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Transport of SO_2 and aerosol over the Yellow sea

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Abstract

Aircraft measurements of air pollutants were made to investigate the characteristic features of long-range transport of sulfur compounds over the Yellow Sea for the periods of 26–27 April and 7–10 November in 1998, and 9–11 April and 19 June in 1999, together with aerosol measurements at the Taean background station in Korea. The overall mean concentrations of SO_2, O_3 and aerosol number in the boundary layer for the observation period ranged 0.1–7.4 ppb 32.1–64.1 ppb and 1.0–143.6 cm^{-3}, respectively. It was found that the air mass over the Yellow Sea had a character of both the polluted continental air and clean background air, and the sulfur transport was mainly confined in the atmospheric boundary layer. The median of SO_2 concentration within the boundary layer was about 0.1–2.2 ppb. However, on 8 November, 1998, the mean concentrations of SO_2 and aerosol number increased up to 7.4 ppb and 109.5 cm^{-3}, respectively, in the boundary layer, whereas O_3 concentration decreased remarkably. This enhanced SO_2 concentration occurred in low level westerly air stream from China to Korea. Aerosol analyses at the downstream site of Taean in Korea showed 2–3 times higher sulfate concentration than that of other sampling days, indicating a significant amount of SO_2 conversion to non sea-salt sulfate during the long-range transport.

Keywords: Long-range transport; Aircraft measurement; SO_2; Sulfate; Ozone; Aerosol

1. Introduction

Sulfur compounds are especially important in East Asia in view of the fact that energy use and anthropogenic emissions are projected to increase, and they can be transported long distance up to several thousand kilometers. Sulfate aerosol forms in the atmosphere by oxidation of SO_2 either in the gas phase and the aqueous phase of cloud or on the aerosol surface.

Aerosol sulfate has been identified as an important contributor to the scatter of sunlight, and a major component of cloud condensation nuclei in the atmosphere (Lelieveld and Heintzenberg, 1992). The present global radiative mean forcing due to anthropogenic aerosols is estimated to be between -0.3 and -3.5 W m^{-2}, which is comparable to the forcing by greenhouse gases of between 2.0 and 2.8 W m^{-2} (Houghton et al., 1996). The mean negative radiative forcing by sulfate in the Northern Hemisphere alone has been estimated at about -1.1 W m^{-2} (Charlson et al., 1991).

Aerosol formation during the long-range transport can change the distribution of rainfall, impact the fishery by changes in deposition of trace metals from acidified dust and industrial emissions, and affect adversely human health by increasing acidic fine particles downwind of major sources (IGAC, 1998).

The subject of long-range transport of pollutants is in the early stage of study in Northeast Asia (Xiao et al., 1997). However, some studies have been done. These include sulfur deposition and source–receptor relationships using the RAINS-Asia model (Carmichael and Arndt, 1996), the dependency of SO_4^{2-}/SO_2 ratio on

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the air mass trajectory (Hatakeyama et al., 1997), and the plume-like transport of SO_2 (Hayami et al., 1997). In addition, some recent progress has been made in the regional databases focusing on O_3 along with a lot of trace gas species and aerosols over the Pacific ocean (Thornton et al., 1999; Talbot et al., 1997; Fenn et al., 1999; Board et al., 1999) in the program of PEM experiments. Furthermore, there is a growing evidence that chemical reactions on mineral aerosol surfaces may be an important pathway for the sulfate formation (Zhang et al., 1994, 1999; Dentener et al., 1996). However, the lack of field observation data hampers a better understanding of the potential role of heterogeneous reactions on the aerosol surface.

The pathway of sulfur transport and transformation requires the vertical profiles of air pollutants not influenced by the local pollutant sources. For this purpose, aircraft measurements have been conducted to understand the long-range transport process of sulfur compounds over the Yellow Sea, together with aerosol sampling at the Taean background station in Korea. A special emphasis is put on the episodic case of the elevated SO_2 level and enhanced aerosol concentration that were observed on 8 November, 1998.

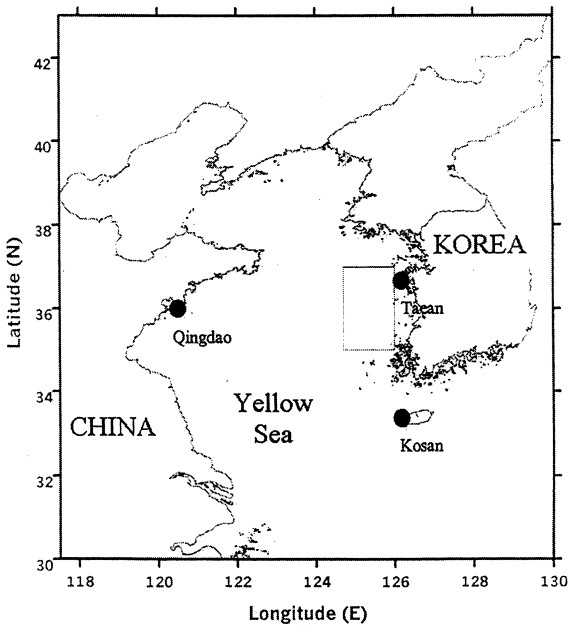

Fig. 1. Aircraft flight area (rectangle) over the Yellow Sea between China and Korea, in 1998 and 1999.

2. Measurements

A Chieftain aircraft (USA PIPER Co.) served as the measurement platform for this study. The total of 14 flights were carried out on 26–27 April and 7–10 November 1998, and 9–11 April and 19 June 1999 below 3 km above the sea level (Fig. 1). The sampling domain was chosen in the area of 124.3–126.0 E and 35.0–37.0 N so as to be located in the downstream side of the main source of China and away from the local emission source. Each flight took 2.5–3.5 h and was flown within and above the mixed layer height.

The aircraft used in this survey was a 10-manned twin propeller type whose engine capacity is 350 HP, and the dimension of $10.55 \times 3.96 \times 12.40$ m with the normal cruising speed of 370 km h^{-1}. The sampling nozzle was designed to minimize the loss of sampling compounds and attached to the bottom of the aircraft. The inverter (Avionic, 3 kw) was used to convert 12 V DC into steady 115 V AC current. Moreover, mass flow controller (MFC, Tylon) was supplemented to the sampling system to keep the flow rate constant against the change of altitude.

Sulfur dioxides were measured with Thermo-43C (Trace level) by UV fluorescence, oxides of nitrogen with Thermo-42C (Trace Level) by ozone chemiluminescene, and ozone with Thermo-49C by UV photometric method. Aerosol number concentration in 5 size bins ranging 0.3–10 μm was measured continuously using an optical particle counter (Rion, KC-01C). Analyzed gas data have been averaged over 1-min interval, but aerosol number concentration over 2-min interval. The geographical position of the aircraft was determined by GPS III pilot (Garmin) system.

SO_2 and NO_x were also routinely calibrated with the span gases. Detection limits for SO_2 and NO_x are 0.2 ppb for the case of 10-s average and 0.05 ppb for the case of 2-min average. The ozone monitor with a detection limit of 1 ppb, was also calibrated before and after each flight. All calibrations were performed through the complete Teflon inlet line. The analyzed gas and geographical location data were stored in the portable computer. The general descriptions of relevant aircraft instrumentation were shown in Table 1. Typical flight routes consisted of vertical profiles up to 3 km and horizontal transections along the longitude over the Yellow Sea.

Aerosol samplings were carried out at the Taean surface site which is located in the western tip of Korea (36°44′N, 126°08′E), 20 m above the mean sea level (Fig. 1). This site is the nearest monitoring station from the Shandong peninsula of China and from the aircraft flight area (Fig. 1). A low-volume air sampler with an impacter of a cut size of 2.5 μm was used to collect aerosol. The flow rate was 16.7 l min^{-1}, and daily sampling started about 0900 LST and lasted for 24 h during the period of the flight mission. Samples were taken predominantly during the non-precipitating periods.

Table 1
Descriptions of relevant instruments equipped in the aircraft

Parameter	Operation principal/instrument model	Range and unit	Response time	Precision
SO_2	UV Fluorescence/Thermo-43C Trace	$\leqslant$ 100 ppb	80 s (10 s avg.)	0.2 ppb (10 s avg.)
O_3	UV Photometric/Thermo-49C	$\leqslant$ 1000 ppb	20 s (0–95%)	1 ppb
NO_x	Chemiluminescence/Thermo-42C Trace	$\leqslant$ 100 ppb	60 s (10 s avg.)	0.05 ppb (2 min. avg.)
Particle number	Optical Particle Counter/Rion Co., KC-01C	$\leqslant$ 1000 cm^{-3}	2 min	0.3–10 μm (5 channel)
Position and altitude	Global Positioning System/Garmin, GPS-II	Latitude (N), longitude (E), altitude (m)	15 s	1–5 m
Temperature	Thermister/TRH-5S	− 20–80°C	1.5 s	0.2°C
Relative humidity	Hygrometer/TRH-5S	0–100%	1.5 s	0.5%

Analyzed species were ion components such as SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, K^+, Ca^{2+}, Na^+ and Mg^{2+}. Water soluble species were extracted with 20 ml of distilled and deionized water for 60 min. Concentrations of SO_4^{2-}, NO_3^- and Cl^- were determined by ion chromatography (Dionex-500). Concentrations of K^+, Ca^{2+}, Na^+ and Mg^{2+} were analyzed by atomic absorption spectroscopy (Varian, SpectraAA 800), while that of NH_4^+ was determined by indophenol colorimetric method (Perkin-Elmer, Lambda 20).

3. Results and discussion

3.1. Meterological field

The study domain can be influenced by various air masses with different pollutant levels. Heavily industrialized regions are located to the west in the Shandong peninsula and to the southwest in the Shanghai area from the sampling domain. The streamline analyses on 850 hPa for 26–27 April and 7–10 November 1998 have been done to identify the air mass origin and is shown in Fig. 2. The predominant airflow consisted of northwesterlies, followed by westerlies and southwesterlies, suggesting that most of air masses traveled from China to the Korean peninsula through the Yellow Sea. On 8 November 1998, the low-level westerly strengthened in association with the anticyclonic flow over Southern China and the cyclonic circulation over Manchuria (Fig. 2d). The episodic transport of a high-SO_2 plume was detected on this day.

The boundary layer depth was identified using the vertical profiles of potential temperature and mixing ratio of water vapor, measured by the radiosonde system (USA, AIR). The soundings at the time of 1500 LST on 7–10 November 1998 were displayed in Fig. 3. The potential temperature profiles (Fig. 3) showed that the boundary layer height ranged 1000–2000 m during this period. However, the height of the boundary layer on 8 November 1998 was about 1000 m that is the lowest value during this period, but the mixing ratio of water vapor in the boundary layer was the highest (6–8 $g\,kg^{-1}$). The wind speed in the boundary layer on that day increased to 13–15 $m\,s^{-1}$, twice stronger than the previous day with the wind direction shifting from the northwesterly to southwesterly-westerly. At the Taean site, temperature dropped by 2–5.5°C in the next day in association with the passage of a cold front with a relative humidity decrease of 30% (Korean Meteorological Agency (KMA), 1998).

3.2. General features

The mean and standard deviations of SO_2, O_3 and aerosol number concentrations in the study domain for

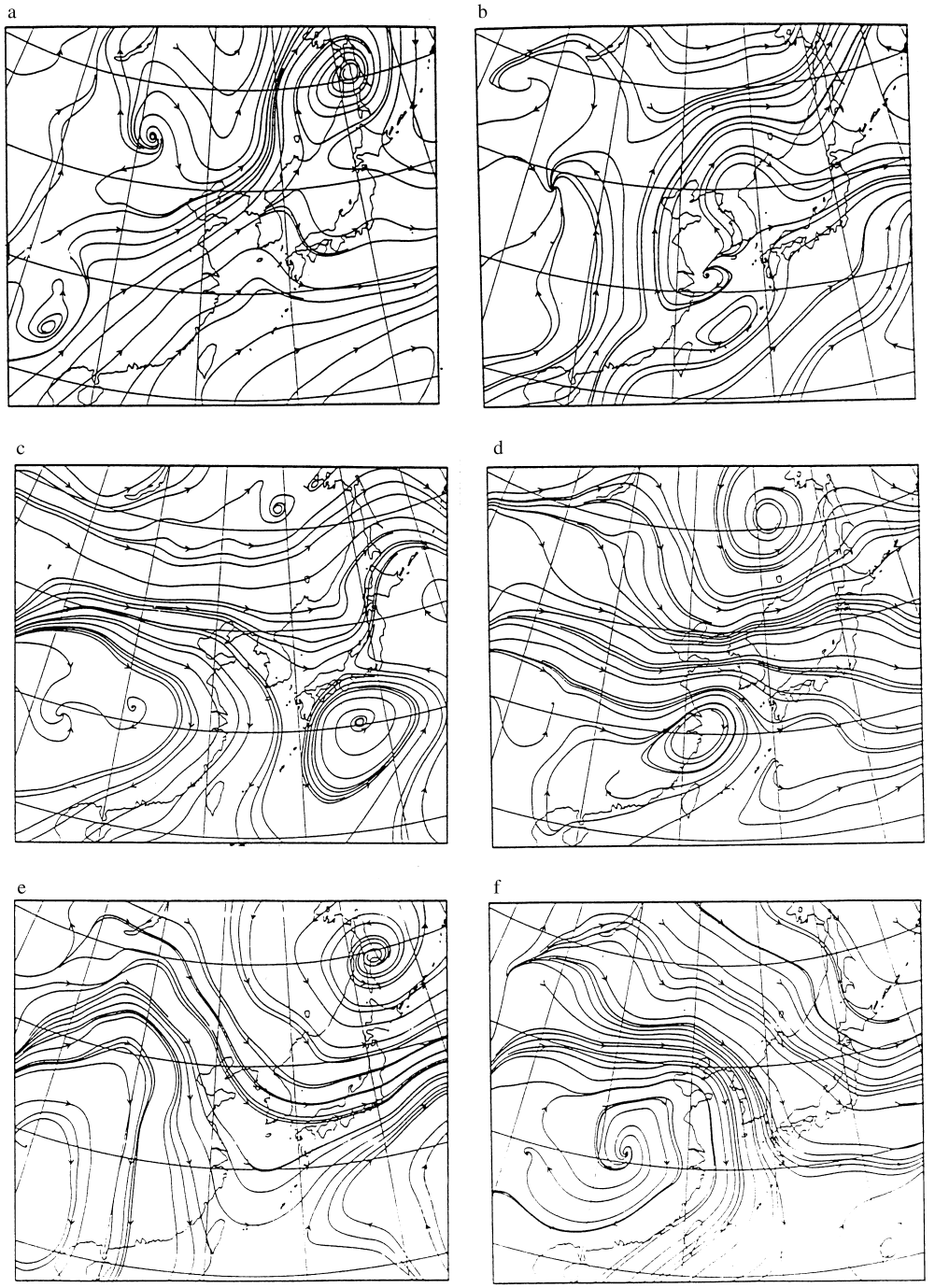

Fig. 2. The streamline analysis charts of 850 hPa on (a) 26, (b) 27 April, and (c) 7, (d) 8, (e) 9, and (f) 10 November 1998.

each aircraft measurement are given in Table 2. The average concentrations of SO_2 for each flight ranged 0.1–7.4 ppb. These values except on 9–11 April 1999 were higher than the observed mean value of 0.93 ppb over the East Sea between Korea and Japan by Hatakeyama et al. (1997). But, similar SO_2 concentration was observed at Kosan which is one of the background monitoring stations in Korea. The average concentration of SO_2 was

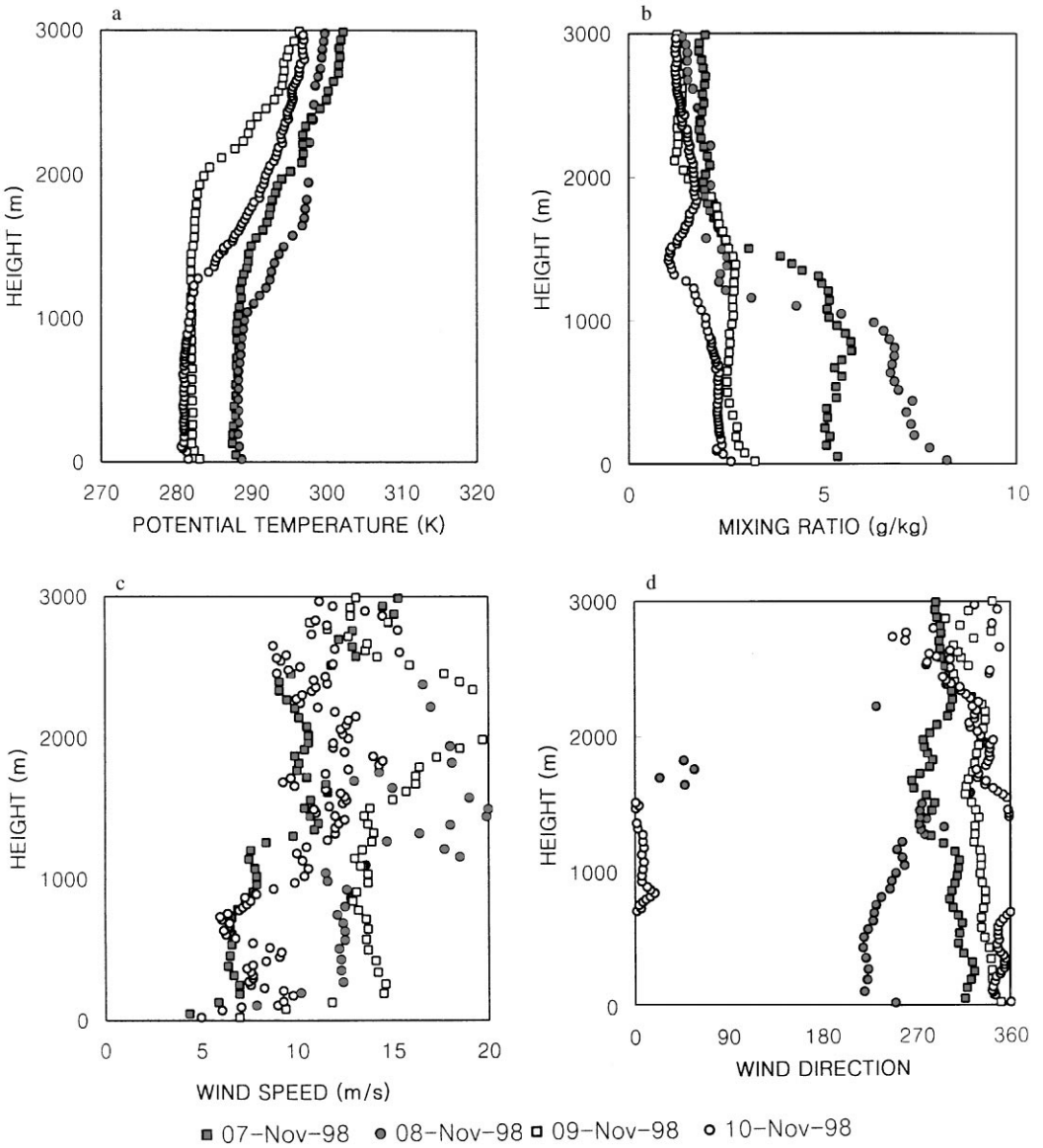

Fig. 3. Vertical profiles of (a) potential temperature, (b) mixing ratio of water vapor, (c) wind speed, and (d) wind direction for the period of 7–10 November 1998.

approximately 1 ppb during March and April 1992 at Kosan, Korea (SERI, 1993). This value is higher than the observed value of 0.2 ppb in Oki island, Japan in November 1992 (Hatakeyama, 1994). Over the remote Pacific troposphere, surface level SO_2 concentration was reported less than 30 ppt for Amsterdam Island (Putaud et al., 1992), Cape Grim (Ayers et al., 1997), and over the Southern Oceans south of Tasmania (DeBruyn et al., 1998). Some vertical profiles of SO_2 by Andreae et al. (1988) showed no clear vertical gradient with the concentrations less than 30 ppt. Therefore, it was clear that transport of SO_2 from anthropogenic activity on the northeast Asian continent was generally contributing a significant amount of SO_2 over the Yellow sea.

On 8 November 1998, a high concentration of SO_2 plume was observed with the mean concentration of 7.4 ppb. However, the SO_2 concentration significantly decreased (about 0.1 ppb) when the slowly moving cyclone passed over the Yellow Sea on 9–10 April 1999.

The O_3 concentration during the aircraft measurement period ranged 32.1–64.1 ppb. These values were comparable or slightly higher than the values reported from the mountainous sites in Japan and marine sites located at similar latitude. At Oki island in Japan, the

Table 2
Summarized results of aircraft measurement for the sampling periods[a]

Flight no.	Date	Flight time	Flight area			SO_2 (ppb)	O_3 (ppb)	Particle no. (cm^{-3})
			Latitude (N)	Longitude (E)	Altitude (m)			
1	1998.4.26	11:37–12:40	36.0–37.0	125.0–126.0	1012 (6.4)	1.0 (0.1)	61.4 (3.3)	8.1 (2.8)
2	1998.4.26	16:25–17:00	36.0–37.0	125.0–126.0	3014 (8.0)	1.1 (0.1)	55.7 (3.8)	12.5 (8.8)
3	1998.4.27	10:30–11:10	36.3–37.0	125.3	300–1200	1.3 (0.5)	58.1 (3.1)	30.4 (11.1)
4	1998.4.27	14:30–15:00	36.4–37.0	125.3	300–1200	1.4 (0.3)	63.9 (2.6)	32.7 (26.3)
5	1998.11.7	15:03–16:15	36.0–37.0	125.0–126.0	1008 (3.9)	1.1 (0.3)	53.3 (2.0)	45.5 (25.7)
6	1998.11.8	11:10–11:44	36.0–37.0	125.0	500–1000	7.4 (3.2)	42.3 (3.2)	90.5 (48.4)
	1998.11.8	11:45–12:06	35.0–36.0	125.0	2000–3000	2.3 (0.6)	52.8 (4.7)	4.5 (5.1)
7	1998.11.8	15:16–15:42	35.0–36.0	125.0	500–1000	3.5 (1.6)	32.1 (1.3)	109.5 (42.2)
	1998.11.8	15:43–16:15	36.0–37.0	125.0	2000–3000	1.3 (0.6)	52.1 (12.3)	17.1 (6.8)
8	1998.11.9	14:32–15:48	36.0–37.0	125.3	500–2000	1.3 (0.4)	48.6 (2.2)	4.0 (1.3)
9	1998.11.10	14:30–15:03	36.3–37.0	125.3	500–1200	—[b]	54.2 (1.2)	7.6 (2.2)
	1998.11.10	15:04–16:04	36.3–37.0	125.3	1500–3000	1.9 (0.5)	52.4 (2.8)	1.5 (0.8)
10	1999.4.9	10:50–11:30	35.3–37.0	124.4	500–1800	0.2 (0.05)	61.1 (4.0)	19.4 (11.5)
11	1999.4.9	15:06–15:40	35.0–36.3	124.4	1000–1800	0.2 (0.03)	51.7 (1.7)	1.4 (3.6)
12	1999.4.10	15:55–17:03	35.0–37.0	125.0–126.0	1022 (45)	0.1 (0.13)	59.8 (2.0)	11.1 (8.8)
13	1999.4.11	15:00–15:38	36.4–37.0	125.0	300–1500	0.1 (0.06)	64.1 (2.1)	0.94 (0.75)
14	1999.6.19	14:36–15:13	35.4–37.0	125.2	1019 (117)	1.1 (0.4)	63.9 (7.1)	143.6 (72.8)

[a]The values in parenthesis denote standard deviation.
[b]—, Not analyzed.

average O_3 concentration was 36.1 ppb in September and 42.2 ppb in October 1991 (Akimoto et al., 1996). High ozone concentrations (Gregory et al., 1996; Talbot et al., 1997) are found in stratospherically influenced air (>120 ppb) and in plumes originating from land sources (70–100 ppb), whereas relatively low concentrations are in clean air (40–60 ppb), and in the marine boundary layer (20–30 ppb). The observation results over the Yellow Sea are almost similar to the clean air, but higher than those of the marine boundary layer.

The aerosol number concentration varied significantly from 1.0 to 143.6 cm^{-3}. The particle size range of aerosols measured in this study was 0.3–10 μm. However, these values were larger than those (24.1–45.7 cm^{-3}) at Ogasawara Hahajima island over the northwestern Pacific ocean (Nagao et al., 1999). The number concentration of aerosols varied remarkably with height. A similar vertical distribution pattern was observed in the Southwest Pacific for the aerosol particles with the particle size of 0.5–3.12 μm (Kristament et al., 1993).

The vertical profiles of SO_2, O_3 and aerosol number concentrations are shown in Fig. 4. The SO_2 concentration decreased with height as observed over the East Sea between Korea and Japan (Hatakeyama et al., 1997). But the decreasing rate with height was much larger than that observed in clean background air. The vertical profiles of SO_2 and aerosol number concentrations (Figs. 4a and b) showed that the high concentrations were observed in the boundary layer especially on 8 November and the very low background concentrations were observed in the rest of sampling periods, suggesting that the Yellow Sea is frequently influenced by the polluted air masses passing through continental China. It is worthwhile to note that the high concentrations of SO_2 and aerosol number were mainly confined in the atmospheric boundary layer as seen in Figs. 3a, 4a and b.

The SO_2 and aerosol particle concentrations tended to decrease with height similar to that of water-vapor concentration (Figs. 3b, 4a and b). However, the O_3 concentration changed little with height (Fig. 4c). The similarity in profiles of water vapor, SO_2 and aerosols is due to their own sources on the surface and removal processes aloft (Kleinman and Daum, 1991).

3.3. The episode of high sulfur transport

The percentiles of SO_2, O_3 and aerosol number concentrations in the boundary layer of all flight periods are shown in Fig. 5. The median of SO_2 concentration for the aircraft measurement period ranged about 0.1–2.2 ppb. But, on 8 November 1998, 1-min average SO_2 concentration increased up to 12.0 ppb in the boundary layer with a significant increase of particle number concentration of almost 2.5 times higher than that of the previous day (7 November 1998). However, O_3 concentration decreased remarkably. This enhanced SO_2 concentration occurred in the low-level westerlies in association with the anticyclonic flow over Southern China and the

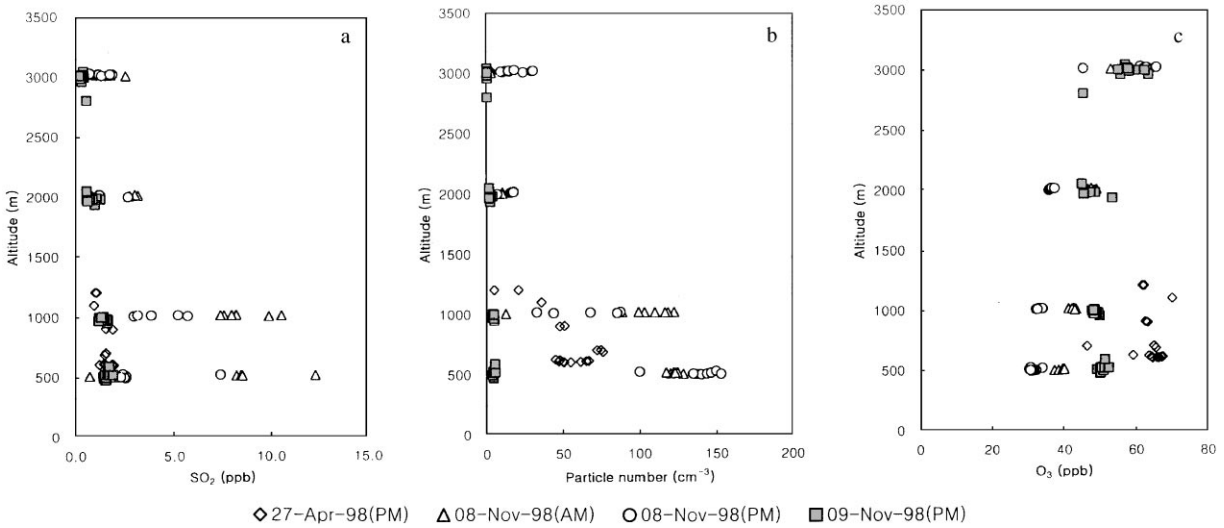

Fig. 4. Vertical profiles of (a) SO_2, (b) aerosol number, and (c) O_3 concentrations for the aircraft measurement period in 1998.

cyclonic circulation over Manchuria (Fig. 2). The yearly mean concentration of SO_2 during 1998 in Seoul, the Capital city of Korea, was 8 ppb (MOE, 1998). The above observed high concentration of SO_2 was almost comparable to that of urban atmosphere. Although, SO_2 high concentrations (0.5 ppb) from continental East Asia were encountered over the South China sea, the Sea of Japan, and off the west coast of Japan (Thornton et al., 1999), these values were more than 1 order lower than this episode over the Yellow sea.

It is worthwhile to note that the ozone concentration remarkably decreased from 52–63 to 32–41 ppb on 8 November 1998 while SO_2 and aerosol number concentrations significantly increased (Fig. 5). After this episode of high SO_2 plume transport, SO_2 and aerosol number concentrations decreased remarkably with northerly and northwesterly flow in association with the passage of a cold front.

Park (1998) showed that the Shandong peninsula had a high density of SO_2 emission, that is, the annual emission amount per 10 000 km^2 of SO_2 in 1996 was more than 0.1 Mt. This region is located just 300–500 km west of the Taean in Korea. Therefore, a favorable meteorological condition could lead to transport sulfur compounds from the Shandong region to the west coast of Korea within a day.

In summary, the evidence of long-range transport of SO_2 was observed in favorable meteorological conditions, such as the 'transport corridor' from source area to the sampling domain.

3.4. Aerosol and ozone

Once the gaseous SO_2 is emitted, about half of it decays in the process of dry deposition and the rest is oxidized in the plume or ambient atmosphere to sulfuric acid aerosols or sulfate by reactions occurring in the gas phase, in the liquid phase, and on the surfaces of solids (Finlayson-Piits and Pitts, 1986).

The analysis results of fine particle measured at the Taean background site during 25–28 April and 7–10 November 1998, and 9–11 April and 19 June 1999 are displayed in Fig. 6. The overall mean of mass concentration was 13.0 $\mu g\ m^{-3}$. Anion concentrations showed similar variation to those of cation. The non-sea-salt (nss) sulfate are estimated based on sodium ion. The overall mean nss-sulfate concentration was 2.84 $\mu g\ m^{-3}$, which was equivalent to 50% of the total water soluble ion concentration by mass. The contribution of nss-sulfate was much higher than the averaged nss-sulfate concentration measured at various places in the Pacific Ocean (0.2–1.5 $\mu g\ m^{-3}$) and that of PM2 particles at remote marine regions (about 1 $\mu g\ m^{-3}$, Kim et al., 1998). Averaged ratio of nss-sulfate to total sulfate was 0.95, indicating possible contributions of the anthropogenic sources to nss-sulfate during the long-range transport. The overall average nss-SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+} concentrations for the observation period were 2.84, 0.70, 0.53, 1.61, 0.41, 0.30, 0.02 and 0.10 $\mu g\ m^{-3}$, respectively.

Because the sampling site is located in the marine environment, the contribution to nss-sulfate concentration from the biogenic sources should be evaluated. Unfortunately, the sulfur species from the biogenic sources were not measured during the experimental period. However, during the PEM-West A measurement, methane sulfonic acid and nss-sulfate concentrations in aerosol were measured at the Kosan site between April 1992 and February 1993 (Arimoto et al., 1996). Kosan is another background site in the southern part

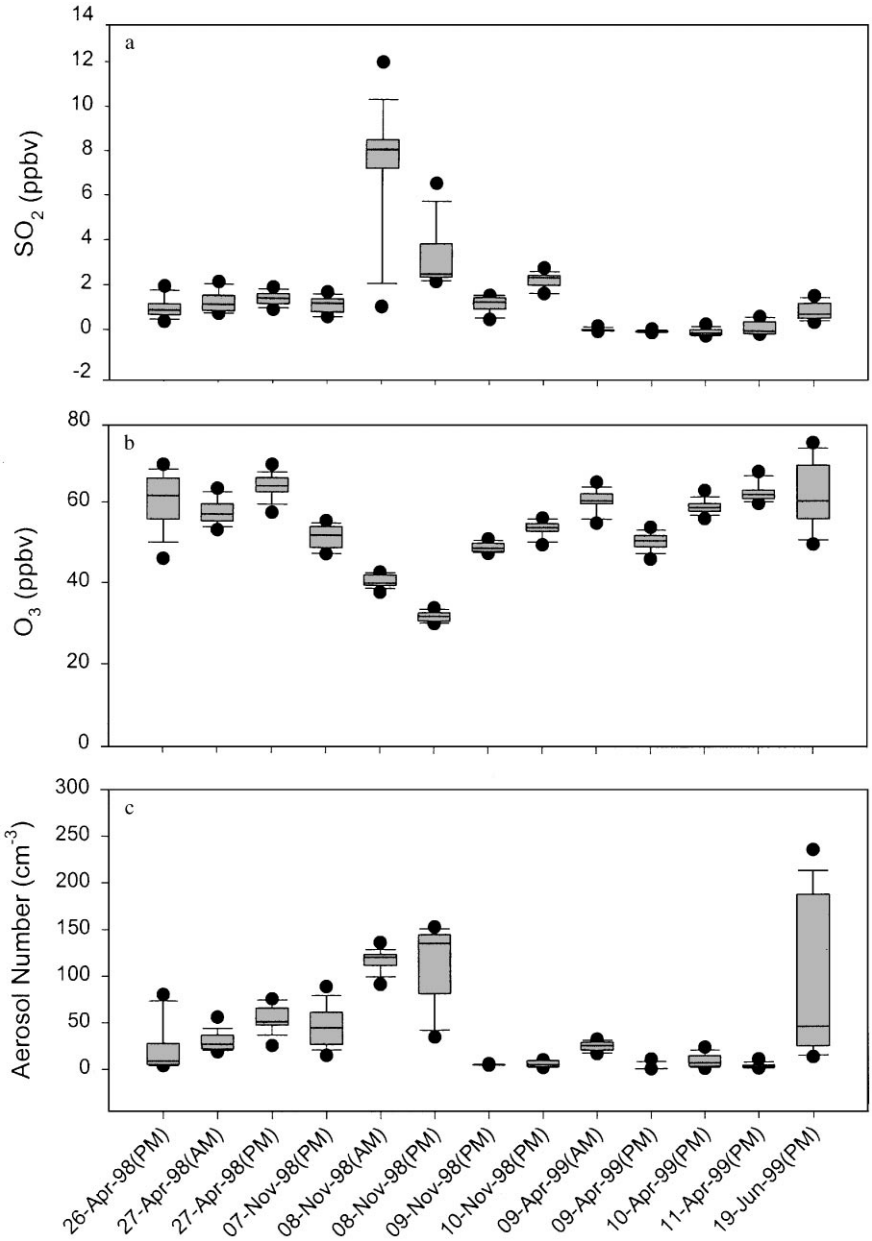

Fig. 5. The box plots of (a) SO_2, (b) O_3, and (c) particle number concentrations for all aircraft measurements from April 1998 to August 1999. The lowest dots, lower outliers, lower box lines, middle box lines, upper box lines, upper outliers, the highest outliers indicate 5, 10, 25, 50, 75, 90 and 95% percentiles, respectively.

of Yellow sea (Fig. 1). They showed that the biogenic fraction of the nss-sulfate was about 11% which is a much smaller amount compared with the remote marine area.

Nss-sulfate concentrations at Taean increased up to almost 9.0 $\mu g\,m^{-3}$ during the episode of high sulfur transport on 8 November 1998 which was 2–3 times higher than the other sampling days along with the increase of dust concentration. As previously described in Section 3.3, aerosol number concentrations from the aircraft measurements on the episodic day was also increased. The positive correlation between nss-sulfate concentration and aerosol number concentration suggests that nss-sulfate contributed to the formation of fine particles through a new particle formation of sulfuric acid vapor and a condensational growth of preexisting aerosol.

The SO_2 and dust concentrations obtained from Taean monitoring site during the aircraft measurement

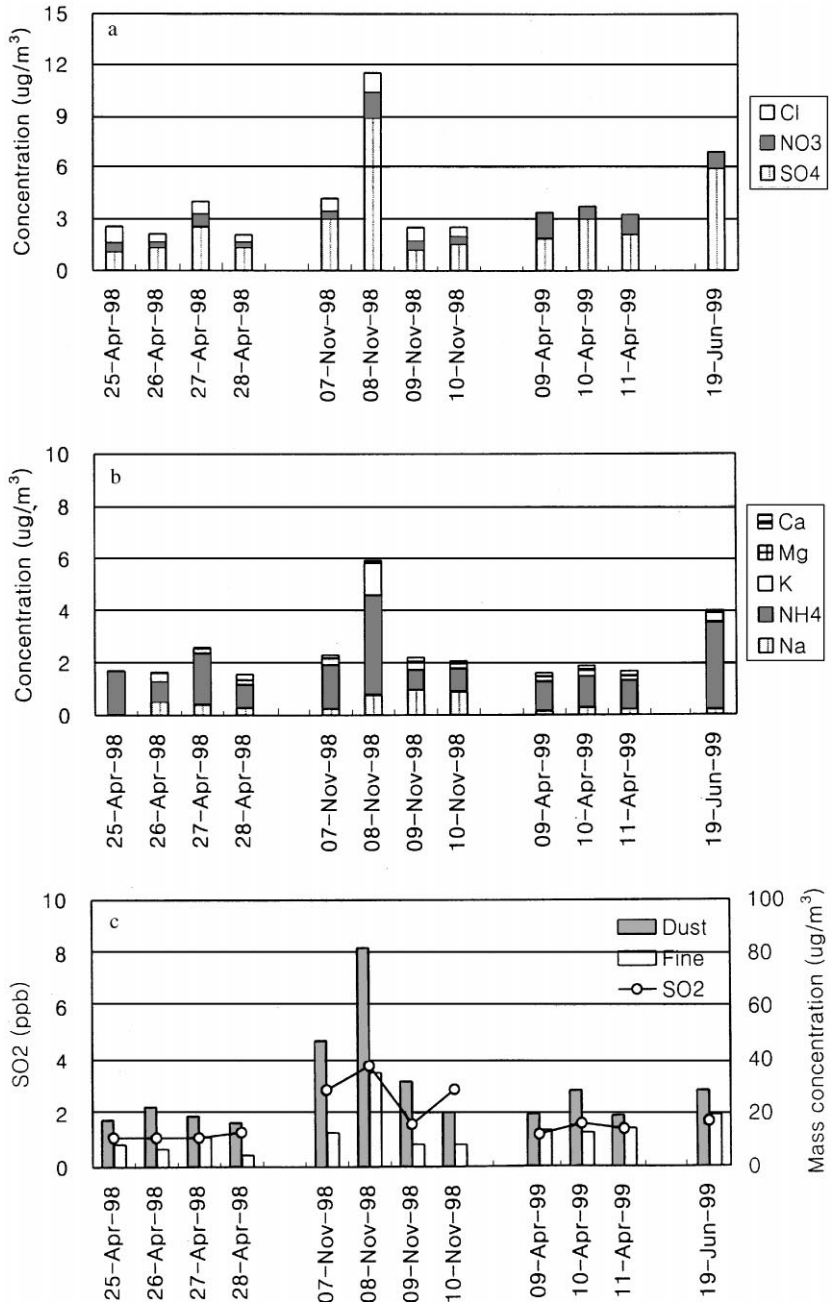

Fig. 6. Variations of daily average concentrations of (a) anions, (b) cations, and (c) SO_2, fine aerosol, and dust measured at the Taean background station during 25–28 April and 7–10 November 1998, and 9–11 April and 19 June 1999.

period are shown in Fig. 6c. O_3 was not measured during this period. Dust concentration increased up to 82.1 μg m^{-3} on 8 November, which was 2.8 times higher than the average level (29.5 μg m^{-3}), while SO_2 increased slightly to 3.8 ppb compared to the average value (1.8 ppb). O_3 concentration from the aircraft measurement remarkably decreased about 10–20 ppb (Table 2), contrary to the increase of dust concentration, implying the possible direct uptake of O_3 on the dust surface.

Similar conclusion was reached for the simultaneous measurements of dust and ozone at a height of about 2 km above the surface in Japan during May 1987

(Zhang et al., 1994). Dentener et al. (1996) suggested that direct O_3 uptake on dust may also be responsible for the tropospheric O_3 decrease. The box model for the dust–gas-phase interactions also explained that the decrease in O_3 concentration results mainly from the direct uptake of O_3 (Zhang and Carmichael, 1999).

4. Conclusion

Air pollutants were measured using an aircraft (Cheftain, PIPER Co.) to investigate the characteristic features of long-range transport of sulfur compounds over the Yellow Sea for the periods of 26–27 April and 7–10 November in 1998, and 9–11 April and 19 June in 1999, together with aerosol measurements at the Taean background station in Korea.

The overall mean concentrations of SO_2, O_3 and aerosol number in the boundary layer for the observation period ranged 0.1–7.4 ppb, 32.1–64.1 ppb and 1.0–143.6 cm^{-3}, respectively. The air mass over the Yellow Sea had a character of both the polluted continental air and clean background air, and the Yellow Sea is frequently influenced by the polluted air masses originating from continental China. The sulfur transport was mainly confined in the atmospheric boundary layer. The median of SO_2 concentration within the boundary layer was about 0.1–2.2 ppb. However, on 8 November 1998, the mean concentrations of SO_2 and aerosol number increased up to 7.4 ppb and 109.5 cm^{-3}, respectively, in the boundary layer. On the other hand, O_3 concentration decreased remarkably. This enhanced SO_2 concentration occurred in the low-level westerly air stream from China to Korea. Aerosol analyses at the downstream site of Taean in Korea showed 2–3 times higher sulfate concentration than that of other sampling days, indicating a significant amount of SO_2 conversion to nss sulfate during the long-range transport.

The present study was undertaken mainly to document an episodic case of high SO_2 and aerosol concentrations through aircraft measurement. A possible interaction between aerosol and tropospheric ozone during the long-range transport is inferred but cannot explain in detail the chemical mechanism. Further measurements of aerosols and gas-phase species including SO_2, NO_x, O_3 and VOCs are required to understand the role of aerosol in tropospheric chemistry.

Acknowledgements

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