Number-size Distributions of Aerosol Particles in the Free Troposphere over the Northwestern Pacific Ocean—Influence of Asian Outflow and Tropical Air Transport

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Abstract

Number-size distribution of aerosols and its spatial variation, were observed in the free troposphere up to 11 km altitude over the northwestern Pacific Ocean, in the Pacific Atmospheric Chemistry Experiment (PACE)-7 campaign in February 2000. Characteristics of the size distributions were observed in the relation to the different air mass. The spatial difference in the features of size distributions was clearly divided by the location of subtropical front. Over the extratropical region, the mode radii were large (0.03–0.06 μm), and the concentrations of aerosols in the accumulation mode (0.15 < r < 0.5 μm) were high, suggesting a strong influence from anthropogenic particles from the Asian continent. On the other hand, the size distributions in the subtropical air mass had the mode radii of 0.01–0.03 μm, and showed low concentrations of accumulation mode particle. Moreover, high number concentrations of very fine particles (0.004–0.01 μm radius), were distributed at upper altitudes (> 8 km) over 15–31°N latitude. These aerosols were assumed to be mainly sulfuric acid particles transported from the tropics. Size distribution at the center of subtropical jet streams was slightly aged, suggesting influence from the continent. However, it is basically resemble to the size distributions measured in the upper troposphere over the subtropical region. On the basis of the trajectory analysis, these aerosols would have been transported from the tropical upper troposphere, to the middle latitudes by Hadley circulation, and then mixed into the subtropical jet streams.

1. Introduction

Atmospheric aerosols influence climate through direct and indirect radiative forcing (Charlson et al. 1992). Industrial areas in East Asia are one of the strongest sources of SO2 (Kato and Akimoto 1992), and the emission is expected to increase (International Energy Agency 2002). Influence of these particles is not limited over the source regions, and spread widely over the North Pacific Ocean (Andreae et al. 1988; Kaneyasu et al. 2000). Asian desert and loess areas are considerable sources of mineral particles. The mineral dust particles were frequently observed in Korea (e.g., Chun et al. 2001) and Japan (e.g., Iwasaka et al. 1983; Tanaka et al. 1989; Okada et al. 1990), and detected over the western Pacific Ocean (Ikegami et al. 1997), the middle Pacific Ocean (Uematsu et al. 1983; Okada et al. 1990) and North America (Isono et al. 1971; Husar et al. 2001). These results provide insights into the long-range transport of particles from the Asian continent.

However, the transport of anthropogenic aerosols from the Asian continent has not been studied in detail as compared to that of mineral
particles. Since the western rim of the northern Pacific Ocean is an entrance of particles and gases into the Pacific Ocean from the Asian continent, transportation of the materials over the area is important. In order to clarify the long-range transport of anthropogenic particles from the Asian continent, the vertical distributions of aerosols should be measured in the free troposphere, up to the tropopause, because long-range transport would depend on the strong westerly winds, centered in the upper troposphere.

As another issue, aerosols from natural sources are also important over the Pacific Ocean. Dominant constituents of submicron aerosol particles of 0.1–1 μm radius, in the middle troposphere over the western Pacific Ocean, were sulfate, sulfuric acid and sea salt (Ikegami et al. 1993 and 1997). Particles smaller than 0.1 μm radius are expected to be mainly composed of sulfuric acid and sulfate. Since these fine particles will have capabilities as cloud condensation nuclei (CCN), they play an important role in indirect radiative forcing. The new particles formed in the tropical upper troposphere (Clarke and Kapustine 2002), were considered to be transported to the higher latitudes by Hadley circulation, with the growth to larger particles. The processes supply cloud condensation nuclei, in remote marine boundary layer (Raes 1995). Zaizen et al. (1996) observed high number concentrations of fine sulfuric acid particles in the middle troposphere over subtropical subsidence regions. These processes cannot be detected with remote sensing, because the very fine particles are not optically active. In spite of the importance, the in situ observation in the upper altitude was very limited. Moreover, the subtropical jet streams, and subtropical front, possibly play an important role in the transportation of aerosols. However, they are not clarified up to now.

The aim of this paper is to obtain the spatial characteristics of number-size distribution of aerosols in the submicron size range down to 0.004 μm radius, over the northwestern Pacific Ocean and to assess the influence of Asian outflow and tropical air transport based on the aircraft observation conducted in February 2000, which was carried out as pre-ACE/Asia (Asian Pacific Regional Aerosol Characterization Experiment).

2. Observation

The Pacific Atmospheric Chemistry Experiment (PACE)-7 campaign was conducted at altitudes below 11 km, over the regions shown in Fig. 1, during the period of 11–21 February 2000. Vertical soundings below 11 km altitude, were conducted over the areas to the northwest of Naha, the northwest of Iwojima and the east of Saipan as shown by short lines in Fig. 1. In the soundings, level flights were conducted along the lines at four altitudes (2 km, 5 km, 8 km and 11 km). In this paper, the results obtained from the observations carried out over the areas south of 45°N are shown. Each flight leg was at an altitude of 5 km, 8 km and 11 km. Dates and altitudes of the flight missions were listed in Table 1.

Figure 2 shows a vertical cross section along longitude of 140°E at 00:00 GMT on 14 February 2000. Flight paths to the south of Nagoya were plotted by heavy dot lines with arrows. Wind direction over the whole regions, north of approximately 20°N, was west. Two cores of subtropical jet streams existed at 30°N (11 km altitude), and at 25°N (9 km altitude over Iwo-
Table 1. List of flight missions conducted south of 45°N during the PACE-7 campaign.

<table>
<thead>
<tr>
<th>No.</th>
<th>Flight mission</th>
<th>Date</th>
<th>Time (GMT)</th>
<th>Altitude (km)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Nagoya- Naha</td>
<td>13 Feb.</td>
<td>03:00-</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>local</td>
<td></td>
<td>05:20-</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Naha</td>
<td>13 Feb.</td>
<td>06:40-</td>
<td>&lt;11</td>
</tr>
<tr>
<td></td>
<td>local</td>
<td></td>
<td>09:50-</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Naha- Iwojima</td>
<td>14 Feb.</td>
<td>00:30-</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>local</td>
<td></td>
<td>02:30-</td>
<td></td>
</tr>
<tr>
<td>4</td>
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<td>14 Feb.</td>
<td>03:45-</td>
<td>&lt;11</td>
</tr>
<tr>
<td></td>
<td>local</td>
<td></td>
<td>06:55-</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>Iwojima- Nagoya</td>
<td>14 Feb.</td>
<td>07:55-</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>09:05-</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>Nagoya- Petero</td>
<td>17 Feb.</td>
<td>00:40-</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>Pavlovsk- Monbetu</td>
<td></td>
<td>04:15-</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>Petero</td>
<td>19–20 Feb.</td>
<td>23:40-</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td>Pavlovsk-</td>
<td></td>
<td>01:33-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Monbetu- Nagoya</td>
<td>19 Feb.</td>
<td>01:40-</td>
<td>5</td>
</tr>
<tr>
<td>8</td>
<td>Monbetu- Nagoya</td>
<td></td>
<td>03:35-</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>Nagoya- Saipan</td>
<td>19 Feb.</td>
<td>04:25-</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>local</td>
<td></td>
<td>08:25-</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>Saipan</td>
<td>20 Feb.</td>
<td>00:05-</td>
<td>&lt;11</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>03:25-</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>Saipan- Nagoya</td>
<td>21 Feb.</td>
<td>00:00-</td>
<td>11</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>03:55-</td>
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</tr>
</tbody>
</table>

jiima). Over Iwojima (approximately 25°N latitude), subtropical front existed at approximately 5 km altitude. The meteorological field is a typical one in winter over this region.

3. Instruments

An aircraft, Grumman Gulf Stream-II was used for the observation. Sample air was introduced to the instruments through an isokinetic intake (inner diameter 5.4 mm; outer diameter 5.8 mm) projecting into the air stream from the aircraft fuselage. The distance between the intake and aircraft fuselage is 200 mm. The outer and inner diameters of the intake increase linearly to 32 and 20 mm at the distance of 45 mm from the intake tip, respectively. The sample air was then introduced through the tube (inner diameter of 20 mm) into a manifold (60 mm in diameter and 290 mm in length) in the cabin of the aircraft. From the manifold, sample air was introduced into the instru-

ments. Volumetric air flow-rate through the intake was 250 l min⁻¹.

The deposition rate of aerosols by inertia to the walls of tubing, was estimated by a laboratory experiment. The deposition rate was obtained as a function of aerosol radius by comparing the size distributions before and after passing through the tubing using the same air velocity as the observation on board. The rate of loss by diffusion was estimated theoretically as the same way noted in Zaizen et al. (1996), because of difficulty of experimental estimation. The observed size distributions were corrected using these loss rates. In addition, the observed size distributions had been modified by relative humidity, because the sample air would be dried after the introduction to the manifold by the difference of temperature between the cabin and ambient air. Most of the measurements were carried out in relatively dry conditions (relative humidity was less than 70%). The modification of particle size caused by the temperature difference cannot be ignored when
relative humidity was higher than 70%. Such a high humidity was observed over Saipan, and below 2 km altitude over Naha and Iwojima.

Number-size distribution of aerosols, in the radius range of 0.05–5 μm was measured using an optical particle counter (OPC), (PMS model PCASP-X), which can count particles in 31 bins. The measuring interval was 10 sec. In this paper, the size range of 0.15–0.5 μm radius is called accumulation mode. Particle size measured as the OPC was calibrated using standard polystyrene latex (PSL) particles of four sizes (0.2, 0.3, 0.5 and 1 μm radius). The refractive index (real part) of PSL is approximately 1.6, whereas most of the values of atmospheric aerosols are 1.4–1.7, which decrease with increasing relative humidity. The difference causes measurement error of particle size. According to the theoretical calculations by Kanagawa (1970), magnitude of the error is assumed to be 0–30% for particles with reflective index of 1.33. Light absorption by elemental carbon, and mineral particles, also potentially cause incorrect measurement of particle size. However, the influence to the observed size distributions is considered to be not large, because the number fractions of these particles were small.

For the measurements in the smaller size ranges (0.004–0.05 μm radius), a system with a differential mobility analyzer (DMA) (TSI model 3071A) and a condensation particle counter (CPC) (TSI model 3010) were used. Size distribution of aerosols was calibrated by laboratory study using monodisperse aerosols prepared by another DMA. Detection efficiency was measured as a function of particle size. It was 0.19 and 0.77 for the particles of 0.005 μm and 0.03 μm radius. On the observation, size distribution was measured every three minutes. Number concentration of particles larger than 0.004 μm radius, was monitored separately using a CPC (TSI model 3760), with the time interval of two seconds. In this paper, this concentration is called the concentration of total particles.

Morphological analysis of individual aerosol particles collected on carbon-covered nitrocellulose (collodion) films with a two-stage impactor was conducted using a transmission electron microscope. In this paper, some electron micrographs of submicron particles collected with the impactor of 0.5-mm diameter jet are shown.

In addition to these aerosol measurements, SO2 mixing ratio was measured with a SO2 analyzer (Thermo Environmental Instruments model 32S). The detection limit is 0.1 ppbv. Isentropic backward trajectories of air parcels were calculated with a global objective analysis data (GANAL) by the Japan Meteorological Agency (longitudinal grid number is 288, latitudinal grid number 145, number of layers 17 and time interval 6 h) with the time step of 10−3 days. Air temperature and dew point temperature were measured using Rosemount temperature sensor and EGG dew point sensor, respectively. Mixing ratios of CO, O3 and other trace gases observed in this campaign were summarized in Sawa et al. (2004).

4. Results

4.1 Vertical sounding over Naha

Figure 3 shows the vertical profiles of aerosol concentrations (3a), air and dew point temperature (3b), and mixing ratio of SO2 (3c) observed over Naha (flight No. 2 in Table 1). The observational area was located approximately 500 km from the coast of the Asian continent. The observation was carried out along a line perpendicular to the dominant wind direction (northwest). The length of the line was approximately 400 km (Fig. 1). Ascending and descending flights were conducted at different locations.

A temperature inversion existed at approximately 4 km altitude, suggesting the top of the mixed layer. SO2 mixing ratio was remarkably high (approximately 7 ppbv at 2 km altitude) in the mixed layer. Note that typical mixing ratio of SO2 in clean marine boundary layer, is between 20 and 50 pptv (for example, Andreade et al. 1988). Aerosol concentrations are also high in the mixed layer. At the higher altitudes (5–11 km), particles in the larger size bins (> 0.05 μm radius) decrease with increasing altitude. On the contrary, the number concentrations of total particle increased at altitudes above 6 km. This means very fine particles (0.004 < r < 0.05 μm) increased, with increasing altitude in the free troposphere.

At 8–11 km altitude, the number concentrations of particles in the radius ranges of 0.05–0.15 μm, and 0.15–0.25 μm observed during the
descending flight, were much higher than those observed during the ascending flight. The descending flight was at approximately 29°N, and the ascending flight was at approximately 26°N latitude. Though not shown here, the latitudinal locations of subtropical front over the longitude (125–130°E) did not differ largely from those at 140°E shown in Fig. 2. Therefore, the flight path at 8 km altitude crossed the subtropical front. The large variation was considered to be due to the presence of the front.

Figure 4 shows the number-size distributions of aerosols observed at all altitudes over Naha. Each size distribution was the average for 10–15 minutes. Two size distributions were obtained at each altitude, except for 11 km. These size distributions showed differences in the characteristics with altitude. Concentrations in the accumulation mode are high, especially at 2 km altitude (Fig. 4d). The size distribution of aerosols could be fitted well with a log-normal function, with a center radius of 0.06 μm. On the other hand, the size distributions observed at 8 km (Fig. 4b) and 11 km altitude (Fig. 4a) have maximum concentrations at approximately 0.02 or 0.03 μm radius, along with quite low number concentrations in the accumulation mode. Size distributions observed at 5 km altitude (Fig. 4c) differ from both size distributions observed at 2 km and 11 km altitudes. Mode radius is at approximately 0.04 μm, and size distribution in the smaller size ranges, than the mode radius, resembles that observed at 8 km altitude. In the larger size ranges than the mode radius, number concentrations are higher than those observed at 8 km altitude.

Figure 5 shows the electron micrographs of aerosol particles collected at 5 km and 11 km altitude over Naha. Most particles at 11 km altitude exhibit satellite droplet rings at the collection surface (Fig. 5a). The morphology in-
Fig. 4. Number-size distributions of aerosols observed at altitudes 11 km (a), 8 km (b), 5 km (c) and 2 km (d) during the flight No. 2 over Naha.

dicates the presence of sulfuric acid (Frank and Lodge 1967). This kind of particles was also dominant in the samples collected at 8 km altitude. On the other hand, mineral and soot particles were found in the sample collected at 5 km (Fig. 5b), suggesting the influence of air from the continental surface.

Figure 6a shows the isentropic backward trajectories of air parcels from 2 km, 5 km, 8 km and 11 km altitudes from the observation area. Figure 6b shows altitudes of the trajectories. The air parcel at 2 km altitude was from low altitudes over the coastal regions of eastern Asia. This confirms that the air at the lower altitude was strongly influenced by anthropogenic pollution from the continental surface. The air parcels at the higher altitudes (5 km, 8 km and 11 km), were also transported from the Asian continent. The extent of the continental influence would tend to be low at higher altitude (8 km and 11 km), whereas the influence was apparently found in the sample collected at 5 km altitude, as shown in Fig. 5b. The vertical transportation of polluted air was possibly linked with the height of mixed layer in the coast regions of the Asian continent.
Fig. 5. Electron micrographs of particles collected at 10.8–10.9 km altitude over Naha (26.3°N, 125.7°E–29.1°N, 127.7°E) during the period of 07:05–07:34 GMT on 13 February 2000 (a), and at 5.1–5.4 km altitude over Naha (26.8°N, 126.0°E–29.3°N–127.8°E) during the period of 08:13–08:43 GMT on 13 February 2000 (b). The scale is 1 μm.

(Sawa et al. 2004). Air parcel at 11 km altitude was transported from the tropical Indian Ocean.

4.2 Vertical sounding over Iwojima
Figure 7 shows the vertical profiles of aerosol number concentrations (7a), air and dew point temperature (7b), and SO₂ mixing ratio (7c) observed over Iwojima (flight No. 4). Iwojima is located approximately 1500 km west of Naha (flight No. 2 in Table 1). The height of the mixed layer was approximately 4 km, similar to Naha. Vertical distributions of the number concentrations of particles larger than 0.05 μm radius are similar to those observed over Naha. The number concentrations were high at altitudes lower than 4 km, and then decreased with increasing altitude up to 11 km altitudes. Similar to Naha, the number concentrations observed at 8–11 km altitude during the descending flight (at approximately 29°N), were higher than those observed during the ascending flight (at approximately 26°N).

Although the total particle concentrations below 3 km altitude are lower than those over Naha (which are higher than 1000 cm⁻³), the number concentrations at the higher altitudes, are almost the same as those at the same altitude over Naha. Number concentrations of par-
particles in the larger size bins, show quite similar vertical profiles to those over Naha. These results suggest that the distribution of aerosols was relatively uniform in longitudinal direction. The enhancement of very fine particles at higher altitudes is also found over Iwojima. 

SO₂ mixing ratios at the four altitudes, were approximately half of the values over Naha.

Though not shown here, the number-size distributions observed over Iwojima were quite similar to those over Naha. The number concentrations of particles in the accumulation mode, were high in the low altitudes (especially at 2 km altitude). Mode radii in the size distributions at the high altitudes were small (approximately 0.02 μm at 11 km altitude).

These similarities of the vertical profiles, and size distributions to those over Naha, are considered to be due to the location of Iwojima which is leeward of Naha at each altitude. Backward trajectory of air parcel from each altitude over Iwojima, was passing over the observation area of Naha, and the extended line is similar to the backward trajectory from Naha (not shown). On the basis of air velocities observed, time duration for the transportation of air from Naha to Iwojima was expected to be 6 h at 11 km altitude and 48 h at 2 km altitude, respectively.

4.3 Vertical sounding over Saipan

Figure 8 shows the vertical profiles of aerosol concentrations (8a), air and dew point temperature, observed over the area to the east of Saipan (flight No. 11). As found in the figure, values of high relative humidity suggest the presence of several cloud layers (0.5 km, 6 km and 8 km altitude). The vertical distribution of aerosols would be influenced by cumulus convection. The number-concentrations of particles larger than 0.05 μm radius, show the enhancement at several altitudes (for example, approximately 8 km), and the horizontal variations seems to be large. However, the concentrations
are basically high at the lower altitudes. On the contrary, the number-concentrations of total particles at altitudes higher than 6 km, are approximately $10^3 \text{ cm}^{-3}$, which are higher than the values at the lower altitudes (several $10^2 \text{ cm}^{-3}$). The spatial differences in number-concentration of total particles, were relatively small during the flight as compared to those over Naha.

Figure 9 shows the number-size distributions observed over Saipan. The size distributions observed at 11 km (9a), 8 km (9b) and 5 km (9c) altitudes are similar in shape. The maximum concentrations existed in the radius range of 0.02–0.03 $\mu$m, and the number concentrations of aerosols in the accumulation mode were low in these size distributions. On the other hand, the size distributions observed at 2 km altitude (Fig. 9d) exhibit aged features. According to the backward trajectories, the air parcels at 8 km and 11 km were transported by westerly winds, whereas the air parcels at 5 km and 2 km were transported from the central Pacific Ocean by easterly winds. The reason why the size distribution at 2 km altitude, is different from the others is considered to be due to influence of air, from the marine boundary layer (MBL). Aerosols in the MBL generally have relatively large mode radius, and contain high number concentration of accumulation mode particles, even in remote regions (Heintzenberg et al. 2004).

4.4 Size distributions observed in subtropical jet streams

Figure 10 shows the size distributions observed at 11 km altitude during the flight path Saipan-Nagoya (flight No. 12). These size distributions are similar to those observed in the upper troposphere over Naha, Iwojima and Saipan. Maximum number concentrations, existed in the radius range of 0.02–0.03 $\mu$m ra-
dius, and the concentrations of aerosols in the accumulation mode were low. The size distribution at 31°N measured at the core of jet streams, shows relatively large mode radius (approximately 0.04 μm), and high concentration of accumulation mode particles as compared with the other distributions shown in Fig. 10.

Figure 11a shows the backward trajectories of air parcels from the flight path. Figure 11b shows altitudes of the trajectories. According to the trajectories, the air parcels at approximately 15–25°N latitude are transported directly from the tropical upper troposphere over the Pacific Ocean, or through southeastern Asia. Air parcels at the higher latitudes are originated from the tropical areas over the Indian Ocean, or the African continent. Namely, the air parcels transported from the tropical upper troposphere, to the middle latitudes by Hadley circulation, and then mixed into subtropical jet streams. These flow patterns are also found in the climatological average flow field, at 200 hPa in winter (for example, Newell et al. 1972).
Fig. 10. Number-size distributions observed at 11 km altitude during the flight from Saipan to Nagoya (flight No. 11).

Fig. 11. Isentropic backward trajectories of air parcels from the flight path Saipan-Nagoya (flight No. 11) at 11 km altitude (a), and altitudes of the trajectories (b).

4.5 Latitudinal distributions of aerosol
The meridional distributions of the number concentrations of accumulation mode particles, and total particles, are shown in Figs. 12a and b, respectively. Each point indicates the average number concentration during 10 minutes. All data acquired in different longitudes were indicated. This analysis is considered to be meaningful, because aerosol distributions in longitudinal direction would be relatively uniform. The meridional distributions are closely corresponding with the location of subtropical front. Below 4 km altitude, the number concentrations of accumulation mode particles are especially high (> 10 cm\(^{-3}\)) over the extratropical region, suggesting a strong influence of aerosols from the Asian continent (Fig. 12a). On the other hand, the low concentrations are found over the subtropical region. At 4–8 km altitude, the concentrations over the extratropical region are also higher than those over the subtropical region. In the upper troposphere (> 8 km), the latitudinal variation is not large over the regions south of tropical jet streams.

Distribution of total particle (Fig. 12b) is different from the distribution of accumulation mode particles. High concentrations are found in lower altitudes in the extratropical region, however, it is remarkable that most of the concentrations are higher than 1000 cm\(^{-3}\) in the upper troposphere over 15–33°N. The relatively low concentrations are found in the middle troposphere over the extratropical region and low altitude over the subtropical region.

5. Discussion
The observational results suggest that characteristics of aerosol size distributions over the northwestern Pacific Ocean (15–40°N) were clearly characterized by the transport history of air. Air mass in the extratropical region was strongly influenced by pollution of anthropogenic particles, from the Asian continent. This is confirmed with several results; that is, (1) mixing ratio of SO\(_2\) was high (several ppbv) below 5 km altitude over both Naha and Iwojima, (2) soot particles were found in the regions, and (3) high CO mixing ratios observed in the regions (Sawa et al. 2004). Characteristics of the size distributions observed in the regions were (1) low number fractions of very fine particle, (2) large mode radii and (3) high concentrations of accumulation mode particles. These features are supposed to be due to anthropogenic particles transporting from eastern Asia. Large production of sulfuric acid vapor,
by the oxidation of SO$_2$ and heterogeneous condensation of the sulfuric acid vapor, possibly cause the rapid growth of very fine particles to larger particles. Consequently, the number concentration of very fine particle decreases, and accumulation mode particle increases. Although high number concentrations of total particles were observed on some occasions, new particle formation is considered to be not active in the free troposphere over the region, because many very fine particles were not observed. However, a skirt of ultra-fine mode was observed at the altitude close to cloud top (2 km) over Naha (Fig. 4). There is a possibility of homogeneous nucleation in the vicinity of clouds, as suggested by Hegg et al. (1990).

On the other hand, different types of average size distributions were observed in subtropical region, and the upper troposphere. Mode radii were generally small (0.02–0.03 μm), and accumulation mode particles showed low concentrations. These features would be due to insufficient precursor gases like anthropogenic SO$_2$. Growth of particles is restricted by this reason. Vertical distribution of the number concentration of total particles suggests the presence of downward flux of very fine particles. It is consistent with the assumption that the particles are transported by Hadley circulation.

Aerosols in the upper troposphere contained large number fractions of very fine particles. According to the analysis by electron micros-
copy, these particles contain sulfuric acid. Backward trajectories suggested that the air parcels in the region were mostly from the tropics. Moreover, extremely low mixing ratios of ozone, observed at 11 km altitude over Naha and Iwojima, confirm the transport of the air mass from the tropics (Sawa et al. 2004). These results suggest that the very fine particles were relatively young particles, formed by homogeneous nucleation of sulfuric acid vapor in the tropical upper troposphere. Backward trajectories of air parcels showed that the time duration for the transport of air from the tropics to the observation site, is approximately 6–10 days. The mode radii of the aerosols observed (0.02–0.03 μm) were consistent with the growth time by coagulation and condensation, with ambient SO₂. Similar size distributions were also observed at the center of the subtropical jet stream. However, the size distribution shows a slightly large mode radius, and high concentrations of accumulation mode particles, as compared with other size distributions observed at the same altitudes over the regions south of the subtropical front. On the basis of the backward trajectories, it is considered that air from the continent will also influence the size distribution.

Newly formed particles at tropical upper troposphere, are assumed to be sulfuric acid particles (Clarke and Kapustin 2002). The analysis using electron micrograph showed that most of the particles observed at upper troposphere over the western Pacific Ocean also contained sulfuric acid. However, organic carbons are also possible constituents, because it is difficult to detect them by analysis. At least, influence of organic carbons from fossil combustion, or biomass burning, is considered to be small, because soot was not detected in most of the samples collected in the regions.

6. Conclusions

Number-size distribution of aerosols, and its spatial variation, were observed in the free troposphere up to 11 km altitude over the northwestern Pacific Ocean during the Pacific Atmospheric Chemistry Experiment (PACE)-7 campaign in February 2000.

Features of aerosol size distributions were different in different air masses and clearly divided by subtropical front. In the extratropical region, the characteristics were (1) low number fractions of very fine particles, (2) large mode radii and (3) high concentrations of accumulation mode particles. These features were considered to be due to the growth of aerosols by anthropogenic SO₂ from eastern Asia. Aerosols in the subtropical region had features (1) small mode radii, (2) low concentrations of accumulation mode.

High number concentrations of particles, with large fractions of very fine particles and small mode radii (0.01–0.03 μm) were distributed from 15–31°N in the upper troposphere. These particles were considered to be newly formed particles in the tropical upper troposphere, by homogeneous nucleation. Although the size distribution observed at the center of the subtropical jet streams had slightly aged features, it is basically similar to the size distributions observed in the upper troposphere over the subtropical region. On the basis of the trajectory analysis, aerosols would have been transported from the tropical upper troposphere to the middle latitudes by Hadley circulation, and then mixed into the subtropical jet streams. The results obtained in this study suggest that particles over the tropics potentially would spread efficiently in global scale through jet streams.

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