Long-range transportation of polyclic aromatic hydrocarbons and nitropolycyclic aromatic hydrocarbons from China to Japan

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In East Asia, which has been undergoing a rapid increase of population and economic development, large amounts of pollutants are released into the air and water. Among the pollutants in the atmosphere, several polycyclic aromatic hydrocarbons (PAHs) and nitropolycyclic aromatic hydrocarbons (NPAHs) are thought to be carcinogenic or mutagenic or have endocrine disrupting activity. PAHs and NPAHs are released in the exhaust from the combustion of petroleum and coal. Coal consumption in China is the largest in the world and represents over 75% of the energy source in China. However, in spite of the much higher concentration of atmospheric PAHs in China than that in Japan, there has been no report concerning the long-range transportation of atmospheric PAHs and NPAHs from China to Japan.

Airborne particulates were collected at Wajima Air Monitoring Station (Nisifuta-machi, Wajima City, Ishikawa Prefecture, Japan), which is located on the Noto peninsula 2.1 km south from the Japan Sea coast, in the path of winter northwest winds and year-round west jet stream that blow from China. No major emission sources of PAHs and NPAHs are near the station. Airborne particulates were collected by a high volume air sampler with a quartz filter from September 17, 2004 to September 16, 2005, including the central heating period of China (October 15 - April 15) and a period of yellow sand (Kosa) was detected at Wajima (March 11 - May 6, 2005). The filter was changed every week. PAHs and NPAHs were analyzed by HPLC with fluorescence and chemiluminescence detection, respectively.

All of the PAH groups (4-, 5- and 6-ring PAHs) and airborne particulates (AP) at Wajima tended to increase during the period October 15 to November 19, 2004 (Figure 1). The concentrations remained high until March 18, 2005 and then decreased. The period of the high concentrations of PAHs approximately coincided with the heating period of China (from October 15, 2004 to April 15, 2005 for a part of the Northern China and from November 15, 2004 to March 15, 2005 for the rest area of Northern China). From March 11 to May 6, 2005, yellow sand (Kosa) transported from the Asian Continent was detected at Wajima. During this period the concentration of AP was also high.

3- and 4-ring PAHs, which have relatively high vapor pressures, were previously shown to be distributed in the gas phase at ambient temperature. On the other hand, most of the 5- and 6-ring PAHs were found in the particulate phase, regardless of the ambient temperature. The concentrations of 5- and 6-ring PAHs, which didn't depend on the meteorological factors, remained at higher levels during the heating period of China (Figure 1).

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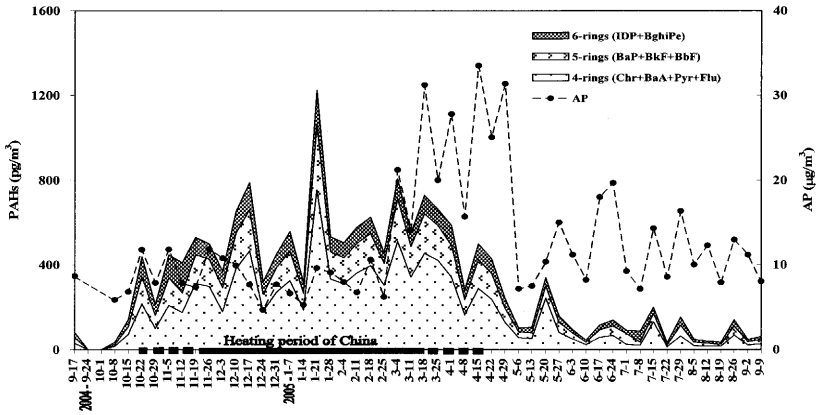

Figure 1 Weekly variations of PAHs and AP at Wajima during the sampling period. (The date means the first day of a week's sampling.)

A cluster analysis of the data collected at Wajima, Shenyang (winter) and Kanazawa (winter) indicated that the samples formed two large clusters (Figure 2). For the Wajima samples that were collected in the heating period of China, all except the 3 December sample were included in Cluster 1 with Shenyang (winter), suggesting the composition of PAHs of the above samples at Wajima was similar to that at Shenyang in winter. The rest of samples at Wajima were similar to that at Kanazawa (winter) since both of them were included in Cluster 2.

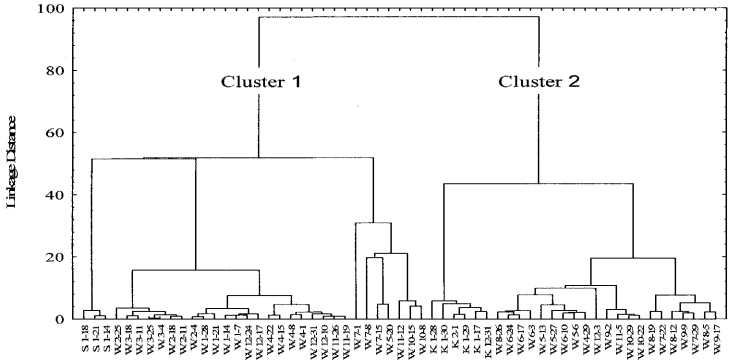

Figure 2 Cluster analysis of atmospheric PAHs and NPAHs at Wajima based on Ward's method and city-block (Manhattan) distance. W: Wajima, K: Kanazawa, S: Shenyang. (Values of Kanazawa and Shenyang cited from Ref. 1)

In this work, the [1-NP]/[Pyr] ratio was calculated to identify the origin of particulates collected at Wajima. The mean and median values at Wajima (0.011, 0.008) during the heating period of China were much smaller than those at Kanazawa (0.235, 0.220)[1)] but close to those at Shenyang (0.004, 0.004)[1)].

We calculated the 4-day back trajectories at 1000 m above ground level of the air to Wajima during the sampling period. Most of the traces of the samples from November 19, 2004 to March 18, 2005, which corresponds to the heating period of China, passed through Northeast China which has a high density of population and industry. The atmospheric pressure pattern in northeast Asia in winter is usually characterized by a high pressure area in the west and a low pressure area in the east. This is thought to result in long-range transportation of air from the Asian continent to Japan.

Higher atmospheric PAH concentrations were observed at Wajima during the heating period of China. A cluster analysis showed that the composition of PAHs at Wajima, during the heating period of China, was close to that at Shenyang but not to that of Kanazawa. The [1-NP]/[Pyr] ratios of Wajima were much smaller than those at Kanazawa but close to those of Shenyang. A back trajectory analysis indicated that the air was transported from Northeast China over the Japan Sea. These results strongly suggest that PAHs and NPAHs emitted in China were transported over long distances to Japan.

References

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