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Aircraft Observation of Aerosol and Cloud-droplet Properties over the East China Sea Influenced by the Outflow of Asian Polluted Air

Deepak Aryal[1,*], Yutaka Ishizaka[2], Kikuo Okada[3]

[1]Central Department of Hydrology and Meteorology, Tribhuvan University, Kirtipur, Kathmandu, Nepal
[2]Division of General education, Aichi Gakuin University, 12 Araike, Iwasaki-cho, Nisshin 470-0195, Japan
[3]Meteorological Research Institute, 1-1 Nagamine, Tsukuba, Ibaraki 305-0052, Japan
*Corresponding author: daryal1@hotmail.com

Abstract Airborne measurements of aerosol particles and cloud microstructures were made over the ocean around the south of Kyushu Islands of Japan during the Asian Atmospheric Particulate Environmental Change Experiment 3/Asia Pacific Regional Aerosol Characterization Experiment (APEX-E3/ACE-Asia) during the period of 17 March to 13 April 2003. Results demonstrated that polluted air from the Asia continent could penetrate several hundreds of kilometers over the oceans and clouds forming in that air had significantly altered microphysical properties. Based on the number concentration of aerosol particles with diameters between 0.3 and 5 μm, two cases were investigated: 22 March 2003 was termed a "clean" case and 12 April 2003 as a "polluted" case. Single particle analysis of particles was also carried out by electron microscopy. The particles in the polluted marine boundary layer were characterized by the presence of sulfate particles with traces of potassium and heavy metals. The cloud droplets in the polluted marine boundary layer exhibited larger number concentrations than those in the clean boundary layer, along with the decrease in the droplet size. Present study demonstrated that polluted air from the Asia continent could penetrate several hundreds of kilometers over the oceans and clouds forming in that air had significantly altered microphysical properties.

Keywords: *East China Sea, Size distribution, Cloud droplet, Biomass burning, single particle analysis*

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1. Introduction

Aerosol particles can affect the radiation balance due to their ability to absorb and scatter solar radiation (direct effect) [4]. On the other hand, these particles play important roles in both liquid condensation and freezing working as cloud condensation nuclei (CCN) and ice nuclei (IN) [29], which modify the optical properties of clouds (indirect effect) [1,5,32]. The direct and indirect radiative effects of aerosol particles depend on their size distribution, chemical composition, mixing properties and shape. These aerosols have an effect on the tropospheric chemistry by providing media for various diverse reactions and serving as carriers for the chemical species. [9].

The emission of gases and particles over East Asia has been increasing due to the rapid development of industrial activities in recent years. This area has been characterized by high emission of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) in the last decade and the trend is expected to be continued [1,13,20]. It has been predicted that the area will become the largest sources of NO_x and SO_2 in the world within the first decade of 21st century [12]. In addition, Kajii et al. [19] found that significantly high concentrations of gases (NO_x, SO_2 and other hydrocarbons) in the westerly wind from the Asian continent, which passed through a lower boundary layer over the Yellow Sea and the southern part of the Korean Peninsula. Hatakeyama et al. [15] also indicated on the basis of aircraft observations that East Asian is a source of high concentrations of gases and particulate pollutants.

The results from the Indian Ocean Experiment (INDOEX) certainly dealt with Asian outflow. A major objective of the INDOEX involves the characterization of the extent and chemical composition of pollution outflow from the Indian subcontinent during the winter monsoon. An intensive field study of atmospheric chemistry to elucidate the impact of anthropogenic emissions on the marine atmosphere is the recent international initiative of the International Global Atmospheric Chemistry Program/East Asian North Pacific Regional Experiment (IGAC/APARE) [30]. The main focus of the APARE is to study the chemical processes occurring in and around the Asian subcontinent, long-range transport of atmospheric species over the northwestern Pacific Rim and to estimate the magnitude of the impact of human activities on the marine atmosphere over this region.

There have been many observations, in other areas than Asia, documenting the affect of anthropogenic particulate matter on radiative properties of clouds and precipitation processes in clouds that may lead to changes in the dynamical processes in clouds such as affecting cloud lifetimes. For example, investigators [10,16,21] report increased CCN led to higher droplet concentrations and narrower droplet spectrum (which manifested itself as a higher cloud albedo) that suppressed drizzle formation and, perhaps, produced longer lasting stratiform clouds.

However, these observations over the areas downstream of the Asian continent are still limited. Thus, knowledge on the spatial distribution of aerosol characteristics (number concentration, size distribution, elemental composition) and the microstructure of clouds over the areas influenced by the outflow of Asian pollution is necessary to clarify the behavior of the atmospheric aerosols in the marine boundary layer and their effects on cloud microstructure. As a part of the Asian Atmospheric Particulate Environmental Change Experiment 3/ Asia Pacific Regional Aerosol Characterization Experiment (APEX-E3/ACE-Asia) during the period of 17 March to 13 April 2003, aircraft observations were made over the sea around Kyushu Islands of Japan. The aim of this paper is to present the characteristic features of the aerosol size distribution and the elemental composition of individual particles observed over the marine atmosphere and their effects on cloud microstructure.

2. Experiments

Aircraft observations of meteorological parameters, aerosol particles and cloud droplets to the south of Kyushu Island in Japan were performed in the spring (March and April) in 2003. The location of the observational area is as shown in the Figure 1.

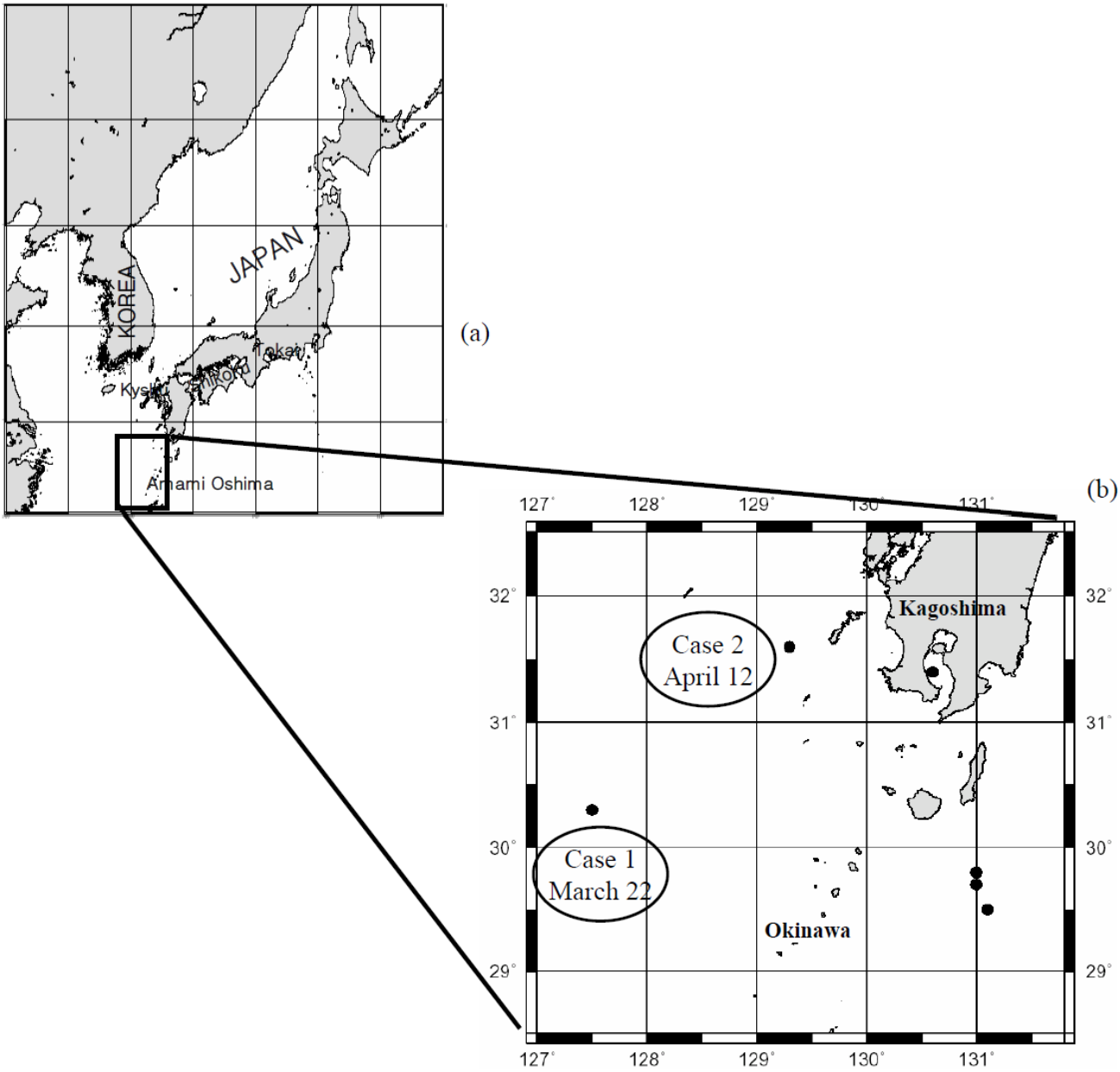

Figure 1. Location of the observation area. The observations of aerosols and cloud droplets were carried out at points marked by closed circles in Figure 1b

The dots in the enlarged figure were the aerosol and cloud observation points. Thirteen flights were performed as described in Table 1.

Table 1. Flight data and cloud information

Date	Flight No.	Location		Alt (Km)	LPC (cm^{-3})	Obs. Time(JST)	cloud base (m)	cloud top (m)
		Lat (^{0}N)	Long (^{0}E)					
17-Mar-03	C404-1	30.8	129.4	5	180	12:40-15:30	650	1350
20-Mar-03	C404-2	30.4	130	5	532	13:00-16:00	900	1150
22-Mar-03	C404-3	30.3	127.7	4.5	160	10:35-13:35	1200	1800
26-Mar-03	C404-4	29.8	130.9	3	180	12:08-15:08	850	1175
29-Mar-03	C404-5	31.3	129.7	4	400	10:39-12:53	950	1100
31-Mar-03	C404-6	29.7	130.9	5	550	12:40-15:50	650	2400
2-Apr-03	C404-7	30.6	129.2	5	440	12:42-15:05	900	1350
5-Apr-03	C404-8	29.1	130.5	5	350	09:43-12:59	800	1900
8-Apr-03	C404-9	30.3	129.3	3	2000	10:22-13:23	625	1700
9-Apr-03	C404-10	29.8	131	4	1088	12:22-15:44	1100	1650
10-Apr-03	C404-11	29.7	131	5	1000	10:15-13:08	975	1400
12-Apr-03	C404-12	31.8	129.3	4	1700	10:18-12:47	450	800
13-Apr-03	C404-13	32	128.5	5	1400	10:18-13:17	N/A	N/A

A Cessna 404 TITAN aircraft was the platform for the observations. The following instruments were onboard: The size distribution of aerosol particles was measured with a laser particle counter (LPC) (Model: TF-500, KANOMAX). The optical system of the LPC is similar to an optical aerosol counter (OPC). The LPC uses light-scattering technique to measure the size spectrum of the optically effective particles, which usually ranges from approximately 0.3 to 10 μm in diameter. The light source is a laser diode (output power: 30 mW, wavelength: 780 nm). Wide-angle (near 300°) scattering light by a particle was collected using an elliptical mirror in shape and scattering intensity was measured with a photo diode. Optically measured particle sizes depend on the complex refractive index of aerosols. Size distributions obtained directly with the optical instrument are equivalent to those calibrated with a known refractive index of m= 1.60 and no absorption for polystyrene latex spheres. The size distribution was measured in a dry condition (e.g., [34]).

A forward scattering spectrometer probe (model FSSP-100, PMS) was used to measure the size distribution of cloud droplets. The size range was 2 to 47 μm in diameter and divided into 15 channels.

Total temperature sensor (Model 102 Deiced, Rosemount Engineering Company, US) was used to measure air temperature. The range of temperature is –70 to +350°C and is capable of measuring the temperature from the surface to 100,000 feet of altitude.

Dew point temperature was measured with a thermoelectric dew point hygrometer system (Model 1011A, General Eastern Instrument Corporation, US). The system is designed to obtain in flight static-pressure measurements of the prevailing dew point or frost point temperature. The system is capable of measuring dew points from 0°C to +50°C and frost points from 0°C to –75°C. The accuracy of the temperature measurement was ± 0.5°C above 0°C and ± 1°C below 0°C. The response time of sensor mirror was 1°C s^{-1}. The aerological data over Kagoshima, which is located close to our observational areas, obtained by the Japan Meteorological Agency (JMA) were also used to augment the airborne measurements.

In order to examine the physical and chemical properties of individual aerosol particles, particles were collected directly on electron microscope nickel grids covered by a carbon-coated Formvar film. The air introduction system used to lead ambient air into the in-cabin is described in detail in Zhang et al. [35]. Air was introduced into an in-cabin manifold through an inlet system (inside diameter of the inlet: 2.5 mm). The system was fixed below the aircraft cabin facing forward to the flight direction and aligned parallel to the aircraft axis. The inlet is similar to a porous diffuser inlet showed by Huebert, B.. To avoid flow turbulence at the inlet and minimize the effect of particle collisions on the nozzle edge, a supplementary air suction system was attached around the inlet. Air introduced to the manifold was distributed to a laser particle counter and a particle collection impactor through straight TYGON tubes. The inlet and the manifold were made of stainless steel, which should minimize the influence of electrical charges on the smoothed wall. Surplus air at the inlet was thrown away from the manifold using the air exhaust system. The flow speed into this system was adjusted to keep isokinetic flow at the inlet nozzle. This assured that flow got into the inlet without experiencing turbulence at the inlet entrance. The flight speed during the observation period was from 70 ms^{-1} to 85 ms^{-1} with an average of 77 ms^{-1}. So the air flow entered the inlet tip at about 77 ms^{-1}; subsequently the conic diffusing section of inlet decelerated the axial velocity to 4.5 ms^{-1}. Aerosol particles were collected from the manifold with an impactor using a 1-mm diameter jet at an airflow rate of about 3 lpm. Sampling time of aerosol particles using an impactor was roughly decided according to the aerosol concentrations measured with the LPC.

In order to investigate the shape and volume of the individual particle, in the laboratory collected particles were shadowed by a Pt/Pd alloy at an angle of 26.6^o (arctan 0.5) resulting in a shadow length equal to twice the height of the particle. The individual (shadowed) particles were examined with a transmission electron microscope (Model H-600, Hitachi Ltd., Tokyo, Japan). Negative films taken at 2000 magnification were processed with a resolution of 1000 dpi in a computer through scanner (Cannon Cano Scan 9900F). Further the particles were examined using an energy-dispersive X-ray (EDX) analyzer (Kevex, sigma II) through a Kenvex UTW (ultra-thin window) detector to obtain the elemental composition

of individual particles. X-ray spectrum was obtained from the center of a particle at an accelerating voltage of 50 kV, with the counting time of 100s. Weight percentages of the elements Na, Mg, Al, Si, S, Cl, K, Ca, Zn and Pb in individual particles were obtained using the quantitative EDX analysis.

Isentropic backward-trajectory analyses (although not shown the figures) were carried out in order to estimate the transport path of the air parcels from which the aerosol particles were collected. The trajectories were calculated for 72 hours using the NOAA-HYSPLIT4 model (Hybrid Single Particle Lagrangian Integrated Trajectory) Model, 1997. Web address:http://www.arl.noaa.gov/ready/hysplit4.html, NOAA/ARL, Silver Spring, MD).

3. Results and Discussion

3.1. Number Concentrations of aerosol Particles Measured by the Laser Particle Counter

Figure 2 shows the number concentrations of aerosol particles measured by the LPC (0.3 to 5 μm) during all the flight campaign. The concentrations were measured at three altitudes: 500 m, around 1500 m and above 3000 m for each flight. Particles concentrations during the first half of the observation were around 200-400 cm^{-3}, with the exception of the concentrations on 22 and 26 March (less than 100 cm^{-3}). During the second half of the observation (8 - 13 April), the concentrations were about three times greater than those observed during the first half.

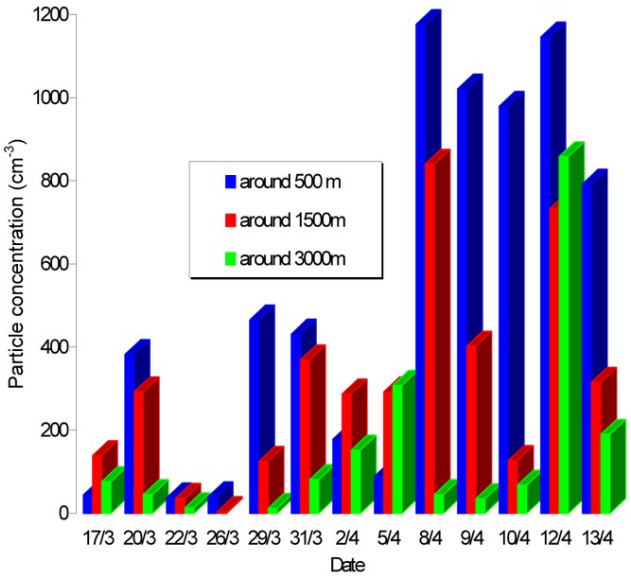

Figure 2. Number concentrations of aerosol particles measured by the laser particle counter in the two diameter ranges > 0.3 μm

These results suggest different types of air masses influenced the measurement site near Kyushu Islands. In particular, during the second half of the observations, according to air mass back-trajectories polluted air masses were found to be frequently transported from central China and southeast China to the observational areas. Thus, the whole set of aerosol spectra were subdivided into clean (March) and polluted (April) time periods.

For this study, two flights were chosen that highlighted the dichotomy between a "clean" and a "polluted" air mass. The number concentration and elemental composition of aerosol particles in the marine boundary layer on 22 March, hereafter denoted as the "clean" case, had low concentrations of aerosol particles and a continentally influenced boundary layer on 12 April, hereafter denoted as the "polluted" case, had high concentrations of aerosol particles.

3.2. Synoptic Situations for the "clean" and "polluted" Cases

During the "clean" case, the Kyushu region was covered by clouds associated with an extratropical cyclone off the Coast of Shikoku Island. In the morning, the front associated with the cyclone was off the coast of Shikoku Island affecting the atmosphere of the observation areas but the weather improved from overcast to scattered conditions in the afternoon. Observed surface and upper air winds were N and NW. Low clouds were observed over Amami Oshima Island.

During the "polluted" case, a high-pressure system was located to the east of Japan. A stationary front was located from off Tokai to south China. Prevailing winds were westerly from the surface to the upper air. The Amami Oshima and Okinawa regions which are well south of the observation areas were experiencing heavy rain.

The vertical profiles of air temperature and dew point temperature from the aircraft are shown in Figure 3 (a) and (b). A cloud layer was observed between the altitude of 1200 and 1800m on 22 March and between 450m and 800m on 12 April. An inversion layer was observed at the altitude of 1800m on 22 March (Figure 3a) and at 870m on 12 April (Figure 3b). Dew point was almost identical with the values of temperature up to 1800m and diverges significantly on 22 March showing the level of clouds. The large decrease in dew point can be seen around 1000 m on 12 April. These data were later verified with he data received by rawinsonde launched at Kagoshima near the observation area at 00:00 UTC (+9 hours to JST) by JMA.

3.3. Number-size Distribution of Aerosol Particles

Figure 4 shows the size distributions and number concentrations of particles for the “clean” case (Figure 4a) and for the “polluted” case (Figure 4b) measured with the LPC. The measurements were made below cloud base (1200 m and 450 m, respectively) and near cloud top (1800 m and 800m, respectively). It can be seen in the figure that the size distributions below cloud base and near cloud top are similar for each case. However, the number concentrations for the “polluted” case were significantly higher than those for the “clean” case.

According to the trajectory analyses (though figures are not shown), air parcels arriving on 22 March approached the observational areas from the north mainly through the atmosphere over the Sea of Japan, whereas the air parcels arriving on 12 April were transported from the China continent. Hence, the number concentrations during the “polluted” case are the largest because they would be influenced by the outflow of anthropogenic aerosols from the China continent.

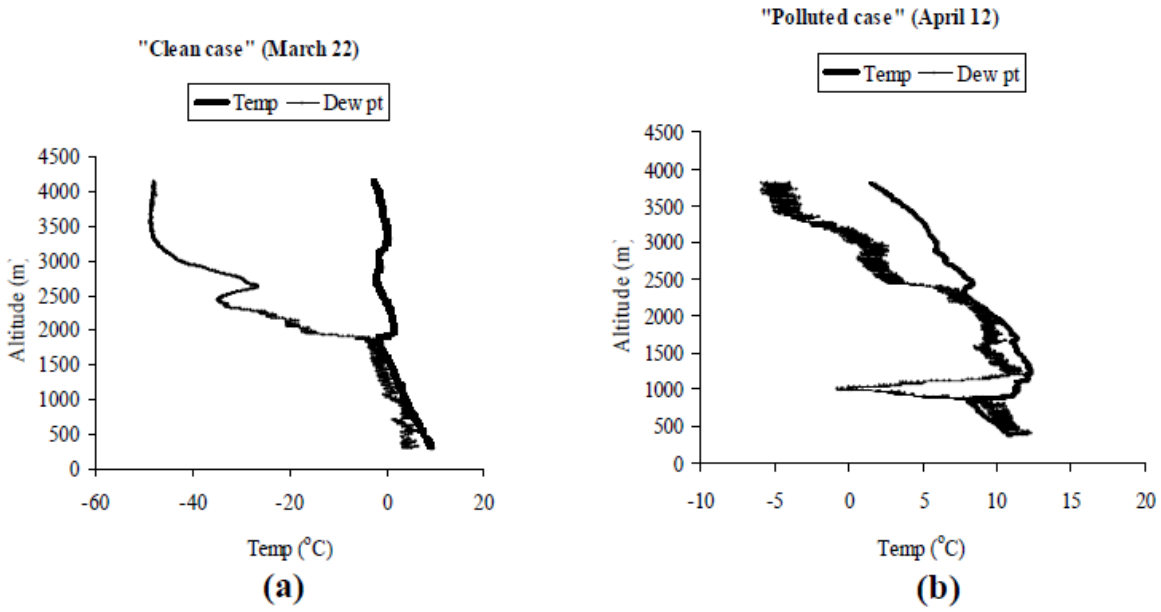

Figure 3. Vertical profiles of temperature (°C) and dew point temperature (°C) observed by aircraft on 22 March "clean case" (a) and 12 April 2003 "polluted case" (b)

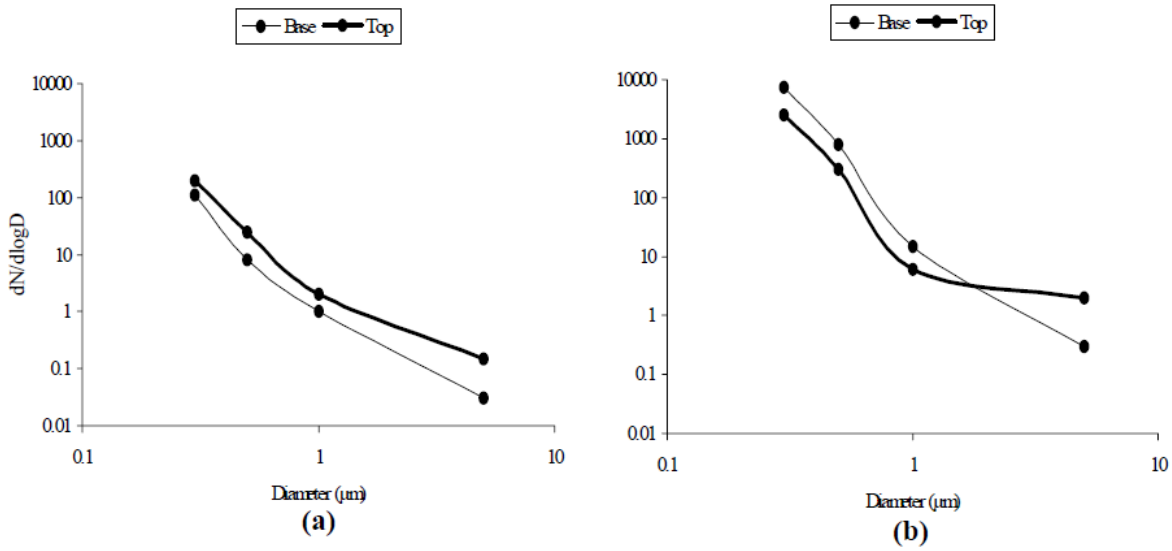

Figure 4. Number-size distributions of aerosol particles below cloud base and near cloud top on 22 March "Clean case" (a) on 12 April, 2003 "polluted case" (b)

3.4.1. Particles collected during the "clean" case

In order to investigate the aerosols which serve as CCN we have collected aerosol samples near the clouds (i.e., below and above). Figure 5 (a) shows the electron micrograph of aerosol particles collected at 11:52- 11:54 JST at an altitude of about 1200 m near but below cloud base on 22 March. It can be seen, aerosol particles larger than 1 μm diameter are composed of electron-opaque material surrounded by small round spots. Figure 5 (b) illustrates the EDX spectrum of particle A in Figure 5 (a). The particles were mainly composed of sodium and chlorine. Thus, the particles were hygroscopic and were in the aqueous phase before being collected on the microscopic grid; their shapes were no doubt spherical in the air. Though not shown here, at this location (below cloud base) measured fine particles (< 1 μm diameter) usually contained sulfur which is termed as S-rich particles.

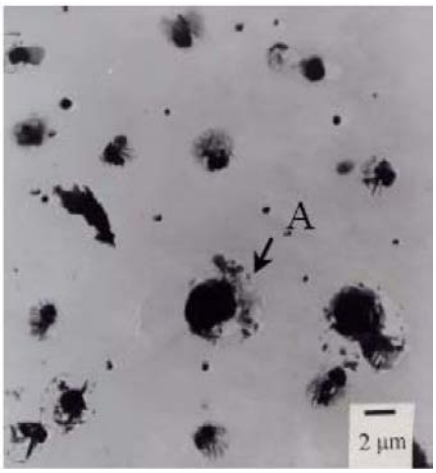

Figure 5. (a) Electron micrograph of aerosol particles collected at 11:52 JST at an altitude of about 1200 m near cloud base on 22 March, "Clean case"

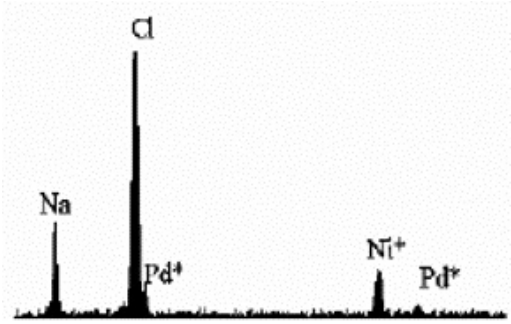

Figure 5. (b) EDX spectrum of the particle A, element with* is mainly due to the Pt/Pd alloy and the collecting surface

Figure 6 (a) shows the morphology of particles collected near the cloud top but out of cloud (1750 m altitude) at 11:56-11:59 JST on 22 March. The EDX spectrum showed that most of the fine particles were mainly S-rich particles, Figure 6 (b). Photograph shown in Figure 6 (a), of S-rich particles did not show satellite droplet rings on the collection surface. Since the droplet rings are characteristics of sulfuric acid particles (e. g., [11]) the absence of droplets rings suggests that those particles were sulfate particles. The particles which showed morphology similar to sea-salt particles found in Figure 5 (a) were very few. The presence of such S-rich particles in the atmosphere is mainly due to the emission of gaseous aerosol precursors and the consequent gas to-particle conversion [27].

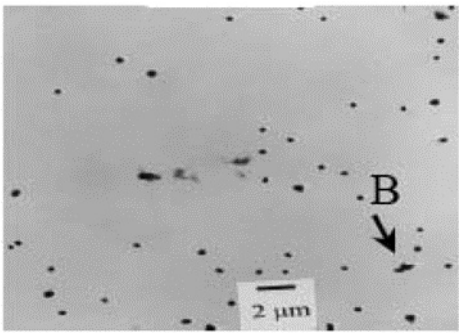

Figure 6. (a) Micrograph of aerosol particles collected at 1750m near cloud top on 22 March, 2003 “Clean case”.

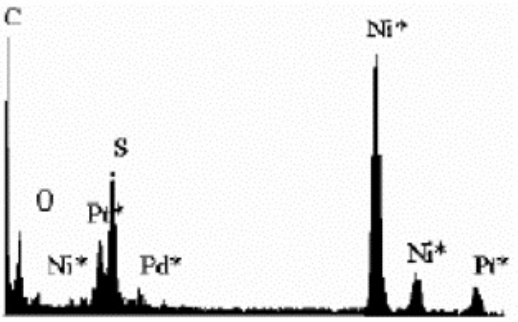

Figure 6. (b) EDX spectrum of the particle B.

3.4.2. Particles Collected during the "polluted" Case

Figure 7a shows the morphology of aerosol particles collected below cloud base on 12 April at 11:34-11:36 JST. Many particles can be seen on the film. In order to get the exact EDX spectrum we have avoided areas where particles were superimposed. The altitude of the collection was about 450m. Particle was S-rich with a small peak of K {for example, the EDX spectrum of particle C shown in Figure 7 (b)}. Particles shown in Figure 7 (a) would be in the diameter range less than 1 μm in the atmosphere; the particles are “flattened” on the collection surface making their diameter appear larger than in the actual atmosphere Figure 8 (a) shows the morphology of aerosol particles collected near the cloud top but in cloud-free air at an altitude of 1100 m at 11:49-11:52 JST on 12 April. The particle density on the collection surface was slightly smaller at cloud top than at cloud base. Figure 8 (b) shows the EDX spectrum of particle D. It shows that the presence of a large S peak together with a small peak of K.

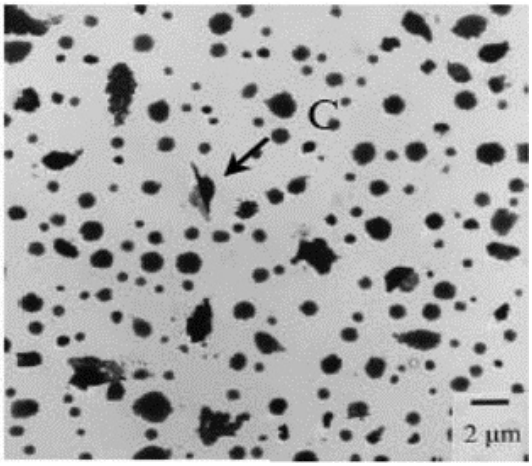

Figure 7. (a) Electron micrograph of aerosol particles collected at an altitude of about 450m near cloud base on 12 April “Polluted case”,

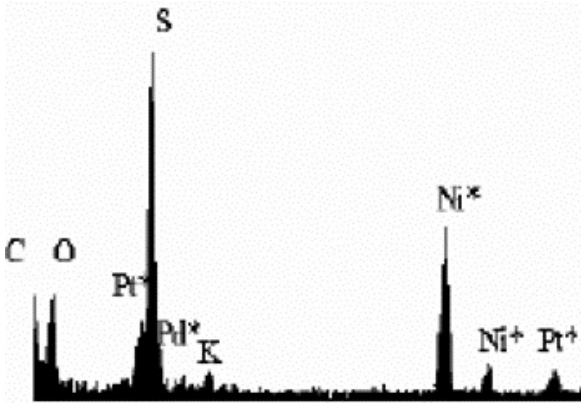

Figure 7. (b) EDX spectrum of the particle C.

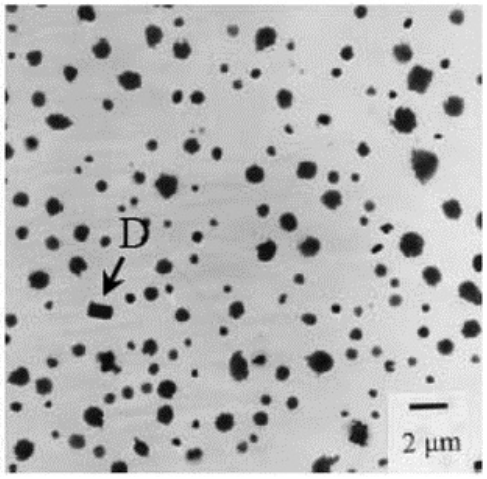

Figure 8. (a) Micrograph of aerosol particles collected at 1100m near cloud top on 12 April, 2003, “Polluted case”

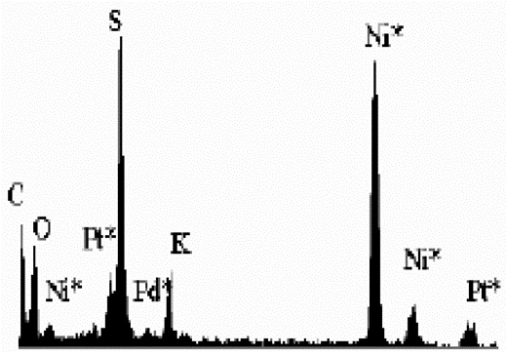

Figure 8. (b) EDX spectrum of the particle D.

The presence of S-rich particles with K is considered to be anthropogenic aerosol particles transported over our observational area. The presence of K in submicrometer S-rich particles has been described to be an indicator of biomass/biofuel combustion (*e.g.;* [3,17,24]). By biomass burning, submicron KCl or K_2SO_4 particles are formed through homogenous processes as shown by Christensen and Livbjerg [6]. Also, S-rich particles, which contain K, have been detected in the polluted atmosphere of China where they have been associated with anthropogenic biomass burning [14,36]. Further, Okada et al., [25] analyzed aerosol samples collected in the atmosphere of the Tibetan plateau and they reported that 20-30% of sulfur containing particles contained potassium suggesting the origin of biomass burning. Moreover, Ikegami et al. [18] observed S-rich particles with K at an altitude of 5-6 km over the western Pacific under the condition of strong westerly winds. They suggested that these S-rich particles were transported from the continental areas.

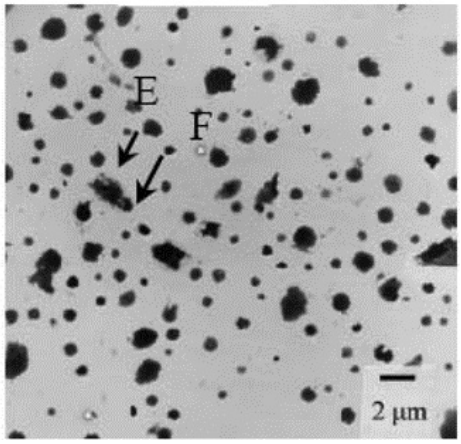

Figure 9. (a)) Electron micrograph of aerosol particles collected near cloud base on 12 April, "Polluted case"

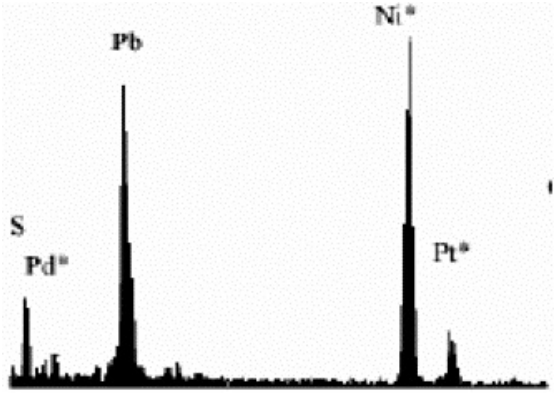

Figure 9. (b) EDX spectrum of the particle E

Figure 9 (a) shows the aerosol particles collected at an altitude of 1150 m at 12:03-12:06 JST on polluted case. Figure 9(b) and (c) show the EDX spectra of particles E and F respectively. Particle E and F were found together like a single particle. Lead (Pb) was detected together with S near the cloud top. Although not shown here heavy metal Zn was also detected around the same location. The presences of these particles were also reported in a polluted case [31,33]. Zn is mainly emitted from iron, still or Ferro alloys plants [26]. Hence, the presence of these particles in the observation site also implies the transport of anthropogenic particles from the continental areas. Though not shown here, the samples on 12 April showed the presence of modified sea-salt particles collected under the cloud base (450 m) and near the cloud top (1100 m). The weight ratios of Cl/Na in the particles tended to decrease with increasing the weight percentages of S. Modified sea salt particles were not observed on 22 March above the clouds or beneath the clouds.

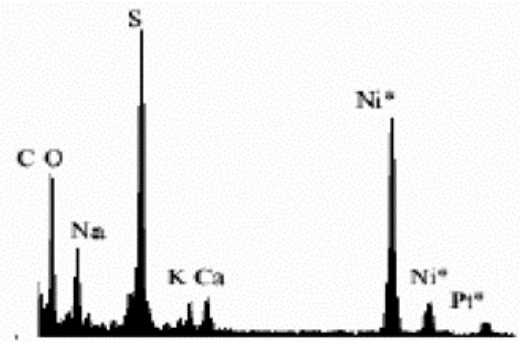

Figure 9. (c) EDX spectrum of the particle F

Thus, this information on the elemental composition and the back trajectories demonstrates that anthropogenic aerosols covered the observational area around Kyushu Island on 12 April while many fewer anthropogenic aerosols, if any, covered the observational area on 22 March.

3.5. Size Distribution of Cloud Droplets

The differences between the aerosol particles in the "clean" and "polluted" cases have been illustrated. We applied the extraction (water dialysis) of water-soluble material to particles. The method is to remove water-soluble material in particles through collection film (e.g., [22,23]), We found that the dialysis was not successful because the collecting surface (formvar film) could not permit to remove water-soluble material. However, the particles examined exhibited the morphological changes in a high humidity, indicating the composition of water-soluble crystals indicating the most of the particles would be hygroscopic and, hence can serve as cloud condensation nuclei (CCN). Here, we investigate the cloud droplet measurements in order to detect the impact of the particles on cloud properties.

Figure 10 (a) and (b) show the vertical profiles of liquid water content (LWC) in the two cases. It can be seen LWC were about three times higher in the "polluted case" (April 12) than in "clean case" (March 22). Figure 11 shows the number-size distributions of cloud droplets in the clean and polluted cases. In the polluted case (April 12), the number concentration of smaller cloud droplets

was higher than that in clean case (March 22). However, larger cloud droplets appeared in higher concentrations in clean case than in polluted case. These results indicate that large amount of anthropogenic S-rich particles (probably sulfate particles) influenced strongly the microstructure of clouds. To further establish this claim, numerical simulation is need to be done of cloud and precipitation formation using clean and polluted CCN spectra inferred from the aerosol particle physical and chemical measurements. For example, Cooper, et al., [8] provide such a numerical model.

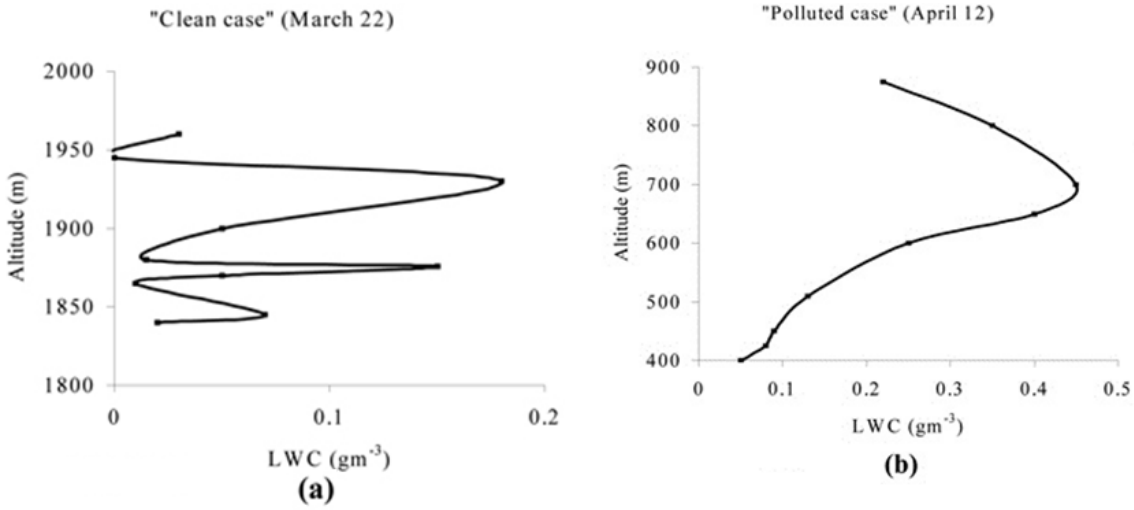

Figure 10. (a) and (b) Liquid water contents (LWC) in clouds for the "clean" and "polluted" cases

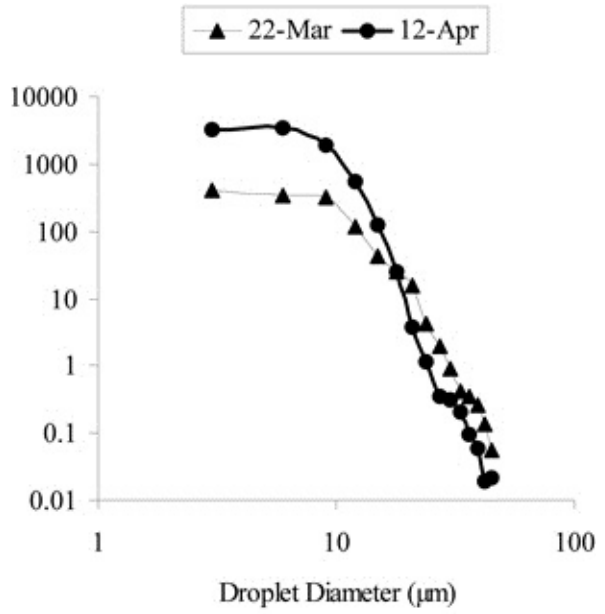

Figure 11. Number-size distribution of cloud droplets measured in cloud in the vicinity of the East China Sea by the FSSP-100 PMS probe

4. Summary and Conclusions

A study of aerosol particles and cloud microstructures has been undertaken using the data from the APEXE3 field campaign in the spring of 2003 south of Kyushu Islands, Japan. A clean and a polluted case were selected for detailed study to identify the differences in the aerosol particle characteristics and the cloud droplet physical characteristics. The following results were obtained from the analyses of the number concentrations, size distributions and elemental compositions of aerosol particles and the cloud droplet size distributions and liquid water contents. Sea-salt particles were found mainly below the cloud base in the clean case. S-rich particles were found above the cloud top. The presence of such S-rich particles in the atmosphere is mainly due to the emission of gaseous aerosol precursors and the consequent gas-to-particle conversion. Sulfur- and potassium-containing particles with traces of heavy metals were found in the polluted marine boundary layer. Results on the elemental composition and the back trajectories demonstrated that these particles were transported from east China and would be primarily from anthropogenic biomass/biofuel combustion. Modified sea-salt particles were also observed in this day. The cloud that formed in the clean marine boundary layer contained fewer and larger droplets that the clouds that formed in the polluted boundary layer. Thus, the anthropogenic particles from east China were responsible for the change in the size distribution of cloud droplets through acting as cloud condensation nuclei. Here we provide evidence that human activities such as industrial emission and biomass burning modified the physical and chemical properties of the aerosol population and led to important change in cloud properties. The extent to which these changes will affect the energy and water cycle in East Asia requires considerable additional study. For example, the response of precipitation, cloud fraction and indirect radiative forcing to the altered cloud droplet properties reported here require a systematic experimental and theoretical evaluation beyond the scope of this paper.

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