Identification of Long-range Transported Polycyclic Aromatic Hydrocarbons in Snow at Mt. Tateyama, Japan

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ABSTRACT

Snow layers, L-I to -III, collected from the wall of a snow pit at Murododaira on Mt. Tateyama, Japan, were melted and filtered into soluble (S) and particulate (P) fractions by a glass filter (pore size: 0.5 μm). The total concentration of the polycyclic aromatic hydrocarbons (PAHs) was highest in L-I. In this layer, PAHs with 5–6 rings, which exist mainly in particulate matter (PM) in the atmosphere, were almost completely in the P-fraction. PAHs with 4 rings, which exist in both the particle and the gas phase in the atmosphere, exhibited higher or equal concentrations in the S-fraction. Finally, non-sea salt SO_4^{2-} , which exists as sulfur oxides in the atmosphere, was filtered into the S-fraction. The air mass for L-I, traced via back trajectory, passed through northeastern and central China. These results suggest that PAHs transported from China to Japan precipitated in snow at Mt. Tateyama. In the melted snow, PM-associated PAHs were primarily in the P-fraction, but lower molecular-weight PAHs were partly in the S-fraction. Gas-phase PAHs were also found in the S-fraction. Furthermore, the high concentration of non-sea salt Ca^{2+} in the S-fraction of L-I implies the presence of Asian Dust.

Keywords: Snow; Long-range transported particulate matter; Polycyclic aromatic hydrocarbon; Inorganic ion; Mt. Tateyama.

INTRODUCTION

Asian Dust (yellow sand) and sulfates are long-range transported from the Asian continent to Japan over the Sea of Japan (Iwasaka *et al.*, 1988; Nishikawa *et al.*, 1991). Recently, we collected total suspended particulate matters (TSP) at Wajima Atmospheric Monitoring Site (WAMS), Kanazawa University, on the Noto Peninsula, Japan, downstream of the westerlies from the Asian continent and analyzed polycyclic aromatic hydrocarbons (PAHs) and nitropolycyclic aromatic hydrocarbons (NPAHs) in the TSP. Atmospheric PAH and NPAH concentrations at WAMS increased in winter season every year. Back trajectory and chemical compositional analyses supported that the increase of the PAH concentrations was caused by long-range transported particulate matters (PM) emitted from combustion of fossil fuels and/or biomass in China (Yang *et al.*, 2007; Hayakawa *et al.*, 2011; Tang *et al.*, 2015). As fine PM with

a diameter of not more than 2.5 μm ($\text{PM}_{2.5}$) is carcinogenic to human (IARC, 2013), the effect of long-range transported PM-associated PAHs on human health has attracted much attention.

Mt. Tateyama (3,015 m above sea level), one of the highest mountains on the west coast of Honshu, Japan, is about 110 km southeast of WAMS (Fig. 1). Murododaira, a flat area whose altitude is 2,450 m near the summit of Mt. Tateyama, is covered with deep snow every winter (Fig. 2), suggesting that snow in Mt. Tateyama could be useful for studying long-range transported atmospheric pollutants. Several pollutants such as Asian Dust and sulfur oxides in snow in this area were reported (Honoki *et al.*, 2001; Osada *et al.*, 2004; Watanabe *et al.*, 2011a).

Thermal-optical analysis measures elemental and organic carbons in PM. However, this method is not specific for PM from combustion of organic matters. Moreover, PAHs with 5 rings and more exist in PM in the atmosphere, while PAHs having 4 rings or fewer exist in PM and/or gas phase (Araki *et al.*, 2009). This suggests that the separate determination of PAHs in PM and the gas phase gives us valuable information concerning not only the long-range transport of combustion PM but also atmospheric behaviors of PAHs. Trace levels of PAHs can be quantified by several

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Fig. 1. Map of Mt. Tateyama in East Asia.

Fig. 2. (A) Murododaira and (B) the Snow Pit I, II and III are snow layers collected (Characteristics are described in Table 1).

methods including gas chromatography-mass spectrometry, liquid chromatography-mass spectrometry and high-performance liquid chromatography with fluorescence detection (HPLC-FLD) (Hayakawa *et al.*, 2017). However, to the best of our knowledge, there is not any report concerning the distribution of PAHs in snow at Mt. Tateyama. In this report, the PAHs in snow were separated into PM and soluble fractions by filtration, and PAHs and inorganic ions were determined in the fractions by the combined use of HPLC-FLD and ion-chromatography (IC) to identify PM-associated PAHs in snow at Mt. Tateyama. By using results, the atmospheric behavior of PAHs and

inorganic pollutants long-range transported over the Sea of Japan was discussed.

METHODS

Sampling and Treatment

The snow pit, 6.36 m deep, was dug at Murododaira on Mt. Tateyama in mid-April, 2015 (Fig. 2(B)). Snow blocks (each 2–3.5 kg) were collected from three layers (L-I through -III from the ground) on the inside wall and stored in cooling boxes. After returned to the laboratory at Kanazawa University, they were melted in glass beakers at

room temperature. The heights, colors, volumes and PM concentrations of the three samples are described in Table 1.

Each sample solution was filtered with a GC50 glass fiber filter (pore size: 0.5 μm , diameter: 47 mm; Advantec, Tokyo, Japan) to obtain soluble (S) and particulate (P) fractions. An aliquot (100 μL) of the filtrate (S-fraction) was injected into the IC to determine inorganic ions. The remaining filtrate was spiked with ethanol solution containing pyrene- d_{10} (Pyr- d_{10}) and benzo[*a*]pyrene- d_{12} (BaP- d_{12}) as internal standards, and then the solution was passed through an Empore C₁₈ disk (diameter: 47 mm; 3M Series, Saint Paul, USA). PAHs adsorbed on the disk were eluted in dichloromethane. After evaporating the eluate, the precipitate was dissolved in *n*-hexane. The solution was treated with a Sep-Pak Silica Cartridge (6 cc; Waters, Milford, USA) and then PAHs were eluted with acetone/*n*-hexane. After evaporating the eluate, the precipitate was dissolved in ethanol. This solution was used for determining PAHs in the S-fraction. After spiking the same internal standards as for the filtrate, the precipitate on the glass filter was extracted with benzene/ethanol. The benzene solution (the P-fraction) was washed with alkaline and acidic solutions and water, successively. After the benzene solution was evaporated, the precipitate was dissolved in ethanol. This solution was used for determining PAHs in the P-fraction.

Quantification of PAHs and Inorganic Ions

An HPLC-FLD (LC-20 Series; Shimadzu, Kyoto, Japan) was used for determining fluoranthene (FR), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), dibenz[*a,h*]anthracene (DBA), benzo[*ghi*]perylene (BghiPe) and indeno[1,2,3-*cd*]pyrene (IDP). The analytical column was an Inertsil ODS-P (4.6 i.d. $\times$ 250 mm; GL Sciences Inc., Tokyo, Japan) and the mobile phase was an acetonitrile/water mixture with a gradient mode. The flow rate of the mobile phase was 1 mL min⁻¹. The time program of FLD was set to detect at the optimum excitation and emission wavelengths for each PAH. Other conditions were the same as in our previous report (Tang *et al.*, 2017). Inorganic ions were analyzed with a 761 Compact IC equipped with a conductivity detector (Metrohm Japan; Tokyo, Japan). Lithium (Li⁺), sodium (Na⁺), ammonium (NH₄⁺), magnesium (Mg²⁺) and calcium (Ca²⁺) cations were determined with a Shodex IC YK-421 analytical column (4.0 i.d. $\times$ 250 mm; Showa Denko, Tokyo, Japan) with 5 mM tartaric acid/1 mM dipicolin acid (1:1) as an eluent. Fluoride (F⁻), chloride (Cl⁻), nitrite (NO₂⁻), nitrate (NO₃⁻), bromate (Br⁻), phosphate (PO₄³⁻) and sulfate

(SO₄²⁻) anions were determined with a SI-90 4E analytical column (4.0 i.d. $\times$ 250 mm; Showa Denko, Tokyo, Japan) with 1.8 mM Na₂CO₃/1.7 mM NaHCO₃ (1:1).

Analyses

Concentrations of non-sea-salt sulfate (nssSO₄²⁻) and calcium (nssCa²⁺) were calculated as follows:

$$\text{nssSO}_4^{2-} = \text{SO}_4^{2-} - (\text{SO}_4^{2-}/\text{Na}^+)_{\text{seawater}} \times \text{Na}^+ \quad (1)$$

$$\text{nssCa}^{2+} = \text{Ca}^{2+} - (\text{Ca}^{2+}/\text{Na}^+)_{\text{seawater}} \times \text{Na}^+ \quad (2)$$

where (SO₄²⁻/Na⁺)_{seawater} and (Ca²⁺/Na⁺)_{seawater} are the concentration ratios (equivalent ratios) of SO₄²⁻ to Na⁺ (0.12) and Ca²⁺ to Na⁺ (0.044) in seawater, respectively (Keene *et al.*, 1986).

Mt. Tateyama weather, snowfall/depth, and snowcap data were obtained from the Toyama local meteorological office, Japan Meteorological Agency (<http://www.jma-net.go.jp/toyama>). 3-day back trajectories were calculated with NOAA HYSPLIT Trajectory Model provided by National Oceanic and Atmospheric Administration (<https://ready.arl.noaa.gov/hypub-bin/trajtype.pl>).

RESULTS AND DISCUSSION

PM Concentration and Color

Murododaira typically accumulates snow from early late October to late April and snow usually melts by summer. The colors of the bottom layer (L-I), the middle layer (L-II) and the top layer (L-III) were light black, light brown and white, respectively (Fig. 2(B)). The PM concentrations were 3.4, 3.12 and 1.94 mg L⁻¹, respectively, and the fiber filters were black, black and grey, respectively. These results suggested that L-I and -II contain Asian Dust and/or combustion PM (Table 1).

PAHs

The concentrations of the 10 PAHs were highest in L-I (Table 2). The data in Tables 1 and 2, when taken together, suggest that L-I was most contaminated with dust and gas exhausted from combustion. In this layer, the PAHs distributed differently between the S- and P-fractions. The concentrations of the 4-ring PAHs (FR and Pyr) were higher in the S-fraction than in the P-fraction. Concentrations of BaA and Chr were higher in the P-fraction but comparable to those in the S-fraction. More than 75.5% of the 5-ring PAHs (BbF, BkF, BaP and DBA) and most (more than 97%) of the 6-ring PAHs (BghiPe and IDP) were in the P-fraction. This result showed that the percentage

Table 1. Snow Samples Collected from the Snow Pit.

Layer	Height from ground (m) ^a	Color (Snow/Filter) ^b	Volume (L)	PM (mg L ⁻¹)
III	5.90–6.10	White/Gray	2.95	1.94
II	5.33–5.39	Brown/Black	3.12	3.12
I	4.13–4.20	Black/Black	3.40	3.40

^a Total depth of snow pit: 6 m 36 cm.

^b Color of snow layer/color of glass filter.

Table 2. Concentrations of PAHs in the Snow.

Layer	Fraction	FR	Pyr	BaA	Chr	BbF	BkF	BaP	DBA	BghiPe	IDP
III	S	2.63	2.39	2.49	1.59	1.15	0.34	0.30	0.01	0.01	0.01
	P	1.30	0.96	0.32	0.85	1.36	0.43	0.56	0.02	0.16	0.88
	Total	3.93	3.35	2.81	2.44	2.51	0.77	0.86	0.03	0.17	0.89
II	S	1.93	2.00	0.89	0.95	0.43	0.11	0.13	0.02	0.03	0.01
	P	0.60	0.42	0.12	0.28	0.40	0.11	0.18	0.01	0.02	0.27
	Total	2.53	2.42	1.01	1.23	0.83	0.22	0.31	0.03	0.15	0.28
I	S	39.0	19.5	2.01	10.4	3.03	0.98	1.51	0.01	0.03	0.01
	P	17.2	11.0	3.29	11.9	15.4	4.70	4.65	0.06	1.12	2.82
	Total	56.2	30.5	5.30	22.3	18.4	5.68	6.16	0.07	1.15	2.83

Unit: ng L⁻¹, S: soluble fraction, P: particulate fraction. Total: S + P.

of PAHs in the P-fraction increased with molecular weight. A similar tendency was observed in L-II and -III, although concentrations of PAHs were lower. The finding that the higher-molecular weight PAHs were mainly in the P-fraction is evidence that the PM in the snow originated from combustion.

Inorganic Ions

The chromatograms revealed Na⁺, NH₄⁺, Ca²⁺, Cl⁻, NO₃⁻ and SO₄²⁻ as major inorganic ions and K⁺, Mg²⁺ and F⁻ as minor ions in the three snow layers (Table 3). The concentrations were generally highest in L-I and lowest in L-III. The exceptions were Cl⁻, which was highest in L-II and SO₄²⁻, which was lowest in L-II. Concentrations of NH₄⁺ and SO₄²⁻, which mainly originate from human activities such as fertilizers and combustion of organic matter, were very high in L-I, which suggests that L-I was contaminated with combustion gas. The concentration of Na⁺, a marker of sea salts, was highest in L-I. The concentration of nssCa²⁺, a marker of soil, which is calculated from the concentration of Na⁺ in clean ocean, was highest in L-I, suggesting that L-I contained sea salts and soils such as Asian Dust. The very high concentration of nssSO₄²⁻ in L-I provides further evidence of combustion PM in this layer.

Back Trajectory Analysis

It is not easy to define the dates of snowfall at the snow pit, because Murododaira is a remote isolated place covered with deep snow during winter season. We tried to estimate the dates by using records of weather, snowcap, and snowfall/depth of Mt. Tateyama from the Japan Meteorological Agency and the analytical results obtained above. The dates of snowfall for L-I and -II were estimated to be February 23–24 and March 15–21, 2015, respectively. 3-day back trajectories of air mass during February 23–24, 2015, passed through northeastern and central China, including deserts which tend to emit Asian Dust, and

megacities which emit combustion PM and gas (Fig. 3(A)). During this period, an Asian Dust storm arrived at Toyama, supporting the idea that the dust storm was responsible for the high concentration of PM in L-I. We collected TSP samples on the Noto Peninsula from 2004 to 2014 and determined PAHs and NPAHs. The atmospheric PAH and NPAH concentrations increased during the period from mid-October to mid-April every year and the TSP concentration often increased in the period from late winter to early spring (Yang *et al.*, 2007; Hayakawa *et al.*, 2011; Tang *et al.*, 2015). As a possible reason for the large ratio (89%) of [nssSO₄²⁻]/[SO₄²⁻] in L-I, the emission of volcanic gas near the snow pit might be considered (Watanabe *et al.*, 2016). These observations explain why the concentrations of PAHs, nssCa²⁺ and nssSO₄²⁻ were high in L-I.

The back trajectories of air masses during March 15 and 18–20, 2015, came not from China but from the Yellow Sea, East China Sea and Sea of Japan. As a result, L-II contained sea salts but not combustion PM or gas (Fig. 3(B)). This is why the air masses of this period did not contain high concentrations of PAHs.

Atmospheric Behavior of PAHs over the Sea of Japan

A triple-nested two-dimensional dynamic cloud model demonstrated that in winter season, clouds form over the Sea of Japan 50–150 km leeward of the Asian continent and gradually develop in height and convective activity. Snow clouds strengthen around 30 km off the western coasts of Honshu and Hokkaido, Japan. The dominant precipitation is snow over the mountain area along the western coasts (Murakami *et al.*, 1994), where SO₄²⁻ and Asian Dust long-range transported from China were detected in the snow (Honoki *et al.*, 2001; Osada *et al.*, 2004; Watanabe *et al.*, 2011). Snow clouds contain PM-associated PAHs and gas-phase PAHs over the Sea of Japan according to this mechanism.

The solubility of PAHs in water increases with decreasing

Table 3. Concentrations of Inorganic Ions in the Snow.

Layer	Na ⁺	NH ₄ ⁺	Ca ²⁺	nssCa ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	nssSO ₄ ²⁻
III	0.24	< LOQ	0.12	0.11	0.19	0.07	0.18	0.16
II	0.32	< LOQ	0.20	0.18	1.08	0.35	0.05	0.01
I	0.42	0.57	0.26	0.24	0.73	1.40	1.37	1.31

Unit: mg L⁻¹ in S-fraction. < LOQ: detected but lower than quantification limit.

Fig. 3. Back Trajectory Analysis (A) February 23–24, 2015 (L-I); (B) March 15–21, 2015 (L-II).

molecular weight. The ratios (%) of PAHs in the P-fraction to the total PAHs in the melted snow (closed circles in Fig. 4) decreased with the increase in octanol/water partition coefficient (K_{ow}), where K_{ow} of each PAH was obtained from the literature (Feng *et al.*, 2007). On the other hand, semi-volatile PAHs are distributed between the PM and gas phases in the atmosphere. The percentage of PAHs in the gas phase increases with decreasing molecular weight. The percentages of PM-associated PAHs in the atmosphere were calculated from the literature (Araki *et al.*, 2009) and are shown as open circles in Fig. 4. It is important to consider the distributions of PAHs not only in the atmosphere but also in snow and melted snow. PAHs with 6 rings, such as IDP and BghiPe, whose K_{ow} are 6.6 and 7.1, respectively, exist in the P-fraction in the filtrate of melted snow. However, for PAHs with 5 rings or fewer, such as BaP and Pyr, the percentages of PAHs in the P-fraction decreased as K_{ow} decreased from 6.1 to 5.2. By contrast, in the atmosphere, not only 6- and 5-ring PAHs but also 4-ring PAHs (BaA and Chr) with K_{ow} over 5.9 exist in the PM phase, but the percentages of PAHs in the PM phase were significantly smaller for the other 4-ring PAHs, such as Pyr and FR, whose K_{ow} values are both 5.2. There is a large difference in the distribution of 5-ring PAHs between the PM-phase in the atmosphere and the P-fraction in the filtrate of melted snow.

As a possible reason for this difference, the following behavior of PAHs may be considered:

where p and g are the PM and gas phases in the atmosphere,

and p and s are the P- and S-fractions in the snow solution. As shown in Fig. 5, the gas-phase PAH (PAH_g) in the atmosphere is absorbed by the snow as PM-unassociated PAH (PAH_s) (Eq. (3)). The PM-associated PAH in the atmosphere (PAH_p) is also absorbed by the falling snow. Then PAH_p moves mainly into the P-fraction (PAH_p) but partly into the S-fraction (PAH_s) in the case of PAHs with 5 rings or fewer (Eq. (4)). These results suggest that the gas-phase PAH in the atmosphere exists as the S-fraction in snow clouds, while PM-associated PAH exists as the P-fraction in snow clouds but dissociates partly to the S-fraction in the melted snow.

It is interesting that PAHs in the P-fraction were observed in all layers. At the time of snow sampling, L-III was collected as a blank snow, because this layer was uncolored. However, the concentrations of PAHs were higher in L-III than in L-II, suggesting that PAHs exist in the free atmosphere as background pollutants.

CONCLUSIONS

Three snow layers, L-I to -III, were collected from the wall of a snow pit, 6.36 m deep, at Murododaira on Mt. Tateyama, Japan. Each melted snow sample was separated into soluble (S) and particulate (P) fractions with a 0.5 μm glass fiber filter. Ten PAHs, with 4–6 rings, and the inorganic ions were determined by the combined use of HPLC-FLD and IC.

The concentrations of PAHs were highest in L-I, in which almost all of the PAHs with 5–6 rings were in the P-fraction. A similar tendency was observed in L-II and -III, although the PAH concentrations were lower. Furthermore, nsSO_4^{2-} existed only in the S-fraction. The air mass for L-I, traced via back trajectory, passed through northeastern and

Fig. 4. Distribution of PAHs vs. Vapor Pressure. Vapor pressures (Pa at 25°C) of PAHs used were cited from the report by Feng *et al.* (2007).

Fig. 5. Atmospheric Behavior of PAHs over the Japan Sea.

central China. These results indicated that PAHs, transported long-range from China to Japan, precipitated in snow on Mt. Tateyama.

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