Aerial Observation of Aerosols Transported from East Asia — Chemical Composition of Aerosols and Layered Structure of an Air Mass over the East China Sea

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ABSTRACT

Aerial observation was conducted during spring (March–April) 2008 over the East China Sea to analyze the transport of atmospheric pollutants from East Asia and the transformation processes during transport. Concentrations of sulfur dioxide (SO_2), ozone (O_3), total reactive nitrogen (NO_y), carbon monoxide (CO), and black carbon (BC) were measured on board the aircraft, and chemical analyses of aerosols were performed subsequently by high-volume filter sampling of aerosols and ion chromatography and inductively coupled plasma-mass spectroscopy (ICP-MS) analyses. The distributions of gases and aerosols clearly suggested a layered structure to the air mass, in good agreement with predictions of the CFORS (Chemical weather FORecast System) model. The non-sea-salt (nss)-[sulfate (SO_4^{2-})]/([SO_2] + nss[SO_4^{2-}]) ratio value, which increased from north to south along the aircraft flight track, indicated oxidation of SO_2 to form sulfuric acid during transport. In addition, pronounced chlorine loss suggested that substantial production of acidic substances occurred in the air mass as it moved away from China. The negative correlation between the Cl^-/Na^+ ratio and the nss–sulfate concentration suggested that the reaction of sulfuric acid with sea salt was the main cause of the strong chlorine loss. On 28 March a very clear layered structure of the air mass was observed that could be clearly explained by the CFORS computer simulation model. Therefore, here the observation results obtained on 28 March 2008 are reported.

Keywords: Long-range transport; East Asia; Ionic species; Metallic components; Chlorine loss.

INTRODUCTION

Anthropogenic emissions have been increasing in East Asia as a result of rapid economic growth in the region. The emitted atmospheric pollutants have been observed even in North America, and they affect the climate of the whole Northern Hemisphere (UNECE, 2007). Both the global and regional aspects of the long-range transport of pollutants are very important, and several cooperative international experiments have thus been conducted in the western Pacific region (Huebert *et al.*, 2003; Parrish *et al.*, 2004; Nakajima *et al.*, 2007). The current authors have also reported the results of aerial observations of atmospheric pollutants in a source region in China (Hatakeyama *et al.*, 2005; Wang *et al.*, 2006;

Wang *et al.*, 2007; Wang *et al.*, 2008), and a receptor region (Hatakeyama *et al.*, 1995a, b; Hatakeyama *et al.*, 1997; Hatakeyama *et al.*, 2001; Hatakeyama *et al.*, 2004). Transformation of pollutants during their long-range transport is an important problem from the perspective of regional air quality and acid rain. To evaluate the transport of atmospheric pollutants from East Asia to Japan and to analyze the transformation processes occurring during the transport, an aerial observation campaign called LEXTRA (Lagrangian EXperiment on long-range TRansported Aerosols) was carried out in March–April 2008 over the East China Sea. On 28 March in particular a very clear layered structure of the air mass was observed that could be clearly explained by the CFORS (Chemical weather FORecast System) computer simulation model. Therefore, we report here the LEXTRA observation results obtained on 28 March 2008. Results obtained on other days will be reported elsewhere.

Oxidative aging of sulfur and nitrogen during long-range transport is a major problem, particularly as it contributes to the formation of acid rain. Therefore, we also analyzed

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the non-sea-salt (nss)-[SO_4^{2-}]/([SO_2] + nss-[SO_4^{2-}]) ratio along the flight track as an indicator of oxidative aging. In addition, we analyzed chlorine loss, which can be an indicator of acid formation during the transport of a polluted air mass, to aid in the elucidation of processes yielding acidic substances in air.

EXPERIMENTAL METHODS

Observation Flights and Aircraft

Aerial sampling was carried out from an air base in Fukue (32.75°N, 128.68°E) (Nagasaki Prefecture), where a ground-based observation station for atmospheric research is also located. Aerial observations were carried out between Fukue and Cape Hedo (26.87°N, 127.02°E) (Okinawa Prefecture) on 28 and 31 March and on 1 and 2 April 2008 (the data from 31 March and 1 and 2 April are referred to only briefly in the current paper). Round-trip level flights were made from Fukue to Cape Hedo (Fig. 1), where another ground-based atmospheric observation station (Cape Hedo Atmosphere and Aerosol Monitoring Station; CHAAMS) (Takiguchi *et al*., 2008) is located. The southbound flight was at 500 m above sea level and northbound flight at 2000 m above sea level. The vertical distribution of gases and aerosols above Cape Hedo were observed by making circular flights at altitudes of 500, 1000, 1500, 2000, and 3000 m above sea level. Vertical distribution flights at 500, 1000, 2000, and 3000 m above sea level were also made between Okinawa and Yoron Island and the aircraft flew straight for 15 min at each altitude from east to west or from west to east.

The aircraft employed was a Beechcraft Kingair 200T chartered from Diamond Air Service, Inc. (Nagoya, Japan). Four air-intake inlets were installed on top of the aircraft. Two 1" OD (1" = 2.54 cm, approx.) forward inlets were installed for aerosol sampling, and one 1/4" forward inlet was installed for monitoring all the gases except for NO_y. The air stream was introduced into a glass manifold to reduce the line velocity, and the air sample was supplied to O_3, SO_2, and CO analyzers from this manifold. A 1/4" backward inlet was installed for monitoring NO_y.

Species Observed

Gaseous species: Ozone was monitored with a UV absorption ozone analyzer (Model 49, Thermo Environmental

Instrument). The precision of the instrument is ± 1.0 ppb, and the instrument was calibrated in the laboratory before and after the experiment with a commercial ozone calibrator (Model 49PS, Themo Environmental Instrument).

SO_2 was monitored with a UV pulse fluorescence SO_2 analyzer (Model 43S, Thermo Environmental Instrument). The precision of the instrument is ± 1.0 ppb, and the instrument was calibrated in the laboratory before and after the experiment with a standard gas of 1 ppm (Taiyo Nippon Sanso) after being diluted to 10 ppb with the use of a calibrator (Model 102, Themo Environmental Instrument).

Total nitrogen oxides (NO_y) were measured with an ozone-chemiluminescence NO–NO_x analyzer (Model 42i, Thermo Environmental Instrument; modified for NO_y measurements, Yamamoto *et al*., 2001). In this analyzer, a molybdenum converter operated at 530°C converts all nitrogen oxides to NO. In order to avoid the adsorption of NO_y species, such as gaseous nitric acid, onto the surface of the tubing, the converter was installed on the ceiling of the aircraft, just after the backward air inlet on the ceiling of the aircraft, and heated to 530°C. The main uncertainty of the NO_y concentration arises from the NO calibration of the O_3 chemiluminescent detector (± 10%) (Sadanaga *et al*., 2008; Yuba *et al*., 2010). The precision of the NO_y analyzer was estimated to be within ± 2% (1σ) (Sadanaga *et al*., 2008).

CO was measured using a CO analyzer (Model 48C, Thermo Environmental Instruments). Since the background level of the CO analyzer signal changes according to water content and temperature, zero air generated by heated Pt converter (Model 96, Thermo Environmental Instruments) was measured periodically. In every 20 minutes, zero air was measured for 5 min and ambient air was measured for the other 15 min. The CO analyzer was calibrated using CO standard gas (CO 1.8 ppm, Taiyo Nippon Sanso) before and after the flights. Accuracy is estimated to be 97%, and precision is estimated to be 95%.

Particulate species: TSP samples were collected with a high-volume tape sampler (Model-191 HVTAS, Kimoto, Osaka, Japan) using a Teflon roll filter (Poreflon Membrane filter, Sumitomo Electric Industry, 100 mm × 10 m, pore size 5.00 μm). During the level flights, 9–10 samples were collected during 15–20 min of sampling at each altitude. During the flights for vertical distribution observations, one sample was obtained at each altitude during a sampling period of about 15 min. Particle mass concentration and particle number concentration were monitored with a simple particle mass monitor (Sibata GT-321; estimating aerosol mass on the basis of optical particle counting) and an optical particle counter (Sibata GT-521).

Temperature and relative humidity were also monitored on board the aircraft. The sensors for the thermometer and hygrometer (Vaisala HMT333) were installed in a shallow pit on the bottom of the aircraft.

Aerosol Chemical Analyses

Teflon membrane filters used for collecting aerosol samples were cut into two pieces for the analyses of ionic and metallic species. For the analyses of ionic species, the first piece of each filter was extracted for 60 min by supersonic

Fig. 1. Observation area and a typical flight track during the LEXTRA aerial observations over the East China Sea.

extraction with ultrapure water in a 5-mL PET test tube. After the extraction and filtration, the extracted sample was first analyzed for anions (NO_2^-, NO_3^-, SO_4^{2-}, Cl^-, and Br^-) with an ion chromatograph (Hitachi D-7000) equipped with an autosuppressor (Alltech ERIS 1000 HP) and an electrical conductivity detector (Hitachi L-7470).

After the anion analyses, the remaining extract was analyzed for cations with a polarized Zeeman atomic absorption spectrophotometer (Hitachi Z-2010). Ca^{2+} was measured in flame atomic absorption mode, and Na^+ and Mg^{2+} were measured in graphite furnace mode.

For the analyses of metallic elements, an inductively coupled plasma–mass spectrometer (Agilent 7500) was employed after extraction of the second filter piece with hydrofluoric acid. The measured elements were Li, Na, Mg, Al, K, Ca, V, Cr, Mn, Fe, Co, Cu, Zn, Ga, As, Se, Rb, Sr, Ag, Cd, In, Sb, Cs, Ba, Hg, Tl, and Pb.

CFORS and Back-trajectory Analyses

The CFORS model (http://cfors.riam.kyushu-u.ac.jp/~cfors/index.html) was developed by Uno *et al.* (2003), and the simulations during the experiment were carried out by the National Institute for Environmental Studies, Japan. The flight track was chosen based on the CFORS prediction of air mass transport during the week before the observation. After the observation, the transport and distribution of pollutants such as SO_2, SO_4^{2-}, and dust were reanalyzed for comparison with the observed data based on NCEP (National Centers for Environmental Prediction) reanalysis data.

Back-trajectory analyses were carried out with the HYSPLIT4 model from the U.S. National Oceanic and Atmospheric Administration (NOAA) (Draxler and Rolph, 2003). The initial altitude and calculation time were set to the observation altitude and 72 h, respectively.

RESULTS AND DISCUSSION

Southbound Flight
Horizontal Distribution of Gaseous and Particulate Species at 500 m above Sea Level

The concentrations of most gases and the particle number and mass concentrations increased during the southbound flight from Fukue to Okinawa at 500 m (Fig. 2).
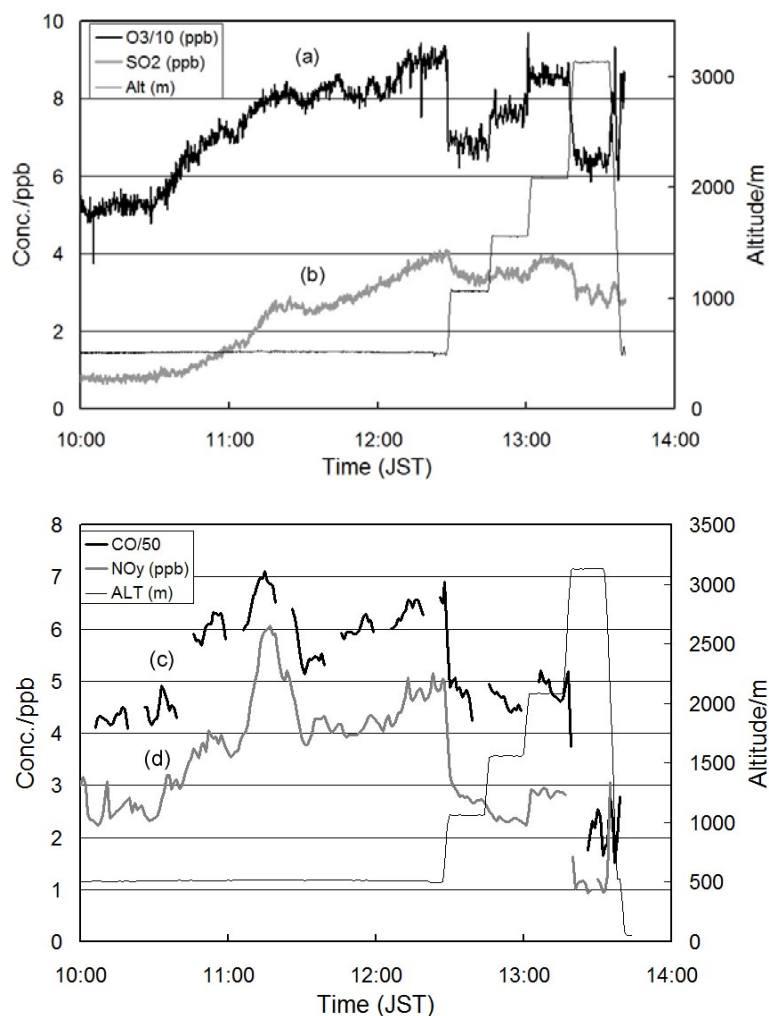

Fig. 2. Distributions of (a) O_3, (b) SO_2, (c) CO, and (d) NO_y during the southbound flight and vertical distribution flight on 28 March 2008.

Moreover, clear, high-concentration peaks were observed at around 11:15 Japan Standard Time (JST) (~30°N, 128°E) in ozone, SO_2, CO, NO_y, and particles. Fig. 3 shows the distribution of sulfate forecasted by CFORS over the East China Sea at noon (JST) on 28 March 2009. A high-concentration belt of pollutants or dust extending from the Shanghai area across northern Okinawa to southern Honshu Island, Japan, is clearly visible. This high-concentration belt of atmospheric pollutants is situated approximately along latitude 28°N. This situation was brought about by an air mass containing a high concentration of pollutants being pulled northeastward out of the Shanghai area to the area south of the Korean Peninsula by a migrating low-pressure system. The horn-shaped air mass thus formed was then pushed southward by the southward movement of the low-pressure front. This horn-shaped air mass movement is one of the weather patterns that transport high concentrations of atmospheric pollutants from the Asian continent (Uno *et al.*, 1998).

The CFORS model results also show the vertical distribution of pollutants. A north–south cross section of the air mass simulated by CFORS (Fig. 4) shows a gradual increase in the concentration of SO_2 from 32°N to 28°N along longitude 128.5°E. This model result is consistent with our observed results. Moreover, the high concentration of SO_2 reaches over 2000 m at 28°N (Fig. 4). As discussed above, since this high-concentration belt was at first driven by the movement of a low-pressure system, up drafted SO_2 remained within the belt at high altitudes. The back-trajectory analysis also show that the air mass containing high concentrations of pollutants passed over the Shanghai area in China (Fig. 5).

Oxidative Aging of Sulfur Oxides along the Flight Track

During the long-range transport of the air mass, compounds in the air mass became oxidized. Fig. 6 shows the change in the ratio of the concentration of nss-sulfate to the sum of the concentrations of SO_2 and nss-sulfate (nss-$[SO_4]/([SO_2] + nss-[SO_4])$). This ratio shows the extent of oxidation of sulfur oxides.

This ratio can be used for a simple estimation of air mass age. The aircraft passed point A (32.0°N, 127.1°E) at 10:30 JST and point B (27.0°N, 128.3°E) at 12:30 JST at 500 m above sea level.

The nss-$[SO_4]/([SO_2]+nss-[SO_4])$ ratio at those two points was 0.25 and 0.50, respectively. The oxidation rate of SO_2 has been reported as $3.7 \pm 1.9\%$ 1/h in summer and $1 \pm 0.5\%$ 1/h in winter (Calvert and Stockwell, 1984). Based on this, we assumed a reaction rate constant of 0.02 1/h for SO_2 oxidation in air, and further assumed that the only sulfur oxide initially present was SO_2, enabling us to roughly estimate that the age of the air mass at point A was 14 h and at point B it was 35 h. Since the aircraft took 2 h in our experiment to fly from point A to point B, the difference in the air mass age between the two points is 19 h. Back-trajectory analyses by the NOAA HYSPLIT model give similar transport times for air masses arriving at point A at 10:30 and at point B at 12:30 after departing the Chinese coast.

Sulfate Formation and Chlorine Loss

As described in section 3.1.2, sulfur dioxide underwent oxidation during the passage of the air mass from north to south (Fig. 7). The concentration of nss-sulfate increased as the airplane traveled southward at the altitude of 500 m, and the concentration of sea-salt sulfate was lower at altitudes above 500 m. These results suggest that SO_2 oxidized to form sulfate and sulfuric acid. The formation of sulfuric acid can be explained as follows. A pronounced chlorine loss took place in the air mass during transport. As the Mg/Na ratio was close to that of seawater, the aerosols sampled at 500 m contained sea-salt particles, but the Cl/Na ratio was very low (average, 0.039, versus 1.79 in seawater; Millelo, 1996) (Table 1). Therefore, only 1.2–2.0% of chlorine remained in the sea-salt particles.

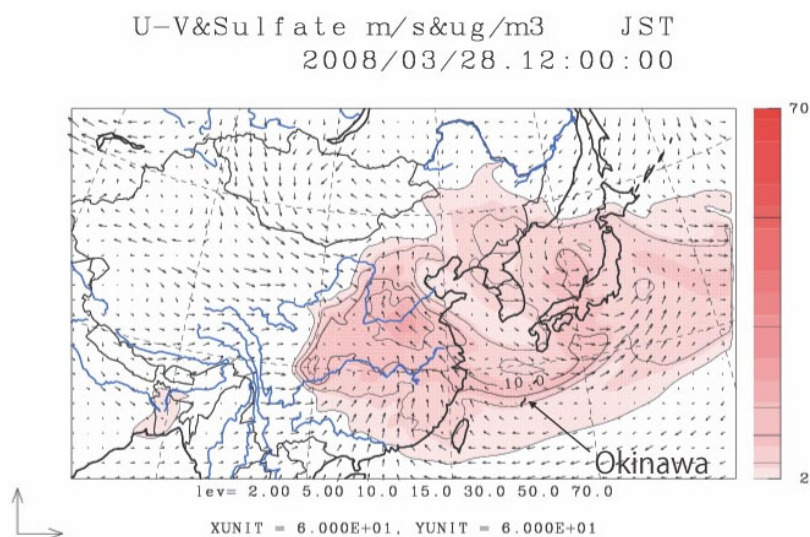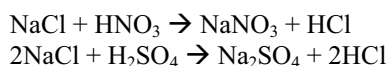The chlorine loss reaction usually takes place as follows.

$$NaCl + HNO_3 \rightarrow NaNO_3 + HCl$$
$$2NaCl + H_2SO_4 \rightarrow Na_2SO_4 + 2HCl$$

U-V&Sulfate m/s&ug/m3   JST
2008/03/28.12:00:00

lev= 2.00 5.00 10.0 15.0 30.0 50.0 70.0

XUNIT = 6.000E+01, YUNIT = 6.000E+01

Fig. 3. Simulated distribution of sulfate (contours) at noon (JST) on 28 March 2008, calculated by the CFORS model.
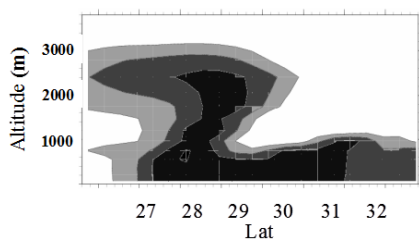
Fig. 4. North–south cross section of the spatial distribution of SO$_2$ calculated by the CFORS model. Black: > 2 ppb, dark gray: 1–2 ppb, gray: 0.5–1 ppb, white: < 0.5 ppb.

Nitric acid is usually well neutralized with calcium compounds within the air mass transported long range over the East China Sea (Hatakeyama, 2004). Therefore, the contribution of sulfuric acid, which is formed by the oxidation of SO$_2$ during the transport, should become larger.

The Cl/Na ratio was negatively correlated with the nss-sulfate concentration (Fig. 8), but a similar clear negative correlation was not found between Cl/Na and the nitrate concentration (not shown). Arakaki *et al.* (2006) reported a similar negative correlation between Cl/Na and SO$_4$ in Okinawa, Japan, and they concluded that the observed
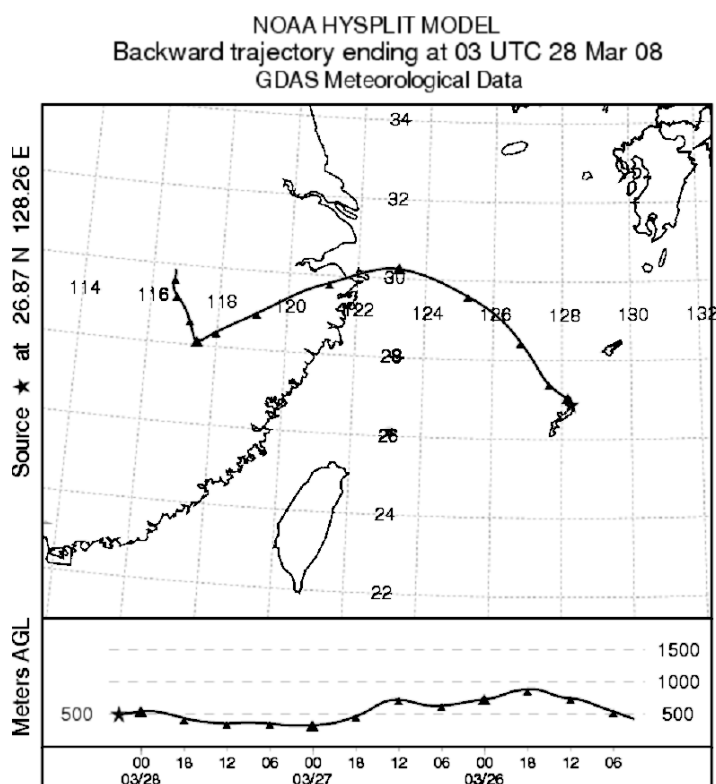
Fig. 5. Back-trajectory of the air mass arriving at 500 m above Cape Hedo, Okinawa at 3:00 UTC (12:00 JST). Top panel, map view; bottom panel, vertical cross section.
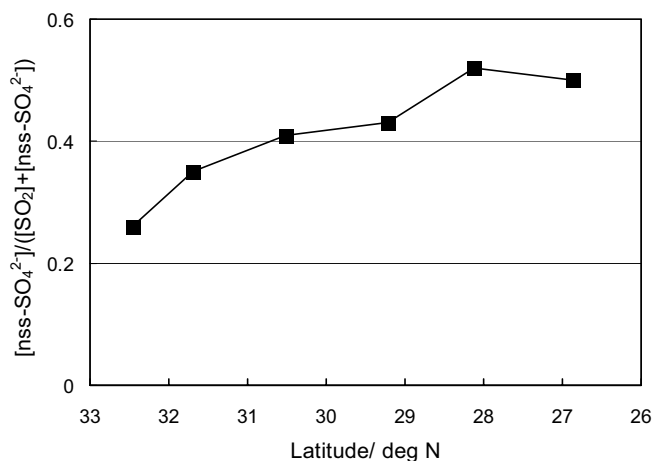

Fig. 6. Changes in the ratio of the concentration of nss-sulfate to the sum of the concentrations of SO$_2$ and nss-sulfate with latitude.
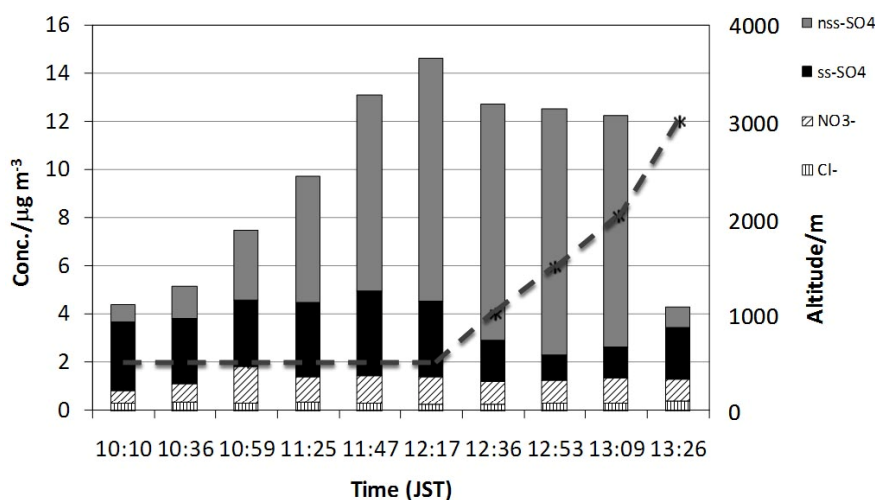
Fig. 7. Concentrations of nss-sulfate, ss-sulfate, and nitrate contained in the aerosols. The dashed line shows the measurement altitude.

Table 1. Cl^-/Na^+ and Mg^{2+}/Na^+ ratios in aerosols collected on board the aircraft on 28 March 2008.

Sample No.	Cl^-/Na^+	Mg^{2+}/Na^+
0328F-1	0.028	0.123
0328F-2	0.035	0.062
0328F-3	0.029	0.122
0328F-4	0.029	0.1
0328F-5	0.022	0.116
0328F-6	0.022	0.116
0328F-7	0.039	0.071
0328F-8	0.075	0.076
0328F-9	0.062	0.088
0328F-10	0.051	0.058
Mean	0.039	0.093
SD	0.018	0.026
Sea water ratio	1.79	0.12

chlorine loss was due to the reaction of sea salt with sulfuric acid. However, the lowest Cl/Na ratio that they observed was 0.26, which is about ten times the ratio found in this study. On the days other than 28 March, the Cl/Na ratios were much higher, ranging from 0.17 to 0.73. Therefore, the air mass observed on 28 March was unusual. Although nitric acid may have played some role in the chlorine loss, a reaction between sea salt and sulfuric acid was likely the main cause of the chlorine loss from sea-salt particles on 28 March.

Vertical Structure above Cape Hedo, Okinawa

The vertical distribution of ozone above Cape Hedo, Okinawa, is shown in Fig. 9. The concentration of ozone was high at 500 m, low at 1000 m and 1500 m, high again at 2000 m, and low again at 3000 m. Many of the other species, particularly those of anthropogenic origin, showed a similar vertical distribution (data not shown). This distribution indicates a very clear structure, which is quite consistent with the vertical profile predicted by CFORS (see Fig. 4, at about 27°N).
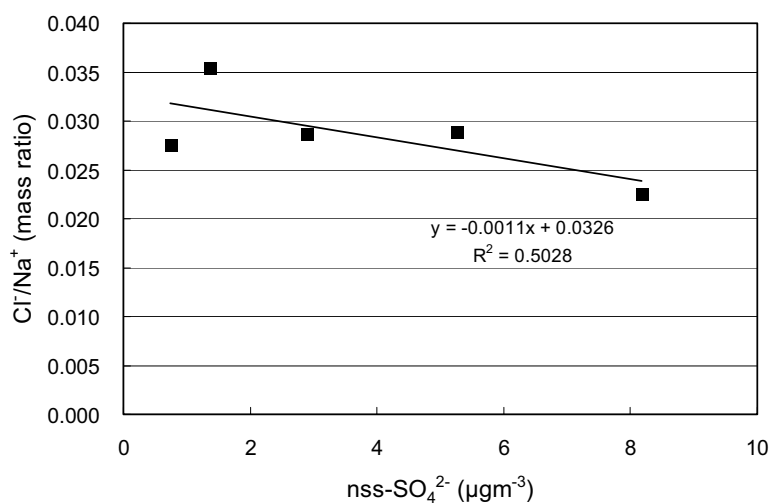

Fig. 8. Correlation between the Cl/Na ratio and the nss-sulfate concentration in aerosols collected during the northbound flight at 500 m above sea level on 28 March 2008.
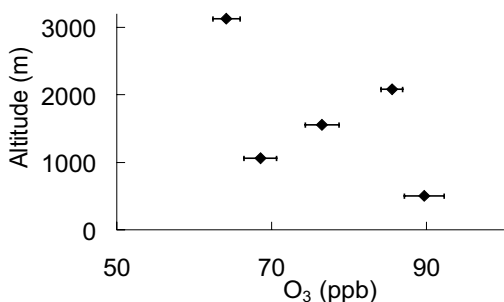
Fig. 9. Vertical distribution of O_3 above Cape Hedo, Okinawa, on 28 March 2008. Error bars show 1σ of O_3 concentrations measured during the circular flight at each altitude.

Back-trajectory analyses (Figs. 5 and 10) suggest that the air mass component at each altitude came from a different area. The air mass at 500 m passed near Shanghai 27 h before arriving at the observation point at the altitude of 650 m (Fig. 5). Therefore, this air mass brought pollutants emitted in eastern China, one of the largest emission source areas in China.

The air mass at 1000 m had circulated above the East China Sea for more than 72 h (Fig. 10(a)), and thus it was relatively clean because it had remained above the ocean for a long time.

The air mass at 2000 m had passed south of Shanghai 28 h previously at the altitude of 2000 m (Fig. 10(b)), where it presumably acquired anthropogenic pollutants.
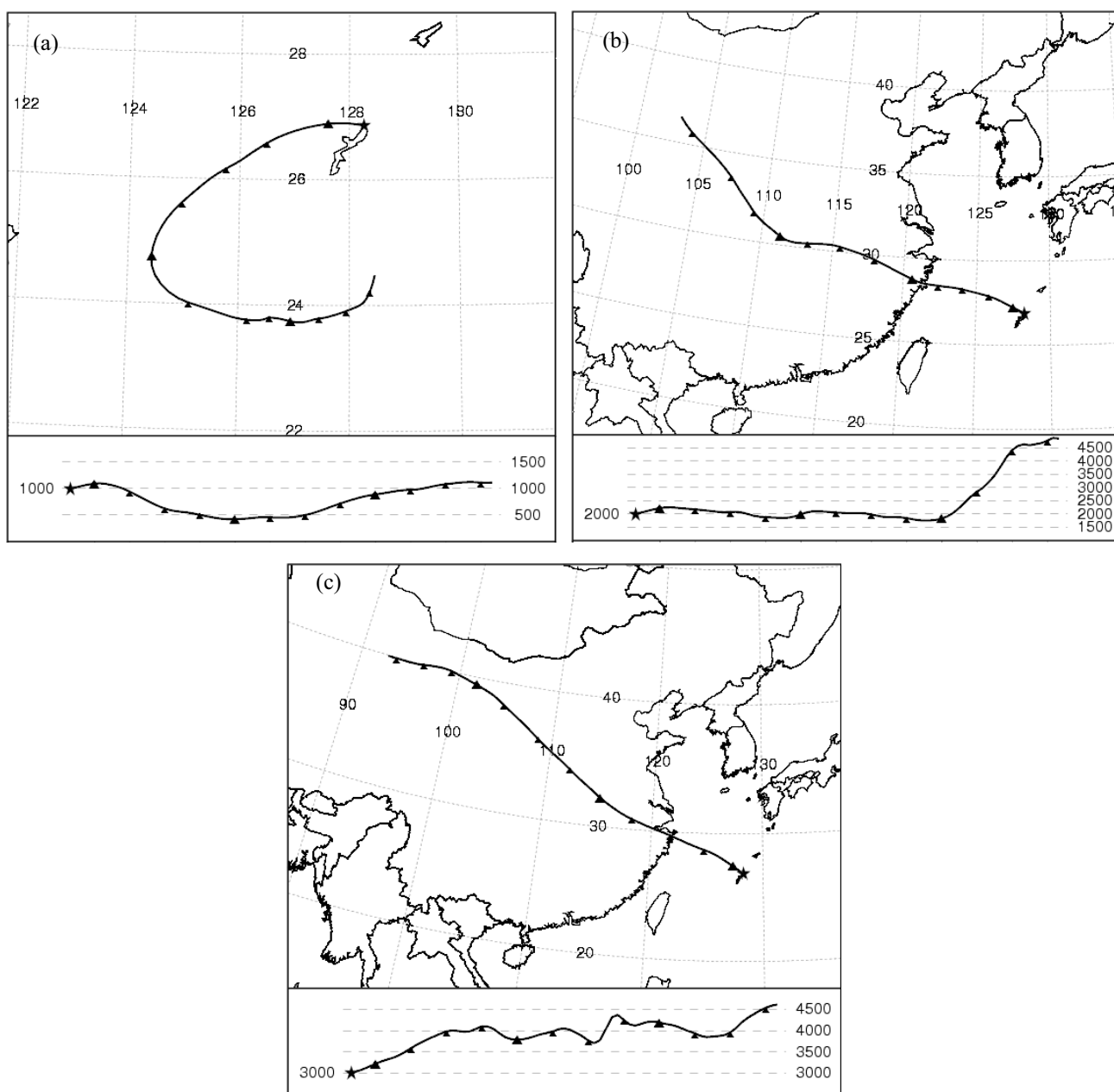

Fig. 10. Back-trajectories of air masses arriving at (a) 1000 m, (b) 2000 m, and (c) 3000 m above Cape Hedo, Okinawa, at around 12:00 JST on 28 March 2008.

The air mass at 3000 m originated in arid areas of western China at high altitude (~4500 m) and moved quickly southeastward (Fig. 10(c)), and thus it probably contained dust particles. This possibility is supported by the particulate results. At 3000 m the mass concentration of coarse particles (PM$_{10}$) was higher compared with those at

2000 m whereas that of fine particles (PM$_{2.5}$) was lower (Fig. 11). The Ca^{2+} concentration also increased at 3000 m, indicating that dust particles were transported above 3000 m (data not shown). The model simulation results also support this hypothesis. As shown in Fig. 12, the concentration of particles at around 3000 m increased on 27–28 March. Thus,
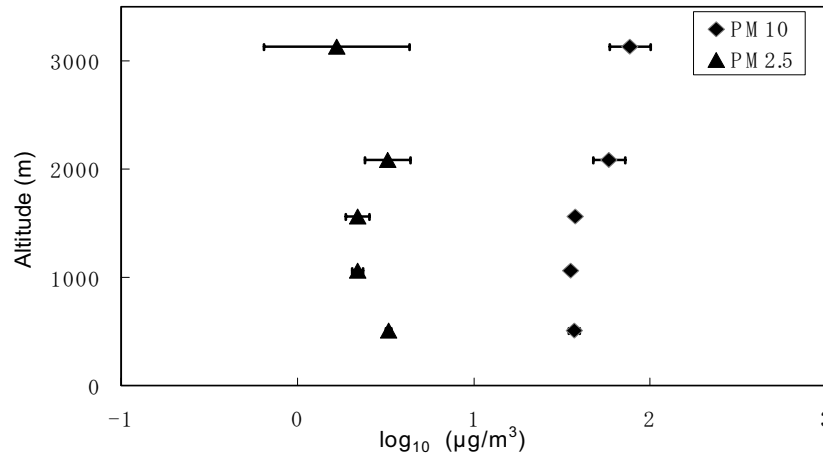

Fig. 11. Vertical distributions of PM$_{10}$ and PM$_{2.5}$ above Cape Hedo, Okinawa, on 28 March 2008. Error bars show 1σ of PM concentrations measured during the circular flight at each altitude.
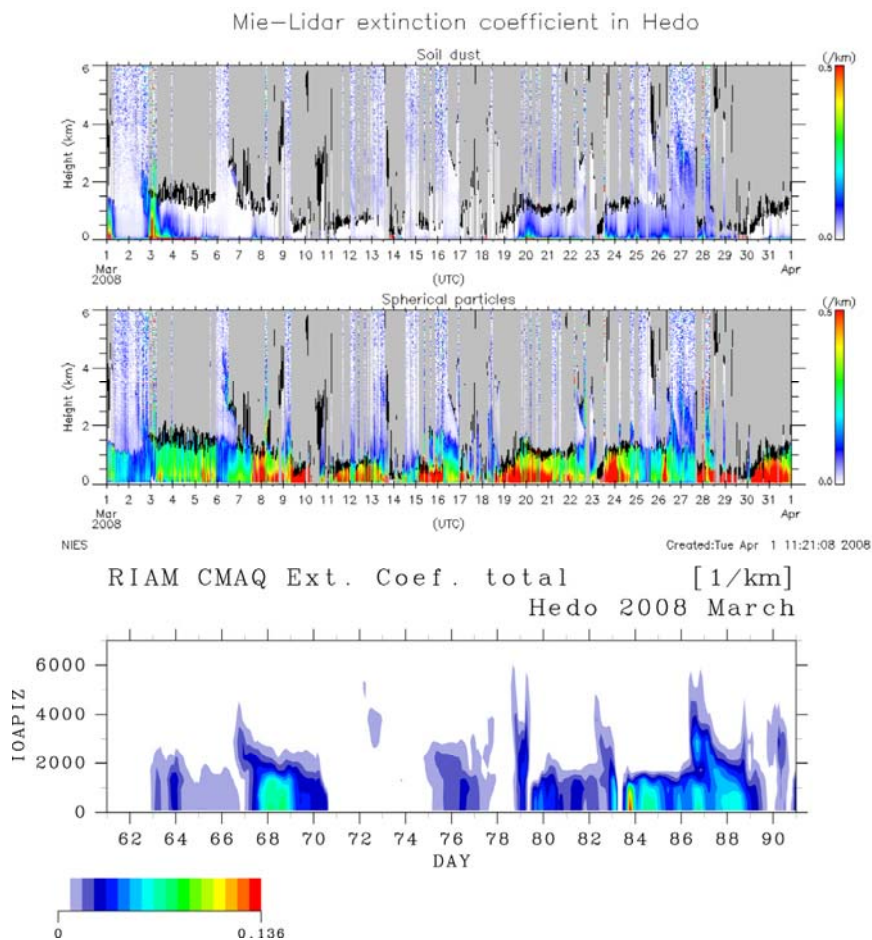

Fig. 12. Lidar observation (top and middle panels) and model analysis data (bottom panel) of extinction coefficients above Cape Hedo in March 2008 (28 March = Julian day 88).

we infer that dust particles transported from the Asian continent were present at 3000 m. In addition, the sea-salt sulfate (ss-sulfate) concentration at 3000 m was high, as shown in Fig. 7, although total concentration of ions was lower than those at other altitudes. That seems curious, because sea salt does not go up so high (King and Mather, 1976). This should be the result of the high concentration of Na^+ contained in the particles collected at 3000 m. As argued above, the particles at 3000 m were mostly transported from arid areas different from those at other altitudes. Na was reported to exist in TSP during dust storms (Zhao *et al.*, 2011) as well as in PM_1 in a semi-arid area (Shen *et al.*, 2009). Particles evolved in desert areas contain halite (Okada and Kai, 2004). Even at the sea level in Jeju Island, Korea chloride concentration in coarse particles is much higher during a dust event than during a high-particle-mass event without dust (Stone *et al.*, 2011), which indicates the presence of halite in Asian dust particles. It is thus possible that the concentration of ss-sulfate could be overestimated to some extent.

Northbound Flight

The return trip from Okinawa to Fukue on 28 March was a level flight at 2000 m above sea level. Concentrations of gaseous species, such as O_3 and CO (Fig. 13(a)), showed quite similar peaks between 29.5°N and 31°N. Even the fine structure of the peak was similar among the different species, indicating that a fine structure existed in the horizontal distribution of pollutants. Although nss-SO_4 and elemental Pb (one of markers for anthropogenic elements; e.g. Zhang *et al.*, 2010) showed peaks at the same location (Fig. 13(b)), NO_3 and elemental Al did not peak at that location (not shown). These results clearly suggest the presence of an air mass rich in anthropogenic pollutants at that location. Back-trajectory analysis indicated that the air mass had passed over the Shanghai–Hangzhou area at a rather lower altitude (~1000 m) 24–30 h previously (Fig. 14).

The horizontal distributions of PM_1, $PM_{2.5}$, PM_7, and PM_{10} showed slightly different peak locations between fine (PM_1 and $PM_{2.5}$) and coarse (PM_7 and PM_{10}) particles. Fine particles showed a peak in the south (~30°N), and coarse particles in

the north (~32°N) (data not shown). Anthropogenic species, such as nss-SO_4 and Pb, decreased very drastically after peaking (Fig. 13). In contrast, nitrate and Ca^{2+} showed neither clear peaks nor drastic decreases (not shown). This result can be explained by nitrate being contained in coarse particles and sulfate in fine one, during long-range transport (Takiguchi *et al.*, 2008). Uematsu *et al.* (2002) reported that air masses containing dust push Chinese coastal air masses rich in anthropogenic pollutants out to sea. It is thus reasonable to assume that anthropogenic pollutants should arrive at the observation area earlier and be found in the southern part, whereas dust-containing air should arrive later and be found in the north.

CONCLUSION

Large-scale transboundary air pollution was found on 28 March by aerial observation of air pollutants, including aerosols. A layered fine structure of the air mass was clearly observed in the vertical distribution of pollutants. Anthropogenic pollutants and dust-containing air masses showed separate horizontal distributions. Our data suggested that SO_2 became oxidized during transport, resulting in the production of sulfuric acid. A pronounced chlorine loss also took place; most of the chlorine in sea salt was released (only 1.2–2.0% remained), probably because the sulfuric acid produced reacted with NaCl.

The air mass observed on 28 March 2008 over the East China Sea was rich in anthropogenic pollutants transported from East Asia, and photochemical oxidation reactions took place within the air mass during its transport.

In this aerial observation the air mass transported by a horn-type transport (Uno, 1998) was found to form a very fine vertical and horizontal structure, and this information can be used to further refine chemical transport models to simulate the long-range transport of atmospheric pollutants.
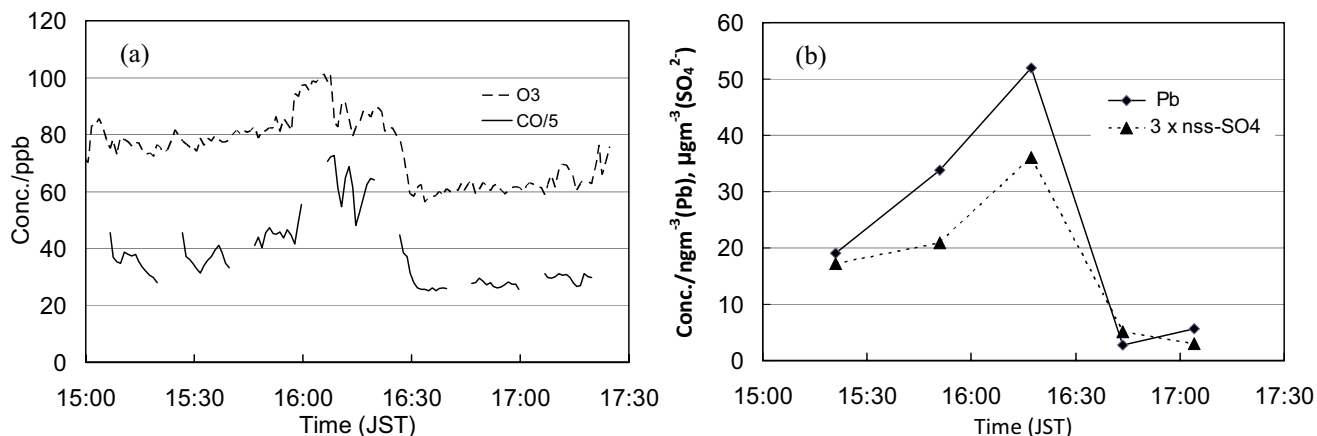
ACKNOWLEDGMENTS

Fig. 13. Variations in (a) gases and BC as well as (b) the aerosol chemical components nss-sulfate and elemental Pb during the northbound flight at 2000 m above sea level on 28 March 2008.
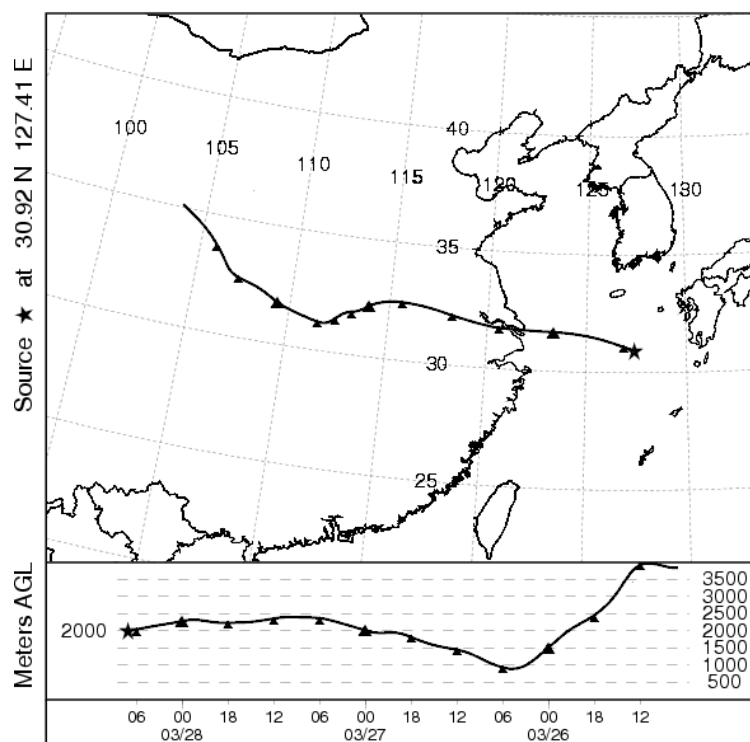
Fig. 14. Back trajectory of the air mass arriving at 2000 m, 30.9°N, 127.4°E, at 16:00 JST on 28 March 2008.

REFERENCES

Arakaki, T., Kuroki, Y., Okada, K., Nakama, Y., Ikota, H., Kinjo, M., Higuchi, T., Uehara, M. and Tanahara, A. (2006). Chemical Composition and Photochemical Formation of Hydroxyl Radicals in Aqueous Extracts of Aerosol Particles Collected in Okinawa. Japan. *Atmos. Environ.* 40: 4764–4774.

Calvert, J.G. and Stockwell, W.R. (1984). Mechanism and Rates of the Gas-Phase Oxidations of Sulfur Dioxide and Nitrogen Oxides in the Atmosphere, In *SO₂, NO and NO₂ Oxidation Mechanisms: Atmospheric Considerations*, Calvert, J.G. (Eds.), Butterworth, Boston, p. 54.

Draxler, R.R. and Rolph, G.D. (2003), HYSPLIT (Hybrid Single-Particles Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://www.arl. noaa.gov/ready/hysplit4.html), NOAA Air Resources Laboratory, Silver Spring, MD.

Hatakeyama, S., Murano, K., Bandow, H., Mukai, H., and Akimoto, H. (1995a). High Concentration of SO₂ Observed over the Sea of Japan. *Terr. Atmos. Ocean. Sci.* 6: 403–408.

Hatakeyama, S., Murano, K., Bandow, H., Sakamaki, F., Yamato, M., Tanaka, S. and Akimoto, H. (1995b). 91 PEACAMPOT Aircraft Observation of Ozone, NO$_x$, and SO₂ over the East China Sea, the Yellow Sea, and the Sea of Japan. *J. Geophys. Res.* 100: 23143–23151.

Hatakeyama, S., Murano, K., Mukai, H., Sakamaki, F., Bandow, H., Watanabe, I., Yamato, M., Tanaka, S. and Akimoto, H. (1997). SO₂ and Sulfate Aerosols over the Seas between Japan and the Asian Continent. *Earozoru Kenkyu* (*J. Aerosol Res. Jpn.)* 12: 91–95.

Hatakeyama, S., Murano, K., Sakamaki, F., Mukai, H. Bandow, H. and Komazaki, Y. (2001). Transport of Atmospheric Pollutants from East Asia. *Water Air Soil Pollut.* 130: 373–378.

Hatakeyama, S., Takami, A., Sakamaki, F., Mukai, H., Sugimoto, N., Shimizu, A. and Bandow, H. (2004). Aerial Measurement of Air Pollutants and Aerosols during March 20–22, 2001, over the East China Sea. *J. Geophys. Res.* 109: D13304, doi: 10.1029/2003JD004271.

Hatakeyama, S., Takami, A., Wang, W. and Tang, D. (2005). Aerial Observation of Air Ppollutants and Aerosols over Bo Hai, China. *Atmos. Environ.* 39: 5893–5898.

Huebert, B. J., Bates, T., Russell, P.B., Shi, G., Kim, Y.J., Kawamura, K., Carmichael, G. and Nakajima, T. (2003). An Overview of ACE-Asia: Strategies for Quantifying the Relationships between Asian Aerosols and their Climatic Impacts, *J. Geophys. Res.* 108: 8633, doi: 10.1029/2003JD003550. And Papers Compiled in the Collective Volume for "ACE-Asia: Characterization of Asian Aerosol and their Radiative Impacts on Climate."

King, W.D. and Mather, C.T. (1976). The Spatial Distribution of Salt Particles at Cloud Levels in Central Queensland. *Tellus* 28: 11–23.

Millelo, F.J. (1996). *Chemical Oceanography*, CRC Press, Boca Raton, Tokyo.

Nakajima, T., Yoon, S., Ramanathan, V., Shi, G., Takemura, T., Higurashi, A., Takamura, T., Aoki, K., Sohn, B., Kim, S., Tsuruta, H., Sugimoto, N., Shimizu, A., Tanimoto, H., Sawa, Y., Lin, N., Lee, C., Goto, D. and Schutgens, N. (2007). Overview of the Atmospheric Brown Cloud East Asian Regional Experiment 2005 and a Study of the Aerosol Direct Radiative Forcing in East Asia. *J. Geophys. Res.* 112: D24S91, doi: 10.1029/2007JD009009. And Papers Compiled in the Collective Volume for "Asian Aerosol Studies from the First Phase Regional Experiments of Atmospheric Brown Cloud (ABC) Project."

Okada, K. and Kai, K. (2004). Atmospheric Mineral Particles Collected at Qira in the Taklamakan Desert, China. *Atmos. Environ*. 38: 6927–6935

Parrish, D.D., Kondo, Y., Cooper, O.R., Brock, C.A., Jaffe, D.A., Trainer, M., Ogawa, T., Hübler, G. and Fehsenfeld, F.C. (2004). Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) and Pacific Exploration of Asian Continental Emission (PEACE) Experiments: An Overview of the 2002 Winter and Spring Intensives, *J. Geophys. Res.* 109: D23S01, doi: 10.1029/2004JD004980. And Papers Compiled in the Collective Volume for "Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) and Pacific Exploration of Asian Continental Emissions (PEACE) Experiments."

Sadanaga, Y., Yuba, A., Kawakami, J., Takenaka, N., Yamamoto, M. and Bandow, H. (2008). A Gaseous Nitric Acid Analyzer for the Remote Atmosphere Based on the Scrubber Difference/NO-Ozone Chemiluminescence Method. *Anal. Sci.* 24: 967–971.

Shen, Z., Cao, J., Tong, Z., Liu, S., Reddy, L.S.S., Han, Y., Zhang, T. and Zhou, J. (2009). Chemical Characteristics of Submicron Particles in Winter in Xi'an. *Aerosol Air Qual. Res*. 9: 80–93.

Stone, E.A., Yoon, S.C. and Schauer, J.J. (2010). Chemical Characterization of Fine and Coarse Particles in Gosan, Korea during Springtime Dust Events. *Aerosol Air Qual. Res*. 11" 31–43.

Takiguchi, Y., Takami, A., Sadanaga, Y., Lun, X., Shimizu, A., Matsui, I., Sugimoto, N., Wang, W., Bandow, H. and Hatakeyama, S. (2008). Transport and Transformation of Total Reactive Nitrogen over the East China Sea. *J. Geophys. Res.* 113: D10306, doi: 10.1029/2007JD009462.

Uematsu, M., Yoshikawa, A., Muraki, H., Arao, K. and Uno, I. (2002). Transport of Mineral and Anthropogenic Aerosols during a Kosa Event over East Asia, *J. Geophys. Res*. 107: 4059, doi: 10.1029/2001JD000333.

UNECE (2007). Hemispheric Transport of Air Pollution 2007 (Air Pollution Studies No. 16), United Nations Publication, New York.

Uno, I., Carmichael, G.R., Streets, D.G., Tang, Y., Yienger, J.J., Satake, S., Wang, Z., Woo, J.H., Guttikunda, S., Uematsu, M., Matsumoto, K., Tanimoto, H., Yoshioka, K. and Iida, T. (2003). Regional Chemical Weather Forecasting System CFORS: Model Descriptions and Analysis of Surface Observations at Japanese Island Stations during the ACE-Asia Experiment. *J. Geophys. Res.* 108: 8668, doi: 10.1029/2002JD002845.

Uno, I., Ohara, T. and Murano, K. (1998). Simulated Acidic Aerosol Long-range Transport and Deposition over East Asia—Role of Synoptic Scale Weather Systems, In *Air Pollution Modeling and its Application,* Vol. 22, Grying, S.E. and Chaumerliac, N. (Eds.), Plenum, New York, pp. 185–193.

Wang, G., Kawamura, K., Hatakeyama, S., Takami, A., Li, H. and Wang, W. (2007). Aircraft Measurement of Organic Aerosols over China. *Environ. Sci. Technol*. 41: 3115–3120.

Wang, W., Liu, H., Yue, X., Li, H., Chen, J., Ren, L., Tang, D., Hatakeyama, S. and Takami, A. (2006). Study on Acidity and Acidic Buffering Capacity of Particulate Matter over Chinese Eastern Coastal Areas in Spring. *J. Geophys. Res.* 111: D18207, doi: 10.1029/2005JD006753.

Wang, W., Ma, J., Hatakeyama, S., Liu, X., Chen, Y., Takami, A., Ren, L. and Geng, C. (2008). Aircraft Measurements of Vertical Ultrafine Particles Profiles over Northern China Coastal Areas during Dust Storms in 2006. *Atmos. Environ.* 42: 5715–5720.

Yamamoto, M., Tamaki, M., Bandow, H. and Maeda, Y. (2001). HNO$_3$ Analyzer by Scrubber Difference and the NO-ozone Chemiluminescence Method. *Atmos. Environ.* 35: 5339–5346.

Yuba, A., Sadanaga, Y., Takami, A., Hatakeyama, S., Takenaka, N. and Bandow, H. (2010). Measurement System for Particulate Nitrate Based on the Scrubber Difference NO-O$_3$ Chemiluminescence Method in Remote Areas. *Anal. Chem.* 82: 8916–8921.

Zhang, R., Shen, Z., Chen, T., Zhang, M. and Liu, Y. (2010). The Elemental Composition of Atmospheric Particles at Beijing during Asian Dust Events in Spring 2004. *Aerosol Air Qual. Res.* 10: 67–75.

Zhao J., Zhang, F., Xu, Y., Chen, J., Yin, L., Shang, X., Xu, L. (2011). Chemical Characteristics of Particulate Matter during a Heavy Dust Episode in a Coastal City, Xiamen, 2010. *Aerosol Air Qual. Res.* 11: 299–308.