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Particulate polycyclic aromatic hydrocarbons in the urban Northeast Region of China: Profiles, distributions and sources

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ABSTRACT

In this study, concentrations of polycyclic aromatic hydrocarbons (PAHs) associated with PM_{10} were measured to examine the status, characteristics and sources of atmospheric PAH pollution in the industrial Northeast Region of China. Mean concentrations of total PAHs were 65.5, 40.0, 73.0 and 436.7 ng m^{-3} in the four seasons respectively. The calculated BaPeq concentrations in winter all exceeded the national standard, imposing serious PAH exposure risk. PAH concentrations varied between the cities, but PAH concentrations in different functional areas within a city did not show significant difference. In general, particulate PAH profiles were dominated by 4- and 5-ring compounds. Elevated proportions of 3-ring PAHs and 5-ring PAHs were found in winter and in summer respectively. Diagnostic ratios and principal component analysis (PCA) were used to identify potential sources of PAHs. Coal combustion activities were the main contributors of particle-associated PAHs in this region.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of ubiquitous persistent organic pollutants (POPs), primarily derived from incomplete combustion of fossil fuels and biofuels (U.S. National Academy of Sciences, 1983). PAHs were widely studied with focus on their health-related impacts, and many of them were proved or possible mutagens and carcinogens (Ianistcki et al., 2009; Nisbet and LaGoy, 1992; US EPA, 1984). Atmospheric PAHs present in both particle and gas phases, while carcinogenic species are mainly associated with the particle phase (Dickhut et al., 2000; Zhang et al., 2009). In addition, most of the carcinogenic PAH species are concentrated in the fine size and ultra fine size of particulate matters (Venkataraman et al., 1994). Those particulate matters could be inhaled into the deeper respiratory tract regions (Cecinato et al., 1999). As a result, particulate PAHs has been a serious concern to both scientists and policy makers. In addition to localized risk, PAHs can disperse regionally and intercontinentally through atmospheric transportation (Lohmann et al., 2007). Recently, high levels of atmospheric PAH concentrations and emission rates have been reported in developing countries especially in China (Bourotte et al., 2005; Sharma et al., 2007; Zhang and Tao, 2009), which has caused health concerns for a large population (Li et al., 2005; Xu et al., 2006).

The Northeast Region of China (NRC) encompasses three big northeastern provinces, i.e. Liaoning, Jilin and Heilongjiang and east area of Inner Mongolia (Fig. 1). This region is bordered by Russia, Mongolia and North Korea to the north, northwest and southeast, respectively. It covers an area of 1,250,000 km^2 and occupies 13% of the national total land area, with a population of about 120 million. NRC has the earliest heavy industry foundation in China, characterized by resource-reliant leading industries including iron/steel, coal, crude oil, machinery, metallurgy and chemical production. As an important producer of energy and raw material, NRC played an important role in the national economic development and industrialization process. However, the traditional economic growth mode not only depletes the un-reproducible natural resources but also causes severe environmental problems including deteriorated air, water and soil quality. NRC is one of the regions with the highest PAH emission intensity and population-normalized emission in China (Xu et al., 2006), which poses significant health risk for people both within this region and in adjacent areas. Based on the previous knowledge and emerging demand, a region-wide survey of atmospheric PAHs was, for the first time, initiated to fully understand the status and characteristics of PAH pollution, especially focus on the more

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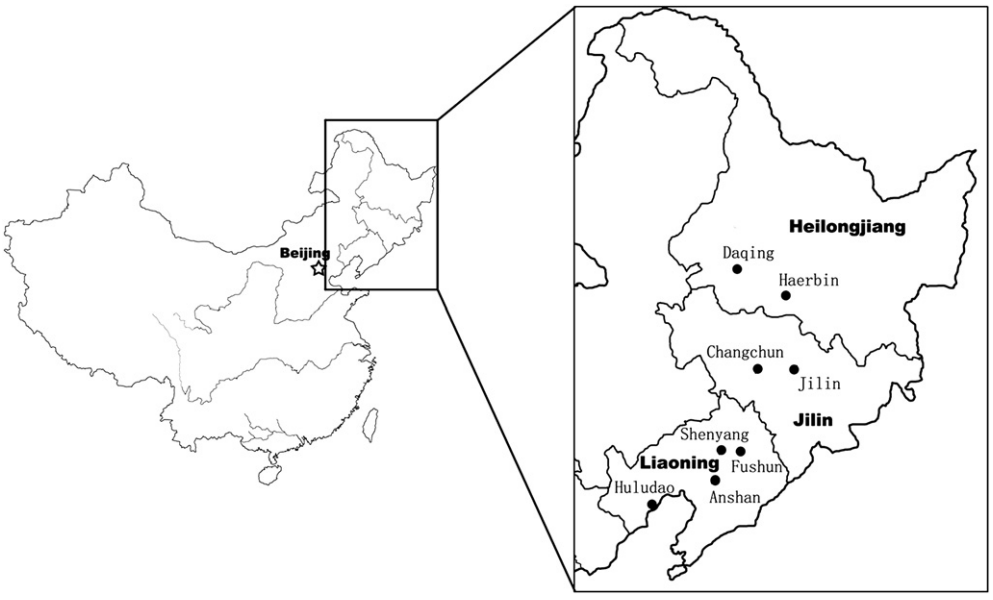

Fig. 1. Location of the Northeast Region and the eight typical industrial cities studied.

carcinogenic particulate PAHs in this heavy industry impacted area. Four intensive field campaigns were performed during April 2008 to January 2009 at 30 urban sites located in eight industrial cities. In this study, observation of the concentrations and profiles of particulate PAHs and their temporal and spatial distributions were presented, major contributing sources are identified and quantified using the multivariate method. This information was vital in terms of pollution control and risk management.

2. Experimental

2.1. Studied area and sampling procedure

The cities within this region are mostly sprawled around local mineral field. Nearly one third of the typical resource cities in China are located in the northeast region. Location and information of the eight typical industrial cities examined in this study were shown in Fig. 1 and Table 1. Most of the studied areas are located on flat plains, except Fushun and Jilin are mountainous. All the cities experience a continental monsoon climate of middle latitude temperate zone, characterized by a long and chilly winter (about five to six month) when coal are heavily used for heating purpose.

A sampling network composed of 30 urban sites was designed and set with three or five sites in each city (Table 1). The sampling sites were selected from the existing automatic air quality monitoring sites and located in different functional areas, i.e. industrial, residential, cultural, mixed and relatively clean areas. Sampling devices were placed on rooftops of two- or three-story monitoring buildings. The sampling inlets were located approximately 8–12 m above the ground level. Four intensive field campaigns were carried out in the spring (April 15–20, 2008), summer (July 21–26, 2008), autumn (October 10–16, 2008), and winter (January 12–17, 2009). The seasonal differences in ambient temperature were large, with the minimum temperature (−5–20 °C) in January and the maximum temperature in July (20–26 °C). Air sampling was almost simultaneously conducted at the 30 sites in each campaign. The 24 h integrated PM_{10} (aerodynamic diameter $<10\,\mu m$) samples were taken continuously for five days.

Particulate PAHs were captured on prebaked (800 °C, 3 h) quartz-fiber filters (Φ90 mm, Pall Life Sciences, USA) using median-volume PM samplers (TH-150CIII, China) operated at a flow rate of

Table 1
Description of the eight cities examined in this study.

City	Location	Urban area	Population	Main industry	Sampling site[a]
Shenyang	41°11′–43°02′N, 122°25′–123°48′E	3945 km^2	7.8 million	Equipment manufacturing, metal smelting, medical	Industrial (2), residential (1), cultural (1), clean site (1)
Fushun	41°41′–42°38′N, 123°39′–125°28′E	675 km^2	1.4 million	Coal mining, petrochemical, fine chemicals, aluminum	Industrial (1), residential(2)
Anshan	40°27′–41°34′N, 122°10′–123°13′E	624.3 km^2	1.3 million	Ion and steel, minerals processing	Industrial (1), residential (2)
Huludao	39°59′–41°12′N, 119°12′–121°02′E	2303 km^2	1.6 million	Petrochemical, equipment manufacturing, zinc	Industrial (1), residential (2)
Changchun	43°05′–45°15′N, 124°18′–127°02′E	4906 km^2	4.9 million	Automobile, medical, food, photoelectronic	Industrial (1), residential (2), cultural (1), clean site (1)
Jilin	42°31′–44°40′N, 125°40′–127°56′E	3636 km^2	2.4 million	Petrochemical, metallurgy, automobile, carbon production	Industrial (1), residential(1), clean site(1)
Harbin	44°04′–46°40′N, 125°42′–130°10′E	7086 km^2	4.8 million	Equipment manufacturing, medical, food, petrochemical	Industrial (1), residential(2), cultural(1), mixed site(1)
Daqing	45°46′–46°55′N, 124°19′–125°12′E	5107 km^2	1.2 million	Oil extraction, petrochemical	Industrial (2), mixed site(1)

[a] With the number of this type of site in parentheses.