameter of 7.75 mm) into

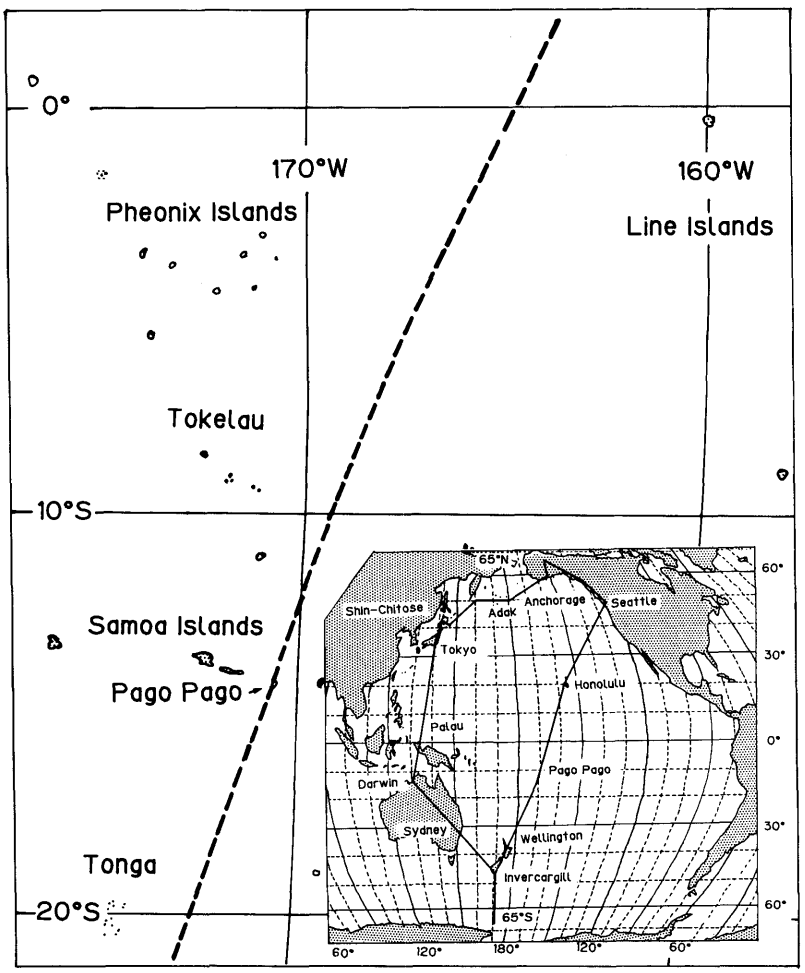

Fig. 1. Routes of the INSTAC-II flight. Measurements were made at an altitude of approximately 12 km. The results shown in this paper were obtained over the tropical Pacific Ocean.

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a manifold (60 mm in diameter and 290 mm in length) in the cabin of aircraft for measuring aerosol size distribution and collecting aerosol particles. There is a tube bend between the sampling inlet and manifold. The diffusion losses for particles of $\geqslant 0.1$ μm can be neglected through all the tube. However, inertial losses by the bend used in our observation were evaluated to be 1% for particles of 0.15 μm, 21% for those of 1 μm and 59% for those of 2 μm radius (Pui et al., 1987).

Collections of individual aerosol particles were made using an automatic two-stage impactor (one with jet diameter of 1.0 mm and another of 0.5 mm) made by the Meteorological Research Institute. Each of the impactor is composed of two jets. The low-pressure impactor of 0.5-mm diameter jet has 50% cutoff radius of 0.05 μm. Whereas, the impactor of 1.0-mm diameter jet has 50% cutoff radius of 0.15 μm. The collecting surface used is a nitrocellulose (collodion) film covered with carbon. The particles collected were examined with an electron microscope (Hitachi, H-600 and H-6010) in order to obtain the size and morphology. Also, the electron beam was irradiated at the center of a particle at an accelerating voltage of 50 kV. An X-ray spectrum was obtained by using an energy-dispersive X-ray (EDX) analyzer (Kevex, delta 5) through a Kevex UTW (ultra thin window) detector. The counting time was 50 s. Since the collecting surface used in the present study is the thin film mentioned above, elements of carbon (C), nitrogen (N) and oxygen (O) in the particles could not be determined exactly. However, for example, the detection of carbon in soot particles with radii greater than approximately 0.2 μm is possible on the basis of the close comparison of EDX spectrum between the particle and collecting film (e.g.,, Okada et al., 1992). Quantitative analysis of elemental composition in individual particle was carried out by using thin foil method (Cliff and Lorimer, 1975). The method was tested for artificial NaCl particles of 0.4–1 μm radius. The weight ratios of Cl/Na showed no size dependence although the ratios were scattered rather in wide range of ± 10%. The detector window thickness of 6 μm was adopted. The quantitative analysis was carried out using Kevex Quantex^{TM} software for energy-dispersive microanalysis. The weight percent of elements detected was calculated on the basis of characteristic X-ray fitted with a Gausian distribution, not the peak intensity. Note that the ratios of Cl/Na less than 1.5 was not obtained in laboratory generated sea-salt particles of 0.2–0.9 μm radius.

Number concentrations of aerosols with radii between 0.15 and 5 μm were measured with an optical particle counter (Dansangyo Co. Ltd., Model PM-730-NS15P) which has a 15-channel discriminator. The counter was calibrated by using polystylene latex particles before the observation.

Latitude, longitude, altitude, airspeed and static temperature were measured by the aircraft navigation system.

3. Results

3.1. Features of spatial change in aerosol number concentration

Fig. 2a shows the spatial distributions of aerosol number concentrations along the northbound flight during the period from 01:32 GMT to 03:17 GMT on 3 March. The aerosol radius range was classified into 4 classes: $0.15 \leqslant r < 0.25$ μm, $0.25 \leqslant r < 0.5$ μm, $0.5 \leqslant r < 1$ μm and 1 μm $\leqslant r$, where r is particle radius. Peaks of the aerosol concentrations are observed intermittently over the region of 12°S–8°S and they are 1–2 orders of magnitude larger than those measured in other regions. Since much cumulonimbus clouds existed over the area of 12°S–8°S, the aircraft often entered the clouds. The high concentrations were observed during the penetration of the upper part of cumulonimbus clouds. Although the ambient air temperature was -52°C, the temperature of the sample air in the optical counter was 27°C Both cloud-interstitial particles and cloud-droplet residues could be measured in clouds by the optical particle counter. Since we did not use the air inlet which can separate these particles from ambient air, the size distributions of cloud-active and -inactive particles in clouds could not be obtained separately from our observation.

Average number-size distributions of particles are shown in Fig. 2b. The distribution A is obtained from the in-cloud data at 11.1°S, 9.8°S and 8.7°S where the concentrations showed peak values. Whereas, the distribution B is that from the cloud-free data at 11.6°S, 10.6°S and 9.1°S. As found clearly, average concentrations in distribution A are approximately 1–2 orders of magnitude larger than those in distribution B in the entire

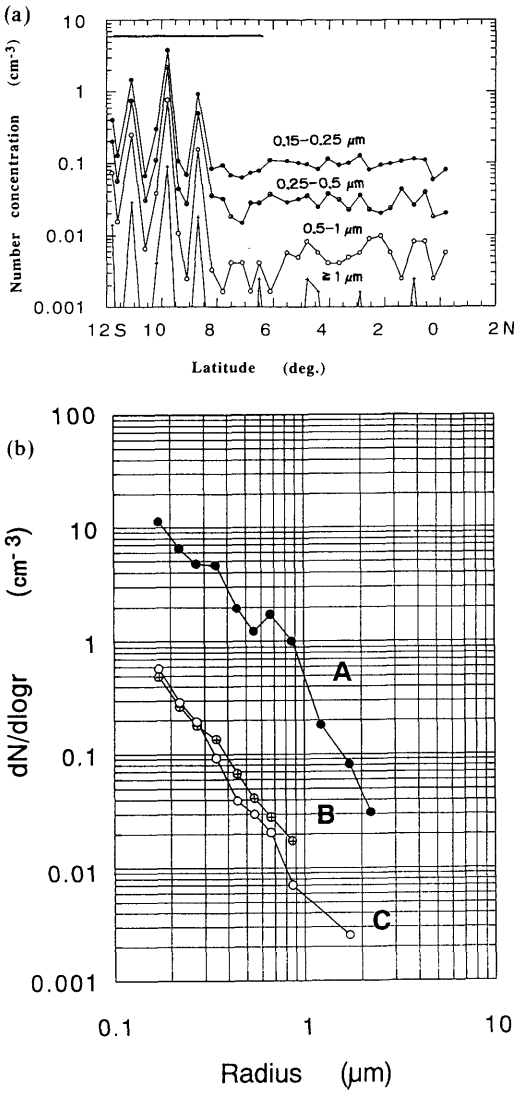

Fig. 2. Spatial change in number concentrations of aerosols from 12°S to 0°N measured at an altitude of 11.2 km during the period from 01:32 GMT to 03:17 GMT on 3 March 1990 (a). The observation was carried out in the northbound flight. The aerosol radius range was classified into 4 classes; $0.15 \leqslant r < 0.25$ μm, $0.25 \leqslant r < 0.5$ μm, $0.5 \leqslant r < 1$ μm and 1 μm $\leqslant r$, where r is particle radius. Area of the sampling of aerosol particles shown in Figs. 3 and 4 was indicated by solid line. Average number-size distributions of aerosols (b). Distribution A was obtained in the cloudy atmosphere (11.1°S, 9.8°S and 8.7°S). Distribution B was that from the cloud-free atmosphere (11.6°S, 10.6°S and 9.1°S). Distribution C was from the data from the cloud-free atmosphere of 6.1°S to 1.4°S.

size ranges. Distribution C is that obtained from the data in the cloud-free atmosphere of 6.1°S–1.4°S where number concentrations were relatively uniform. Distribution B exhibits nearly same features as distribution C. The mass concentration of aerosol particles of 0.15–1 μm radius was evaluated on the basis of number-size distribution by assuming that the density of particles is 2 g cm^{-3}. Also, particle loss in the sampling system was considered in the calculation. The assessed mass concentration for distribution A was 1.94 μg m^{-3} which is approximately 50 times greater than that for distributions B (0.045 μg m^{-3}) and C (0.030 μg m^{-3}).

3.2. Composition of particles

Fig. 3a shows electron micrograph of aerosol particles collected over the region from 11.8°S to 6.5°S (see Fig. 2a). Particle shown by an arrow has satellite droplet rings around a central part on the collecting surface, which is the characteristic morphology of sulfuric-acid containing particle (e.g.,, Frank and Lodge, 1967). However, most of the particles reveal morphological features other than sulfuric acid. In order to clarify the composition, an EDX analysis is applied to the particle marked by A. The X-ray spectrum is shown in Fig. 3b. The spectrum clearly shows the dominant peaks of Na and Cl, along with small peaks of Mg and S. Note that a peak of O would be partially originated from the collecting film. Therefore, this particle is identified to be sea salt. The X-ray spectrum from the particle B is also shown in Fig. 3c, indicating the presence of Na, Mg, S, Cl and Ca. This particle is also sea salt. However, the intensity of Cl relative to that of Na is low as compared with that for particle A.

On the basis of the results of the morphological examination and X-ray analysis, the number fractions of five types of particles in the radius ranges from 0.05 μm to 1 μm are summarized in Table 1. The results show high number proportions of sea salt; that is, sea-salt particles are detected in high number fractions of more than 0.64 in the radius range of 0.1–1 μm, whereas, sea-salt particles of 0.05–0.1 μm radius occupy about 40% of particles. Such a high proportion of sea salt was also found for particles of 0.08–0.3 μm in the middle tropical troposphere (Ikegami et al., 1993). The fraction of sulfuric-acid containing particles tends to increase with decreasing radius, ranging from about 0.1

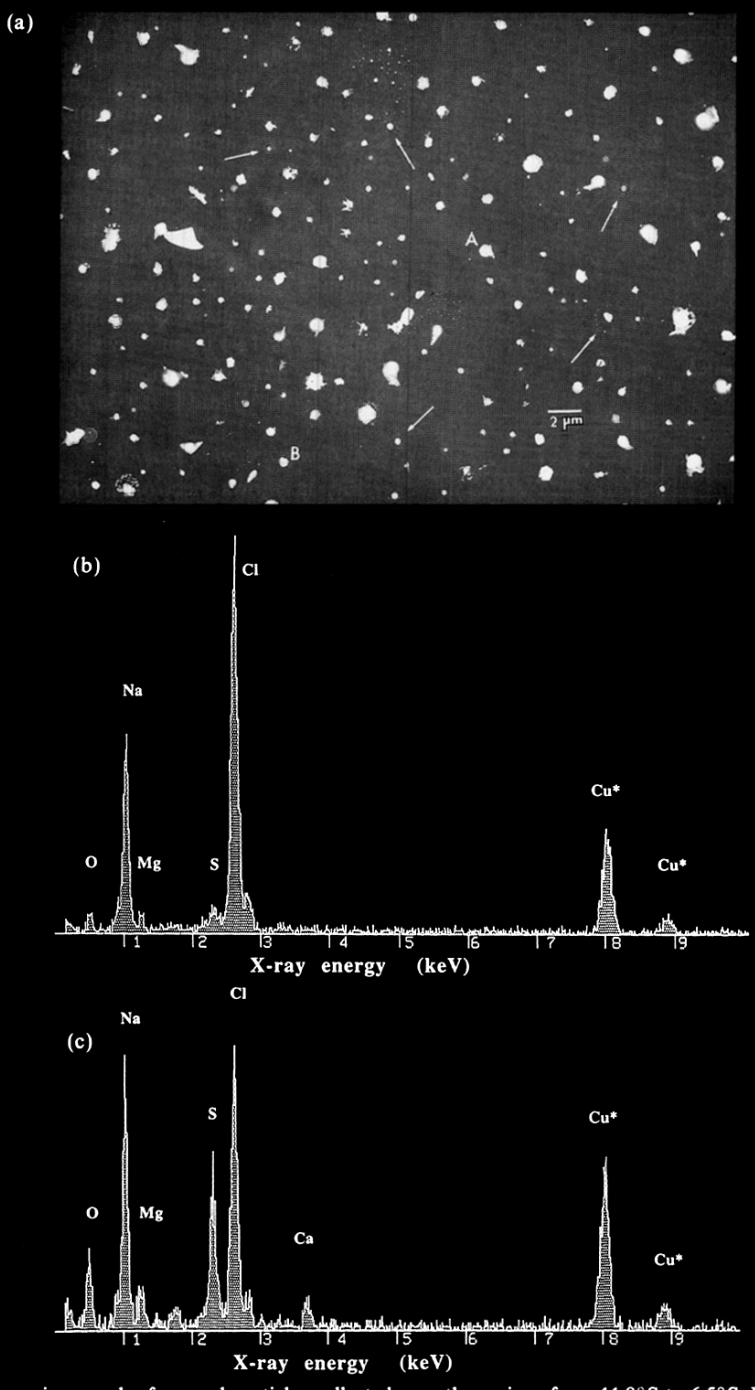

Fig. 3. Electron micrograph of aerosol particles collected over the regions from 11.8°S to 6.5°S. Particles shown by arrows contain sulfuric acid. The collection was carried out from 01:30 GMT to 02:20 GMT on 3 March 1990 (a). X-ray spectra from particles A (b) and B (c) are also indicated. Peaks by Cu* are caused by the copper electron microscopic grid. The ordinate in the spectra is the count of characteristic X-ray.

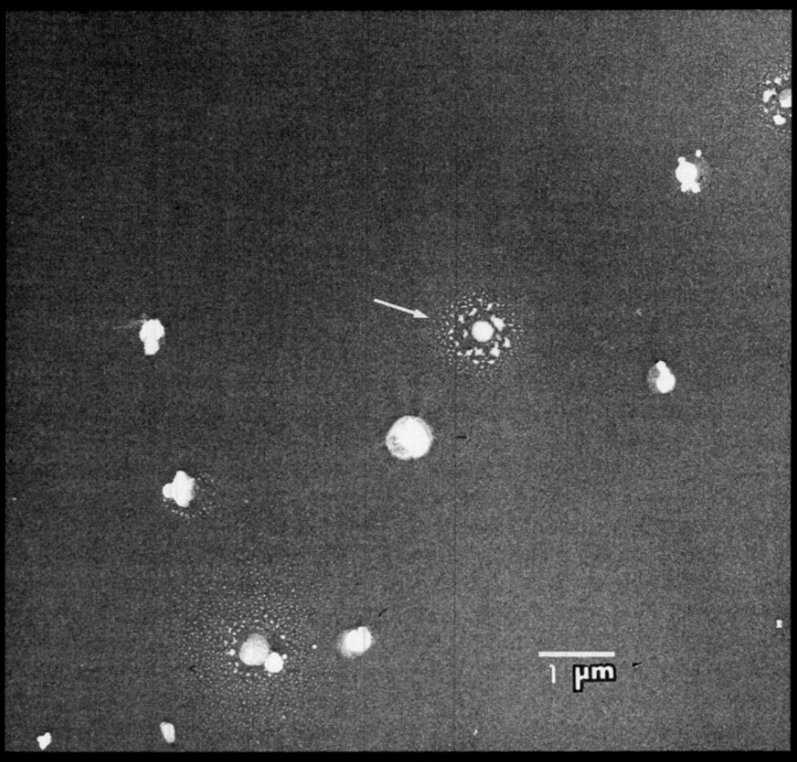

Fig. 4. Electron micrograph of aerosol particles obtained from the same sample as that in Fig. 3a (a). Sulfuric-acid containing particles indicated by an arrow, which show satellite droplet rings on the collecting surface, can be seen. X-ray spectrum from the central part (inclusion) of the sulfuric acid particle (indicated by an arrow in Fig. 4a) was obtained by subtracting the spectrum from the collecting surface (b).

to 0.4. On the other hand, fractions of mineral particles increase with increasing radius in the range of 0.1–1 μm, ranging from about 0.03 to 0.17. Particles showing aggregations of small electron-dense spherules, identified to be soot by morphological identification (e.g.,, Ogren and Charlson, 1983; Okada et al., 1992), constitute about 2% of the particles of 0.1–0.5 μm radius. Ammonium sulfate is present in a very low fraction. Note that ammonium particles were identified by morphological identification (Heard and Wiffen, 1969). Particles which could not be classified are termed "other particles" in Table 1.

Fig. 4a shows electron micrograph of particles taken from the same sample as that shown in Fig. 3a. A particle containing sulfuric acid is shown by an arrow. Fig. 4b shows the X-ray spectrum obtained from the particle (indicated by an arrow in Fig. 4a). The spectrum exhibits the presence of Na and S without Cl. X-ray analysis was applied to 25 sulfuric-acid containing particles in the radius range of 0.1–0.5 μm. It is found from the examination that 44% of the particles contained Na and S without Cl. Although sea-salt components were found, these particles were classified into sulfuric acid in Table 1.

Though not shown here, sulfuric-acid containing particles were dominant in the submicron size range over the cloud-free regions from 8°S to 7°N where aerosol spatial distributions were relatively uniform with low concentrations.

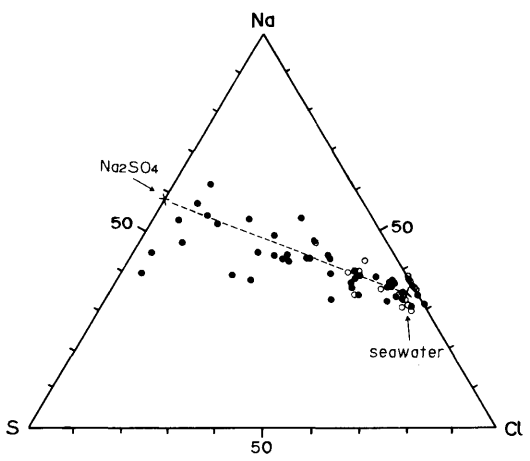

Fig. 5. Relative weight ratios of Na, Cl and S in individual sea-salt particles. The ratio for larger particle of ≥ 0.3 μm radius is indicated by a open circle. The ratio for smaller particle of <0.3 μm radius is shown by a closed circle.

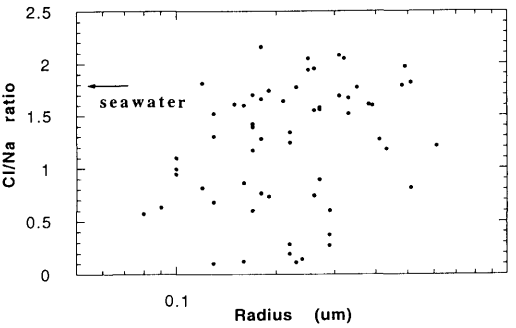

Fig. 6. Cl/Na weight ratio as a function of sea-salt particle radius. Seawater value of Cl/Na ratio is 1.8.

3.3. *Relative weight ratios of Na, Cl and S*

Fig. 5 illustrates relative weight ratios of Na, Cl and S in sea-salt particles. Particles with weight ratios of Na/(Na + Mg + S + Cl + K + Ca) more than 0.3 are identified to be sea salt. However, particle with satellite droplet rings on the collecting surface were not identified to be sea salt. The number of the particles used in Fig. 5 is 61. All the particles have radii less than 1 μm. The ratio for larger particle of ≥ 0.3 μm radius is indicated by a

Table 1. *Number of particles in the radius ranges from 0.05 μm to 1 μm; the number fraction for each radius range is shown in parenthesis*

Type of particles	Radius range 0.05–0.1 μm	0.1–0.5 μm	0.5–1 μm
sea-salt	18 (0.42)	188 (0.64)	8 (0.67)
sulfuric acid	18 (0.42)	58 (0.20)	1 (0.08)
mineral	0 (0)	10 (0.03)	2 (0.17)
soot	0 (0)	7 (0.02)	0 (0)
ammonium sulfate	1 (0.02)	0 (0)	0 (0)
others	6 (0.14)	31 (0.11)	1 (0.08)
total	43	294	12

open circle, whereas, the ratio for smaller particle of $<0.3\ \mu m$ radius is shown by a closed circle. As found from Fig. 5, the ratios are not identical to the seawater ratio marked by a cross and are distributed around a broken line between the seawater and Na_2SO_4 values. This indicates the increase in Cl deficiency with increasing excess sulfur. In order to assess the Cl deficiency as a function of radius more clearly, the relation between Cl/Na weight ratio and particle radius is shown in Fig. 6. Although the data are scattered in a wide range, Cl/Na ratios tend to decrease with decreasing radius. For small particles with radii less than 0.3 μm, 48 % of the sea-salt particles had Cl/Na ratios less than 1.

4. Discussion

High aerosol number-concentrations were observed during in-cloud flight and were 1–2 orders of magnitude larger than those in cloud-free atmosphere. This indicates that the high concentrations were realized in association with the presence of convective clouds. Moreover, much sea-salt particles were collected during the areas with large variabilities of aerosol concentrations. From the images obtained by the Geostationary Meteorological Satellite (GMS) at 03 GMT on 3 March, much convective activity occurred in the observation areas.

Although the shattering of droplets at sampling inlet will occur (e.g., Hudson and Frisbie, 1991), the process may not produce high concentrations of small sea-salt particles (residues) in our observation because we used a sharp-edged air inlet. Also, breakup of large cloud droplets in convective clouds will supply small sea-salt particles (residues). However, the present study can not assess the effect of this process on the measured concentration increase. One of other possible explanation is considered as follows. Wind velocities near the sea surface should be $>10\ m\ s^{-1}$ in the presence of severe convective clouds (e.g., Houze, 1977; Ishihara and Yanagisawa, 1982). Consequently, sea-salt particles are generated at a large rate and transported into clouds by upward air-motion. Because sea-salt particles act as efficient cloud condensation nuclei, they could form cloud droplets. During the in-cloud flight, cloud droplets as well as cloud-interstitial aerosol particles would be sampled in the air intake. It is suggested that high concentrations of aerosols measured at approximately 12 km altitude were realized mainly upon the evaporation of cloud droplets which contained sea salt.

In the marine atmosphere, non sea-salt sulfate particles were measured especially in the Aitken size range (Mészáros and Vissy, 1974). However, as indicated in the previous section, number fractions of sea salt in the radius ranges of 0.05–0.1 μm and 0.1–0.5 μm were 0.42 and 0.64, respectively. Cipriano et al. (1983) pointed out on the basis of laboratory experiments that Aitken particles could be formed from the seawater. Hence, there is a possibility that many Aitken sea-salt particles were generated from the sea surface below severe convective clouds. This may lead to the results that the concentrations of sea salt exceed those of non sea-salt sulfate particles in the radius range of $\leqslant 0.5\ \mu m$.

Modification of sea salt in the atmosphere has been studied by many researchers by using a bulk analysis (Marten et al., 1973; Okada et al., 1978; Hitchcock et al., 1980) and by a single particle analysis (Van de Vate et al., 1978; Miura et al., 1991; Mouri and Okada, 1993). In the coastal atmosphere where most of the researches were carried out, chlorine deficiency (loss) was detected mainly in submicron sea-salt particles. As shown in the previous section, most of submicron sea-salt particles had Cl/Na weight ratios less than seawater value of 1.8. The ratios lower than 1 were found especially in the radius range below 0.3 μm. From the relative weight ratios of Na, Cl and S (Fig. 5), the Cl weight ratios decreased with increasing the amount of excess S. Moreover, the ratios were located along a line which suggests the formation of Na_2SO_4. Thus, sea-salt particles should be modified mainly by chemical reactions with sulfuric acid. Mouri and Okada (1993) have shown that modified sea-salt particles with radii $r < 0.5\ \mu m$ were present in the surface atmosphere over the western Pacific Ocean. They found that the number fraction of modified sea-salt particles (Cl/Na < 1) was small with values from 0.02 to 0.18 in the radius range of 0.1–0.5 μm. Consequently, sea-salt particles collected in our aircraft observations would have been modified during vertical transport in convective clouds.

Recently, Koga et al. (1993) carried out measurements onboard a ship in the Pacific Ocean and

found high atmospheric dimethylsulfide (DMS) concentrations of approximately 95 pptv around at the region of 5° S and 160° W which is nearly the same oceanic area as our aircraft observation. They also found high concentrations of DMS in the surface seawater south of the equator along 160° W–180°. It is suggested that the main origin of sulfuric acid detected in our aircraft observation is likely to biogenic SO_2 formed from DMS oxidation (Lovelock et al., 1972).

Sulfuric-acid containing particles were also present externally with sea-salt in the sample. Since the sample used in this research were obtained in a wide region of 11.8° S–6.5° S as shown in Fig. 2a, particles in the cloudy and cloud-free atmosphere were collected in the same sample. In the cloud-free atmosphere between clouds, the aerosol concentrations were nearly equal to those in the cloud-free regions of 8° S to 7° N where sulfuric-acid containing particles were dominant in the submicron size range. Hence, there is a possibility that these sulfuric-acid particles collected were present in the cloud-free atmosphere.

However, as shown in the previous section, the X-ray analysis of sulfuric acid particle showed that the particles contained Na and S without Cl occupied 44% of the particles. This suggests the importance of the heterogeneous formation of sulfuric acid on sea-salt particles through clouds in the tropical free troposphere. Note that one cannot rule out a possibility that sulfuric acid particles with sea salt inclusion would be formed through the coalescence between cloud droplets with sea salt and those with sulfuric acid.

5. Conclusions

In the upper troposphere (11.2 km altitude) over the tropical Pacific Ocean, high number concentrations of aerosols were observed intermittently with space, sunchronized with the presence of deep convective clouds. Sea-salt particles were present in number fractions of 0.42 (0.05–0.1 μm radius) and 0.64 (0.1–1 μm radius) during the in-cloud flight.

The EDX analysis showed that the weight ratios of Cl/Na in individual submicron sea-salt particles tended to decrease with increasing excess sulfur. Also, the Cl deficiency (loss) was largest in the smaller sea-salt particles. Moreover, it is suggested that sea-salt particles play an important role in the heterogeneous formation of sulfuric acid particles in the tropical free troposphere.

Present research suggests the importance of deep convective clouds in assessing the changes in the spatial distribution and features of aerosol particles in the upper troposphere.

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REFERENCES

Blifford, I. H., Jr. and Ringer, L. D. 1969. The size and number distribution of aerosols in the continental troposphere. *J. Atmos. Sci.* **26**, 716–726.

Cadle, R. D., Lazrus, A. L., Pollock, W. H. and Shedlovsky, J. P. 1970. The chemical composition of aerosol particles in the tropical stratosphere. *Proc. Meeting on Tropical Meteorology*, American Meteorological Society, Honolulu.

Cipriano, R. J., Blanchard, D. C., Hogan, A. W. and Lala, G. G. 1983. On the production of Aitken nuclei from breaking waves and their role in the atmosphere. *J. Atmos. Sci.* **40**, 469–479.

Cliff, G. and Lorimer, G. W. 1975. The quantitative analysis of thin specimens. *J. Microscopy* **103**, 203–207.

Dinger, J. E., Howell, H. B. and Wojciechowski, T. A. 1970. On the source and composition of cloud nuclei in a subsident air mass over the North Atlantic. *J. Atmos. Sci.* **27**, 791–797.

Frank, E. R. and Lodge, J. P., Jr. 1967. Morphological identification of airborne particles with the electron microscopy. *J. Microscopie* **6**, 449–456.

Gillette, D. A. and Blifford, I. H., Jr. 1971. Composition of tropospheric aerosols as a function of altitude. *J. Atmos. Sci.* **28**, 1199–1210.

Heard, M. J. and Wiffen, R. P. 1969. Electron microscopy of natural aerosols and the identification of particulate ammonium sulfate. *Atmos. Environ.* **3**, 337–340.

Hitchcock, D. R., Spiller, L. L. and Wilson, W. E. 1980. Sulfuric acid aerosols and HCl release in coastal atmosphere: evidence of rapid formation of sulfuric acid particulates. *Atmos. Environ.* **14**, 165–182.

Hofmann, D. J., Rosen, J. M., Pepin, T. J. and Pinnick, R. G. 1975. Stratospheric aerosol measurements I: time variations at Northern Midlatitudes. *J. Atmos. Sci.* **32**, 1446–1456.

Houze, R. A., Jr. 1977. Structure and dynamics of a tropical squall-line system. *Mon. Wea. Rev.* **105**, 1540–1567.

Hudson, J. G. and Frisbie, P. R. 1991. Cloud condensation nuclei near marine stratus. *J. Geophys. Res.* **96**, 20795–20808.

Huebert, B. J. and Lazrus, A. L. 1980. Bulk composition of aerosols in the remote troposphere. *J. Geophys. Res.* **85**, 7337–7344.

Ikegami, M., Okada, K., Zaizen, Y. and Makino, Y. 1993. Aerosol particles in the middle troposphere over the northwestern Pacific. *J. Meteor. Soc. Japan* **71**, 517–528.

Ishihara, M. and Yanagisawa, Z. 1982. Structure of a tropical squall line observed in the western tropical Pacific during MONEX. *Pap. Meteor. Geophys.* **33**, 117–135.

Iwasaka, Y., Yamato, M., Imasu, R. and Ono, A. 1988. Transport of Asian dust (KOSA) particles; importance of weak KOSA events on the geochemical cycle of soil particles. *Tellus* **40B**, 494–503.

Koga, S., Tanaka, H. and Hayashi, M. 1993. Dimethylsulfide measured in the western Pacific and southern Indian Ocean. *J. Meteor. Soc. Japan* **71**, 183–194.

Lechner, I. S., Fisher, G. W., Larsen, H. R., Harvey, M. J. and Knobben, R. A. 1989. Aerosol size distributions in the southwest Pacific. *J. Geophys. Res.* **94**, 14893–14903.

Lovelock, J. E., Maggo, R. J. and Rusmussen, R. A. 1972. Atmospheric dimethyl sulphide and the natural sulphur cycle. *Nature* **237**, 452–453.

Martens, C. S., Wesolowski, J. J., Harriss, R. C. and Kaifer, R. 1973. Chlorine loss from Puerto Rican and San Francisco Bay area marine aerosols. *J. Geophys. Res.* **78**, 8778–8792.

Meszaros, A. and Vissy, K. 1974. Concentrations, size distribution and chemical nature of atmospheric aerosol particles in remote oceanic areas. *J. Aerosol Sci.* **5**, 101–109.

Miura, K., Kumakura, T. and Sekikawa, T. 1991. The effect of continental air mass on the modification of individual sea-salt particles collected over the coast and the open sea. *J. Meteor. Soc. Japan* **69**, 429–438.

Mouri, H. and Okada, K. 1993. Shattering and modification of sea-salt particles in the marine atmosphere. *Geophy. Res. Lett.* **20**, 49–52.

Okada, K., Ikegami, M., Uchino, O., Nikaidou, Y., Zaizen, Y., Tsutsumi, Y. and Makino, Y. 1992. Extremely high proportions of soot particles in the upper troposphere over Japan. *Geophys. Res. Lett.* **19**, 921–924.

Okada, K., Ishizaka, Y., Masuzawa, T. and Isono, K. 1978. Chlorine deficiency in coastal aerosols. *J. Meteor. Soc. Japan* **56**, 501–507.

Ogren, J. A. and Charlson, R. J. 1983. Elemental carbon in the atmosphere: cycle and lifetime. *Tellus* **35B**, 241–254.

Parameswaran, K., Rose, K. O. and Krishna Murthy, B. V. 1991. Comparison of aerosol extinction profiles from lidar and SAGE II data at a tropical station. *J. Geophys. Res.* **96**, 10861–10866.

Patterson, E. M., Kiang, C. S., Delany, A. C., Wartburg, A. F., Leslie, A. C. D. and Huebert, B. J. 1980. Global measurements of aerosols in remote continental and marine regions: concentrations, size distributions, and optical properties. *J. Geophys. Res.* **85**, 7361–7376.

Pui, D. Y. H., Romay-Novas, F. and Liu, B. Y. H. 1987. Experimental study of particle deposition in bends of circular cross section. *Aerosol Sci. Technol.* **7**, 301–315.

Takeda, T., Nagaya, K. and Iwasaka, Y. 1979. Lidar observations of aerosols in the upper troposphere. *J. Meteor. Soc. Japan* **57**, 362–366.

Van de Vate, J. F., Plomp, A., Jong, C. and Vrins, E. L. M. 1978. A battery-operated portable unit for electrostatic and impaction sampling of ambient aerosol for electron microscopy. *Staub-Reinhalt. Luft* **38**, 53.

Yamato, M. and Ono, A. 1989. Chemical and physical properties of stratospheric aerosol particles in the vicinity of tropopause folding. *J. Meteor. Soc. Japan* **67**, 147–165.