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Source apportionment of polycyclic aromatic hydrocarbons in the Dahuofang Reservoir, Northeast China

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Abstract Polycyclic aromatic hydrocarbons (PAHs) in 24 surface sediments from the Dahuofang Reservoir (DHF), the largest man-made lake in Northeast China, were measured. The results showed that the concentrations of 16 US EPA priority PAHs in the sediments ranged from 323 to 912 ng/g dry weight with a mean concentration of 592 ± 139 ng/g. The PAH source contributions were estimated based on positive matrix factorization model. The coal combustion contributed to 31 % of the measured PAHs, followed by residential emissions (22 %), biomass burning (21 %), and traffic-related emissions (10 %). Pyrogenic sources contributed ~84 % of anthropogenic PAHs to the sediments, indicating that energy consumption release was a predominant contribution of PAH pollution in DHF. Compared

with the results from the urban atmospheric PAHs in the region, there was a low contribution from traffic-related emissions in the sediments possibly due to the low mobility of the traffic-related derived 5+6-ring PAHs and their rapid deposition close to the urban area.

Keywords Source apportionment · PAHs · Sediments · Dahuofang Reservoir · Northeast China

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are compounds typically containing two to eight aromatic rings. PAHs have been of special concern for several decades due to their potential risk to human health and environment. Early in the 1970s, the US Federal Water Pollution Control Act (1972) and the US Clean Water Act (1977) recognized PAHs as a substantial health hazard, and the US Environmental Protection Agency (US EPA) classified 16 PAHs as priority pollutants. Sanctorum et al. (2011) estimated that 16 PAH emissions amounted to 183 tons in 1996, 182 tons in 2000, and 200 tons in 2008 in Flanders, Belgium, mainly due to the traffic emission. There has been a growing concern for increasing PAH emissions in China for the last two decades due to a rapid urbanization and industrialization (Liu et al. 2005, 2009; Guo et al. 2006). Sixteen PAH emissions were estimated to be

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114 Gg in China in 2004, accounting for 22 % of total 16 PAH emissions in the world (Zhang and Tao 2009).

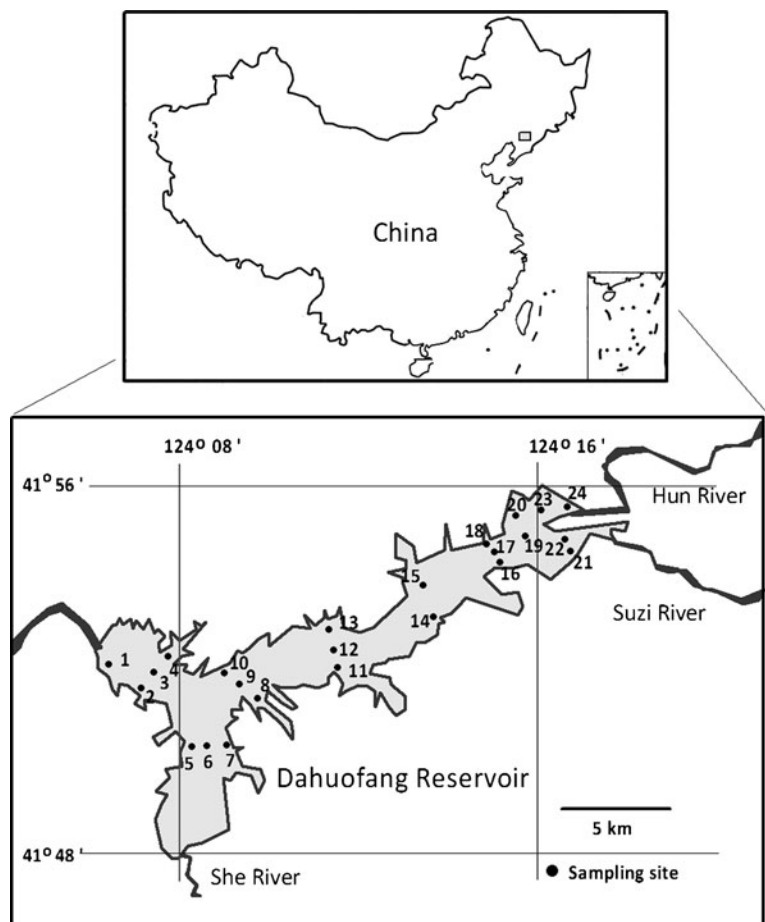
The determination of various potential sources and quantification of each source contribution for PAHs are of a great challenge when the mixed sources are involved. Diagnostic ratios of special PAH compounds have been proven to be useful in source identification in sediments (Socolo et al. 2000; Yunker et al. 2002; Mai et al. 2003; Sanctorum et al. 2011). In recent years, receptor models have been successfully applied for quantitative source apportionment of PAHs in the atmosphere and in the sediments (Lee et al. 2004; Okuda et al. 2010).

Liaoning province located in the southern part of Northeast China is one of the most important industrial bases in China, covering a wide range of industries such as machinery, electronics, metal refining, petroleum, chemical industries, construction materials,

coal, and so on. In 2009, the GDP of Liaoning province ranked the seventh in China, and energy consumption at 0.16 billion tons of standard coal accounted for approximately 5.6 % of energy use in China (LPBS 2009). The Dahuofang Reservoir (DHF) is located in the east-central Liaoning province (Fig. 1), and it is the largest man-made lake in Northeast China. Recent reports pointed out that the remote/urban lakes could act as the natural traps for anthropogenic pollutants such as PAHs (Moriwaki et al. 2005; Barra et al. 2006). Thus, DHF in the Northeast China is an ideal area to track the regional PAH sources.

The objectives of this study are (1) to quantitatively apportion the sources of PAHs in the sediments of DHF using a positive matrix factorization (PMF) model, and (2) to evaluate the difference in PAH congener patterns in atmosphere and sediments in the DHF region and in their sources.

Fig. 1 The location of the Dahuofang Reservoir in China and the locations of the sampling sites in the reservoir



Materials and methods

Sampling

DHF is located in the northeastern part of China among 41°41' N–41°57' N and 124°05' E–124°20' E as shown in Fig. 1. The length of the reservoir is about 35 km with the largest width about 4 km and minimum 0.3 km. Its area is 114 km² with the drainage area of 5,437 km².

Twenty-four surface sediment samples were collected using a grab sampler in January, 2010, and the locations of sampling sites are shown in Fig. 1. The top 3-cm layer was carefully removed with a stainless steel spoon for subsequent analysis. All sediment samples were wrapped in aluminum foil and stored at -20°C until analysis.

PAH analysis

The PAH analysis procedure followed that described by Guo et al. (2006). Briefly, homogenized samples were freeze-dried and ground. About 10 g of the sample was spiked with a mixture of recovery standards of five deuterated PAHs (naphthalene-d₈, acenaphthene-d₁₀, phenanthrene-d₁₀, chrysene-d₁₂, and perylene-d₁₂). The samples were extracted with dichloromethane in a Soxhlet extractor for 72 h, with activated copper added to remove the sulfur in the samples. The extract was concentrated and fractionated using a silica–alumina (1:1) column. PAHs were eluted using 35 ml of hexane/dichloromethane (1:1). The PAHs fraction was concentrated to 0.5 ml, and hexamethylbenzene was added as internal standard. The mixture was further reduced to 0.2 ml and subjected to GC-MS analysis. An agilent 5975C mass spectrometer interfaced to an Agilent 7890 gas chromatography was used to analyze the samples. The GC was equipped with a DB-5MS capillary column (30 m long×0.25 mm inside diameter×0.25 μm film thickness), with He as carrier gas. The chromatographic conditions were as follows: injector temperature, 290°C; detector temperature, 300°C; temperature program, 60°C (3 min), 60–290°C at 3°C/min, held at 290°C for 20 min. The PAHs quantified were as follows: naphthalene (Nap), acenaphthylene (Ac), acenaphthene (Ace), fluorene (Fl), phenanthrene (Phe), methylphenanthrene (MP), dimethylphenanthrene (DMP), anthracene (Ant), acephenanthrylene (Acp),

fluoranthene (Flu), pyrene (Pyr), retene (Ret), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), Benzo[e]pyrene (BeP), indeno[1,2,3-cd]pyrene (IP), dibenz[a,h]anthracene (DBA), perylene (Pery), and benzo[ghi]perylene (BghiP). Procedural blanks, standard-spiked matrix, and parallel samples were analyzed for quality assurance and control. Procedural blank samples contained no detectable amount of the target PAHs. Recoveries of PAH compounds in spike matrix sample ranged from 85 to 95 %. The relative precision of parallel samples was below 15 % (n=4). Recovery was 80±7 % for naphthalene-d₈, 82±10 % for acenaphthene-d₁₀, 89±9 % for phenanthrene-d₁₀, 94±6 % for chrysene-d₁₂, and 97±9 % for perylene-d₁₂. PAH concentrations were recovery corrected.

Data analysis by PMF model

Detailed concept and application of PMF source apportionment has been described in EPA PMF 3.0 Fundamentals & User Guide (www.epa.gov/heads/products/pmf). PMF is a PCA-based receptor model with non-negative constraints that involve solution of quantitative source apportionment by oblique solutions in a reduced dimensional space (Paterson et al. 1999). The PMF model defines the concentration matrix of chemical species measured at receptor sites as the product of source composition and contribution factor matrix with a residue matrix. In principle, the PMF model is based on the following equation:

$$X_{ij} = \sum_{k=1}^p A_{ik} Fk_j + R_{ij} \tag{1}$$

where X_{ij} is the concentration of the j th congener in the i th sample of the original data set; A_{ik} is the contribution of the k th factor on sample i ; Fk_j is the fraction of the k th factor arising from congener j ; and R_{ij} is the residual between X_{ij} and the estimate of X_{ij} using p principal components.

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{X_{ij} - \sum_{k=1}^p A_{ik} Fk_j}{S_{ij}} \right)^2 \tag{2}$$

Where S_{ij} is the uncertainty of the j th congener in the i th sample of the original data set containing m congeners and n samples. Q is the weighted sum of

squares of differences between the PMF output and the original data set. One of the objectives of PMF analysis is to minimize the Q value.

Before the analysis, undetectable value (dull value) was replaced with concentration value of one half the method detection limit. Uncertainty file should be provided to give the model an estimate of the confidence the user has in each value. In this study, the adopted uncertainty was taken as value of 20 % measured concentration based on the results from regularly analyzing a standard reference material (Mai et al. 2003).

Results and discussion

PAH concentrations in surface sediments

The concentrations of PAHs are summarized in Table S1. All PAH concentrations were calculated on a dry weight basis. The total concentration of 16 PAHs in the sediments ranged from 323 to 912 ng/g with a mean concentration of 592 ± 139 ng/g. The main 16 PAHs found in this study were Phe>Flu>Pyr>Fl>Ant>BaA in a decreasing order, and those compounds comprised 71 % of total 16 PAH concentrations (Fig. 2). As shown in Fig. 3, the highest concentration of 16 PAHs was found in the She River mouth (912 ng/g, in site 5). Besides, the higher concentration was observed in the Hun River mouth stations (708 ng/g, in site 24) in the upper half of the reservoir, and decreased with distance away from the Hun River mouth. The lowest concentration of 16 PAHs was detected in the middle of DHF (323 ng/g, in site 14). There are three tributaries including Hun River, Suzi

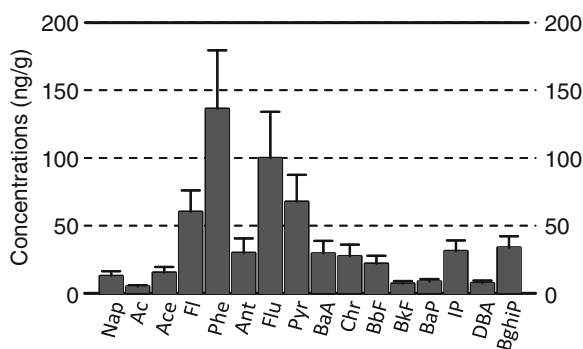
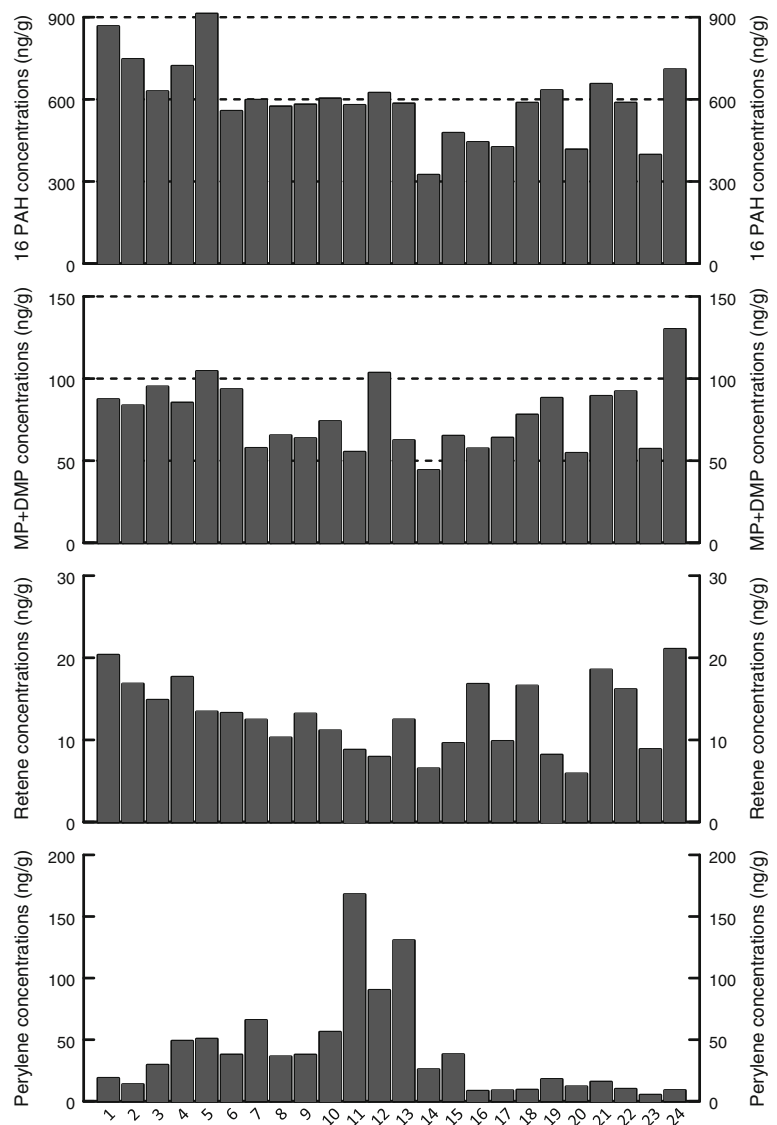


Fig. 2 Sixteen individual PAH congener concentrations assessed in sediments of DHF

River, and She River. The water inflow from Hun River, Suzi River, and She River are accounting for 52, 38, and 10 % of the total river water inflow into the reservoir, respectively. The results from Liaoning Environment Protection Bureau water quality monitoring program for Tenth Five Year Plan indicated that the COD, BOD₅, and heavy metals (chromium, lead, and arsenic) pollution was mainly derived from those river waters (FEPMS 2006). Therefore, the river runoff input was considered as the major contribution of PAHs in the reservoir. The 16 PAH levels in this study were lower than those detected in sediments of Nansi Lake (160–32,600 ng/g), North China (Li et al. 2009), and Taihu Lake (1,207–4,754 ng/g), East China (Qiao et al. 2006). Compared with river sediments, the 16 PAH level in this work was lower than those detected in the sediments of the Pearl River, South China (Luo et al. 2008), and Haihe River, North China (Jiang et al. 2007), but comparable with those from the Qiantang River, East China (Chen et al. 2007), Yangtze River and Yellow River (Hui et al. 2009). DHF as the important part of the Daliao River Basin, the concentrations of 16 PAHs were low to moderate contamination level in the whole Daliao River Basin (Guo et al. 2007; Men et al. 2009).

MP and DMP are occurring naturally in crude oil, coal and wood, as well as released by the incomplete combustion of organic materials. The sum concentrations of MP and DMP varied 44–130 ng/g in the sediments with a mean value of 77 ± 25 ng/g. MP and DMP concentrations in the sediment from DHF in the study were significantly lower than the data reported in the Pearl River Delta in China (Mai et al. 2001), Maryut Lake in Egypt (Barakat et al. 2010), and rivers and estuaries in Malaysia (Zakaria et al. 2002), which were seriously contaminated with MP and DMP of petrogenic origin. Ret was found at low levels in sediments of DHF (5.8–21.0 ng/g). Some studies reported that the Ret and its precursors were derived from natural sources (Hites et al. 1980; Wakeham et al. 1980). However, a few reports showed an anthropogenic input of Ret (Mai et al. 2003). The distribution trend of Ret was basically consistent with that of the 16 PAHs in this study, suggesting that Ret was possibly indicative of anthropogenic origin rather than diagenetic products. Pery concentration was found to vary drastically from 5 to 167 ng/g in the sediments, and the percentage of Pery relative to 16 PAHs in the sediments were >10 % in the sites 11, 12, and 13.

Fig. 3 Horizontal distributions of 16 PAH concentrations, MP+DMP, Retene, and Perylene in surface sediments in DHF



According to Hites et al. (1980), whenever Pery concentration is higher than 10 % of the total parent PAHs, it can be considered as originating from the diagenetic processes of organic matter in anoxic conditions.

Identification of sources from PMF model

PMF was developed in the early 1990s and has been most often applied to atmospheric data sets. PMF offers an advantage over PCA in that it can estimate the contribution from the specific source of individual sample. Besides, the user does not need to provide source profiles of target compounds. These properties

make PMF well suited for the studies of the sources apportionment in sediments. In recent years, PMF has been successfully applied to spatially distributed data sets to apportion the sources of PAHs in sediments (Lin et al. 2011).

In this study, 22 species of PAHs of 24 samples were used as the input data. The factor number ranged from three to eight was tried in order to choose the “optimal solution”. When the number of factors for PMF was six, the $Q(E)$ value range from 165.79 to 165.88, which was slightly lower than theoretical $Q(E)$ value of 168, indicating that the six factors were suitable to subsequent analysis. The values of slope and r^2 were 0.77–0.98 and 0.73–0.98, indicating the

modeled and actual concentrations fit well and six factors can fully explain the data information. Additionally, six-factor solution was adopted which gave the most stable results and the easily interpretable factors based on the background of PAH sources in the Liaoning Province (Tian et al. 2009; Wang et al. 2009; Liu et al. 2010).

Factor profiles for the six-factor solution of PMF are shown in Fig. 4. The first factor that accounts for 16 % of the total variance is dominated by Pery, which are believed to be diagenetic source (Fig. 5).

Factor 2 is higher loaded on BghiP and IP. BghiP, IP, and BaP are the dominant PAHs in exhaust emissions. The use of BghiP as a marker for traffic-related emissions has been reported by several works (Harrison et al. 1996; Nielsen 1996). This factor is responsible for

10 % of the total variance and could be identified as traffic-related emissions.

Factor 3 accounting for 21 % of the total variance is predominately composed of Phe, Fl, and Flu, with moderate loadings of Acp and Pyr. High emissions of Phe, Flu, and Pyr were observed in particulate matter of wood burning (Freeman and Cattell 1990). Additionally, Acp was identified as tracers of wood burning, and only presented in this factor (Yunker et al. 2002).

Factor 4 explains 22 % of the total variance, and mainly consists of Phe, Flu, BbF, and Pyr representative of residential emissions (Tian et al. 2009). The residential emissions refer to combustion process mainly for residential heating or cooking by usage of coal, wood, and natural gas.

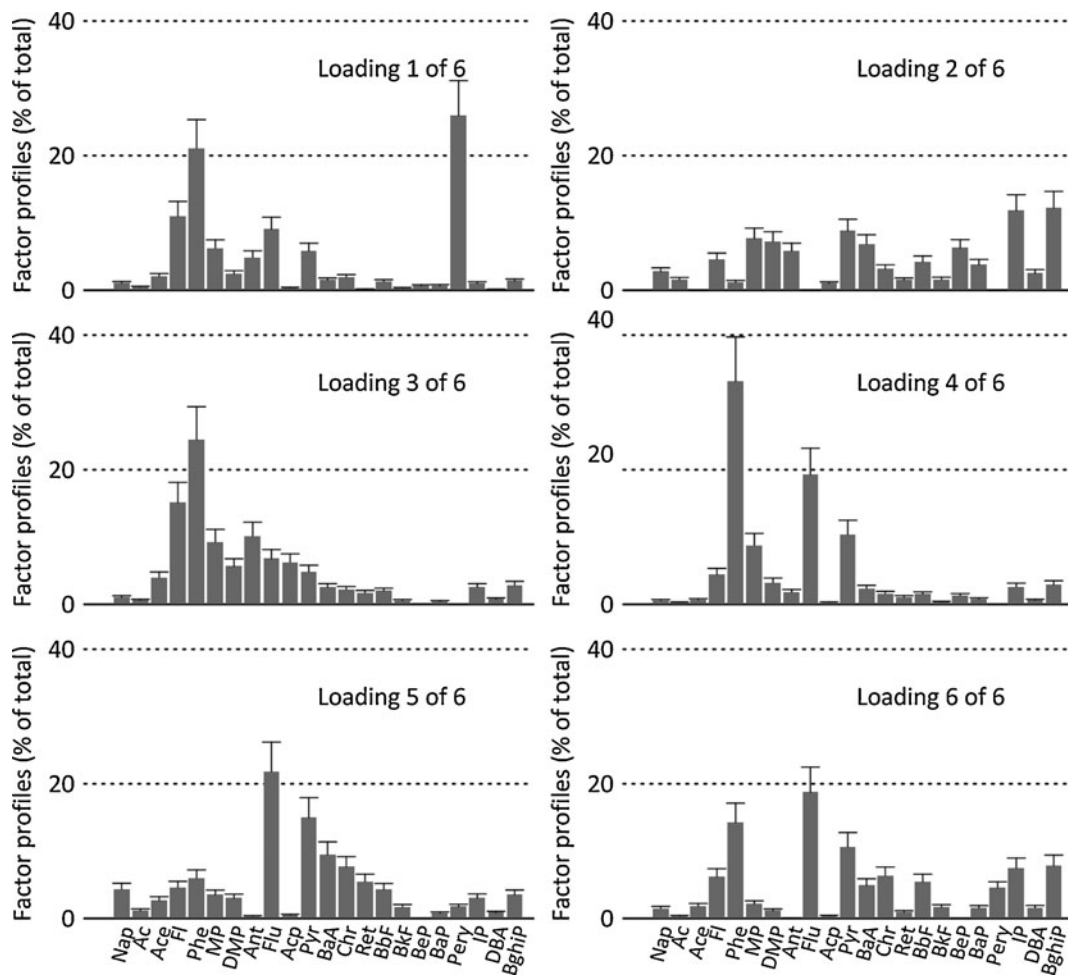


Fig. 4 Factor loadings obtained from PMF model analysis of PAHs in surface sediments of DHF

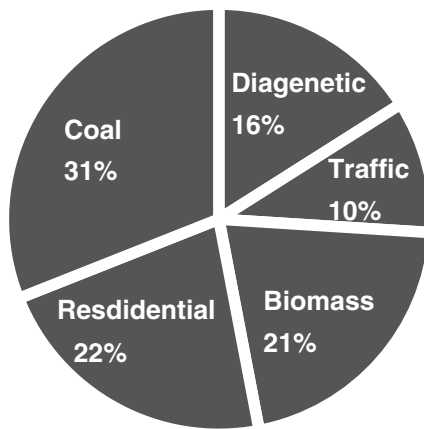


Fig. 5 Source average mass contributions (%) of each factor to total PAHs in surface sediments in DHF

Factors 5 and 6 characterized by four-ring PAHs is responsible for total 31 % of the measured PAHs. Flu and Pyr are usually used as markers for coal combustion (Tian et al. 2009; Wang et al. 2009). A similar profile was provided for the coal combustion in power plant. High emissions of Flu, Phe, and Pyr were also observed from coke ovens. Thus, this factor is the coal-fired power plant/coke oven source which is of representative of high temperature coal combustion process.

Sources contributions

Pyrogenic sources contributed ~84 % of the measured PAHs in the sediments, indicating that release from energy consumption is a predominant contributor of the PAHs in DHF (Fig. 5). The main sources of pyrogenic PAHs were coal-fired sources, residential emissions, wood burning, and traffic-related emissions, and their source contributions were, namely, 31, 22, 21, and 10 %. Overall, the source contribution patterns of pyrogenic PAHs in the sediments of the region were similar to that of the Daliao River Estuary in Liaoning Province. According to the attribution of the PAH sources resulting from the PMF model, the PAHs in the Daliao River Estuary sediments mainly came from coal combustion (43.3 %), biomass burning (24.3 %), and traffic emissions (16.7 %) (Liu et al. 2010).

Coal combustion is the main source of PAHs in the northeast China, and elsewhere in China such as the Pearl River region, South China (Luo et al. 2008) and

Taihu Lake, East China (Liu et al. 2009). Coal combustion provides up to 75 % of China's energy consumption. In Liaoning province, coal consumption was 124.29 million tons standard coal in 2004, accounting for 51.5 % of the total energy consumption (LPBS 2009). Moreover, Ravindra et al. (2008) pointed out that the coal combustion processes produce one to two orders of magnitude more PAHs than comparable release from petroleum product or natural gas usage. Biomass burning was recognized as a significant source of PAH emissions in China (Xu et al. 2006; Lin et al. 2011). According to source emissions inventory, PAHs derived from biomass burning accounted for a comparatively large proportion (40.1 %) (Xu et al. 2006). Additionally, Streets et al. (2001) estimated that contribution from biomass burning accounted for 33 % of total emission based on black carbon emission inventory in Liaoning Provinces. The emission from motor vehicles was the most important source of PAHs in environment. According to source emissions inventory, the PAHs derived from motor vehicles were estimated to be ~16 Gg in China in 2004, accounting for ~14 % of total PAH emissions in China (Shen et al. 2011). Therefore, the source pattern in the sediments of DHF was consistent with the regional energy structure.

Compared with results from atmospheric particle-bound PAHs

In this study, a ratio of traffic-related emissions contribution to coal combustion contribution is equal to 0.32 (10 vs. 31 %) in the sediments in DHF, which was similar to source apportionment results from the sediments of the Liaohe River Estuary (0.28 by 16.7 vs. 59.0 %) (Liu et al. 2010) and the soils from Dalian (0.39 by 28 vs. 72 %) (Wang et al. 2009). Differently, the ratios in the atmospheric particulates are 0.49 and 3.76 in winter and summer in Dalian (Tian et al. 2009), and 0.57 and 3.46 in winter and summer in Harbin (Ma et al. 2009). It was suggested that traffic-related emissions came mainly from the urban areas. Additionally, traffic-related emissions PAHs were mainly consisted of low-volatile PAHs with 5+6 rings, characterized by relatively high temperature of condensation and adsorbed on the airborne particles, and they are classified in the low mobility category subjected to rapid deposition and retention close to their source areas (Van Jaarsveld et al. 1997). By contrast,

coal-combustion-derived PAHs were composed of semi-volatile four-ring or five-ring PAHs. Semi-volatile PAHs can exist in the vapor phase and their gas–particle conversion makes them more mobile in the atmosphere (Lang et al. 2007). Thus, compared with the results from nearby urban atmospheric PAHs, there was a low contribution from traffic-related emissions in the sediments of DHF.

Conclusions

This work reported that concentrations of 16 PAHs in surface sediments in DHF ranged in 323–912 ng/g dry weight (a mean concentration of 592 ± 139 ng/g). The occurrence levels of 16 PAHs in the sediments in DHF were low to moderate level of contamination compared with those found in other aquatic systems in China. The source apportionment result from the PMF model indicated that pyrogenic sources contributed ~84 % of PAHs in the sediments. The main sources of pyrogenic PAHs were coal combustion, residential emissions, wood burning, and traffic-related emissions. The pyrogenic source compositions of PAHs in DHF correspond well to the regional energy consumption structure. There was a low contribution from traffic-related emissions in the sediments compared with the results of nearby urban atmospheric PAHs, suggesting that more traffic-related emissions PAHs from urban areas had a rapid deposition and retention close to their source areas.

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