



# Distribution, Source Apportionment and Risk Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in Surface Sediments at the Basin Scale: A Case Study in Taihu Basin, China

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## Abstract

As a systematic research at basin scale, this study investigated the spatial distribution, source apportionment and ecological risks of eighteen polycyclic aromatic hydrocarbons (PAHs) in surface sediments at different functional regions (rivers, lakes and reservoirs) from Taihu basin. Results showed that the mean values of 18 PAHs (defined as  $\sum_{18}\text{PAHs}$ ) in river sediments (1277 ng/g) was much higher than those observed in lake sediments (243 ng/g) and reservoir sediments (134 ng/g). The accumulation of PAHs in river sediments was largely impacted by the local social-economic development and energy consumption. The positive matrix factorization (PMF) and isomer ratios analysis of PAHs suggest that relative contributions to PAHs in sediments were 15% for gasoline and heavy oil combustion, 9% for oil spills, 30% for coal combustion, 23% for traffic source, and 23% for diagenetic source. Ecological risk assessment based upon risk quotient (RQ) method indicated that sediments at Taihu basin have suffered moderate risk of PAHs.

**Keywords** Polycyclic aromatic hydrocarbons (PAHs) · Taihu basin · Different functional regions · PMF model · Ecological risk assessment

## Introduction

Polycyclic aromatic hydrocarbons (PAHs) are classified as persistent organic pollutants and ubiquitous in the environment (Han et al. 2021). PAHs mainly originate from anthropogenic processes, such as combustion and gasification of wood, incomplete combustion of fossil fuel, vehicular exhausts, and industrial sources (Wang et al. 2021a). In the aquatic environment, PAHs can enter the water phase through atmospheric dry/wet deposition, wastewater discharge, oil leaking, and urban runoff. (Xiang et al. 2018), and then accumulate into sediments due to their hydrophobicity and lipid solubility (Cai et al. 2019). Moreover, as the secondary source, contaminated sediments can release

PAHs into water phase (Han et al. 2021). Therefore, sediments have been found to be a major sink for PAHs in the aquatic environment (Liu et al. 2017).

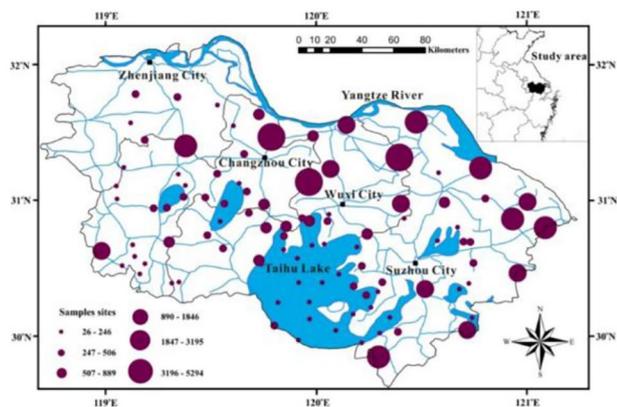
In recent years, the pollution level, spatial variation, sources and ecological risk of PAHs in sediments have received extensive attention (Anyanwu et al. 2020; Wang et al. 2020). However, most of these studies were often limited to collecting samples at one or a limited location in the aquatic system around one city. It may therefore be difficult to accurately understand PAHs pollution level in a large basin around different cities because of the different functionality in these cities. Over the past years, the issue of PAH pollution in sediments has been increasingly severe with the economic and industrial prosperity (Zhang et al. 2021a). Hence, it is necessary to discuss the pollution situation of PAHs of a basin scale in detail.

Taihu basin, located in the Yangtze River Delta (YRD), is the third largest freshwater lake in China, and supplies drinking water, domestic water and industrial water for more than two million people in the YRD (Wang et al. 2021b). As the highly industrialized and populated area, Taihu basin contributes approximately 11% of the gross domestic

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**Fig. 1** Sampling locations of the study area and the concentrations of  $\Sigma_{18}$ PAHs (ng/g) in sediments at the Taihu basin

product (GDP) of China (Xiang et al. 2018). Due to its rapid industrialization and urbanization in the YRD, Taihu basin is faced with more pollution sources than other watersheds. Although PAH pollution has been reported in sediments of Taihu lake (Li et al. 2019), few studies address the PAH pollution in various sediment (e.g. lake sediment, river sediment and reservoir sediment) at the basin scale. Therefore, comparing with other reports, we selected surface sediments at three different functional regions (rivers, lakes and reservoirs), and studied PAH pollution at the large scale of Taihu basin in the present study. The objectives of this study were to (1) investigate the spatial variation of PAHs in sediments at the different functional regions (e.g. river, lake and reservoir) at Taihu basin, (2) evaluate the possible origins of PAHs by positive matrix factorization (PMF) model, and (3) assess ecological risk of sediments associated PAHs in the study area by risk quotient method.

## Materials and Methods

Surface sediment samples (about 5 cm upper layer) from ninety-nine sampling sites located in Taihu Basin were collected with a grab sampler (Fig. 1). These 99 sampling sites were around four cities, i.e., Zhenjiang City, Changzhou City, Wuxi City, and Suzhou City, of which, 40 sites were located in the river region, 48 sites in the lake region, and 11 sites in the reservoir region. Once being collected, the sediment samples were cooled and stored frozen ( $-20\text{ }^{\circ}\text{C}$ ) in laboratory. Before extraction, sediment samples were freeze-dried under the condition of vacuum, and then passed through a 100-mesh sieve, homogenized, and stored in a refrigerator ( $-4\text{ }^{\circ}\text{C}$ ) prior to further analysis.

Eighteen PAHs were analyzed in present study and their abbreviation were listed in Table S1. Using internal standard method, eighteen PAHs in surface sediments were

extracted by Soxhlet extraction method, and purified with anhydrous sodium sulfate (1 cm) and silica/alumina gel (2/1; 40 cm) by a chromatographic column (Azimi et al. 2020). Then, the PAHs were analyzed by Agilent 7890 A gas chromatography-5975 C mass spectrometry (Agilent Technologies, Palo Alto, USA) with a 30 m HP-5 column (0.25 mm i.d., 0.25- $\mu\text{m}$  film thickness, Agilent Technologies) (Zhang et al. 2020). For quality assurance and quality control, a method blank was analyzed along with every 12 sediment samples. Quantification of the target PAHs was performed by an internal calibration method and the correlation coefficients ( $R^2$ ) of nine-point calibration curves (0.01, 0.02, 0.05, 0.1, 0.2, 0.5, 1.0, 2.0, and 5.0  $\mu\text{g}/\text{ml}$ ) were higher than 0.995 for each PAH. The mean recoveries of surrogate standards were  $93.0 \pm 19.6\%$  for  $\text{NaP-}d_8$ ,  $107 \pm 17.6\%$  for  $\text{Ace-}d_{10}$ ,  $114 \pm 16.1\%$  for  $\text{Ph-}d_{10}$ ,  $100 \pm 17.6\%$  for  $\text{Chry-}d_{12}$  and  $96.4 \pm 18.1\%$  for  $\text{Per-}d_{12}$ .

In order to evaluate the possible risk of PAHs in Taihu basin, the risk quotient (RQ) was widely used in recent years (Wu et al. 2022).  $\text{RQ}_{\text{NCs}}$  and  $\text{RQ}_{\text{MPCs}}$  were defined by the following equations:

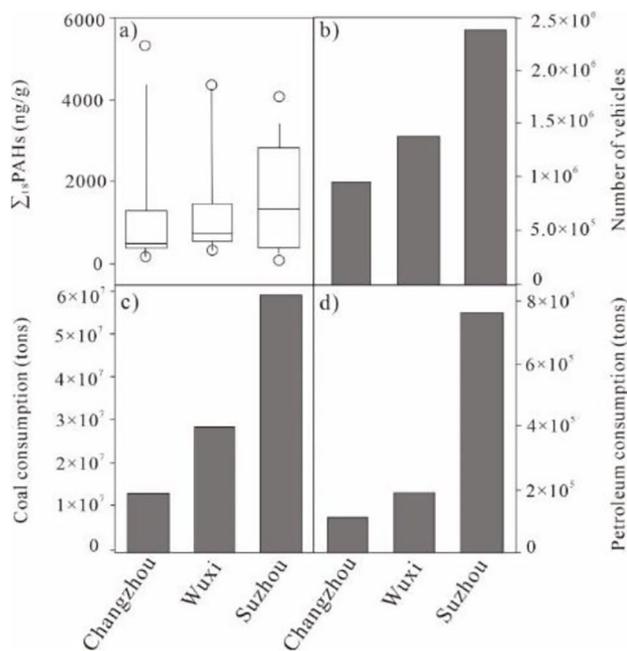
$$\text{RQ}_{\text{NCs}} = \frac{C_{\text{PAHs}}}{C_{\text{QV}(\text{NCs})}} \quad (1)$$

$$\text{RQ}_{\text{MPCs}} = \frac{C_{\text{PAHs}}}{C_{\text{QV}(\text{MPCs})}} \quad (2)$$

Where  $C_{\text{PAHs}}$  is the concentration of certain PAHs in the sediment;  $C_{\text{QV}}$  refers to the corresponding quality values of certain PAHs in the medium; the negligible concentrations (NCs) and the maximum permissible concentrations (MPCs) of PAHs in sediment were used as  $C_{\text{QV}}$  in the present study (Wang et al. 2022). The ecological risk classification of individual PAHs and total PAHs is shown in Table S2.

## Results and Discussion

The total concentrations of  $\Sigma_{18}$ PAHs in sediments at Taihu basin ranged from 26 to 5294 ng/g dry weight (dw), with the mean value of 649 ng/g (Table S3). The mean concentrations in sediments followed the order of river region (1277 ng/g) > lake region (243 ng/g) > reservoir region (134 ng/g) (Fig. 1). The higher pollution of PAHs observed in river sediments might be attributed to the direct inputs from the municipal and industrial sewage (Wang et al. 2018). In addition, due to the large area of the lake region (2427  $\text{km}^2$ ), dilution effects would be the possible reason on the low residual concentrations in sediments from lake and reservoir regions. The PAHs contents in river region were higher than



**Fig. 2**  $\Sigma_{18}$ PAHs contents in river sediments a), number of vehicles b), coal combustion c), and petroleum consumption d) from Changzhou, Wuxi and Suzhou

those in Songhua River, China (Cui et al., 2018), but PAHs contents in lake and reservoirs regions at present study were much lower than those in Chaohu lake (Qin et al., 2014) and Huaxi reservoir, China (Wang et al., 2017).

At the river region, PAH levels in Suzhou and Wuxi city were significantly higher than those in Changzhou city ( $p < 0.05$ ). This phenomenon might be associated with local social-economic development and energy consumption (Niu et al. 2017). The average concentrations of  $\Sigma_{18}$ PAHs accumulated in river sediments from Suzhou, Wuxi and Changzhou cities appeared a similar trend with local coal and petroleum consumption and motor vehicle's number (Fig. 2). These results suggested that the incomplete combustion of fossil fuel could be expected as the main contributors to the sediment-associated PAHs in river region from Taihu basin (Wang et al. 2021c).

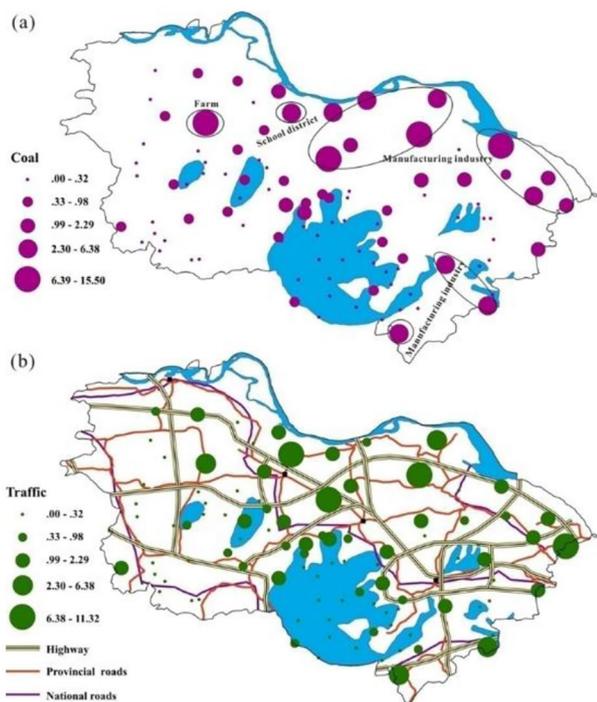
Among individual PAHs, Per, Ret, Flu, and BghiP were predominant compositions of  $\Sigma_{18}$ PAHs in all sediment samples, accounting for 18%, 13%, 8.5%, and 8.2% of total concentrations, respectively. Particularly, Per had the highest average concentration in the river and lake regions. The relative concentrations of Per were greater than 20% of  $\Sigma_{18}$ PAHs contents in 56% of the sampling sites at river and lake regions, indicating significant contribution of diagenetic source (Silliman et al. 1998). Without natural origin derived Per, the compositions of other 17 PAHs in sediments at the three different regions were all dominated by 4 - rings homologues. The results suggested that PAHs in the three

different regions were mainly from the pyrolysis sources (Anyanwu et al. 2020). Furthermore, reservoir sediments contained the highest relative abundance of low molecular weight PAHs (2–3 ring) compared with river and lake sediments, possibly suggesting significant atmospheric deposition or air - water diffusive exchange (Zhang et al. 2021b).

The correlation coefficient for PAHs and TOC in sediments at Taihu basin were shown in Table S4. There were no obvious correlations between the concentrations of  $\Sigma_{18}$ PAHs and TOC in the reservoir and lake regions, but a significantly positive correlation in river region ( $r^2 = 0.35$ ,  $p < 0.05$ ). In detail, the contents of TOC in the lake region were significantly correlated with the concentrations of DBA and BghiP ( $r^2 = 0.43$ ,  $p < 0.01$ ;  $r^2 = 0.39$ ,  $p < 0.01$ , respectively). In the river region, there were significant correlations of TOC with Ac, Py, Fl, Ph, and Ret ( $p < 0.05$ ), while not with other PAHs. Particularly, there was strong evidence that Fl, Ph, and Ret concentrations were associated with TOC contents in river sediments ( $p < 0.01$ ). In summary, the different sediment type revealed different correlativity between TOC and PAHs, which might be attributed to different PAH sources and particulate organic matter emissions from the industries adjacent to the rivers (Li et al. 2022).

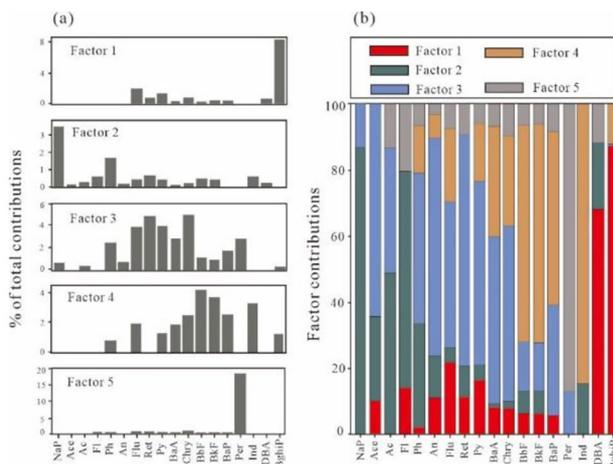
The ratios of Ph/An, Flu/Py, Chry/BaA and Ind/BghiP have been widely used to identify the possible PAHs origins (Han et al. 2021). The values of these isomeric ratios varied from 0.1 to 19.3 for Ph/An, 0.8–19.8 for Flu/Py, 0.2–4.9 for Chry/BaA, and 0–139.8 for Ind/BghiP, respectively. The values of these isomeric ratios in river sediments suggested that more than 80% of river sediment samples contained PAHs originated from pyrogenic related sources (Gao et al. 2018). In addition, the ratios of Ph/An and Chry/BaA in the reservoir and lake regions indicated that more than 60% of sediment samples from the lake and reservoir regions were from pyrolysis related sources (Tucca et al. 2020), but the ratios of Flu/Py and Ind/BghiP suggested these samples were predominantly generated from pyrogenic sources (Zhang et al. 2017). Therefore, PAHs in sediments at the lake and reservoir regions were likely derived from pyrolytic, petrogenic and diagenetic origins. Furthermore, Log-based ratios Ph/An, Flu/Py, Chry/BaA and Ind/BghiP negatively correlated against Log-based concentration of  $\Sigma_{18}$ PAHs ( $p < 0.05$ ), which might suggest that the samples accumulated high concentrations of PAHs were mainly influenced by pyrolytic sources.

The US EPA PMF V5.0 model was also widely used for source identification of PAHs in sediments (Feng et al. 2022), and the detailed principles and information have been described in previous studies (Hopke 2016; Wang et al. 2016). In the present study, the scatter plot of observed and predicted concentrations of  $\Sigma_{18}$ PAH had excellent correlation ( $r^2 = 0.99$ ,  $p < 0.01$ ), suggesting PMF model is a useful



**Fig. 4** Spatial distribution for the source contributions of PAHs in sediments from coal (a) and traffic (b) source

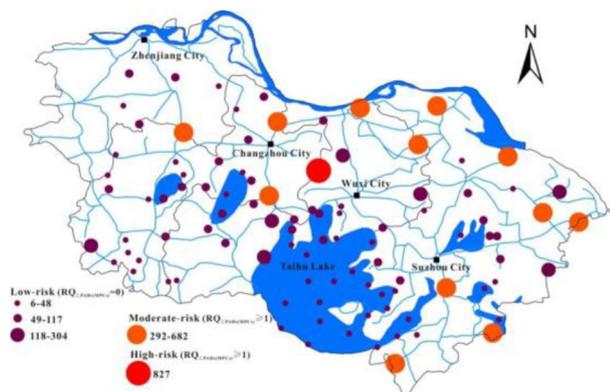
approach to identify the sources of sediment associated PAHs. Five factors were extracted for further analysis and the results of PMF model were shown in Fig. 3a. Factor 1 (accounting for 15% of all factors) could be explained by high loading of BghiP which suggested gasoline emission and heavy oil combustion (Ding et al. 2018). Factor 2 (accounting for 9% of all factors) was identified as petroleum sources because of high loadings of low molecular weight PAHs such as NaP (Ren et al. 2021). In the study area, the petroleum sources were mainly related to oil spills from ships or the pollution discharge of oil. Factor 3 (accounting for 30% of all factors) was predominantly impacted by Ret, Chry, Flu, Py, BaA, Ph, Per, and BaP, which can be related to coal combustion (Wang et al. 2021a). The sampling sites with high contributions of coal combustion in present study were generally related to the manufacturing industry, including textile and paper, raw chemical materials and chemical, nonmetal mineral and metal production (Fig. 4a). Based on the data from statistical yearbook at Jiangsu province, the coal consumption of manufacturing industry accounted for 27% in Zhenjiang City, 56% in Changzhou City, 36% in Wuxi City, and 38% in Suzhou City of the total consumption of coal, respectively. Therefore, the manufacturing industry in Taihu basin was a noticeable contributor to PAH pollution in surface sediments. Factor 4 (accounting for 23% of all factors) had high contribution of BbF, BkF, Ind, BaP, Chry, and moderately by Flu, BaA, Py, and



**Fig. 3** Results of the PMF model: (a) The percentages of logarithmic concentration of PAHs congeners from total contributions for the PMF Factors, and (b) Source contribution percentages from five PMF estimated factors to the 18 PAHs in Taihu basin

BghiP, indicating traffic exhaust emissions. BbF, BkF, Ind, Chry, and BghiP were identified as traffic tunnel markers (Mohammed et al. 2021). With the rapid development of economy and the improvement of living standard in China, the number of vehicles increased at annual rate of 21% in China, and the vehicle exhaust emission is the most important source of PAHs in urban zones (Yusuf et al. 2022). It is clearly observed that the sampling sites of high contribution values from transportation were closely associated with the main roads in the study area, and most of these sites were close to the excellent transportation network (Fig. 4b). Therefore, with the increasing of the number of vehicles, PAHs pollution from the traffic exhaust emission are more serious at Taihu basin. Factor 5 (accounting for 23% of all factors) was characterized by weighted loading of Per, suggesting diagenic processes of terrestrial organic matter in anoxic circumstances (Feng et al. 2022). To sum up, the relative contributions to  $\sum_{18}$  PAHs in sediments at Taihu basin were 15% for gasoline and heavy oil combustion, 9% for oil spills, 30% for coal combustion, 23% for traffic source, and 23% for diagenetic source, respectively.

The contribution of each source to the total predicted concentration in all samples was illustrated in right part of Fig. 3b. The results indicated that NaP (87%), Ac (49%) and Fl (66%) were mainly originated from oil spills from ships or the pollution discharge of oil instead of petroleum combustion. Ace (64%), An (66%), Ret (70%), Py (55%), BaA (51%), and Chry (53%) were predominantly contributed by coal combustion with diagenetic source contributing less than 10%. For BbF, BkF, BaP and Ind, traffic source was the main contributor and contributed more than 50% of the sources of these PAHs. Approximately 68% of DahA and 87% of BghiP came from gasoline emission and heavy oil



**Fig. 5** Ecological risk rank of  $\Sigma_{18}$ PAHs contamination in sediments from Taihu basin

combustion. Coal combustion contributed 46% of Ph and 32% of Ph came from oil spills. In addition, diagenetic source contributed approximately 87% of the source contributions for Per.

The mean values of  $RQ_{(NCs)}$  for individual PAH in all sediments at the three regions were higher than 1.0 except for Ace, Chr, and six-ring PAHs (Table S5). The results indicated that these individual PAH homologues were widely appeared moderate ecosystem risk, but Ace, Chry, and six-ring PAHs had low risk for some sampling points in Taihu basin. In addition, the mean values of  $RQ_{(MPCs)}$  for individual PAH in all sediments at the three regions were less than 1.0, suggesting risk free of PAHs in the study area (Table S5). The characterizations of  $RQ_{\Sigma PAHs (NCs)}$  and  $RQ_{\Sigma PAHs (MPCs)}$  in all sediments at Taihu basin were showed in Fig. 5.  $RQ_{\Sigma PAHs (NCs)}$  in all sediments at Taihu basin were higher than 1.0, and thirteen sampling sites at the three regions were observed with  $RQ_{\Sigma PAHs (MPCs)}$  value higher than 1.0, indicating moderate ecosystem risk of PAHs at Taihu basin. Specially, one sampling site at river region which near Changzhou city had a  $RQ_{\Sigma PAHs (NCs)}$  greater than 800, strongly suggesting high risk by exposure to resided PAHs. In summary, sediments at Taihu basin have suffered moderate risk of PAHs.

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