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REVIEW ARTICLE

Polycyclic aromatic hydrocarbons in sediments of China Sea

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Abstract Increasing pollution pressures were placed in the coastal and estuarine ecosystems in China because of the elevated pollutants discharged from various sources. Polycyclic aromatic hydrocarbons (PAHs) in the environment were closely linked to human activities, which have been intensively studied for their geochemical interest as markers. In this review, the status of PAH contamination in China Sea was assessed by comprehensive reviews of the concentrations, sources, and fates of PAHs in sediments of China Sea. PAH concentrations in China Sea sediments decreased from north to south due to the higher emissions in North China. Atmosphere was probably the main carrier of PAHs in the north due to the higher contents of atmospheric fine particles and higher wind speeds. However, riverine inputs were probably the most important sources of PAHs in the coastal sediments of South China due to higher rainfall.

Keywords Polycyclic aromatic hydrocarbons · China Sea · Coastal sediment · Environmental fate · Source

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Introduction

Polycyclic aromatic hydrocarbons (PAHs), predominantly the incomplete combustion products of coal, coke, and biomass, are ubiquitous contaminants in atmospheric, terrestrial, riverine, and marine environments (Wolska et al. 2012). Some of these compounds (such as benzo[a]pyrene) are environmentally important due to their potential negative effects to mammals and aquatic organisms (Samanta et al. 2002). A number of studies have used PAHs as geochemical markers for evaluating the anthropogenic influences because they are strongly linked to local socioeconomic activities (Yunker et al. 2002).

China is one of the biggest energy-consuming countries with the highest PAH emissions in the world (1.14×10^{11} g in 2004; Zhang and Tao 2009). Almost all of the total annual PAH emissions remained within the boundary of mainland China (Lang et al. 2008a). Approximately 69.9 % of the PAH outflow from mainland China (7.1 % of the total annual emission) reached no further than the mainland China offshore environment. Estuarine and coastal areas are among the most important places for human inhabitants, yet they are often considered as the ultimate sinks of anthropogenic pollutants (Lohmann et al. 2007; Duan et al. 2013). PAHs have placed increasing pressures on coastal and estuarine ecosystems over the past decades because of enhanced human activities in coastal areas (Jiang et al. 2007). Many studies (Liu et al. 2005; Guo et al. 2007a) have shown that the fluxes of PAHs to marine sediments in coastal China inferred from dated sediment cores experienced a huge increase in recent decades, and the profiles of PAH concentrations and compositions revealed the transformation of China's industry structure (from an agricultural economy to an industrial one).

The coastal region is the most densely populated part of the country (Fig. 1). Rapid growth of the economy in China has been coupled with increasing environmental pollution.

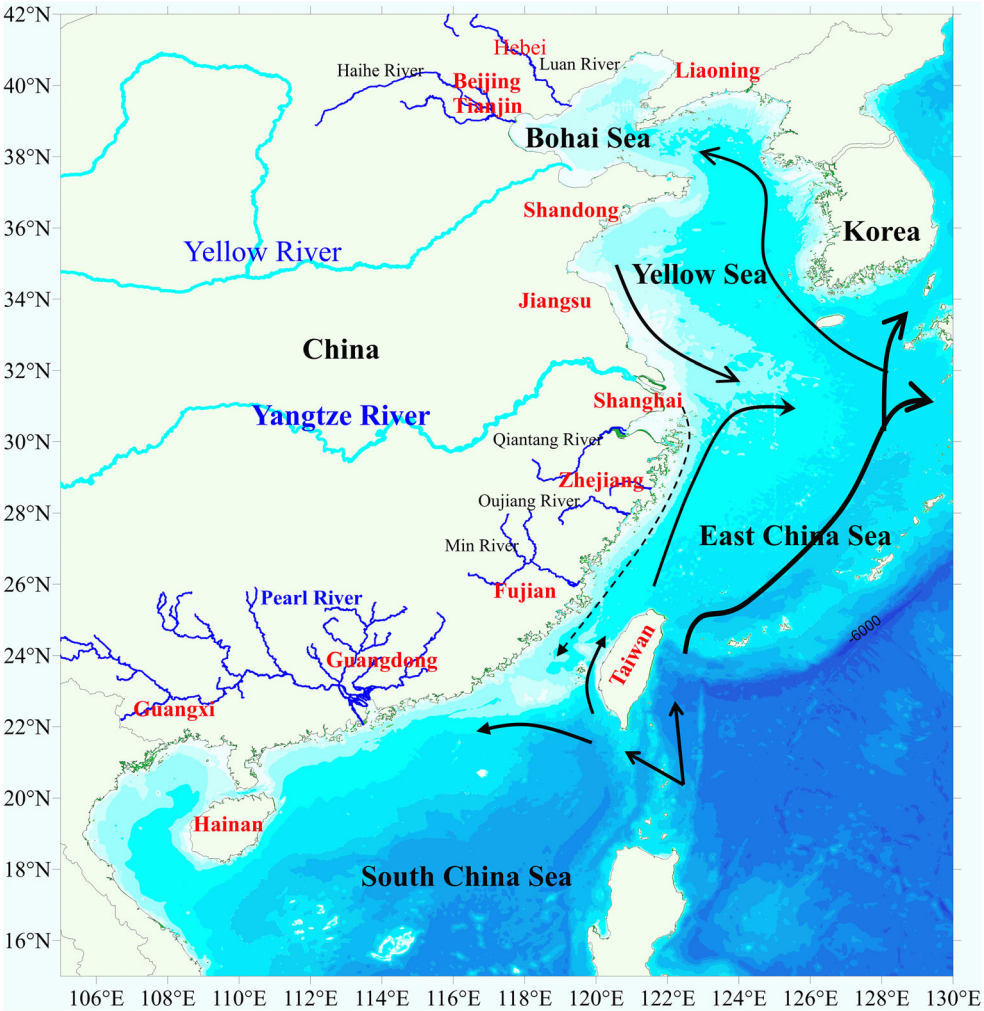

Fig. 1 Geological settings of Chinese coastal zone. Major surface currents are indicated by *arrows*. Gridded bathymetric data are from http://www.bodc.ac.uk/data/online_delivery/gebco/

According to the official reports (National Bureau of Oceanography of China 1997-2014), the coastal environment of China has worsened significantly in recent decades. This phenomenon has attracted broad attention in recent years. The Bohai Bay, Yangtze River estuary, and Pearl River estuary were seriously polluted (National Bureau of Oceanography of China 1997–2014). The three most developed Chinese city groups are located in these places (Beijing–Tianjin–Hebei area, Yangtze River delta, and Pearl River delta; shown in Fig. 1). Therefore, economic development is probably closely related to environment deterioration (Crain et al. 2009). The coastal marine environment has become an important research topic. However, so far, there have been few studies carried out to evaluate the differences of environmental evolution process between the different regions of coastal zone, especially between the North and South China.

Therefore, the main goal of this article was to present the possible influencing factors of marine environment deterioration in different regions of China by taking PAHs as tracers using available data. A fairly comprehensive review of PAHs in surface sediments of China Sea was presented. Distributions, potential sources, and fates of PAHs in coastal China and the relationships between environmental conditions and sources or fates were reviewed.

Occurrences of PAHs in estuarine, coastal, and oceanic sediments of China Sea

Sediments were considered as the final sink of semi-volatile and non-volatile organic pollutant (Nellier et al. 2015). That is, most of the anthropogenic pollutants released in different periods were all recorded in coastal sediments (Mighall et al. 2002). PAHs have been observed to be most concentrated in estuarine and coastal environments near urban centers, where inputs from the watersheds and airsheds are most localized (Yunker et al. 2002; Zakaria et al. 2002). In this paper, PAH concentrations in coastal, estuarine, and oceanic sediments reported in recent years were summarized (Fig. 2; the detailed information was provided in supporting information Table 1).

As shown in Fig. 2, the total PAH concentrations (16 US EPA priority PAHs) in the Haihe River sediments were

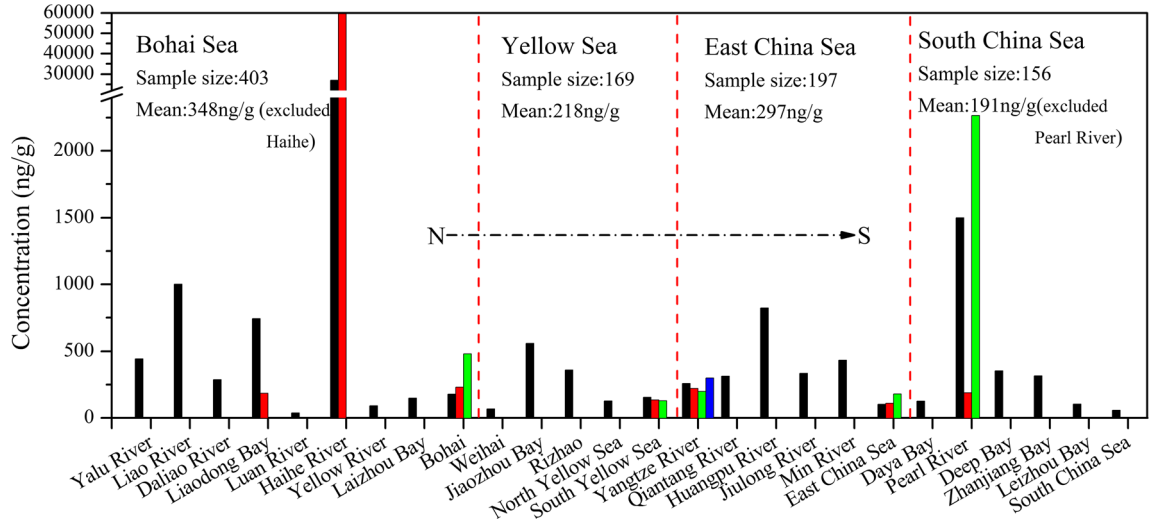

Fig. 2 Distribution of PAHs in estuarine and coastal sediments of China (listed in order of decreasing north latitude; data are from Bouloubassi et al. 2001; Chen et al. 2007; Chen et al. 2012; Deng 2013; Deng et al. 2013; Duan et al. 2015; Guo et al. 2007b; Hu et al. 2011a, b; Huang et al. 2012; Hui et al. 2009; Hung et al. 2011; Jiang et al. 2007; Lang et al. 2012; Li et al. 2012; Li et al. 2015; Lin et al. 2011; Liu et al. 2007; Liu et al. 2008a; Liu et al. 2012; Luo et al. 2006; Mai et al. 2001; 2002; Maskaoui et al. 2002; Men et al. 2009; Qin et al. 2011; Qiu et al. 2009; Shi et al. 2005; Wang et al. 2006; Wu et al. 2003; Yan et al. 2009; Zhang et al. 2004; and Zhang et al. 2005. (The detailed information was provided in supporting information)

extraordinarily higher (27.1 μg/g; Jiang et al. 2007). According to the official report (Ministry of Environmental Protection of the People's Republic of China 2013), Haihe River was the most polluted river in China. The Haihe River is located in northern China and close to one of the most industrialized and urbanized city groups (Beijing–Tianjin–Hebei area). It has a vital action in maintaining the rapid development of this area. Therefore, high evaporation, low precipitation, and high emission (city sewage, industrial and medical waste) generated serious local pollution (Jiang et al. 2007). The Bohai Bay has also been heavily polluted by emissions from the Beijing–Tianjin–Hebei metropolitan area (National Bureau of Oceanography of China 2014). The Pearl River estuary sediments were the second most polluted region by PAHs.

Lower trend from north to south is shown in Fig. 2. Geographically from north to south (Fig. 1), four major geographical regions, Bohai Sea (BS), Yellow Sea (YS), East China Sea (ECS), and South China Sea (SCS), were defined. The average concentrations of PAHs in the BS, YS, ECS, and SCS sediments were 348 (excluding Haihe river), 218, 297, and 191 ng/g (excluding Pearl River), respectively (Fig. 2). Overall, the PAH concentrations were reduced from north to south. This pattern was very similar to the spatial distribution of PAH emission density (shown by Zhang and Tao 2008). It means that PAHs in the sediments of China Sea were closely related to the nearby land-based sources. Besides, PAH concentrations observed in the coastal sediments of the three city groups (Beijing–Tianjin–Hebei area, Yangtze River Delta, and Pearl River Delta) were higher than in the adjacent areas. Therefore, the distance from cities was also probably an important factor which affects PAH concentrations in the coastal sediments.

Compositional characteristics and sources of PAHs in coastal sediments

Industrial processes (such as coal combustion) and other human activities (such as vehicle exhaust) were the largest emissions of PAHs (Wolska et al. 2012; De Souza and Corrêa 2015). Surface runoff and atmospheric deposition were considered as the most important inputs of pollutants into coastal marine environment (Soclo et al. 2000). Human activities and environment conditions can greatly impact the concentrations and distributions of PAHs in the environment. Therefore, source identification was exercise crucial to the control strategies determination for environmental pollutants.

The distribution of individual PAH compounds in sediment samples was generally dominated by the three-, four-, and five-ring species (with a mean of 27.2, 37.5, and 18.8 %, respectively; Fig. 3). However, there were some differences between different regions. As shown in Fig. 3, contents of three- and four-ring PAHs in the BS and YS (with a mean of 72.2 and 69.0 %, respectively) were significantly higher than in the ECS and SCS ($p<0.05$; with a mean of 54.7 and 60.2 %, respectively). However, contents of five- and six-ring PAHs in the BS and YS (with a mean of 20.9 and 24.8 %, respectively) were significantly lower than in the ECS and SCS ($p<0.05$; with a mean of 35.5 and 34.5 %, respectively). Therefore, the proportion of lower molecular weight compounds in sediments of the North China Sea was higher than those in

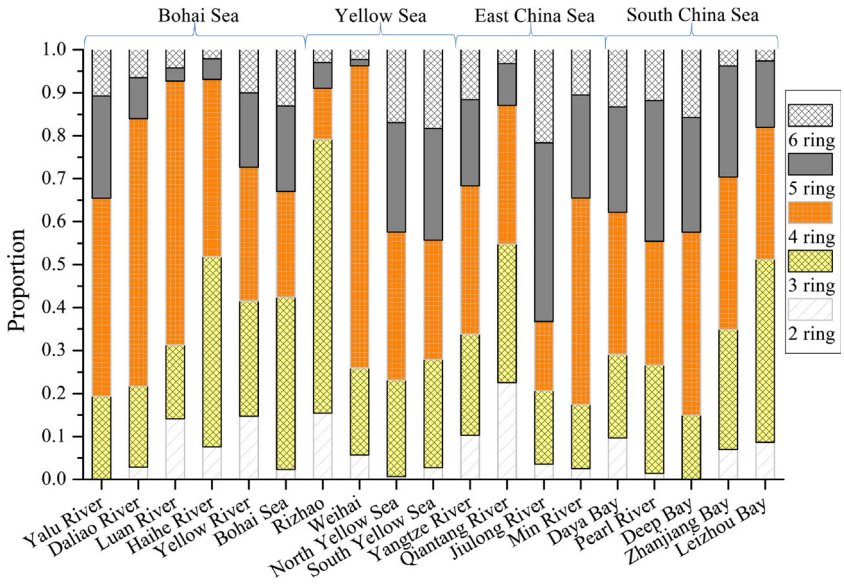

Fig. 3 Compositional characteristics of PAHs in estuarine and coastal sediments of China (listed in order of decreasing north latitude; data are from Wu et al. 2003; Guo et al. 2007b; Jiang et al. 2007; Hui et al. 2009; Chen et al. 2012; Chen et al. 2007; Maskaoui et al. 2002; Zhang et al. 2004; Yan et al. 2009; Luo et al. 2006; Qiu et al. 2009; Qin et al. 2011; Lin et al. 2011; and Huang et al. 2012)

sediments of the South China Sea, and the reverse was true for higher molecular weight compounds. This means that the sources and transportation ways of PAHs in the sediments of the North China Sea were significantly different from that in the South China Sea, as demonstrated by Liu et al. (2012).

Two-ring PAHs were abundant in the samples from the Bohai Bay (Luan River estuary, Haihe River estuary, and Yellow River estuary), Yangtze River estuary, and Qiangtang River estuary. According to previous studies (Yamada et al. 2003; Stogiannidis and Laane 2015), two-ring PAHs were strongly related to petroleum pollution. China's busiest ports (Ningbo–Zhoushan Port, Shanghai Port, and Tianjing Port) are located in these areas. Water transportation may have played an important role on the environment (Li et al. 2010a; Lin et al. 2011). Meanwhile, used crankcase oil was also, probably, an important source of PAHs in the coastal sediments (Zakaria et al. 2002).

Three- and four-ring PAHs in the environment were generally considered to be derived from incomplete combustion of coal, coke, and biomass (Douben Peter 2003). Fluoranthene, acenaphthylene, pyrene, and fluorene were predominant in signals of coal and coke combustion. Benzo[a]anthracene and chrysene were predominant in signals of natural gas combustion (Khalili et al. 1995; Yunker et al. 2002). Incomplete combustion of e-waste (abandoned electronics) also can generate a large amount of three- and four-ring PAH (dominated by phenanthrene and fluoranthene; Yu et al. 2006). In China, the most important energy source is coal, which is used widely for industrial (especially in the steel and power industry) and domestic (e.g., home heating in winter in North China) purposes (Chen et al. 2005). As shown in Fig. 4, the amounts of coal and coke used in North China were much higher than those used in South China. According to Zhang and Tao (2008), the amount of PAHs emission from biofuel combustion in North China was much higher than in other regions. Due to the vast areas of forest in northeastern China (Liaoning, Jilin, and Heilongjiang province; 37 %, more than twice of the average forest cover ratio of China), the emission of PAHs from forest fires in northeastern China made up a contribution of about 15 % of the total emissions of this region during springtime and even more than 25 % in May (Zhang and Tao 2008). Therefore, the higher concentrations (Fig. 2) and the higher proportions of three- and four-ring species (Fig. 3) in North China Sea were probably caused by the higher emissions from the combustions of coal, coke, and biomass.

Vehicle exhaust was an important source of higher molecular weight PAHs. Light-duty vehicles were found to be a significant source of heavier PAHs (such as benzo[g,h,i]perylene), whereas heavy-duty diesel engines were the dominant source of three-ring PAHs (Marr et al. 1999). In addition, atmospheric concentration of benzo[a]pyrene (five-ring PAH) in China was much higher than that reported elsewhere in the world. It may be due to the dominance of coal as a fuel source (Douben Peter 2003). However, six-ring PAHs such as benzo[g,h,i]perylene and indeno[1,2,3-*cd*]pyrene were only detected in the samples of highway tunnel and diesel and gasoline engines rather than coke or wood combustion samples (Khalili et al. 1995). This means that five- and six-ring species were mainly originated from vehicles. There was no significant difference in petroleum consumptions between the north and the south (Fig. 4). Therefore, the higher proportion of five- and six-ring PAHs in the south (Fig. 3) was probably caused by the higher persistence (the degradation of lower molecular weight PAHs by sunlight or microorganism was faster in the south due to the higher temperature; Nadal et al. 2006; Haritash and Kaushik 2009). High evaporation of lower molecular weight PAHs in higher temperature would also contribute to the differences between south and north (Li et al. 2006).

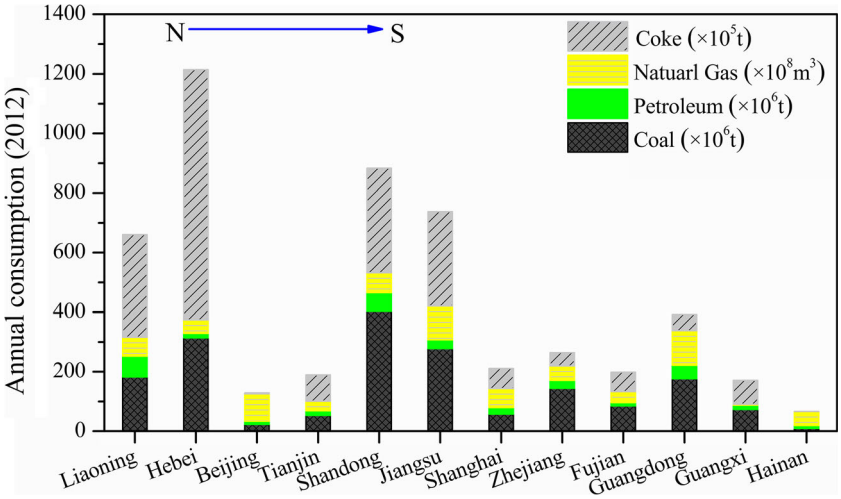

Fig. 4 Annual consumptions of energy products in coastal provinces and directly controlled municipalities (Beijing, Tianjin, and Shanghai) of China in 2012 (National Bureau of Statistics of the People's Republic of China; http://www.stats.gov.cn)

Environmental fate of PAHs in coastal areas of China

The distribution patterns (Fig. 2) and composition characteristics (Fig. 3) of PAHs in the north and south were markedly different, mainly caused by the energy structure difference between the north and south. Differences of environmental fate between the north and south caused by the differences of temperature and/or other factors also played important roles on the distribution of PAHs in coastal China, as discussed above.

Point sources, industrial outfalls, and municipal wastewater facilities were perhaps the most important sources for some estuaries (such as the Haihe River estuary). However, they were trivial in a wider area (Douben Peter 2003). The movement of water derived from watersheds (containing dissolved and particulate-associated pollutant) and atmospheric deposition from airsheds of the coastal ocean (both in precipitation and dry deposition) were primary sources of PAH inputs to the coastal marine environment (Lohmann et al. 2007).

The differences in pattern of air and water samples were clearly observed (Fig. 5). This indicated that air and water played different roles in the geochemical cycles of PAHs in different regions. The PAH distributions in air show a wide range in concentrations (Fig. 5a) and were generally dominated by fluoranthene, phenanthrene, and pyrene. The atmospheric concentrations of PAHs (16 EPA PAHs) increased from north to south, indicating a negative correlation with ambient temperature. Ambient temperature also dictated the energy usage, which in turn would influence the particle loading (Qiu et al. 2003). As shown in Fig. 6, contents of atmospheric fine particles in the cities of North China were significantly higher than those in the cities of South China. The fate of the PAHs in the atmosphere was strongly influenced by whether the PAHs were present in gaseous form or were particle-associated. Most of the PAHs from residential wood combustion (Shen et al. 2013), harbor activities and ship traffic (Donateo et al. 2014), vehicular sources (diesel and gasoline), and coal burning (Teixeira et al. 2012) were associated with fine particulate matter. Small particles were known to remain in the atmosphere for 1 to 2 weeks, thereby allowing long-range transport of particle-associated PAHs (Inomata et al. 2012; Wang et al. 2015).

High molecular weight congeners were mostly present in the fine particulate phase, while lighter PAHs were preferably segregated into the coarse particle matter or associated to the gas phase (Shen et al. 2013). However, a higher percentage of PAHs tend to enrich in coarse particulate and gas phase under higher ambient temperature (Teixeira et al. 2012). PAHs associated to coarse particle or gas phase were more readily photodegraded by sunlight (Bourotte et al. 2005). The atmospheric photochemical reactions will limit the long-range transport of gaseous PAHs (Ram and Anastasio 2009). Therefore, long-range transportation of PAHs was more likely to occur in North China due to the lower ambient temperature.

In winter, air parcels arriving in east coastal China were from the northeast (Yellow Sea) to northwest (inland) directions. Northeast winds from the Yellow Sea were oceanic, but they may not necessarily be clean because the atmosphere over the Yellow Sea could contain continental outflow and polluted discharge from North China (Feng et al. 2006). In the south coastal China, the air parcels were mainly from the South China Sea in the southwest to southeast directions in summer and from the northeast along the Guangdong province coast in winter. Higher wind speeds in North China (70 m above the ground; Fig. 7) also favored the dispersion of pollutants. Therefore, strong winds that blow from the land toward the ocean in North China were conducive to the transportation of pollutants from the land to ocean.

In conclusion, higher loadings of particles and PAHs from coal and coke combustion coupled with lower ambient temperature and strong offshore wind in North China favored the dispersion of PAHs from the land to ocean.

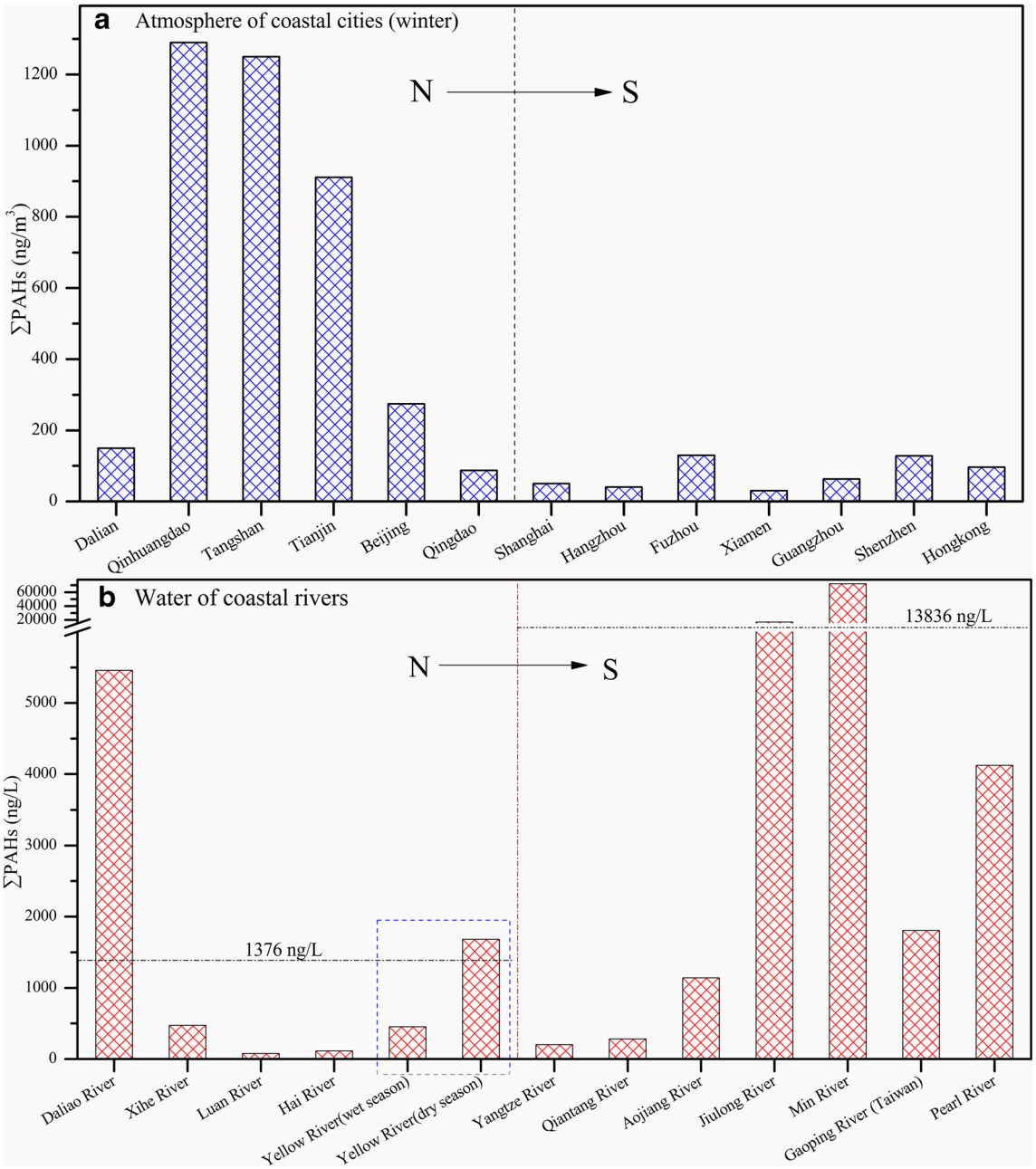

Fig. 5 Distributions of PAHs in atmosphere of coastal cities (**a**, listed in order of decreasing north latitude; data are from Tian et al. 2009; Liu et al. 2008b; Wu et al. 2005; Feng et al. 2006; Guo et al. 2003a; Zhao et al. 2011; Jiao et al. 2009; Liu et al. 2010; Guo et al. 2003b and Yi et al. 2013) and waters from the coastal rivers of China (**b**, listed in order of decreasing north latitude; data are from Guo et al. 2007b; 2011; Li et al. 2010b; Shi et al. 2005; Lang et al. 2008a, b; Zhang et al. 2012; Chen et al. 2007; Li et al. 2010c; Maskaoui et al. 2002; Zhang et al. 2004; Doong and Lin 2004; and Luo et al. 2004)

As shown in Fig. 5b, the total concentrations of PAHs (16 EPA PAHs) in the aqueous phase and suspended particulate matter (SPM) combined ranged from 77.2 to 5458.9 ng/L and 201.1 to 72.3 μg/L in the coastal rivers of North and South China, respectively. In general, concentrations of PAHs in the rivers of South China were much higher than those in the rivers of North China (Fig. 5b).

The most interesting discovery was that the PAH concentrations in the Yellow River in the dry season were higher than those in the flood season (Fig. 5b; Lang et al. 2008b). PAHs in the coastal waters of Gulf of Cambay (India) were also higher in the winter than in the monsoon or summer (Reddy et al. 2005). This indicated that the dilution of PAH concentrations in waters by rain was significant (Olivella 2006). Although rainfall was abundant in south China (Fig. 7), the concentrations of PAHs in the rivers of South China were much higher (Fig. 5b). Therefore, it can be concluded that PAHs were mainly migrated by surface runoff in South China.

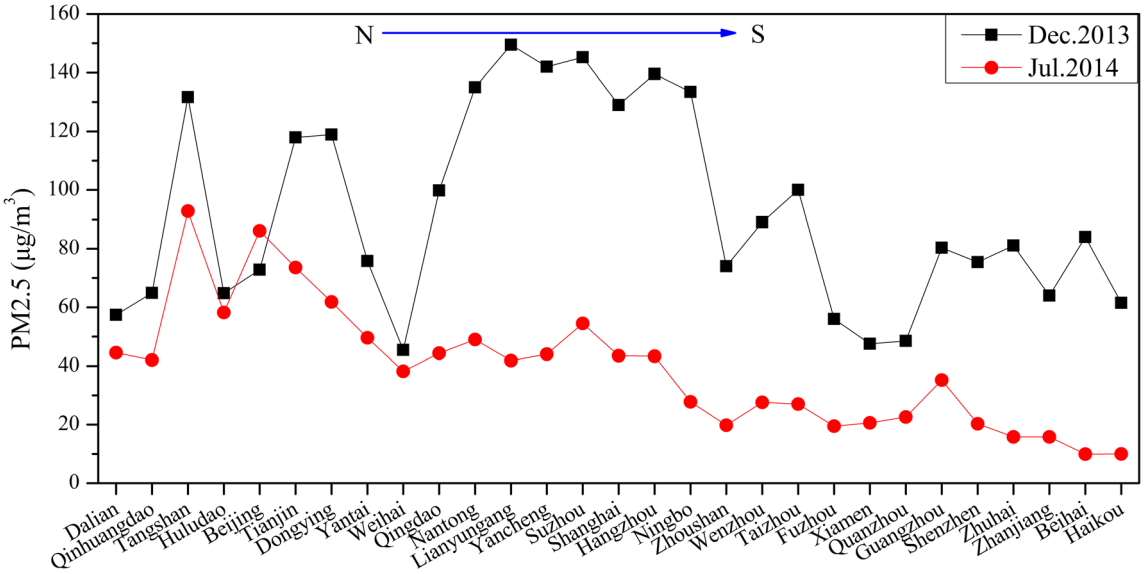

Fig. 6 Contents of atmospheric fine particles ($PM_{2.5}$) in coastal cities of China (data are from http://www.tianqihoubao.com/aqi/)

According to previous studies (Zakaria et al. 2002; Boonyatumanond et al. 2006), the high PAH abundance in the coastal sediments of Thailand and Malaysia were contributed by the rapid transfer of land-based pollutants into aquatic environments by heavy rainfall.

PAH fluxes into the coastal environment of China

Sedimentation accounts for most of the initial removal of PAHs from the water column. Continental shelf sediment was considered as the final sink of persistent organic pollutants. The movement of water derived from watersheds and atmospheric depositions from airsheds of the coastal ocean were primary sources of PAH inputs to the coastal marine environment (Douben Peter 2003). However, it was quite a challenge to evaluate the countrywide riverine and atmospheric PAH inputs because of the different geological settings (i.e., between the north and south) and different numbers of target analytes employed in individual studies (Wang et al. 2007). Based on current available data (Wang et al. 2007; Lang et al. 2008a; Qin et al. 2011; Lin et al. 2013), general fluxes of PAHs in coastal China are shown in Fig. 8.

Based on Fig. 8, atmospheric deposition has an overwhelming advantage over riverine input (5655 tons per year by atmospheric deposition; however, only 232 tons and 33.9 tons per year by Yangtze River and Pearl River, respectively). Previous studies (Lin et al. 2011; Wu et al. 2011) also have indicated that the particle-phase PAHs in the atmosphere play an important role in the source-to-sink process of the pyrogenic PAHs in China Sea. So far, the influence of atmospheric deposition on the coastal environment was hard to assess due to the changeable weather conditions and the obvious regional differences.

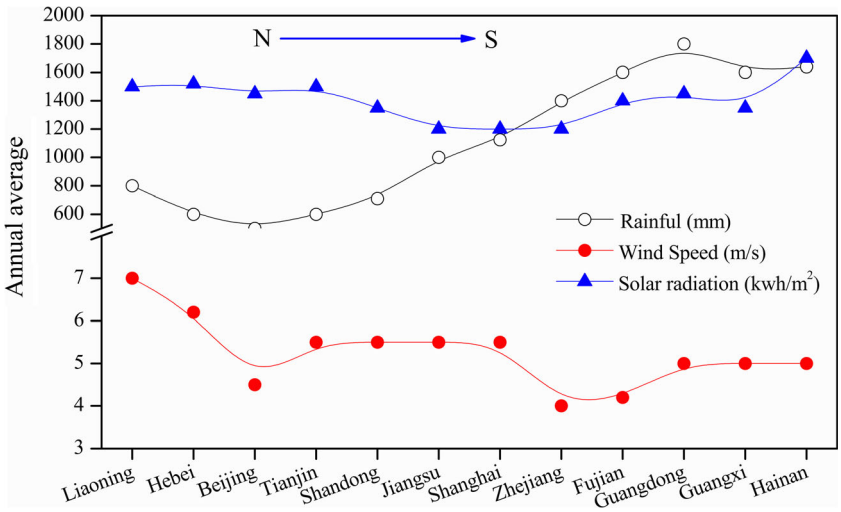

Fig. 7 Climatic conditions of coastal provinces and directly controlled municipalities of China (China Meteorological Administration 2014)

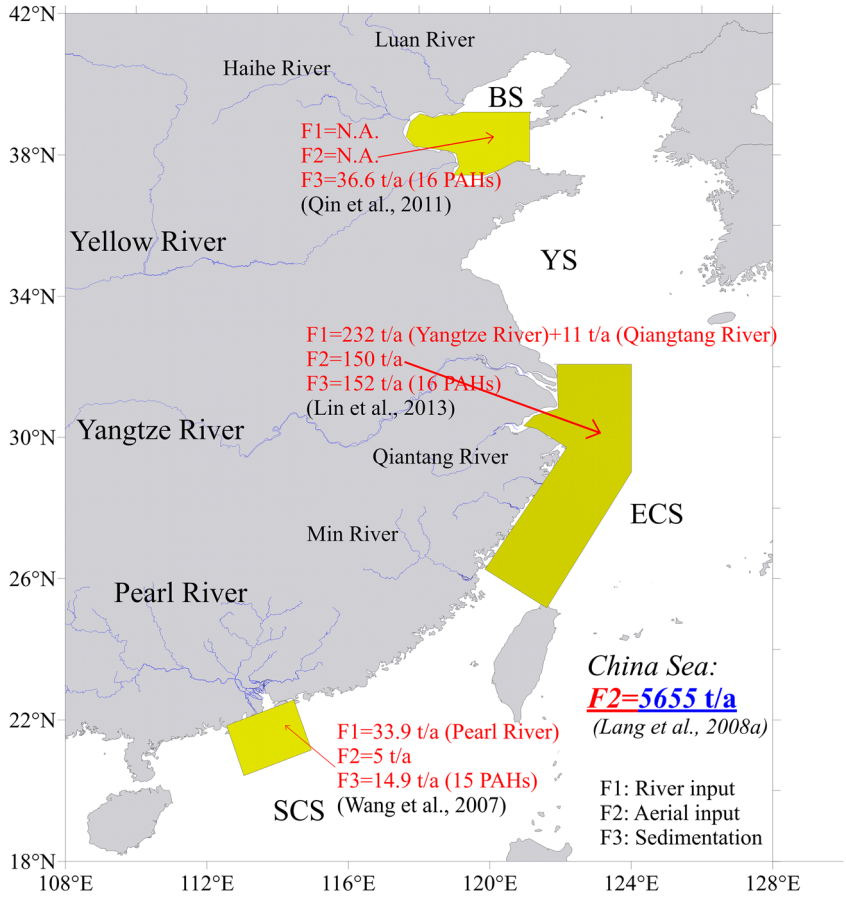

Fig. 8 General fluxes of PAHs in coastal China

The deep-sea was considered as a final sink of semi-volatile and non-volatile organic pollutants (Froescheis et al. 2000); ~38 % of the total annual input of the 16 PAHs into the East China Sea was buried in the Yangtze estuarine–inner shelf (Lin et al. 2013). Based on the radionuclide ($^{210}Pb_{xs}$, $^{234}Th_{xs}$, ^{7}Be) record of the underlying sediments and PAH data, about half of the PAHs introduced to the Portland Harbor may be settling locally, and the remainder was exported to offshore locations (Gustafsson et al. 1998). Based on the results (Froescheis et al. 2000; Gustafsson et al. 1998; Lin et al. 2013), most inputs of PAHs into the coastal sea were probably finally buried in the sediments. As shown in Fig. 8, the contribution of aerial input to concentrations of PAHs in the shelf sediments was far greater than that of riverine inputs (5655 tons/a by atmosphere; 232 and 33.9 tons/a by the Yangtze River and Pearl River, respectively). However, riverine inputs also have significant impact on pollutants in the regions far away from estuary (such as in the central Yellow Sea; Duan et al. 2014); the ocean water even played an important role of OCP sources for the atmosphere (Lin et al. 2012). Therefore, current understanding of the fluxes of PAHs in coastal China remains elusive.

In the current studies (Wang et al. 2007; Lin et al. 2013), the riverine flux was assumed to positively correlate with the regional total emission of PAHs. The flux was estimated based on the monitoring results of only a few times sampling. Therefore, lower frequency monitoring could result in substantial underestimation of flux as the most dynamic periods (such as typhoon events) of transport were missed (Clark et al. 2007). In northern California continental shelf, the net sediment flux during the single 3-day event accounted for seven times the flux observed over an entire year (Ogston et al. 2000). The concentrations of nutrients increased up to three orders of magnitude during a storm event (Correll et al. 1999). The concentrations of pollutants (including dissolved and adsorbed pollutants) also increased markedly (Gardner and Carey 2004; Hwang and Foster 2006). Therefore, the riverine inputs were very likely substantially underestimated, especially in medium and small rivers.

Conclusions

PAH concentrations in coastal China sediments decreased from north to south due to the higher emissions in North China. And the proportions of three- and four-ring PAHs in the sediments of the North China Sea were significantly higher than those in the South China Sea ($p<0.05$). PAHs in the north were highly associated with coal and biomass burning. Atmosphere was probably the main carrier of PAHs in the

north due to the higher contents of atmospheric fine particles and higher wind speeds. Higher proportions of five- and six-ring PAHs in the South China Sea were caused by the degradation and/or volatilization of low molecular` weight PAHs under the higher ambient temperature. Riverine inputs were probably the most important sources of PAHs in the coastal sediments of the South China Sea due to the higher rainfall.

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