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Technical Information

A Comparison of Particulate-Bound Polycyclic Aromatic Hydrocarbons Long-Range Transported from the Asian Continent to the Noto Peninsula and Fukue Island, Japan

Lu Yang[1),*], **Ning Tang**[1),2),3)], **Atsushi Matsuki**[1),2)], **Akinori Takami**[4)], **Shiro Hatakeyama**[5)], **Naoki Kaneyasu**[6)], **Edward G. Nagato**[2)], **Kei Sato**[4)], **Ayako Yoshino**[4)], **Kazuichi Hayakawa**[2)]

[1)]Graduate School of Natural Science and Technology, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan
[2)]Institute of Nature and Environmental Technology, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan
[3)]Institute of Medical, Pharmaceutical and Health Sciences, Kanazawa University, Kakuma-machi, Kanazawa 920-1192, Japan
[4)]National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki, 305-8506, Japan
[5)]Center for Environmental Science in Saitama, 914 Kamitanadare, Kazo, Saitama 347-0115, Japan
[6)]National Institute of Advanced Industrial Science and Technology, 16-1 Onogawa, Tsukuba, Ibaraki, 305-8569, Japan

***Corresponding author.**
Tel: +81-76-234-4942
E-mail: veronicayl@stu.kanazawa-u.ac.jp

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ABSTRACT This is the first comparative study to examine two different background monitoring sites in Japan to compare differences in the concentration and composition of long-range-transported polycyclic aromatic hydrocarbons (PAHs) from the Asian continent. This study chose the Noto Peninsula (the Kanazawa University Wajima Air Monitoring Station; WAMS) and Fukue Island (the Fukue-jima Atmosphere and Aerosol Monitoring Station; FAMS) to compare 10 weekly periods in 2009 and 2010 from autumn to spring, seasons that are strongly influenced by the Asian continent. The PAHs concentration differed significantly for most periods at these two sites. The backward trajectory analysis found that the low height of the air mass may cause the low concentration and the similar air mass condition could result in the similar concentration. The concentration of long-range-transported PAHs depended more on the source of the coal combustion areas such as Northeast China in heating period.

KEY WORDS PAHs, Long-range transportation, Backward trajectory analysis, the Noto Peninsula, Fukue Island

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs), which are ubiquitous environmental pollutants (Hayakawa, 2018, 2016), are mainly formed by the incomplete combustion of fossil fuels and biomass (Rogge *et al.*, 1993). PAHs exist in both the gaseous and particulate phases in the atmosphere. Their gaseous/particulate partitioning depends on several factors, such as the vapor pressure, temperature, concentration, and properties of dust (Araki *et al.*, 2009). Many particulate-bound PAHs are considered to cause respiratory diseases, such as asthma and bronchitis, and cardiovascular diseases, such as hypertension (EPA, 2015). Some PAHs, such as benzo[*a*]pyrene (BaP), are carcinogenic or probably carcinogenic to humans (IARC, 2014). Some previous studies have found that exposure to ambient PAHs has been associated with the cough occurrence in adult chronic cough patients (Anyenda *et al.*, 2016). Furthermore, benz[*a*]anthracene (BaA),

BaP, benzo[*b*]fluoranthene (BbF), benzo[*ghi*]perylene (BgPe), chrysene (Chr) and dibenz[*ah*]anthracene (DBA) have been linked to increased coughing and wheezing in 12-month-old babies (Miller *et al.*, 2004). PAHs research is indispensable also because PAHs, can react with biological systems to induce oxidative stress and endocrine disruption, which affect human health (Hayakawa *et al.*, 2007).

Japan is located leeward of westerly winds from the Asian continent. PAHs and other air pollutants in Japan are not only domestically produced but can also be long-range transported from locations on the Asian continent, such as China, from autumn to the following spring. The long-range transport of air pollutants like PAHs, from the Asian continent to the Sea of Japan and East China Sea has been reported (Tang *et al.*, 2015; Kaneyasu *et al.*, 2014; Yang *et al.*, 2007). In this study, two background monitoring sites on the Noto Peninsula (the Kanazawa University Wajima Air Monitoring Station; WAMS) and Fukue Island (the Fukue-jima Atmosphere and Aerosol Monitoring Station; FAMS), have been established and continuously observed since 2004 and 2009, respectively, to observe long-range-transported airborne particulates from the Asian continent over several years. These two sites are considered suitable for monitoring long-range-transported particulates from the Asian continent because they are distant from local anthropogenic pollution sources (Tang *et al.*, 2015; Kaneyasu *et al.*, 2014; Sato *et al.*, 2013).

To identify the different impacts of long-range transported PAHs on these two different regions, this study used available data from three previous studies conducted at WAMS and FAMS (Tang *et al.*, 2015; Kaneyasu *et al.*, 2014; Ogawa *et al.*, 2012). The periods monitored at the two sites overlap, resulting in the availability of PAHs data for both sites for ten weekly periods covering different seasons in 2009 and 2010. We predict that there may be differences in concentration and composition between these two sites. We hope to find the characteristics of air mass movements from the Asian continent by backward trajectory analysis to determine the reasons resulting in differences between these two sites. This is useful to establish new environmental protection policies in different regions of Japan.

2. MATERIALS AND METHODS

2.1 Monitoring Sites

As shown in Fig. 1, the monitoring site on the Noto Peninsula was WAMS (latitude 37.4°, longitude 136.8°), which has an altitude of 6 m and is located 2.1 km south of the Sea of Japan coast. The monitoring site on Fukue Island was FAMS (latitude 32.8°, longitude 128.7°),

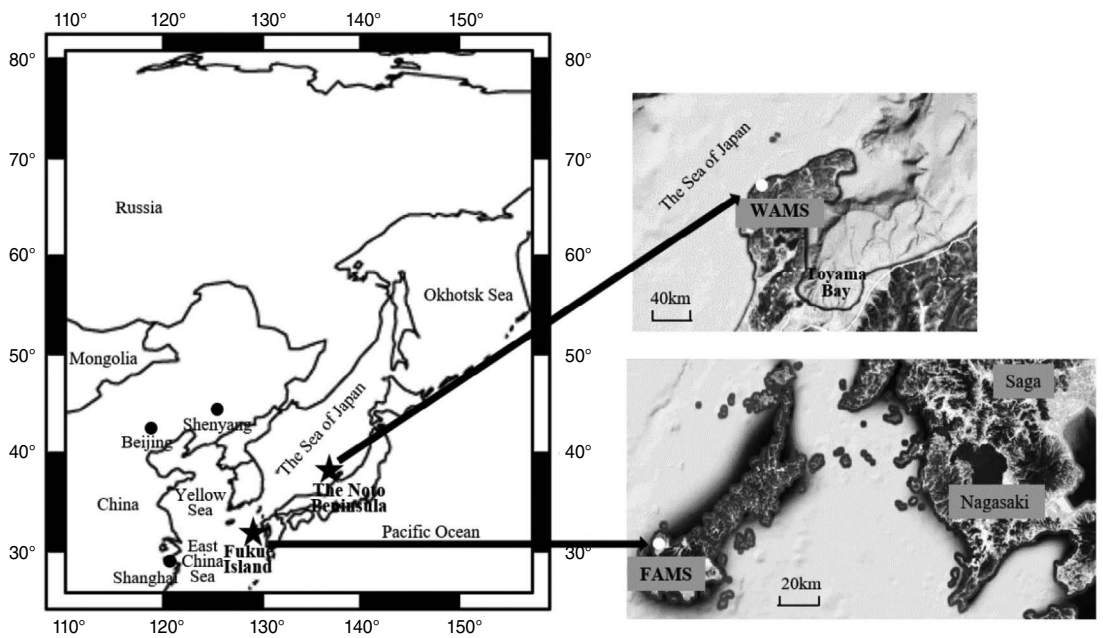

Fig. 1. Map showing the locations of the Noto Peninsula and Fukue Island.

which is mainly operated by the National Institute for Environmental Studies, has an altitude of approximately 300 m and is located 20 km west of a commercial street.

2.2 Sampling and Chemical Analysis

Particulate matters were collected using a high-volume air sampler equipped with a quartz fibre filter at both sites. Nine particulate-bound PAHs with 4 to 6 rings, including fluoranthene (Flu), pyrene (Pyr), BaA, Chr, BbF, benzo[*k*]fluoranthene (BkF), BaP, BgPe and indeno[1,2,3-cd]pyrene (IDP), all of which have been listed as target compounds by the US EPA, were analyzed by HPLC-fluorescence (WAMS) and GC-MS (FAMS). Although the analysis method was different, each method had validation for the result of PAHs. Both methods added internal standards for recovery determinations and quantifications. The mean recovery of internal standards in this study were 100 ± 20% (HPLC-fluorescence) and 100 ± 15% (GC-MS) for target compounds which both in the error range allowed in environmental chemical analysis. On the other hand, one to three field blank samples, which were treated in a similar manner to the samples but were not used for sampling, were prepared during each observation period. No PAH contamination was detected on any of these blank filters. The other detailed pre-treatment methods are given in our previous studies (Tang *et al.*, 2015; Sato *et al.*, 2013). The PAHs data for both sites for ten weekly periods covering different seasons in 2009 and 2010 are summarized in Table 1.

2.3 Data Analysis

The long-range transport of air mass containing PAHs was analyzed based on backward trajectories using the U.S. National Oceanic and Atmospheric Association's HYSPLIT4 model (HYSPLIT-WEB, HYSPLIT Trajectory Model, NOAA-ARL) with 7 paths of daily backward trajectories at a sampling point height of 1000 m (the atmosphere at this height is less affected by the ground mixing layer) and a tracking time of 72 h for each period. This study sought to explain air masses in terms of three main factors: source, velocity, and height. The air-mass velocity was determined from the path length travelled by the air mass in 72 h. The air mass directions were compared to understand why the impacts of PAHs were different between WAMS and FAMS.

3. RESULTS AND DISCUSSION

3.1 Concentration

Table 1 shows the PAHs concentration during ten periods at WAMS and FAMS. The PAHs concentration ranged from 398 to 1040 pg/m^3 at WAMS and from 582 to 2280 pg/m^3 at FAMS. The mean concentration of PAHs at FAMS (1400 ± 650 pg/m^3) was approximately two times higher than that at WAMS (645 ± 198 pg/m^3). This study conducted the significant difference test (*t*-test) for each period at the two sites, and $p < 0.05$ indicates that the PAHs concentrations in two sites differ significantly. The minimum and the maximum FAMS/WAMS ratios of PAHs were 1.00 and 4.17 in periods 3 and 9, respectively. Because it is normal for

Table 1. PAHs concentrations (pg/m^3) during ten periods at WAMS and FAMS.

Year	Period	WAMS*	FAMS*	FAMS/WAMS
2009	3/27–4/2 (period 1)	628	1550	2.47
	4/3–4/9 (period 2)	1040	1830	1.76
	4/10–4/16 (period 3)	583	582	1.00
	10/13–10/19 (period 4)	524	1690	3.23
2010	3/26–4/1 (period 5)	541	956	1.77
	4/2–4/8 (period 6)	678	1860	2.74
	4/9–4/15 (period 7)	565	625	1.11
	10/23–10/29 (period 8)	398	603	1.51
	12/7–12/13 (period 9)	546	2280	4.17
	12/14–12/20 (period 10)	948	2050	2.17
	Mean	645	1400	2.19
	Standard deviation	198	650	0.99

PAHs: Flu + Pyr + BaA + Chr + BbF + BkF + BaP + BgPe + IDP.
*t-test: $p < 0.05$.

the concentration in the same period to differ significantly between different locations, it is important to know the reason for these differences, which can help us to identify the sources and composition changes. Therefore, we chose period 3 and period 9, which had the minimum and maximum FAMS/WAMS ratios, respectively, for further analysis. We also performed a concise analysis of period 7, which had the second-lowest FAMS/WAMS ratio, as shown in Table 1.

3.2 Composition

Fig. 2 shows the percent composition of the nine PAHs during ten periods at WAMS and FAMS. The proportions of the 9 PAHs differed with period and site. Though the 4-ring PAHs (Flu + Pyr + BaA + Chr) had the highest proportions in each period, some compositions changed substantially in some periods, such as the Chr ratio in period 9 and 10 at FAMS, compared with other periods. These results indicate that PAHs compositions not only differ significantly between sites but also differ between periods, even at the same location.

In urban areas, PAHs are mainly emitted from automobiles, power plants, domestic heating sources and industrial processes (Tang *et al.*, 2005). Several PAHs ratios were reported as source markers in a previous study (Khalili *et al.*, 1995). Fig. 3 shows the origin sources estimated by [Flu]/[Flu + Pyr] and [IDP]/[IDP + BgPe] for the 10 periods at the two sites. In this scatterplot, [Flu]/[Flu + Pyr] and [IDP]/[IDP + BgPe] correspond to the X and Y axes, respectively. The source may have been coal or wood combustion if both ratios were over 0.5, or the source was more likely to have been vehicles when both ratios were below 0.5. Otherwise, coal or wood combustion and vehicles were both likely to be sources. Though Table 1 and Fig. 2 show that the concentrations and compositions were different in most periods, the value of both [Flu]/[Flu + Pyr] and [IDP]/[IDP + BgPe] were mostly over 0.5 indicated that the main sources of PAHs at the two sites were coal or wood combustion.

3.3 Backward Trajectories

Backward trajectory analysis is a very important method to discuss origin air mass (Murao, 2011). From the above analysis (section 3.1), the concentrations of PAHs in most periods were not the same at the two sites. Thus, this study mainly analyzed the backward trajectories of periods 3 and 9, which had the minimum and the maximum concentration differences, respectively, at the two sites. For each backward trajectory figure, the upper part shows the horizontal trajectories, and the lower part shows how an air mass starting at an elevation of 1000 m varied in altitude.

As shown in Fig. 4, the backward trajectories showed that the directions of air mass flow to the two sites were very different in period 3. Although the backward trajectories marked (5) to (7) in Fig. 4(a) suggested that on

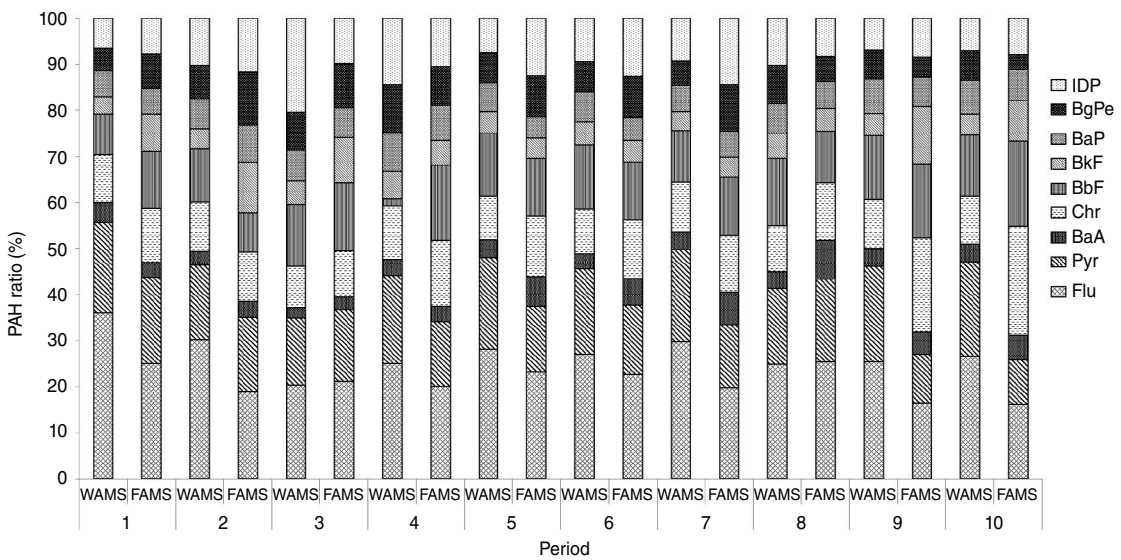

Fig. 2. The percent composition of 9 PAHs in ten periods at the Noto Peninsula and Fukue Island.

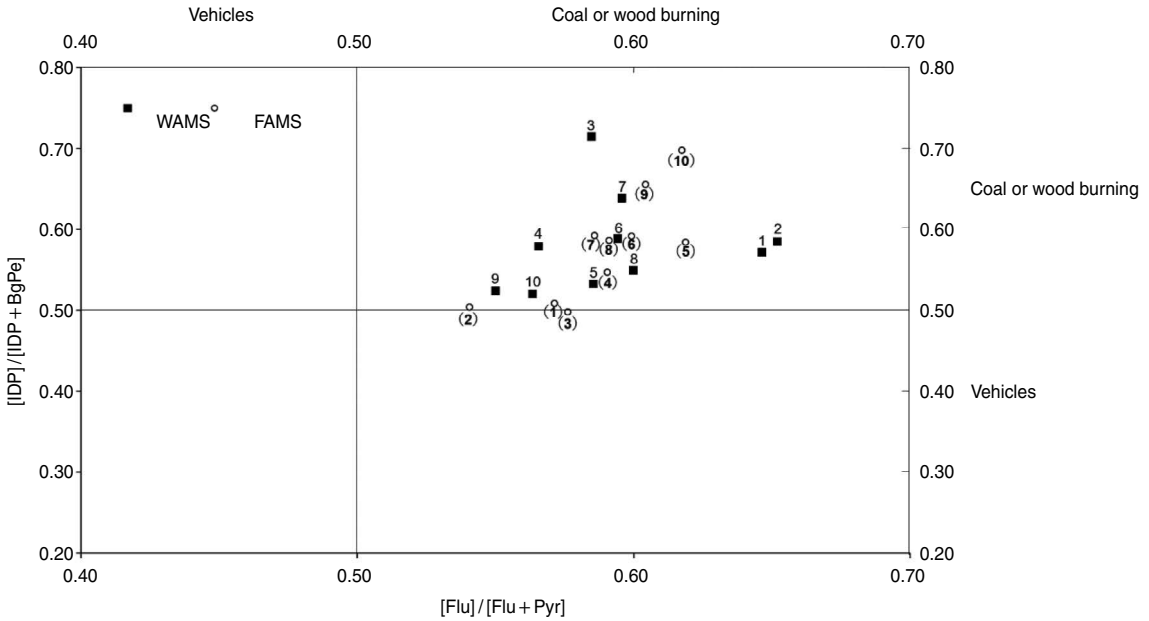

Fig. 3. Origin sources estimated by [Flu]/[Flu + Pyr] and [IDP]/[IDP + BgPe] for 10 periods at two sites. The mark of (○) means FAMS; the mark of (■) means WAMS.

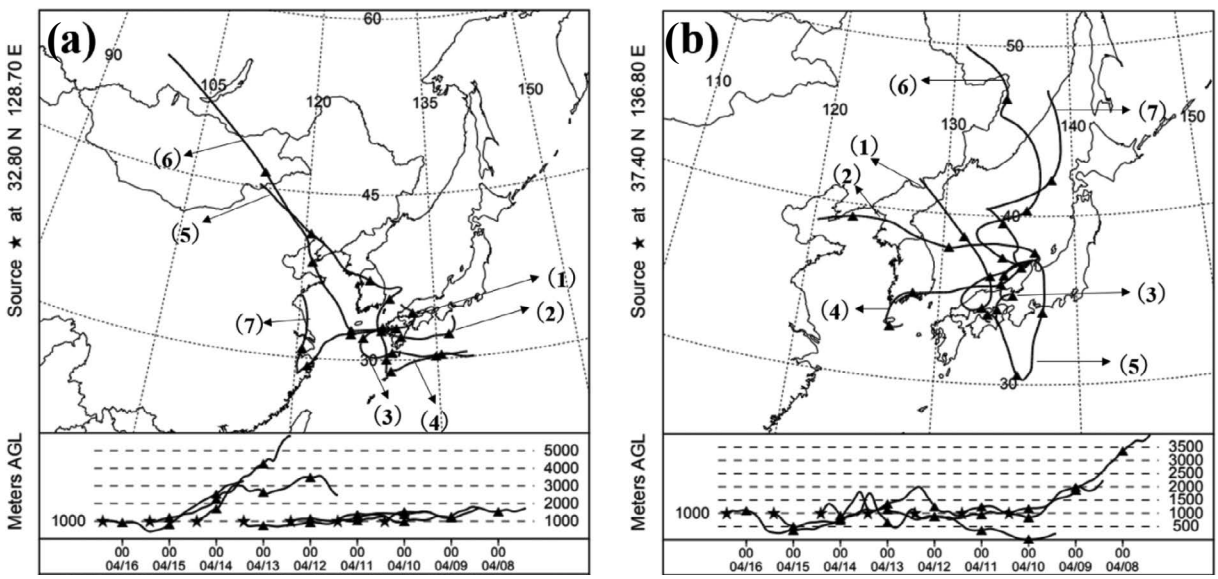

Fig. 4. Backward trajectories at a height of 1000 m flowing to FAMS (a) and WAMS (b). The trajectory of (1) 2009/4/10, (2) 4/11, (3) 4/12, (4) 4/13, (5) 4/14, (6) 4/15 and (7) 4/16 (period 3). (▲) Indicates 24-h data points within a 72-h span. (★) Indicates the start time for each 72-h backward trajectory. AGL means above ground level.

these later days the air mass moved rapidly from the Asian continent to FAMS, trajectories (1) to (4) indicated that the air mass on these days moved very slowly and the height was very low. From a previous study, PAHs concentrations decreased with increasing distance from China, with China > Russia > Korea≈the Sea of Japan > the Pacific Ocean (Sato *et al.*, 2008). Since air mass stay on the Pacific Ocean for a longer period with very few combustion particles, particulates are diluted, resulting in fewer PAHs in the parcel even if PAHs are long-range transported from the Asian continent in the next few days. On the other hand, though

the air mass flow to WAMS came from all regions, such as China, Russia, North and South Korea and Japan, the height of the air mass flow to WAMS was low and had almost no change except for two high air-mass routes from China ((1) and (2)), shown in Fig. 4(b). A low altitude means that particulates containing PAHs are likely to drop during long-range transportation, since air masses from the Asian continent mostly pass through the Sea of Japan. This may explain why the PAHs concentration was also low at WAMS.

Due to the backward trajectories were different between WAMS and FAMS in the period 3, even if we explained why they had a low concentration during this period, it could not explain why PAHs concentration was nearly the same between these two sites (Table 1). For this, we analyzed the period 7 (Fig. 5) briefly which also had a similar concentration and FAMS/WAMS ratio (1.11) relative to period 3. As shown in Fig. 5, period 7 differed from period 3 that the backward trajectories at WAMS were similar to those at FAMS. The air mass was from the ocean and had a relatively low height for the first few days ((1) to (4)) and came from the Asian continent and had a relatively high height for the later few days ((5) to (7)). This observation suggests that the low height of the air mass influenced the long-range transport of particulates and may have caused the low concentrations in period 3 and period 7. The similar air mass condition could result in the similar concentration like period 7.

As shown in Fig. 6(b), it was found that part of the WAMS air mass came from northeast China and the other part from Russia, while all of the FAMS air mass came from a greater height in northeast and central China (Fig. (6a)). In our previous study, coal combustion systems were identified as the main contributor of PAHs in northern Chinese cities such as Shenyang and Beijing in the winter (Tang *et al.*, 2017, 2013; Hattori *et al.*, 2007). Period 9 was in the Chinese heating period (mid-November–mid-March), which meant high PAHs emissions. Backward trajectories indicated that the air flow to FAMS contained much more PAHs because air flow path mostly passed through the areas such as Shenyang, which are well known for coal burning, a process that can increase the number of PAHs found in airborne particulates. According to the data downloaded from Japan Meteorological Agency, it indicated there were three rain days (the precipitation over 20 mm; the maximum was 37 mm) at WAMS, while there was only one rain day (the precipitation was 45 mm) at FAMS in period 9. Therefore, the PAHs concentration was much lower than FAMS (FAMS/WAMS: 4.17) might be due to the wet deposition (rain out and wash out effects) for particulates. It did not consider the factor of wet deposition in period 3 and period 7 because both sites had the

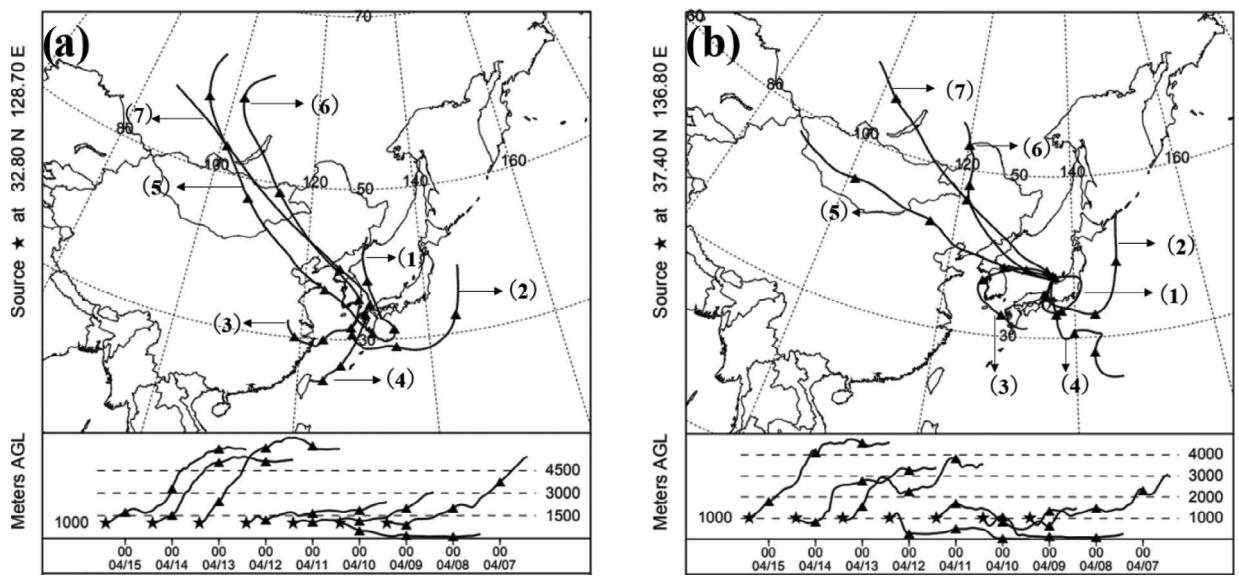

Fig. 5. Backward trajectories at a height of 1000 m flowing to FAMS (a) and WAMS (b). The trajectory of (1) 2010/4/9, (2) 4/10, (3) 4/11, (4) 4/12, (5) 4/13, (6) 4/14 and (7) 4/15 (period 7). (▲) Indicates 24-h data points within a 72-h span. (★) Indicates the start time for each 72-h backward trajectory. AGL means above ground level.

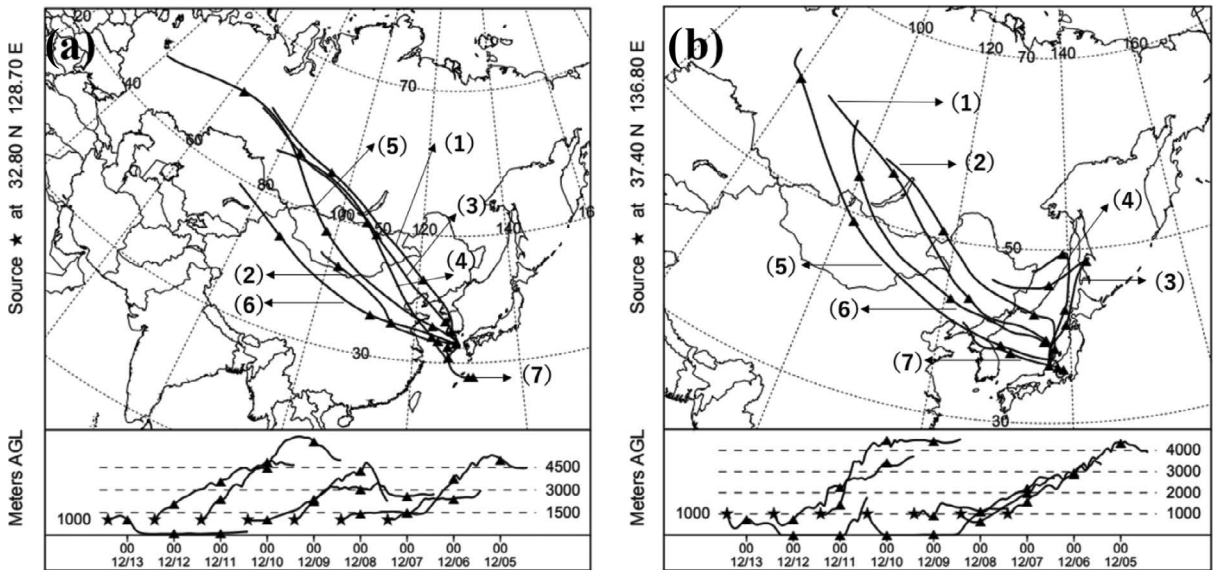

Fig. 6. Backward trajectories at a height of 1000 m flowing to FAMS (a) and WAMS (b). The trajectory of (1) 2010/12/7, (2) 12/8, (3) 12/9, (4) 12/10, (5) 12/11, (6) 12/12 and (7) 12/13 (period 9). (▲) Indicates 24-h data points within a 72-h span. (★) Indicates the start time for each 72-h backward trajectory. AGL means above ground level.

same and only one heavy rain day (period 3: the precipitation over 20 mm; period 7: the precipitation over 35 mm). Although wet deposition is very important, unfortunately, we only did the observation of particulates during these periods. In the future, we prepare to collect dry and wet deposition simultaneously.

4. CONCLUSIONS

Weekly concentrations and compositions of PAHs were compared between the Noto Peninsula and Fukue Island. The PAHs concentration differed significantly for most periods at these two sites. In this paper, the influence of PAHs degradation on compositions during long-range transported from the Asian continent were not large due to winter-specific weather conditions (Yang *et al.*, 2007). Based on the conclusion that the [Flu]/[Flu + Pyr] ratio did not change much during transport, as observed in a previous study (Ogawa *et al.*, 2012), we only used the composition ratios to surmise the origin sources. In the future, we will analyze other components, such as As, Se, water-soluble ions and OC/EC, to more accurately determine emission sources (Shen *et al.*, 2013). On the other hand, because wet deposition is also the important factor for PAHs analysis, we prepare to collect dry and wet deposition simultaneously in the future.

This is the first comparative study about Fukue Island and the Noto Peninsula. Through this study, we determined that air mass movements from the Asian continent were the main reason for these differences. The backward trajectory analysis found that the low height of the air mass may cause the low concentration and the similar air mass condition could result in the similar concentration. The concentration of long-range-transported PAHs depended more on the source of the coal combustion areas in heating period.

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