Pollution, sources, and ecological-health risks of polycyclic aromatic hydrocarbons in coastal waters along coastline of China

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\textbf{ABSTRACT}
This study investigated the distribution, sources, and potential risks of polycyclic aromatic hydrocarbons (PAHs) in coastal waters along over 18,000 km of coastline in China. Concentrations of PAHs in coastal waters ranged from 141.99 to 717.72 ng/L. Approximately 84.38\% of sampling sites were determined at moderate PAH pollution level. PAHs in coastal waters at most of sampling sites mainly originated from combustion based on characteristic ratios of PAHs. Ecological risks posed by PAHs in coastal waters were evaluated as high level at 59.38\% of sampling sites and moderate level at 40.63\% of sampling sites although toxic equivalent quotients of PAHs only ranged from 2.86 to 126.52 ng/L benzo[a]pyrene that was not detected at all sampling sites. Maximal cancer risk/hazard quotient of total PAHs in coastal waters for adults and children reached $6.34 \times 10^{-4}/5.85 \times 10^{-2}$ and $2.25 \times 10^{-2}/7.72 \times 10^{-2}$, respectively. PAHs exerted high cancer risks for children at 31.25\% of sampling sites. Health risks posed by PAHs in coastal waters of this study were higher than those of Japan, Belgium, Greece, Italy, Spain, USA, and Australia, but much lower than those of Singapore, Iran, Brazil, and Egypt. These findings indicate that PAH pollution has become a crucial stress affecting the sustainable development of coastal regions.

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\textbf{Introduction}
Polycyclic aromatic hydrocarbons (PAHs), ubiquitous in the environment, are a group of over 100 individual persistent organic compounds possessing two or more fused aromatic rings (Barro et al. 2009). PAHs have exhibited potential/proven carcinogenicity and genotoxicity (Capone and Bauer 1992; Purcaro et al. 2013), long-distance transportation features (Pandey et al. 2011), high persistence (Akhbarizadeh et al. 2016; Marini and Frapiccini 2013), poor biodegradation (Akhbarizadeh et al. 2016), high toxicity (McGrath and Di...
Toro 2009; Qin et al. 2013), and significant bioaccumulation (Li et al. 2016) to attract increasing attention. Moreover, 16 PAHs have been selected as priority pollutants by the United States Environmental Protection Agency (USEPA) owing to their carcinogenicity, toxicity, and mutagenicity (Zheng et al. 2016; USEPA 2014). Furthermore, health issues including health risks of PAHs have also become scientific and public-concerning hotspot in recent years (Akhbarizadeh et al. 2016; Qamar et al. 2017; Rajasekhar et al. 2018; Sarria-Villa et al. 2016). Therefore, it is critical to explore the potential ecological and health risks posed by PAHs when paying attention to those pollutants in aquatic systems.

It is generally acceptable that PAHs originate from pyrogenic, petrogenic, and diagenetic sources (Li et al. 2015; Mostafa et al. 2009). Thereafter, PAHs are also categorized into pyrogenic group that mainly originates from incomplete combustion of fuels including coal, petroleum, wood, and grass (Birks et al. 2017; Mostafa et al. 2009), petrogenic group that is mainly from petroleum sources such as fuels, crude oil, and lubricants (Birks et al. 2017; Mostafa et al. 2009), and diagenetic group that is derived from biogenic precursors (Mostafa et al. 2009). Different sources address different distribution of PAHs in the environment (Birks et al. 2017) to exert various impacts to the ecosystem and human beings. Therefore, it is of important concern to determine the sources of PAHs in the environment, especially the aquatic systems.

Coastal regions with intensive land-ocean interactions are not only the critical ecologically fragile regions but also the most important regions for human health and social sustainability because over 45% of people around the world live within approximately 100 km of global coastlines (Zhu et al. 2017). Coastal regions cover 13% of the total landmass and contain 40% of population in China (Meng et al. 2017). They are also the areas with the fastest developing pace and the extensive anthropogenic activities in China. Coastal regions are currently facing two critical issues including rapidly increasing human populations and continually compressed ecosystem services in these ecologically fragile areas (Dennison 2008). Environmental pollution has become a crucial stress affecting the critical ecologically fragile regions such as the coastal zone and the Qinghai–Tibet Plateau due to the rapid economic development and extensive anthropogenic activities (Wang et al. 2018; Wen et al. 2018; Wu et al. 2016; Wu et al. 2018; Zhu et al. 2017). PAHs in the waters of some local bays, gulfs, and lakes have exerted considerable ecological risks to the aquatic ecosystems (Agah et al. 2017; Li et al. 2015; Qin et al. 2013; Ranjbar Jafarabadi et al. 2017). Therefore, information on the pollution and ecological-health risks of PAHs in the coastal waters of the coastal zone at a national or continental scale is critical for regional sustainability. The objectives of this study are to investigate the distribution characteristics, analyze the possible sources, and discuss the potential risks of PAHs in the coastal waters along the over 18,000 km of coastline in China. The final aim is to provide comprehensive insight for the environment pollution and sustainable development in the coastal regions.

**Methods and materials**

**Chemicals and reagents**

The 16 priority PAH congeners including naphthalene (Naph), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Anth), pyrene...
(Pyr), fluoranthene (Flt), benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno [1,2,3-cd]pyrene (InP), dibenzo[a,h]anthracene (DbA), and benzo[g,h,i]perylene (BghiP) were analyzed. Standard mixture stock solution of 16 PAH congeners with concentration of 1000 μg/L and mixture stock solution of deuterated internal standards including phenanthrene-d10, chrysene-d12, naphthalene-d8, perylene-d12, and acenaphthene-d10 with concentration of 2000 mg/L were purchased from o2si smart solutions, LLC (Charleston, SC, USA). Dichloromethane, n-hexane, and methanol used for sample preparation and analysis were HPLC grade and obtained from MREDA (Mreda Technology Inc., USA). Anhydrous sodium sulfate was baked at 450°C for 8 h and stored in sealed containers.

**Study area and sample strategy**

Sampling was performed in November of 2017. Mixed-surface coastal water sample with volume of 30 L was composed of six subsamples with volume of 5 L collected along the coastline of each sampling site at the distance interval of 5 m by using pre-cleaned amber glass sample bottles and then being quickly transported back to the laboratory for further analysis. Coastal water samples were collected from 32 sampling sites along Chinese coastline covering all the four coastal regions in China including Bohai Area (B1–B8), Yellow Sea Area (Y1–Y11), East China Sea Area (E1–E6), and South China Sea Area (S1–S7) (Figure S1 in the online supplementary information (SI)). Over 45% of global people live in the coastal regions, suggesting that some functional zones such as estuaries, gulfss, bays, zones, bathing beaches, and ports are critical to human health and ecosystems. Moreover, mariculture has become one of the most important aquacultures in the coastal regions (Gao et al. 2012), illustrating that mariculture also has important impacts on human health and sustainable ecosystems. Therefore, sampling locations involved the main estuaries, gulfss, bays, maricultural zones, bathing beaches, and ports along the coastline in China to represent the important functional zones of the coastal regions affecting the ecosystem and human health.

**Sample preparation and analysis**

The water samples with volume of 1 L were firstly filtered using 0.45-μm membrane filters (Pall Life Sciences, Ann Arbor, MI, USA). Subsequently, the filtrated samples were spiked with a 5 μL of deuterated internal standard mixture solution (100 mg/L) before solid phase extraction (SPE) to compensate the loss of target analytes during the extraction process as well as analysis procedure. The water samples were percolated under very low vacuum through the Oasis HLB 6cc/200 mg cartridges (Waters Corp., Mill-ford, MA) that were conditioned with 10 mL of dichloromethane, 10 mL of methanol, and 10 mL of ultrapure-water. After extraction, 10 mL of dichloromethane was passed through the cartridges to elute and yield a fraction containing PAHs. The eluent was concentrated to nearly dry under a gentle nitrogen stream after passing through anhydrous sodium sulfate and re-dissolved in 1 mL of n-hexane for Gas Chromatography-Mass Spectrometer (GC-MS) analysis. Samples were analyzed by Agilent 7820A GC system (Agilent technologies Inc., Palo Alto, CA, USA) with a M7 single quadrupole MS system from Persee (Beijing, China).
A 30 m × 0.25 mm I.D. DB-5MS column (Agilent J&W Scientific, Folsom, CA, USA) coated with 5% diphenylpoly dimethyl siloxane (film thickness 0.25 μm) was employed for separation of target compounds during analysis. The injector was operating at 280°C in splitless mode. Helium (>99.999% pure) was used as carrier gas at 1.0 mL/min during the whole run. The temperatures of the transfer line and ion source were held at 280 and 230°C, respectively. The column oven temperature program started at 40°C for 3 min, then programed to 200°C at 25°C/min, and held for 5 min, finally to 290°C at 8°C/min rate, and held for 10 min to comprise a total runtime of 35.65 min, including a 7-min solvent cut for all analyses.

All target compounds were identified by full scan mode (m/z 50-400) based on their mass spectra and GC retention times. Subsequent acquisition and quantification were performed by a time scheduled selective ion monitoring (SIM) program. The quantitative and qualitative ions for 16 PAHs were presented in Table S1.

**Quality control and quality assurance**

Quality control procedures in the laboratory included the analyses of method blanks (solvent), spiked blanks (solvent spiked by PAH standard mixture solution), and sample in duplicate. The recovery was checked by analyzing water samples spiked with known amount of PAH standard. Recoveries of 16 PAHs were in the range of 79.68%–116.84%. Concentrations of PAHs were all corrected according to the recoveries of internal standards. The limit of detection (LOD) for PAHs ranged from 0.34 to 3.81 ng/L. The information on retention time, mean recovery, relative standard deviation (RSD), and LOD was shown in Table S1.

**Source apportionment**

Several molecular ratios can serve as indictors to determine the potential sources of PAHs in water (Agah et al. 2017; Akhbarizadeh et al. 2016; Birks et al. 2017; Budzinski et al. 1997; Li et al. 2015; Li et al. 2017; Liu et al. 2016; Ranjbar Jafarabadi et al. 2017; Yuan et al. 2017; Yunker et al. 2002; Zhang et al. 2016; Zheng et al. 2016). This study selected molecular ratios of Flt/(Flt + Pyr), BaA/(BaA + Chry), Anth/(Anth + Phe), InP/(InP + BghiP), Flt/Pyr, Phe/Anth, and LMW (low-molecular-weight PAHs, 2–3 rings)/HMW (high-molecular-weight PAHs, 4–6 rings) to evaluate the origin of PAHs in coastal waters. The diagenetic source of PAHs was not discussed in this study because this source is scarce in the environment (Mostafa et al. 2009) and typical indicator (perylene) that is not listed in the priority pollutants was not monitored. Detailed characteristic values of selected ratios for source apportionment were referred to Table S2.

**Ecological risk assessment**

Potential ecological risks of PAHs in coastal waters were evaluated using risk quotients (RQs). RQs of PAHs were calculated according to the following equations (Cao et al. 2010; Li et al. 2015; Ranjbar Jafarabadi et al. 2017; Sun et al. 2009):
\[
RQ_{i}(NCs) = \frac{C_i}{C_{i,QV}(NCs)}
\]

\[
RQ_{i}(MPCs) = \frac{C_i}{C_{i,QV}(MPCs)}
\]

\[
RQ_{i}(NCs) = \sum_{i=1}^{n} RQ_{i}(NCs) \left( RQ_{i}(NCs) \geq 1 \right)
\]

\[
RQ_{i}(MPCs) = \sum_{i=1}^{n} RQ_{i}(MPCs) \left( RQ_{i}(MPCs) \geq 1 \right)
\]

where \( RQ_{i}(NCs) \) and \( RQ_{i}(MPCs) \) refer to risk quotients based on the negligible concentrations (NCs) and the maximum permissible concentrations (MPCs) of individual PAHs in water, respectively; \( C_i \) refers to the concentration of individual PAHs in water; \( C_{i,QV}(NCs) \) and \( C_{i,QV}(MPCs) \) are the quality values of NCs and MPCs of PAHs in water, respectively; \( n \) is the number of PAH congeners; \( RQ_{i}(NCs) \) and \( RQ_{i}(MPCs) \) refer to sum of individual risk quotient that is equal or greater than 1 based on NCs and MPCs of individual PAHs in water, respectively. The equivalent values of NCs and MPCs of individual PAHs in water and risk classification thresholds are referred to Tables S3 and S4. Pollution of PAHs in water can be classified into four levels according to criterion listed in Table S4 (Li et al. 2015). Therefore, this study also evaluated pollution levels of PAHs in coastal waters.

**Toxic evaluation**

The toxicity of each PAH congener was calculated based on toxic equivalent factor (TEF) method and the total toxicity of PAHs was the sum of individual PAH congener toxicity as the following equation (Cao et al. 2010; Li et al. 2015; Qamar et al. 2017; Zheng et al. 2014):

\[
TEQ = \sum_{i=1}^{n} C_i \times TEF_i
\]

where \( TEQ \) is toxic equivalent quotient; \( TEF_i \) is the toxic equivalent factor of each PAH congener relative to BaP. The \( TEF \) values were listed in Table S4 and the maximum values were taken if \( TEF \) values of the same congener appeared variously in different references.

**Health risk assessment**

Dermal contact such as swimming, surfing, diving, and working in surface seawater is the main health exposure path of PAHs in coastal waters because coastal waters are not suitable to drink. Therefore, this study adopted cancer and non-cancer risks through dermal contact to evaluate the potential health risks of PAHs. The detailed information on cancer risk (CR) and hazard quotient (HQ) of individual pollutant is shown by the following equations (Akhbarizadeh et al. 2016; Rajasekhar et al. 2018; Sarria-Villa et al. 2014):
$\text{CR}(\text{individual}) = \text{DAD} \times \frac{\text{SFO}}{\text{GIABS}} = \text{DA}_{\text{event}} \times \frac{\text{EV} \times \text{ED} \times \text{EF} \times \text{SA}}{\text{BW} \times \text{AT}} \times \frac{\text{SFO}}{\text{GIABS}}$

$= 2 \times \text{FA} \times K_p \times C_W \times \sqrt{\frac{6 \times \tau_{\text{event}} \times t_{\text{event}}}{\pi}} \times \frac{\text{EV} \times \text{ED} \times \text{EF} \times \text{SA}}{\text{BW} \times \text{AT}} \times \frac{\text{SFO}}{\text{GIABS}}$

$\text{HQ}(\text{individual}) = \text{DAD} \times \frac{1}{\text{RfDO} \times \text{GIABS}} = \text{DA}_{\text{event}} \times \frac{\text{EV} \times \text{ED} \times \text{EF} \times \text{SA}}{\text{BW} \times \text{AT}} \times \frac{1}{\text{RfDO} \times \text{GIABS}}$

$= 2 \times \text{FA} \times K_p \times C_W \times \sqrt{\frac{6 \times \tau_{\text{event}} \times t_{\text{event}}}{\pi}} \times \frac{\text{EV} \times \text{ED} \times \text{EF} \times \text{SA}}{\text{BW} \times \text{AT}} \times \frac{1}{\text{RfDO} \times \text{GIABS}}$

$\text{CR} = \sum \text{CR}(\text{individual})$

$\text{HQ} = \sum \text{HQ}(\text{individual})$

where \text{DAD} and \text{DA}_{\text{event}} refer to dermal absorbed dose and absorbed dose per event, respectively; \text{EV}, \text{ED}, and \text{EF} refer to the event frequency, exposure duration, and exposure frequency, respectively; \text{SA} represents skin surface area; \text{BW} is body weight; \text{AT} represents average lifespan; \text{SFO} is oral slope factor; \text{GIABS} refers to the fraction of pollutant absorbed in gastrointestinal tract; \text{RfDO} is oral reference dose; \text{FA} refers to the fraction of absorbed water; \text{KP} stands for dermal permeability coefficient of pollutant; \text{CW} refers to concentration of PAHs in coastal water; \tau_{\text{event}} is lag time per event; \text{t}_{\text{event}} refers to event duration. The values of parameters were obtained from the references (Akhbarizadeh et al. 2016; Man et al. 2013; Rajasekhar et al. 2018; Sarria-Villa et al. 2016; USEPA 2004; USEPA 2016) and listed in Table S5.

To evaluate the level of the health risks posed by PAHs at a global scale, data on the highest concentrations of PAHs in coastal waters of other countries were cited from literatures (Ahmed et al. 2017; Akhbarizadeh et al. 2016; Cocci et al. 2017; Monteyne et al. 2013; Obbard et al. 2007; Sánchez-Avila et al. 2010; Sankoda et al. 2017; Shaw et al. 2004; Silva et al. 2007; Valavanidis et al. 2008; Williams et al. 2017) to assess health risks of PAHs for adults and children living in corresponding countries. The highest concentration of PAHs in this study was adopted to evaluate health risks for comparison.
Results and discussion

**Distribution features of PAHs in coastal waters along Chinese coastline**

The total concentrations of PAHs in coastal waters ranged from 141.99 to 717.72 ng/L with the average value of 390.06 ng/L (Figure 1a). The concentration ranges of 2-ring, 3-ring, 4-ring, 5-ring, and 6-ring PAHs were 1.52-269.52, 23.55-475.28, 17.02-197.12, ND (not detected)-292.45, and 7.63-165.19 ng/L with the average concentrations of 69.32, 129.83, 75.33, 75.06, and 40.53 ng/L, respectively (Figure 1a). Composition proportions of 2-ring, 3-ring, 4-ring, 5-ring, and 6-ring PAHs were in the ranges of 0.47%–43.07%, 13.68%–70.21%, 7.58%–41.52%, 0.00%–44.84%, and 2.17%–39.42% with average values of 16.10%, 34.30%, 20.66%, 19.03%, and 9.91%, respectively. Concentrations of LMW PAHs ranged from 44.95 to 516.43 ng/L with the average value of 199.15 ng/L, while those of HMW PAHs were in the range of 69.12–470.27 ng/L with a mean value of 190.92 ng/L. The maximal proportion of LMW and HMW PAHs reached 77.05% and 70.37% with average proportions of 50.40% and 49.60%, respectively. Interestingly, PAHs in coastal waters near many beaches such as Y5-Y7 and S3-S5 were mainly composed by LMW PAHs which were easy to diffuse in coastal waters from the adjacent area and might be also introduced by pleasure-boats and/or

![Figure 1](image_url)

Figure 1. Concentrations (a) and composition box plot of PAHs (b) in coastal waters of the study area. LP, MP, and HP refer to light pollution, moderate pollution, and heavy pollution, respectively. In each box, the bottom and top of the box illustrate the 25th and 75th percentiles; the mid-line of box means the median value; the small square represents the average value of the target compound; the bottom and top of the whiskers refers to the minimal and maximal concentrations.
aquaculture vessels. Distribution and composition of PAHs in coastal waters along Chinese coastline exhibited significant site-specific feature (Figure 1). PAHs in most of coastal water samples collected from Haihe Estuary (B4) and Shandong Peninsula (B6, B7, B8, Y3, Y4, Y5, Y6, Y7, and Y8) showed high concentrations.

Concentrations of individual LMW PAH congeners ranged from below detection limit (Acy, Ace, Flu, and Anth at multiple sites) to 269.52 (Naph at Y5) ng/L while those of individual HMW PAH congeners ranged from below detection limit (Pyr, Flt, BaA, Chry, BbF, BkF, BaP, InP, and DbA at multiple sites) to 204.90 (BbF at E4) ng/L (Figures 1b and S2). BaP was not detected at all sampling sites, while Naph, Phe, BghiP, InP, and DbA were the most frequently detected with detection percents of 100%, 100%, 100%, 93.75%, and 93.75%, respectively. Generally, 3-ring PAHs served as the dominant congeners of LMW PAHs, contributing to 50.23%–99.00% of LMW PAHs for 75.00% of sampling sites, whereas 4-ring and 5-ring PAHs were the dominant groups for HMW PAHs, contributing to 59.76%–95.09% of HMW PAHs for 93.75% of sampling sites.

Based on pollution evaluation criterion (Li et al. 2015), 84.38% of sampling sites were determined at moderate PAH pollution, while the rest were at light pollution level (Figure 1a). Considering coastal water samples were all collected from the sites with extensive anthropogenic activities, the pollution status deserved more attention.

Source apportionment of PAHs in coastal waters along Chinese coastline

Seven characteristic ratios were adopted for source apportionment of PAHs in coastal waters (Figure 2) to avoid the possible false-positive and false-negative phenomena caused by single index. For ratios with value of 0 or error (divided by 0) due to non-detected congeners, source of PAHs was determined according to the remaining ratios. PAHs of some sampling sites such as B1-B8, Y1-Y2, E4-E6, and S1-S7 mainly originated from coal and biomass combustion based on identical evaluation results of Flt/(Flt + Pry) and InP/(InP + BghiP). PAHs of some sampling sites such as Y4-Y6, Y10-Y11, and E1-E3 possibly originated from combustion of petroleum, coal, and biomass due to inconsistent evaluation results of Flt/(Flt + Pry) and InP/(InP + BghiP). PAHs of Y9 might originate petroleum combustion based on results of InP/(InP + BghiP). The combustion was the main source of PAHs for most of sampling sites along the coastline, indicating the great influence of anthropogenic activities on the pollution of coastal waters. The evaluation results were reasonable because sampling occurred in late autumn and early winter during which combustion frequently occurred for heating. According to previous investigation, combustion had been identified as the main source of PAHs in the South China Sea (Cai et al. 2017).

Potential ecological risks of PAHs in coastal waters along Chinese coastline

Potential ecological risks of individual PAH congeners and total PAHs in coastal waters of the study area were calculated and expressed as risk quotients including \( RQ_{(NC)} \) and \( RQ_{(MPC)} \) (Table 1 and Figure S3). Except BaP that was not detected in this study, \( RQ_{(NC)} \) of individual PAHs ranged from ND to 2049.03 (BbF at E4) with the average values from 1.02 (Chry) to 574.86 (BbF), while \( RQ_{(MPC)} \) of individual PAHs ranged
Figure 2. Cross plots of PAHs for the ratios of (a) Anth/(Anth + Phe) vs. Flt/(Flt + Pyr), (b) BaA/(BaA + Chry) vs. Flt/(Flt + Pyr), (c) InP/(InP + BghiP) vs. Flt/(Flt + Pyr), (d) Phe/Anth vs. Flt/(Flt + Pyr), (e) Flt/Pyr vs. Flt/(Flt + Pyr), and (f) LMW/HMW vs. Flt/(Flt + Pyr).

Table 1. Statistical summary of ecological risk assessment on PAHs in coastal waters.

| Chemical          | RQ_{INCI} or RQ_{INCG} Mean | Min | Med | Max | Mean | Min | Med | Max | Low | Moderate | High |
|-------------------|-------------------------------|-----|-----|-----|------|-----|-----|-----|-----|----------|------|
| Naphthalene (Naph)| 5.78                          | 0.13| 6.31| 22.46| 0.06 | 0.001| 0.06| 0.22| 15.63| 84.38    | 0.00 |
| Acenaphthylene (Acy)| 36.12                       | 0.00| 23.79| 330.16| 0.36 | 0.00| 0.24| 3.30| 18.75| 75.00    | 6.25 |
| Acenaphthene (Ace)| 24.89                        | 0.00| 17.50| 208.28| 0.25 | 0.00| 0.17| 2.08| 25.00| 68.75    | 6.25 |
| Fluorene (Flu)   | 39.66                        | 0.00| 39.77| 279.20| 0.40 | 0.00| 0.40| 2.79| 34.38| 62.50    | 3.13 |
| Phenanthrene (Phe)| 11.91                        | 0.00| 10.75| 29.25| 0.12 | 0.02| 0.17| 2.08| 25.00| 68.75    | 6.25 |
| Anthracene (Anth)| 33.76                        | 0.00| 43.20| 160.50| 0.34 | 0.00| 0.43| 1.61| 37.50| 59.38    | 3.13 |
| Pyrene (Pyr)     | 37.35                        | 0.00| 37.62| 156.07| 0.37 | 0.00| 0.38| 1.56| 18.75| 78.13    | 3.13 |
| Fluoranthene (Flt)| 9.40                         | 0.00| 11.18| 29.29| 0.09 | 0.00| 0.11| 0.29| 25.00| 75.00    | 0.00 |
| Benz[a]anthracene (BaA)| 175.18                   | 0.00| 271.21| 460.70| 1.75 | 0.00| 2.71| 4.61| 46.88| 63.13    | 0.00 |
| Chrysene (Chry)  | 1.02                         | 0.00| 0.00| 20.02| 0.01 | 0.00| 0.00| 0.20| 87.50| 12.50    | 0.00 |
| Benzo[b]fluoranthene (BbF)| 574.86                  | 0.00| 555.30| 2049.03| 5.75 | 0.00| 5.55| 20.49| 28.13| 0.00     | 71.88 |
| Benzo[k]fluoranthene (BkF)| 43.93                     | 0.00| 3.32| 383.15| 0.44 | 0.00| 0.03| 3.83| 40.63| 53.13    | 6.25 |
| Benzo[a]pyrene (BaP)| 0.00                        | 0.00| 0.00| 0.00| 0.00 | 0.00| 0.00| 0.00| 100.00| 0.00     | 0.00 |
| Indeno[1,2,3-cd]pyrene (InP)| 35.84                    | 0.00| 17.19| 128.46| 0.36 | 0.00| 0.17| 1.28| 6.25 | 31.25    | 62.50 |
| Dibenzo[a,lanthracene (DbA)| 31.07                      | 0.00| 12.22| 144.47| 0.31 | 0.00| 0.12| 1.44| 6.25 | 84.38    | 9.38 |
| Benzo[ghi]perylene (BghiP)| 35.51                      | 0.54| 23.56| 147.05| 0.36 | 0.00| 0.24| 1.47| 0.00 | 60.00    | 9.38 |
| ΣPAHs            | 1096.22                      | 470.16| 1102.73| 2491.94| 8.49 | 1.753| 8.47| 22.74| 0.00 | 46.38    | 59.38 |

Note: Min, Med, and Max mean minimal value, median value, and maximal value, respectively.
from ND to 20.49 (BbF at E4) with the average values from 0.01 (Chry) to 5.75 (BbF). According to evaluation criterion (Cao et al. 2010), BaP showed the lowest ecological risks since it was not detected. HMW PAHs including BbF, BaA, InP, DbA, and BghiP were the individual PAH congeners that exhibited high ecological risks to the ecosystems. BbF and BaA exerted high risks to 71.88% and 53.13% of sampling sites when comprehensively considering $RQ_{(NC)}$ and $RQ_{(MPC)}$, respectively. InP, DbA, and BghiP exerted high risks to 12.50%, 9.38%, and 9.38% of sampling sites and moderate risks to 81.25%, 84.38%, and 90.63% of sampling sites, respectively. LMW PAH congeners mainly exhibited moderate ecological risks to the ecosystems.

Figure 3. Toxic equivalent quotients and corresponding compositions for (a) total PAHs, (b) low-molecular-weight (LMW) PAHs, and (c) high-molecular-weight (HMW) PAHs in coastal waters.
Maximal $RQ_{(NC)}$ and $RQ_{(MPC)}$ of total PAHs reached 2491.94 and 22.74 with the average values of 1096.22 and 8.49, respectively. HMW PAHs mainly including BbF and BaA contributed significant proportion to $RQ_{(MPC)}$ of total PAHs. All individual PAH congeners with concentrations exceeding their corresponding NCs contributed $RQ_{(NC)}$ of total PAHs. Total PAHs exerted high risks to 59.38% of sampling sites and moderate risks to 40.63% of sampling sites based on $RQ_{(NC)}$ and $RQ_{(MPC)}$, more serious than individual congeners. Complex relationship might exist among the individual PAHs when they co-occurred in the environment and exerted potential risks to the ecosystems. Therefore, it is more reasonable to use $RQ_{(NC)}$ and $RQ_{(MPC)}$ of total PAHs to explore their potential ecological risks.

Toxicity of PAHs in coastal waters along Chinese coastline

Toxicity of PAHs in coastal waters was expressed as TEQs relative to the reference congener BaP that is a carcinogenic compound (Figure 3). The TEQs of total PAHs ranged from 2.86 to 126.52 ng/L BaP with the average value of 39.99 ng/L BaP. HMW PAHs were the dominant toxicity contributor of PAHs, accounting for 94.41%–99.87% of total TEQs for sampling sites except Y8 where DbA and InP were not detected (Figure 3a). Except six sampling sites (B6, Y3, Y4, Y5, Y7, and S2), 3-ring PAHs served as dominant contributor for toxicity of LMW PAHs in the remaining sampling sites (Figure 3b). These six sampling sites included all types of functional zones, exhibiting site-specific features of toxicity of LMW PAHs. Except eight sampling sites (Y8, Y10, Y11, E4, E5, S1, S3, and S4), 6-ring PAHs accounts for over 50% of toxicity of HMW PAHs with maximal contribution proportion of 99.50% (Figure 3c). For sampling sites including Y10, Y11, E4, E5, S1, S3, and S4, 5-ring PAHs were dominant toxicity contributor of HMW PAHs, while 4-ring PAHs accounted for 73.09% of toxicity of HMW PAHs at Y8. These eight sampling sites included all types of functional zones, also exhibiting site-specific features of toxicity of HMW PAHs.

Owing to TEF values of 1, InP and DbA served as the main toxicity contributors for total PAHs. Moreover, BbF was also an important toxicity contributor due to its relatively high detection frequency, concentrations, and TEF value. Interestingly, BaP that is one of the most common PAHs was not detected in this study. Since BaP is one of the most toxic compounds among PAHs, it is safe to argue that the toxicity of PAHs in the coastal waters along the coastline of China should increase further when BaP existed with relatively high concentrations.

Potential health risks of PAHs in coastal waters along Chinese coastline

Potential health risks of PAHs in coastal waters of the study area for both children and adults were express as cancer risks and hazard quotients (Figures 4, S4, and S5). Cancer risks of total PAHs in coastal waters for adults and children were in the ranges of $8.24 \times 10^{-5} – 6.34 \times 10^{-4}$ and $2.93 \times 10^{-4} – 2.25 \times 10^{-3}$ with the average values of $2.65 \times 10^{-4}$ and $9.40 \times 10^{-4}$, respectively (Figure 4a). Based on ranking criterion (Ge et al. 2013), cancer risks of PAHs in coastal waters at 93.75% and 6.25% of sampling sites for adults were classified into moderate and low levels, respectively. In contrast,
cancer risks of PAHs in coastal waters at 68.75% and 31.25% of sampling sites for children were classified into moderate and high levels, respectively. Moreover, cancer risks of PAHs for children were nearly 3.6 times those for adults, illustrating that children will be more susceptible to potential harm posed by PAHs. Except BaP that was not detected, cancer risks posed by individual PAH congeners for adults/children ranged from negligible/negligible to $2.45 \times 10^{-4}/8.71 \times 10^{-4}$ (BbF) with average values of $(2.38 \times 10^{-6} - 6.88 \times 10^{-5})/(8.45 \times 10^{-6} - 2.44 \times 10^{-4})$, respectively (Figures S4, S5). Except Y8, HMW PAHs served as the dominant health risk contributor in the remaining sites, accounting for 57.36%–95.62% of total cancer risks. Cancer risks posed by 3-ring PAHs accounted for 74.30%–99.87% of those posed by LMW PAHs at all sampling sites while cancer risks of 5-ring and 6-ring PAHs contributed to 59.94%–97.05% of cancer risks posed by HMW PAHs at sites except Y8. Cancer risks of PAHs exhibited the site-specific and compound-specific distribution features so that the risk control measures should be unique for different sites.

Hazard quotients of total PAHs in coastal waters for adults and children ranged from $7.59 \times 10^{-3}$ to $5.85 \times 10^{-2}$ and from $1.00 \times 10^{-2}$ to $7.72 \times 10^{-2}$ with the average values of $2.44 \times 10^{-2}$ and $3.22 \times 10^{-2}$, respectively (Figure 4b), all lower than risk safe threshold value of 1.0. Moreover, hazard quotients of PAHs for children were about 1.3 times those for adults, illustrating cancer risks of PAHs were more serious than non-cancer risks for children who are still more susceptible to non-cancer harm of PAHs than adults. Except BaP that was not detected, hazard quotients of individual PAH congeners ranged from negligible to $2.26 \times 10^{-2}$ and from negligible to $2.98 \times 10^{-2}$ with the average values of $2.19 \times 10^{-4} - 6.34 \times 10^{-3}$ and

![Figure 4. Potential cancer risks (a) and hazard quotients (b) of PAHs in coastal waters.](image-url)
2.89 × 10⁻⁴–8.37 × 10⁻³ for adults and children, respectively (Figures S4, S5). Similar to distribution feature of cancer risks, HMW PAHs mainly contributed to hazard quotients, while 3-ring PAHs served as the dominant contributor for non-cancer risks posed by LMW PAHs and 5-ring and 6-ring PAHs accounted for over 59% of non-cancer risks posed by HMW PAHs.
Comparison on potential health risks of PAHs in coastal waters at a global scale

Concentrations, potential cancer risks, and hazard quotients of PAHs in coastal waters around the world are illustrated in Figure 5. Concentrations of PAHs in coastal waters around the world ranged from 3.56 ng/L (Japan) to 1.25 mg/L (Egypt), showing drastic fluctuation. Accordingly, health risks posed by PAHs in coastal waters around the world also exhibited significant variation. Cancer risks of PAHs ranged from $3.57 \times 10^{-6}$ (Japan) to $7.93 \times 10^{-1}$ (Egypt) for adults and from $1.27 \times 10^{-5}$ (Japan) to $2.82$ (Egypt) for children. Hazard quotients of PAHs ranged from $3.30 \times 10^{-4}$ (Japan) to $73.15$ (Egypt) for adults and from $4.35 \times 10^{-4}$ (Japan) to $96.56$ (Egypt) for children, respectively. PAHs in coastal waters of Egypt posed very high cancer risks and unacceptable non-cancer risks for both adults and children. PAHs in coastal waters of Iran, Singapore, and Brazil posed high cancer risks for adults, while those from Iran, Singapore, Brazil, and China exerted high cancer risks for children. PAHs in coastal waters of Japan, Belgium, Italy, and Australia posed low cancer risks for adults, while those from China, Greece, Spain, and USA exerted moderate cancer risks for adults. PAHs in coastal waters of Japan and Australia posed low cancer risks for children, while those from Belgium, Greece, Italy, Spain, and USA exerted moderate cancer risks for children. Except Egypt, non-cancer risks posed by PAHs in coastal waters from other countries were at acceptable levels. Health risks posed by PAHs in this study were higher than those from Japan, Belgium, Greece, Italy, Spain, USA, and Australia but much lower than those from Singapore, Iran, Brazil, and Egypt.

Conclusions

Concentrations of PAHs in coastal waters along Chinese coastline were in the range of 141.99–717.72 ng/L, exhibiting that moderate and light PAH pollution occurred at 84.38% and 15.62% of sampling sites, respectively. PAHs in most of coastal water samples collected from Haihe Estuary and Shandong Peninsula showed relatively high concentrations. Naph, Phe, BghiP, InP, and DbA were the most frequently detected PAH congeners in coastal waters along the coastline. PAHs in coastal waters at most of sampling sites mainly originated from combustion based on characteristic-ratio source apportionment. PAHs exerted high ecological risks to 59.38% of sampling sites and moderate risks to 40.63% of sampling sites based on risk quotients. Toxic equivalent quotient of PAHs ranged from 2.86 to 126.52 ng/L BaP with InP and DbA serving as main toxicity contributors for total PAHs. Cancer risks of total PAHs in coastal waters for adults and children were in the ranges of $8.24 \times 10^{-5}$–$6.34 \times 10^{-4}$ and $2.93 \times 10^{-4}$–$2.25 \times 10^{-3}$. Cancer risks of PAHs in coastal waters at 93.75% of sampling sites for adults were classified into moderate level, while those at 31.25% of sampling sites for children were classified into high level. Hazard quotients for adults and children ranged from $7.59 \times 10^{-3}$ to $5.85 \times 10^{-2}$ and from $1.00 \times 10^{-2}$ to $7.72 \times 10^{-2}$ to exert acceptable non-cancer risks. Potential health risks of PAHs in coastal waters along coastline of China were higher than those of Japan, Belgium, Greece, Italy, Spain, USA, and Australia, but much lower than those of Singapore, Iran, Brazil, and Egypt. These findings indicate that PAH pollution has become a crucial stress affecting the
sustainable development of the coastal regions. It is urgent to take effective and efficient measures to control PAH pollution in the coastal regions.

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