Long-range transport of fluoride in East Asia monitored at Noto Peninsula, Japan

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ABSTRACT

Airborne particulate matter was collected at Wajima, the Noto Peninsula, Ishikawa, Japan by a high-volume air sampler with a quartz fiber filter every week from September 17, 2004 to September 16, 2005. The filter was newly changed every week. There are no major emission sources of atmospheric pollutants near the sampling site. Water-soluble fluoride anion extracted from the filters was analyzed by ion chromatography. The concentration of non-sea-salt fluoride was higher during the heating period of China (from October 15, 2004 to April 15, 2005), significantly higher during the period of Asian Dust (from the mid of April to the beginning of May in 2005), than that in the other seasons. This result suggests that the main contributor of fluoride in the period of Asian Dust was high-fluoride-concentrated soil from the arid area of China. A noticeable positive correlation ($r = 0.54$, $n = 28$, $p < 0.01$) between the level of non-sea-salt fluoride and PAHs was observed, when only the data obtained during the period of Asian Dust was excluded. In view of the fact that PAHs emitted from Northeast China were long-range transported to Japan during the heating period of China, fluoride emitted from coal combustion long-range transported from the Asian continent to Japan during the same period appears to be another main source of fluoride.

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1. Introduction

Fluorides are contained in soil and ground water. Exposure to high levels of fluoride through drinking water or air results in skeletal fluorosis etc. in humans (Thiessen, 1988; Arnesen and Krogstad, 1998; Fornasiero, 2001). Gaseous and particulate fluorides are emitted into the atmosphere from several sources, such as coal-burning systems, aluminum refining plants, phosphate fertilizer plants, glass works, ceramic industries and steel mills (Okita et al., 1974). Among them, coal-burning power plants produce 82% of the total fluoride in the air in the United States (Wu et al., 2004). China consumes the largest amount of coal in the world and generates over 75% of its energy from coal (Chen et al., 2005). Furthermore, fluoride pollution in China is exacerbated by the use of high-fluoride coal-clay (Wu et al., 2004). Fluoride is mainly derived from coal-burning in Beijing (Wang et al., 2005). Fluoride emitted into the air in the Asian continent is probably transported to Japan over the Japan Sea (Ohizumi and Fukuzaki, 1996; Kido et al., 2001).

Polycyclic aromatic hydrocarbons (PAHs), that also mainly originate from imperfect combustion of fossil fuel, are carcinogenic and/or mutagenic (Ames et al., 1975; Epstein et al., 1979). The main source of PAHs in Japanese cities is automobile emissions (Hayakawa et al., 1995; Murahashi et al., 1999), while in China it is coal-burning (Tang et al., 2005; Hattori et al., 2007).

As our previous research (Yang et al., 2007), airborne particulate matter was collected at the Wajima Air Monitoring Station, on the Noto Peninsula, Japan. This site is located in a less-populated suburban area of Wajima near the Japan Sea coast and has no nearby major sources of air pollutants. This site is meteorologically in the main path of winter northwest winds and a year-round jet stream that blow from the Asian continent to Japan. We recently reported that the concentrations of PAHs at Wajima were much higher during the heating period of China (from October 15, 2004 to April 15, 2005) than those in the other seasons (Yang et al., 2007). A principal component analysis of the PAH composition and a back trajectory analysis indicated that PAHs detected at Wajima mainly originated from Northeast China (Yang et al., 2007).

Although it is possible that some sources (e.g., coal combustion) emit both fluoride and PAHs and probable that both of them are long-range transported from China to Japan. However, there has been no report concerning the seasonal variation and origin of atmospheric fluoride at Wajima. In this study, we examined whether the atmospheric fluoride at Wajima were long-range transported from

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Fig. 1. Location of the sampling sites.

China and whether the fluoride and PAH concentrations were correlated in order to investigate the major contributor of fluoride.

2. Materials and methods

2.1. Sampling

Total suspended airborne particulate matter was collected at Wajima Air Monitoring Station (Nisifutamata-machi, Wajima City, Ishikawa Prefecture, Japan), which is located on the Noto peninsula 2.1 km south of the Japan Sea coast and about 100 km north of Kanazawa (Fig. 1). The elevation of the site is about 60 m. This station is the former National Wajima Acid Rain Monitoring Station of the Ministry of the Environmental Agency, Japan. Noto peninsula is less-populated area and no major emission sources of atmospheric pollutants are near the station. Airborne particulate matter was collected by a high-volume air sampler (AH-600, Shibata Japan) with a quartz filter (8 in. $\times$ 10 in., 2500QAT-UP, Pallflex Products, Putnam, CT, U. S. A.) at a flow rate of 700 L min⁻¹ continuously from September 17, 2004 to September 16, 2005. A new filter was installed every week. The used filter was stored in a freezer (-20°C) until pretreatment. The flow rate of the sampler was calibrated every 1 month.

2.2. Pretreatment and analysis of samples

Ten square inches of each filter was cut into small pieces. The pieces were placed in a 50 mL centrifuge tube containing 20 mL Milli-Q water and extracted ultrasonically for 40 min. After 5-min centrifugation, the supernatant was passed through membrane filters (HLC-DISK 3 Hydrophobic, pore size 0.45 μm , Cica-Reagent) for analysis. Fluoride was analyzed by an ion chromatography system (761 Compact IC, Metrohm), which consisted of an anion separation column (IC SI-90 4E, Shodex), a chemical suppressor and a conductivity detector. The response of the detector was calibrated before the analysis of collected samples. Fluoride was determined with 0.5 mL min⁻¹ of 11.0 mM NaHCO₃/0.3 mM Na₂CO₃ eluent. The limit of detection ($S/N=3$) was 0.003 mg L⁻¹ and 0.07 ng m⁻³. Sodium ion was analyzed by the same ion chromatograph using a cation separation column (Shim-pack IC-C3 (S), Shimadzu) with 0.2 mL min⁻¹ of 2 mM tartaric acid/1 mM dipicolinic acid as an eluent. The limit of detection ($S/N=3$) was 0.14 mg L⁻¹. Blank values were subtracted from sample determinations.

Filters were also analyzed with a scanning electron microscope (SEM) (HITACHI, S-3000N) coupled with an energy dispersive X-ray (EDX) analyzer (HORIBA, EMAX-500).

Fig. 2. Variations of fluoride (□) and nss-fluoride (■) at Wajima. Dates indicate the first day of a 7-day sampling period. $\times$: not detected; Δ : detected but below the detection limit.

Table 1

Average concentrations (ng m^{-3}) and standard deviations (SD) of fluoride in four seasons at Wajima and some other cities.

Sampling site	Autumn		Winter		Spring		Summer		Reference
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	
Wajima	$n=3$		$n=12$		$n=13$		$n=3$		Present work
	0.26	0.18	0.50	0.35	1.27	0.84	0.09	0.06	
Tokyo	–		<100		–		–		Saitoh et al. (2002)
Changchun	–		1076		–		437		Inoue et al. (1995)

Autumn: from Sep. to Nov.; winter: from Dec. to Feb.; spring: from Mar. to May; summer: from Jun. to Aug.

2.3. Back trajectory analysis

The sources of the air samples were predicted with the online program, Meteorological Data Explorer (METEX, <http://cgermetex.nies.go.jp/metex/>), which was developed by the National Institute for Environmental Studies (NIES), Japan. Meteorological data was obtained from the European Centre for Medium-range Weather Forecasting (ECMWF). The system calculated three-dimensional air mass trajectories (height, longitude and latitude) with a time resolution of 6 h and a horizontal resolution $2.5^\circ \times 2.5^\circ$ and vertical resolution was 15 equal levels from surface pressure to 10 hPa.

3. Results and discussion

3.1. Atmospheric concentration of fluoride at Wajima

Soluble fluoride concentrations in airborne particulate matter monitored weekly at Wajima ranged from 0.03 ng m^{-3} in summer to 3.91 ng m^{-3} in spring (130 times higher than the lowest value in summer), with a mean of 0.85 ng m^{-3} . Concentrations of fluoride in 2 autumn samples and 13 summer samples were below the limit of detection (Fig. 2). The higher level at the end of spring is probably due to Asian Dust (three weeks of April 15, 22 and 29, 2005), which will be discussed later. Except for the spring (Asian Dust) samples, the mean concentration of fluoride was the highest in winter, which was about twice the mean value of autumn (0.26 ng m^{-3}) and 6 times higher than the mean value of summer (0.09 ng m^{-3}) (Table 1). The highest fluoride concentrations were observed in Changchun (1076 ng m^{-3} in

winter, Inoue et al., 1995). This was probably because of the wide use of coal heating in north China in winter. The fluoride concentration at Changchun is about 1400 times greater than that at Wajima although fluoride concentration in Tokyo (less than 100 ng m^{-3} in winter, Saitoh et al., 2002) could not be compared with that of Wajima. The values of two other sites are also presented in Table 1 for comparison.

3.2. Non-sea-salt fluoride ion

In order to remove the contribution of sea-salt to the fluoride concentration at Wajima, the concentration of non-sea-salt fluoride (nss-F^-) was calculated by the following equation assuming that the chemical composition of sea-salt particles is the same as that of sea water, and that the soluble sodium ion in particulate samples comes solely from sea-salts (Ohizumi and Fukuzaki, 1996):

$$[\text{nss-F}^-] = [\text{F}^-] - [\text{Na}^+] \times 0.000146$$

Fluoride and nss-fluoride concentrations are compared in Fig. 2. The same trend was observed even after the contributions from sea-salt were subtracted. The percentage contributions from sea-salt were the lowest (5.7%, $n=3$) for the 3 weeks (April 15, 22 and 29) when Asian Dust was transported from the Asian continent to Japan. Except for the above period, sea-salt contributed large but not very much during the heating period of China (from October 15, 2004 to April 8, 2005; 31.8%, $n=20$). From May 6, 2005, the total concentration of fluoride greatly decreased and sea-salt became the main contributor to fluoride (64.0%, $n=6$). In order to find accurate sources of fluoride, the contribution of sea-salt to fluoride should not be ignored. Therefore the following discussion is based on only non-sea-salt values of fluoride.

3.3. Relationships between nss-fluoride, PM and PAHs

The time course of nss-fluoride is compared with those of airborne particulate matter (PM) and PAHs in Fig. 3 and in which data of PAHs are from our previous report (Yang et al., 2007). As it is shown in the figure, nss-fluoride was almost not detected or lower than the detection limit from May to August, 2005, although several samples could not be determined. The concentrations of nss-fluoride were higher from November 19, 2004 to April 29, 2005. This period approximately coincided with the heating period of China (from October 15, 2004 to April 15, 2005), as mentioned above in the part of

Fig. 4. Electron microscope analysis of particulates collected at Wajima. (A) Irregularly shaped mineral particles collected during week of April 15, 2005. (B) Pollen grains collected during week of March 11, 2005.

introduction. It should be emphasized that, from April 15 to April 29, 2005, the nss-fluoride level was extremely high. As shown in the figure, PM also showed a high level during this period, apparently because of an increase in the transport of Asian Dust. Asian Dust storm was detected at Wajima four times during this period (Japan Meteorological Agency <http://www.jma.go.jp/jp/kosa/index.html>). Furthermore, many mineral particles which showed irregular shapes were observed on the filter of week April 15, 2005 (Fig. 4A). A strong positive correlation was observed between nss-fluoride and PM (Fig. 5). However, from March 11 to April 8, 2005, the fluoride level was not so unusually high although PM level was as high as that during the above Asian Dust period. Large numbers of pollen were found during March and April, 2005 (Fig. 4B), suggesting that pollen strongly contributed to the high concentration of PM during the period from March 11 to April 8, 2005. This may be why nss-fluoride didn't increase with PM during that period.

In the week of April 15, 2005 (during the period of Asian Dust), the nss-fluoride concentration increased to its highest level, while PAHs increased only slightly. In mid April, after the end of the heating period in China, the amounts of pollutants including fluoride and PAHs sharply decreased in 3 weeks of April 15, 22 and 29 (Fig. 3). The Asian Dust didn't result in a large increase of PAHs, probably because the emission of PAHs in China had already decreased at that time. On the other hand, the level of nss-fluoride was much higher during the same period, although the emission of fluoride from coal combustion might also have decreased a lot simultaneously. The chemical and physical behaviors of PAHs are much more complicated than those of fluoride. Atmospheric reactions of PAHs, such as radical reactions and photolysis degradation, result in the half lives of PAHs that from a

few days to a few weeks (Arey et al., 1986; Hayakawa et al., 2002). Unlike PAHs, fluoride doesn't have a half-life in the atmosphere. A large amount of fluoride may adsorb to the surface of suspended dust particles during the heating period might still exist in the atmosphere and brought to Wajima by the dust storm.

When all samples are organized into a 3D scatter plot (Fig. 6) by using data of nss-fluoride, PM and PAHs, samples collected during the period of Asian Dust (group 1) are quite different from others (group 2). This indicates that the emission sources of fluoride during the Asian Dust season are different from those of other periods. A dust particle consists of many kinds of minerals; some of them show anion-exchange activity. It has been reported that sulfur dioxide emitted from China is adsorbed on the surface of dust particles as sulfate in the atmosphere over the Japan Sea (Uno et al., 1997; Ohizumi et al., 1997). Asian Dust itself is an important source of fluoride. In the arid inland areas of northwest China, the original site of Asian Dust, fluoride poisoning is a widespread and serious endemic disease (Wang and Cheng, 2000; Han et al., 2004). In those areas, natural saline soil and blown-sand soils containing abundant soluble fluoride are widely distributed and cause fluoride

Fig. 5. Correlations between the concentrations of nss-F⁻ and PM detected at Wajima ($n = 31$).

Fig. 6. 3D scatter plot of all the samples those collected in this study by using the data of nss-fluoride, PM and PAHs ($n = 31$). $\diamond$: samples obtained during the period of Asian Dust (3 weeks of April 15, 22 and 29); $\circ$: samples obtained out of the period of Asian Dust).

poisoning of ground water (Wang and Cheng, 2001). The soluble fluoride content in naturally salinized soils (sodic soils) of the arid area ranged from 5.34 to 12.48 mg kg⁻¹; from 3.0 to 13.6 mg kg⁻¹ for oasis soils, and from 7.6 to 15.2 mg kg⁻¹ for brown sand soil (Zhou and Liu, 1997; Han, 1985). Therefore it is not surprising that fluorides contained in Asian Dust particles were transported from the Asian Continent to Japan. And the dust particles themselves, originated from the arid inland areas of northwest China, were probably the main contributor of fluoride detected at Wajima during the Asian Dust season.

In order to clarify the main contributor of fluoride that detected at Wajima except the period of Asian Dust, the correlation coefficient between nss-fluoride and PAHs was calculated with all the data obtained this time just except those obtained in weeks of April 15, 22 and 29 in 2005, during the period of Asian Dust. As shown in Fig. 7, noticeable positive correlation between the level of nss-fluoride and PAH was observed. This result suggested that the major emission sources of nss-fluoride and PAHs in airborne particulate matter collected at Wajima might be identical. We recently reported that PAHs from coal emission long-range transported from the Asian continent to Japan during the heating period of China (Yang et al., 2007). Therefore, not only natural sources, fluorides exhausted from anthropic emission sources, also had been long-range transported from the Asian continent to Japan like PAHs.

3.4. Back trajectory analysis

The atmospheric pressure pattern of Northeast Asia in winter is characterized by a high pressure area in the west and a low pressure area in the east, which causes the long-range transport of air masses from the Asian Continent to Japan (Terada et al., 2002). Back trajectory analysis is usually used in atmospheric research, for simulating the air transport route (Kato et al., 2004). In this study, 72-h back trajectories of the air samples collected at Wajima were calculated at 1500 m above ground level every day during the whole sampling period (from September 15, 2004 to September 14, 2005). Most of the back trajectories of the samples from October 15, 2004 to April 14, 2005, which corresponded to the heating period of China, passed through areas of high population and industry activity in Northeast China (Fig. 8A). Also, most of the back trajectories of the samples during the period from April 15 to May 5, which corresponded to the Asian Dust season, passed through the Asian continent (Fig. 8B). On the other hand, most of the trajectories during the no-heating period didn't pass through the Asian Continent (Fig. 8C). These results suggest that the atmospheric fluoride at Wajima could be strongly affected by combustion particulates transported from the Asian Continent during the heating period of China.

Fig. 7. Correlations between the concentrations of nss-F⁻ and PAHs detected at Wajima ($n=28$) (not including the data obtained in the period of Asian Dust).

Fig. 8. Back trajectory analysis of air samples. (A) Trajectories during the heating period of China (October 15, 2004 to April 14, 2005). (B) Trajectories during the period of Asian Dust (April 15, 2004 to May 5, 2005). (C) Trajectories during the no-heating period of China (September 17 to April 14, 2004 to May 6 to September 9, 2005).

4. Conclusions

We determined the fluoride concentration in airborne particulate matter collected at Wajima, on the Noto peninsula, Japan. The atmospheric nss-fluoride concentration was higher during the heating period of China. During the period of Asian Dust in spring, the nss-fluoride concentration was especially high, suggesting that Asian Dust strongly contributed to the long-range transport of fluoride. The source appears to be fluoride-high-concentrated soil in the arid area of China. On the other hand, the level of nss-fluoride was positively correlated with PAH levels ($r = 0.54$, $n = 28$, $p < 0.01$) when the period of Asian Dust (April 15 to May 5, 2005) is excluded. We had reported that PAHs exhausted from coal combustion were long-range transported from the Asian continent to Japan during the heating period of China (from October 15, 2004 to April 15, 2005) (Yang et al., 2007). Therefore, like PAHs, nss-fluorides originated from coal combustion and long-range transported from the Asian continent is the main contributor during the heating period. Finally, back trajectory analysis indicated that the air was transported from the Asian continent over the Japan Sea in the heating period and Asian Dust season.

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