

Article

Polycyclic Aromatic Hydrocarbons in Sediments from Typical Algae, Macrophyte Lake Bay and Adjoining River of Taihu Lake, China: Distribution, Sources, and Risk Assessment

Yibo Dong ^{1,2}, Zaisheng Yan ^{2,*}, Huifang Wu ^{1,*}, Guoqing Zhang ^{1,2}, Haichen Zhang ^{1,2} and Mingzhong Yang ^{1,2}

¹ College of Urban Construction, Nanjing University of Technology, Nanjing 211816, China; Dong1006182974@gmail.com (Y.D.); zhang19850055195@gmail.com (G.Z.); whfkh163.com (H.Z.); renzaita2006@126.com (M.Y.)

² State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China

* Correspondence: zshyan@niglas.ac.cn (Z.Y.); whf@njtech.edu.cn (H.W.)

Citation: Dong, Y.; Yan, Z.; Wu, H.; Zhang, G.; Zhang, H.; Yang, M. Polycyclic Aromatic Hydrocarbons in Sediments from Typical Algae, Macrophyte Lake Bay and Adjoining River of Taihu Lake, China: Distribution, Sources and Risk Assessment. *Water* **2021**, *13*, 470. <https://doi.org/10.3390/w13040470>

Academic Editor: Maria

Włodarczyk-Makuła

Received: 25 December 2020

Accepted: 08 February 2021

Published: 11 February 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).

Abstract: Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants in sediments and pose a serious risk for freshwater ecosystems. In this study, sediment samples from 24 sites were collected from the cyanobacterial bloom-occurring, macrophyte-growing lake bay and adjoining river of Taihu Lake. Here, the concentration levels, sources, and risk assessment of 16 priority PAHs in the surface sediments from typical algae, macrophyte lake bay and adjoining river of Taihu Lake, were investigated, and the results were compared with those of previous studies. The total PAH (Σ PAH) concentrations ranged from 4900 to 16,800 ng·g⁻¹ in sediments of the Taihu Lake bay and from 5736.2 to 69,362.8 ng·g⁻¹ in sediments of the adjoining river. The level of PAHs in riverine sediments was significantly higher than those of the Taihu Lake bay, and that of the Dongshan River was significantly higher than that of the Mashan River, while there was no significant difference in the concentrations of PAHs between the cyanobacterial bloom-occurring and macrophyte-growing lake zone. The results indicated petroleum contamination was dominated in the cyanobacterial bloom-occurring, macrophyte-growing lake bay, while PAHs of the riverine sediments derived from petroleum contamination and the combined combustion including wood, coal combustion, and petroleum combustion according to the identification by the molecular diagnostic ratio and principal component analysis (PCA). Sediment risk assessment based on sediment quality guidelines (SQGs) suggested that partial regions of the Taihu Lake bay were subjected to the potential ecological risk of the 3-ring and 5-ring PAHs, and there existed negative effects related to naphthalene pollutant in all survey regions. The adjoining riverine sediments showed a high ecological risk.

Keywords: polycyclic aromatic hydrocarbons (PAHs); sediment; lake bay; adjoining river; distribution; sources; risk assessment

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of persistent organic pollutants that are prevalent in the environment, which could be transported to the lakes and rivers through surface runoff, sewage discharge, atmospheric deposition, and some other means, such as oil leaks [1–4]. PAHs entering the water body are easily adsorbed on the particles and tend to accumulate in the sediments [5]. Hence, sediment is one of the important environmental reservoirs for PAHs [6,7]. Due to their mutagenicity, carcinogenicity, and toxicity, PAHs pollution raises great concerns worldwide [8,9].

Knowledge of the distribution, sources, and risk assessment of PAHs in sediments from typical macrophyte, algae lake bays and adjoining rivers is crucial for effective abatement of pollution and risk [10–12]. Previous studies have found that the sediment-water interfaces in the cyanobacterial bloom-occurring, macrophyte-growing lake zone of Taihu Lake have significant differences in environmental conditions [13,14], which affect the content, distribution characteristics, degradation characteristics, and ecological risks of PAHs in sediments [2,12]. The accumulation and deposition of cyanobacteria in the typical algae lake zone accelerate the migration and transformation process of PAHs into lake sediments [12,15,16], while the growth and decay of aquatic plant residues in the typical macrophyte lake zone also affect the distribution characteristics of PAHs in sediments [17,18]. Thus, it is necessary to investigate the distribution and sources of PAHs in surface sediments from the cyanobacterial bloom-occurring, macrophyte-growing lake bay.

PAHs can be derived from natural or anthropogenic sources, and anthropogenic sources are generally considered to be the main sources of PAHs in the environment [2,8], which was mainly related to industrial and agricultural development and human transportation. Anthropogenic activities such as incomplete combustion of fossil fuels, mass production of coal tar, oil spills, and wood processing have caused a large number of organic pollutants into natural water bodies [19,20]. The potential pollution sources of sampling sites in the rivers surrounding Taihu Lake were mainly shipping activities, industrial shipping, agricultural activity, aquaculture, and domestic sewage from a residence community, but sites in the lake bays were also subjected to aquaculture and shipping activities [2]. Compared to lake bays, rivers surrounding the Taihu Lake are more seriously influenced by anthropogenic activities, and the sources and components of pollutants are more complex. Therefore, studying the pollution status of PAHs in surface sediments of the lake bay and adjoining river can better evaluate the ecological risk of PAHs in the Taihu Lake basin.

In recent years, many studies have been carried out on PAHs in environmental media such as lake, river, and bay sediments [8,21,22]. Qian et al. found that the PAHs pollutants in sediments from different regions of the Haihe River in China were significantly different, and most of them derived from the combined combustion sources, including various combustion processes [23]. Li et al. investigated the source of PAHs in sediments from Chaohu Lake, China by different receptor models combined with the PAHs diagnostic ratios, and the results indicated that the input of PAHs was closely related to coal and wood combustion from the surrounding industrial development [24]. Zhang et al. used three receptor models to determine the distribution of potential sources of PAHs in sediments of the Taihu Lake, and found that vehicle emissions were one of the main sources with higher toxicity risk, followed by coal and wood combustion sources [25]. In addition, studies also focused on the influence of eutrophication on PAHs biogeochemical processes in Taihu Lake [26,27]. However, studies related to the differences of distribution, sources, and risk assessment of PAHs in surface sediments from the cyanobacterial bloom-occurring, macrophyte-growing lake bay and adjoining river have rarely been reported.

In this study, the typical algae, macrophyte lake bays of Taihu Lake (Meiliang Lake bay and East Taihu Lake bay, respectively), and the adjoining rivers (Mashan river and Dongshan river, respectively) were selected for sampling survey. The objective of this study was to determine the concentration levels, distribution, composition, and main sources of PAHs in surface sediments of the cyanobacterial bloom-occurring, macrophyte-growing lake zone of Taihu Lake and the adjoining rivers, and assess their status of the ecological environment risk.

2. Materials and Methods

2.1. Study Area and Sample Collection

The studied Meiliang Lake bay (cyanobacterial bloom-occurring zone in Taihu Lake) is located in 10 km southwest suburb of Wuxi City, within the Jiangsu Province of China

(31°32' N, 120°13' E). It is a scenic spot with a length of 16 km from north to south and a width of 7–10 km from east to west. Mashan river is located in Mashan Town on the south-west side of Meiliang Lake bay, which is an important channel that runs through two lake bays in the northern part of Taihu Lake. There are frequent commercial ships and dense industrial districts around the river. In addition, Meiliang Lake bay is the parking place for cyanobacteria salvage ships. The Dongshan river is an important branch of East Taihu Lake, which runs through Dongshan Town, Suzhou City, Jiangsu Province (31°5' N, 120°24' E). The river flows through many residential and industrial areas, with a total length of about 12 km. It performs a central role as a channel for aquaculture, commercial port traffic, and the ship processing industry, forming a complex estuarine environment around East Taihu Lake. The studied East Taihu Lake bay (macrophyte-growing zone in Taihu lake) is close to the estuary of Dongshan river connecting East Taihu Lake.

According to the vicinity of potential input of PAHs from different pollution sources or anthropogenic pollution sources, surface sediment samples were collected from twenty-four sites located all around the macrophyte-growing, cyanobacterial bloom-occurring lake bay and adjoining rivers of Taihu Lake. The locations of the sampling station were shown in Figure 1. All the surface sediment samples were collected by a stainless-steel grab sampler, placed in polyethylene bags, stored in an incubator paving with ice bags, and transported back to laboratory. At the lab, sediment samples were stored at −20 °C until use.

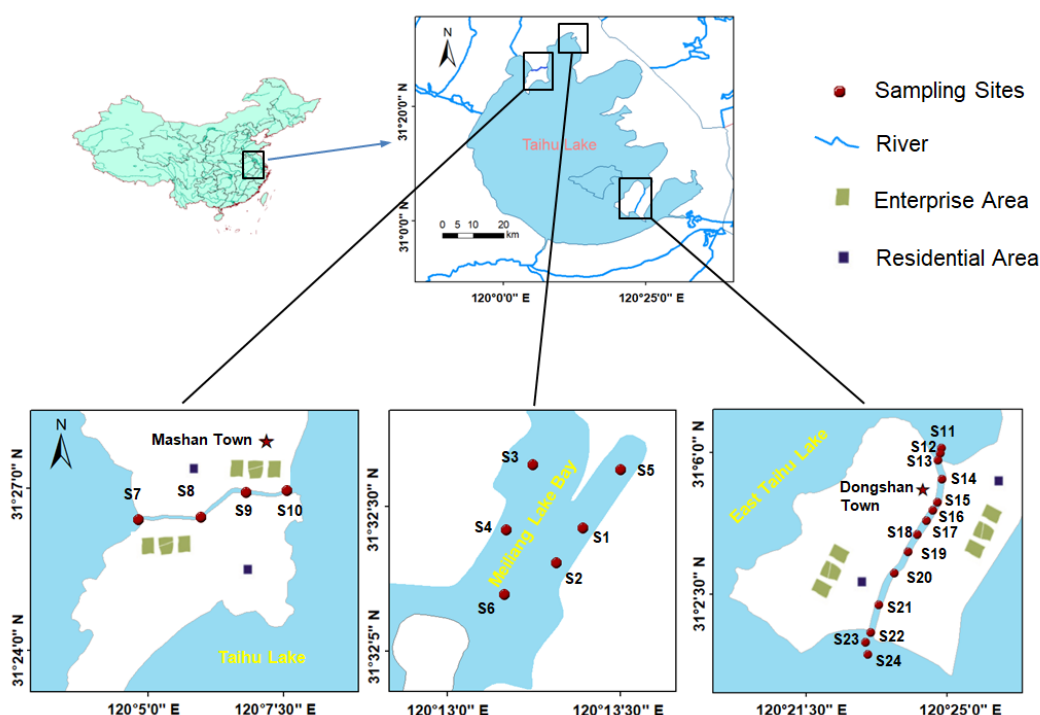


Figure 1. Location of sampling sites in the lake bay and adjoining river of Taihu Lake.

2.2. Sample Preparation and Analysis

The 16 priority PAHs included naphthalene (Nap), acenaphthene (Ana), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flua), pyrene (Pyr), benz(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DbA), benzo(g,h,i)perylene (BgHiP), acenaphthylene (Any), and indeno(1,2,3-cd)pyrene (InP).

For PAHs analysis, the sediment samples were freeze-dried, crushed into powder, and passed through a 100-mesh sieve. PAHs concentrations in sediments were determined according to previous studies [2,3,10,19,28]. Approximately 4 g of sieved sediment

sample combined with equal volume quartz sand and 3 g of copper powder were extracted with accelerated solvent extraction (ASE, Dionex ASE 100) with dichloromethane solvent (chromatographic purity). The extracts were completely condensed to approximately 2 mL, followed by Florisil solid-phase extraction column for clean-up. PAHs fraction was obtained by elution with 70 mL of acetonitrile. The eluent was collected, condensed, and then concentrated to 1 mL under a gentle flow of high-purity nitrogen for analysis.

The levels of PAHs in sediment samples were analyzed with a high-performance liquid chromatograph (HPLC) (Agilent 1200, USA) fitted with a 4.6×250 mm reverse phase C18 column using 8:2 acetonitrile:water (v:v) as the mobile phase at a flow rate of $1 \text{ mL} \cdot \text{min}^{-1}$ at 25°C . The HPLC analysis was performed by fluorescence detector and UV detector. For the fluorescence detector, the excitation wavelength was 260 nm and 290 nm. The emission wavelength was 380 nm and 410 nm, and the excitation and emission wavelengths were changed according to a time program. For UV detector, the detection wavelength was designated as 254 nm. Identification of individual PAHs was based on the comparison of retention time between samples and the standard solution.

2.3. Quality Assurance and Quality Control

The analytical process was subjected to strict quality control procedures for all samples. The peak identification of PAHs was based on the retention time of standard components analyzed with the same operation conditions of instrument. The quantification was performed using a calibration curve established using external standard for each individual PAH. The calibration curves of individual PAH were established according to the peak area through determining a series of concentrations of the standard solutions. Calibration plots had satisfactory linear regression coefficients with R^2 values greater than 0.99. The standard solution was also inserted every tested fifteen samples to recalibrate the retention time of the target compounds. The PAH recoveries were determined by adding the known concentration of standards solutions containing 16 priority PAH compounds. The mean recoveries ranged from $76.9 \pm 7.1\%$ to $88.4 \pm 9.5\%$ for 16 PAHs standards. The detection limits were $0.1\text{--}5.0 \text{ } \mu\text{g} \cdot \text{kg}^{-1}$ for sediment samples. All reported concentrations were within the quality control range, given on a dry weight basis.

2.4. Statistical Analysis

Student's *t*-test was performed to test the differences in the concentrations of PAHs between the cyanobacterial bloom-occurring and macrophyte-growing lake zone. Statistical significance of differences was determined by one-way analysis of variance using SPSS software (IBM SPSS Statistics 19). A $P < 0.05$ was considered significant.

3. Results and Discussion

3.1. Level and Distribution of PAHs in Sediments

The collected samples were surface (0–10 cm) sediments, and PAHs were detected in all the surface sediments at different points in Taihu Lake. The total concentration of 16 priority PAHs in sediments from the macrophyte-growing, cyanobacterial bloom-occurring lake bay of Taihu Lake ranged from 4900 to 16,800 $\text{ng} \cdot \text{g}^{-1}$ dry weight (dw) in sediments, and the total concentration range of 16 PAHs in sediments of the adjoining river ranged from 5736.2 to 69,362.8 $\text{ng} \cdot \text{g}^{-1}$ (dw). The concentrations of PAHs in sediments worldwide were shown in Table 1. The concentrations of Σ PAHs in sediments from typical macrophyte, algae lake bay of Taihu Lake were lower than those from Dianchi, Chaohu and Nansi Lake in China and riverine and estuarine sediments in elsewhere in the world [29–31]. Compared with other studied lakes and bays located in more developed areas around the world, such as Mill River in USA and Tokyo Bay in Japan [32,33], the level of Σ PAHs in sediments of the lake bay in this study was relatively lower.

Table 1. Comparison of polycyclic aromatic hydrocarbons (PAHs) concentrations ($\text{ng}\cdot\text{g}^{-1}$ dry weight (dw)) in surface sediments from different rivers, estuaries, and lakes in the world.

Location	Number of PAHs	ΣPAHs ($\text{ng}\cdot\text{g}^{-1}$ dw)	References
		Range	
Haihe River, China	16	258.77–11,296.66	[34]
Chaohu Lake, China	15	80.82–30,365.01	[30]
Dianchi Lake, China	16	210–11,070	[29]
Erhai Lake, China	16	32420–55,8530	[35]
Liaohe River, China	16	92.2–29,5635.2	[36]
Nansi Lake, China	16	160–32,600	[31]
Nanfei River (Chaohu Lake), China	28	7140–70,500	[30]
Fuhe River (Baiyangdian Lake), China	16	191.6–6360.5	[37]
Mill River, US	16	0–39,000	[32]
Susquehanna River, US	16	74–18,073	[38]
Tyne River, UK	16	9100–23,700	[39]
Tokyo Bay, Japan	16	534–29,2370	[33]
Manzala Lake, Egypt	39	246–9910	[40]
Calabar River, Nigeria	17	1670–20,100	[41]
Lenga Estuary, Chile	16	290–6118	[42]
Bahía Blanca Estuary, Argentina	18	15–10,260	[43]
Meiliang Lake Bay and East Taihu Lake Bay	16	4900–16,800	this study
Mashan River and Dongshan River	16	5736.2–69,362.8	

Based on the Sediment Quality Guideline thresholds (SQG), the level of PAHs pollution can be divided into four categories: (a) low, 0–100 ng/g ; (b) moderate, 100–1000 ng/g ; (c) high, 1000–5000 ng/g ; or (d) extremely high, > 5000 ng/g [44]. According to SQG, the concentration of PAHs in sediments from the adjoining river was at an extremely high level of PAHs pollution in this study. Compared with the adjoining river, the level of PAHs in the sediments from typical macrophyte, algae lake bay of Taihu Lake had lower PAHs concentrations. These results suggest that rivers are essential channels for connecting land and lake ecosystems, which support many human activities exerting anthropogenic stresses [2].

The contents of PAHs in surface sediments from lake bays and adjoining rivers are shown in Figure 2. The highest PAHs concentration was observed at the site from Dazuishan Bridge (S21, 69,362.8 $\text{ng}\cdot\text{g}^{-1}$ dw) of Dongshan river. Dazuishan Bridge was a local aquatic product trading place, which may be greatly affected by oil spills from frequent fishing ships and the fossil fuel, wood, and coal combustion [2,10]. PAHs were present at higher concentrations in sediments from Gangdong Bridge (S16, 57,573 $\text{ng}\cdot\text{g}^{-1}$ dw), Ferry Bridge (S14, 43,217.3 $\text{ng}\cdot\text{g}^{-1}$ dw), Dashui Harbor (S17, 33163.2 $\text{ng}\cdot\text{g}^{-1}$ dw), and Dingxianghu Lake bay (S15, 30193.7 $\text{ng}\cdot\text{g}^{-1}$ dw) than those in sediments from the other sites, which is due to the human activities, such as district for residential areas and ship repairing sites as potential pollution sources. In the Taihu Lake bay, the highest concentration of PAHs was observed at a gate control section of Meiliang Lake bay (S5, 16,800 $\text{ng}\cdot\text{g}^{-1}$) connecting with Liangxi river, and the concentrations at other sites were relatively low (below 10,000 $\text{ng}\cdot\text{g}^{-1}$ dw). For the distribution of PAHs concentration, there was no significant difference in the concentrations of PAHs between the cyanobacterial bloom-occurring and macrophyte-growing lake zone ($P = 0.772 > 0.05$). These results suggested that the distribution of PAHs concentration in sediments is more dependent on specific pollutant sources than lake-zone type or other environmental factors.

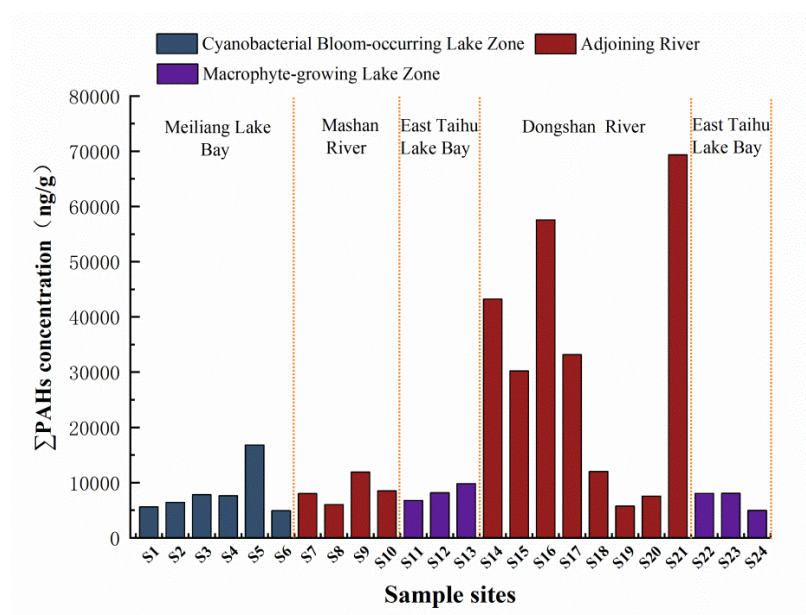


Figure 2. The contents of PAHs in surface sediments of the lake bay and adjoining river.

It was also observed that the concentration of PAHs in sediments of the Dongshan river (S14–S21) was significantly higher than that in the Mashan river (S7–S10), with a significant difference ($P = 0.024 < 0.05$). This may be due to the denser population on both sides of the adjoining river of Taihu Lake in Dongshan Town, which was more affected by residential activities (fossil fuel, wood, and coal combustion) and industrial emissions (diesel and gasoline combustion). In addition, the average concentration of PAHs in sediments from the adjoining river was nearly three times ($P = 0.026 < 0.05$) than those of the macrophyte, algae lake bay, which was due to river ecosystems supporting numerous human activities, such as human infrastructure around a river that exert considerable anthropogenic pressure [1,2]. Pollution entered the river at many different locations (point source of PAHs) along the length of the river. These PAHs contaminants also entered into the sediments of lake bay zones, which might become potential centralized sources of PAHs in these areas.

The composition pattern of PAHs in sediments was shown in Figures 3–4. Based on the number of aromatic rings, the 16 PAHs compounds could be divided into the following five groups as follows:

- (1) two-ring PAHs (Naphthalene, Nap);
- (2) three-ring PAHs (Any, Ana, Flu, Phe, Ant);
- (3) four-ring PAHs (Flua, Pyr, BaA, Chr);
- (4) five-ring PAHs (BbF, BkF, BaP, DbA);
- (5) six-ring PAHs (InP, BghiP) [45].

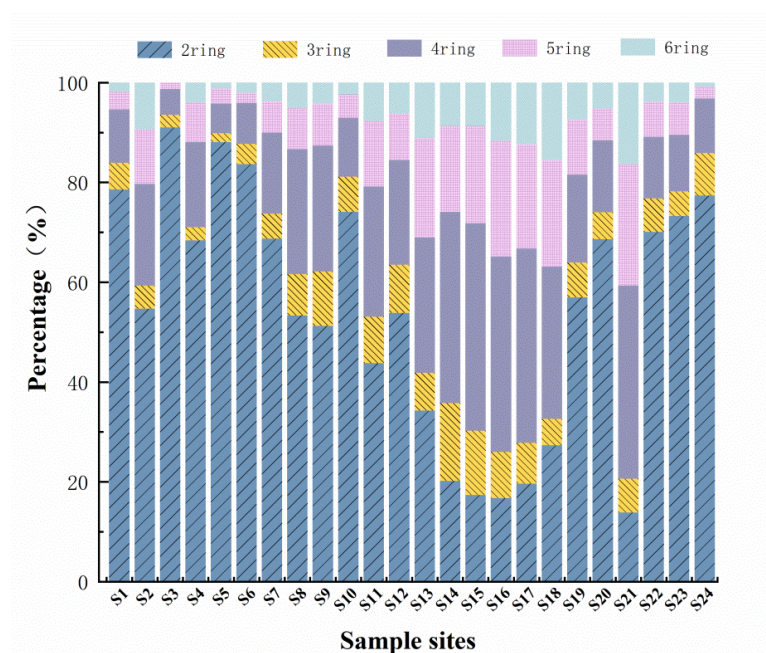


Figure 3. Composition of PAHs in the surface sediment collection from the lake bay and adjoining river.

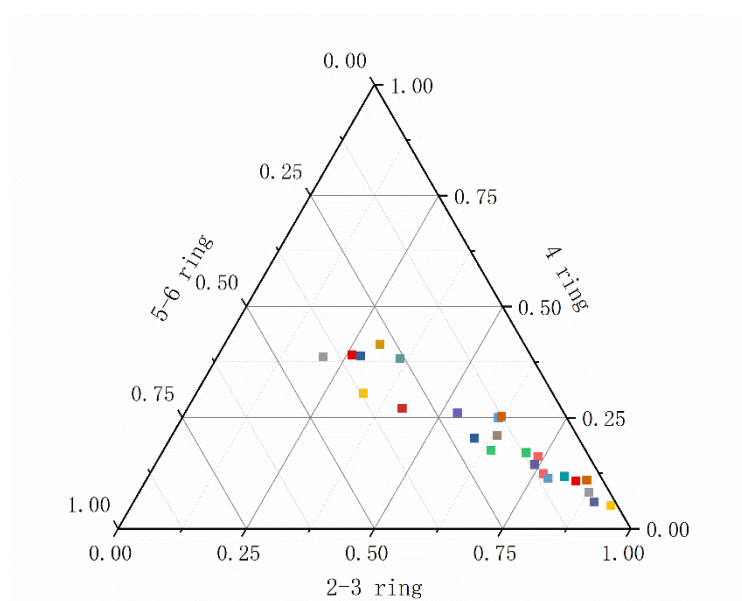


Figure 4. The composition patterns of PAHs by ring size in surface sediments samples. (2-3 rings: sum of naphthalene (Nap), acenaphthylene (Any), acenaphthene (Ana), fluorene (Flu), phenanthrene (Phe), and anthracene (Ant); 4 rings: sum of Flt, pyrene (Pyr), benz(a)anthracene (BaA), and chrysene (Chr); 5-6 rings: sum of benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DbA), benzo(g,h,i)perylene (BghiP), and indeno(1,2,3-cd)pyrene (InP)).

The analysis results showed that the proportion of 2-ring PAHs compounds (Nap, 34–91%) in the macrophyte-growing, cyanobacterial bloom-occurring lake bay was generally higher than that of the adjoining river (14–69%). It can be found that the proportion of 3-ring PAHs in the adjoining riverine sediments was slightly higher than that of the lake bay. The total proportion of the 2–3 ring was higher in the algae, macrophyte lake bay of Taihu Lake, which may be due to oil spill from frequent fishing ships around Meiliang Lake bay and East Taihu Lake bay. The proportions of 4–5- and 6-ring PAHs in sediments

of the adjoining river of Taihu Lake were 12–42%, 5–24%, and 2–16%, respectively, which were higher than those of the lake bay (5–26%, 1–11%, and 1–8% respectively). Human activities such as industrial emissions, burning of fossil fuels, and aquaculture around rivers were important factors that account for a large proportion of the 4–6 ring PAHs [46]. The 2–3 ring PAHs of the S21 sampling collection accounted for a higher proportion, which may be caused by heavy oil spills and tail gas pollution from fishing ships [2].

3.2. Source Appointment of PAHs in Surface Sediments

3.2.1. Source Identification by Isomer Ratios

The ratio method has been widely used to identify the sources of PAHs contamination in the environment [47,48]. In this study, four specific ratios, namely, $\text{Ant} / (\text{Phe} + \text{Ant})$, $\text{Flua} / (\text{Flua} + \text{Pyr})$, $\text{BaA} / (\text{BaA} + \text{Chr})$ and $\text{InP} / (\text{InP} + \text{BghiP})$ were used to analyze the PAHs sources of the sampling sites. In general, the ratio of $\text{Ant} / (\text{Phe} + \text{Ant}) < 0.1$ and the ratio $\text{Flt} / (\text{Flt} + \text{Pyr}) < 0.4$ indicate the PAHs mainly originated from petroleum contamination, while the ratio of $\text{Ant} / (\text{Phe} + \text{Ant}) > 0.1$ is usually considered to be typical of combustion source. The ratio of $\text{Flua} / (\text{Flua} + \text{Pyr})$ or $\text{InP} / (\text{InP} + \text{BghiP}) > 0.5$ indicates that PAHs are mainly derived from the inputs of grass, wood, and coal combustion sources. In addition, the ratio of $\text{Flua} / (\text{Flua} + \text{Pyr}) > 0.4$ and the ratio of $\text{InP} / (\text{InP} + \text{BghiP}) > 0.2$ usually indicates petroleum contamination, while the remaining range of these two ratios is regarded to be related to petroleum combustion source [49].

The ratio calculation results of the present study were shown in Figure 5. Compared to the molecular diagnostic ratios of the characteristic PAHs, $\text{Ant} / (\text{Phe} + \text{Ant})$, $\text{Flua} / (\text{Flua} + \text{Pyr})$, $\text{BaA} / (\text{BaA} + \text{Chr})$, and $\text{InP} / (\text{InP} + \text{BghiP})$ ratios indicated that PAHs in sediments of the macrophyte-growing, cyanobacterial bloom-occurring lake bay of Taihu Lake mainly derived from petroleum contamination and petroleum combustion, which was closely related to the oil spill and tail gas emissions from fishing ships in the area. The ratio calculation results also suggested PAHs in sediments of the Mashan river and Dongshan river originated from biomass, coal combustion, and petroleum combustion. In general, petroleum contamination dominated in the Taihu Lake bay, while the adjoining river was more affected by the combined combustion sources and petroleum contamination, which was related to the complex human activities around the river [50].

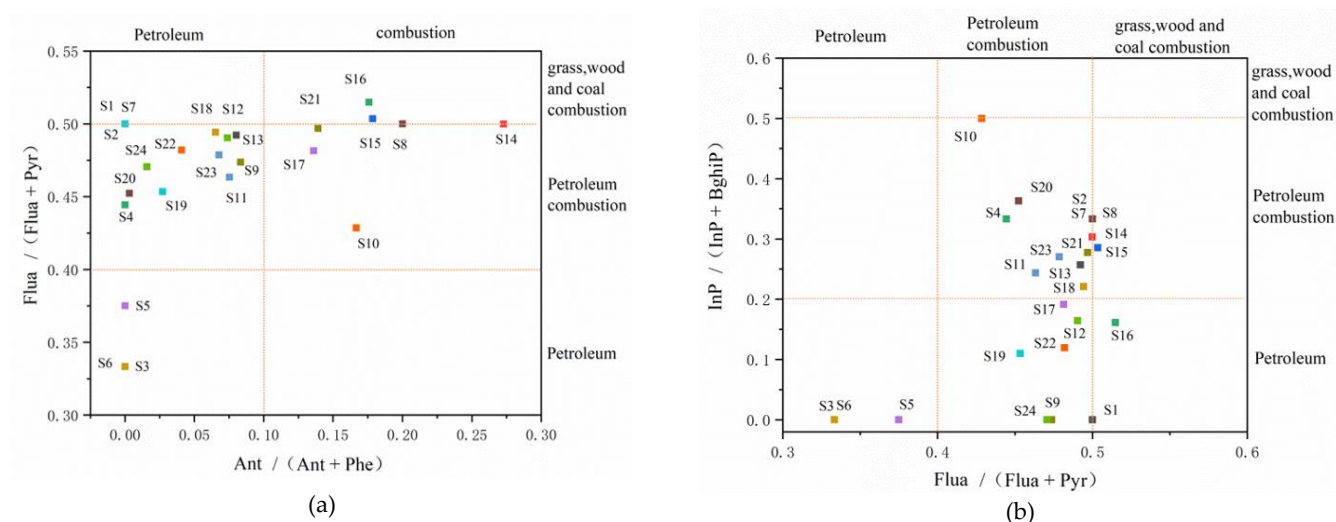


Figure 5. Plots of diagnostic ratios: (a) $\text{Flua} / (\text{Flua} + \text{Pyr})$ versus $\text{Ant} / (\text{Ant} + \text{Phe})$; (b) $\text{InP} / (\text{InP} + \text{BghiP})$ versus $\text{Flua} / (\text{Flua} + \text{Pyr})$ in sediments from the lake bay and adjoining river.

3.2.2. Principal Component Analysis

Principal components analysis (PCA) was performed to further understand the relationship between PAH compositions and the possible chemical sources for various factors (Table 2). The first principal component (PC1) accounted for 60.552% of total variance and the second (PC2) for 31.951%, thus accounting for 92.504% of the total variance. PC1 had heavy loading for most (4–6) rings PAHs compounds and was heavily loaded with Ana. Higher relative abundances of BbF, BkF, BaP, and DbA were suggested to indicate the incomplete combustion of liquid fossil fuels in previous studies [51]. BghiP and InP were considered to be derived from the emissions of gasoline and diesel engines combustion systems [52]. In general, Ana, Flua and Pyr could be designated as the important indicator of wood and coal combustion [53], which could be related to industrial emissions and local enterprise production activities. Based on the results, it was inferred that PC1 referred to the contribution of surrounding industrial pollution and traffic emissions to the import of PAHs. PC2 had a high loading for 3 rings PAHs compounds (Flu, Any, Phe, Ant). Flu, Phe, and Ant mainly derived from activities on petroleum raw materials, such as the volatilization and leakage of petroleum products [54]. This could be due to anthropogenic activities such as ship repairing and oil spills from ships in transport [55]. Thus, PC2 was assigned as a source from petroleum.

Table 2. The component loading after varimax rotation and cumulative variance of principal component analysis (PCA) of PAHs in surface sediment from the lake bay and adjoining river of Taihu Lake.

PAHs	Principal Component	
	PC1	PC2
Nap	0.366	0.343
Ana	0.830	0.376
Flu	0.219	0.959
Any	0.367	0.906
Phe	0.711	0.687
Ant	0.447	0.885
Flua	0.868	0.490
Pyr	0.874	0.482
Chr	0.803	0.570
BaA	0.883	0.466
BkF	0.956	0.284
BbF	0.917	0.390
BaP	0.859	0.501
DbA	0.876	0.470
BghiP	0.956	0.270
InP	0.927	0.256
Eigenvalues	9.688	5.112
Variance %	60.552	31.951
Cumulative variance %	60.552	92.504

Abbreviation: Nap, naphthalene; Ana, acenaphthene; Flu, fluorene; Phe, phenanthrene; Ant, anthracene; Flua, fluoranthene; Pyr, pyrene; BaA, benz(a)anthracene; Chr, chrysene; BbF, benzo(b)fluoranthene; BkF, benzo(k)fluoranthene; BaP, benzo(a)pyrene; DbA, dibenz(a,h)anthracene; BghiP, benzo(g,h,i)perylene; Any, acenaphthylene; InP, indeno(1,2,3-cd)pyrene.

In summary, the results obtained from PCA indicated that surrounding industrial and agricultural production, traffic emissions, and oil leakage from ships were the main sources of PAHs in surface sediments from the study area of Taihu Lake.

3.3. Risk Assessments of PAHs in Surface Sediments

Sediment Quality Guidelines (SQGs) developed on the basis of the Biological Effects Database for Sediments (BEDS) are very useful for the evaluation of the quality of sediment in freshwater and marine environments [56,57]. SQGs provide two guideline values: effects range low (ERL) and effects range median (ERM), which are established using the 10th and 50th percentiles, respectively, in a database of increasing concentrations associated with adverse biological effects. PAHs concentration below the effect range low (ERL) value implies that the sediment PAHs do not adversely affect organisms, whereas PAHs concentration higher than the effect range median (ERM) value could cause frequent damage to the biota. Concentrations values between the ERL and ERM imply a possible effects range within which effects occur either potentially or irregularly [58]. In addition, the risk estimation of PAHs in sediments was also determined according to the Hazard Quotient (HQ) [59–61]. The estimated concentration of each PAH in solution in sediment porewater was divided by its chronic toxicity value to derive a HQ [59–61]. The $HQ \geq 1$ indicate negative effects for exposed receptors, and $HQ < 1$ show that there are no negative effects.

The toxicity assessment form of PAHs in sediments from the macrophyte, algae lake bay and adjoining rivers of Taihu Lake (Table 3) showed that the concentration of Nap at all sampling sites was higher than that of ERM, which indicated that the ecological environment of the area of the lake bay and adjoining river in this study was greatly damaged by the 2-ring PAHs compound (Nap). It may be adversely affected by oil spills from traffic ships in the area and should be paid more attention. The ecological risk of the macrophyte-growing, cyanobacterial bloom-occurring lake zone was generally lower than that of the adjoining river of Taihu Lake, and there existed potential adverse biological effects in partial areas of the lake bay for 3-ring PAHs (Flu, Phe) and 5-ring (BkF and BbF). Except for Any, Ana, and InP, the remaining 13 PAHs all posed high ecological risks to some sites of the adjoining river (mainly Mashan River). This was due to the fact that the surrounding industrial pollution and resident activities had large adverse environmental impacts, and risk monitoring in these areas should be strengthened. In general, the ecological risk of the adjoining river of Taihu Lake was relatively high, and the macrophyte-growing, cyanobacterial bloom-occurring lake zone showed a high potential ecological risk. Furthermore, the results of HQ values (Table 3) showed that the 2-ring PAHs compound (Nap) in sediments would pose negative effects with the HQ value (2.52) in the ecological risk assessment.

Table 3. Toxicity risk assessment for surface sediments from studied lake bay and adjoining river using Sediment Quality Guidelines (SQGs) and Hazard Quotient (HQ).

Com- pound	SQG (ng g ⁻¹)		Range	Mean	<ERL		ERL–ERM		>ERM		HQ
	ERL	ERM			Lake Bay	River	Lake Bay	River	Lake Bay	River	
Nap	160	2100	2951.2– 14,800	5738.2	0	0	0	0	12	12	2.52
Any	16	500	N.D.–167.8	26.5	12	6	0	6	0	0	0.01
Ana	44	640	N.D.–623.4	60.2	12	6	0	6	0	0	0.03
Flu	19	540	N.D.–167.8	90.6	8	4	4	7	0	1	0.04
Phe	240	1500	200–3835.7	1004.3	3	0	9	7	0	5	0.63
Ant	85.3	1100	N.D.–1439.5	179.5	12	4	0	7	0	1	0.09
Flua	600	5100	100–8741.6	1585.3	11	5	1	4	0	3	0.87
Pyr	665	2600	200–8849.1	1627.6	11	5	1	2	0	5	0.9
BaA	261	1600	N.D.–3852.5	761.0	9	5	3	2	0	5	0.47
Chr	384	2800	28–5415.2	921.3	11	5	1	4	0	3	0.58
BbF	320	1880	N.D.–8276.5	1153.1	9	5	3	2	0	5	0.63

BkF	280	1620	2.7–3457.6	588.1	8	4	4	6	0	2	0.35
BaP	430	1600	N.D.–4374	798.6	11	6	1	1	0	5	0.49
InP	-	-	N.D.–3144.1	362.5	-	-	-	-	-	-	0.28
DbA	63.4	260	N.D.–740.4	107.9	11	6	1	1	0	5	0.15
BghiP	430	1600	N.D.–8191.6	1160	11	5	1	2	0	5	0.75

N.D., no detectable; ERL, effect range low; ERM, effect range medium; HQ, Hazard Quotient.

4. Conclusions

In this work, 16 priority PAHs in sediments from the macrophyte-growing, cyanobacterial bloom-occurring lake bay and adjoining rivers in Taihu Lake were investigated, and possible source contributions were estimated based on the isomer ratios diagnoses and PCA analysis. High levels of PAHs were observed in riverine sediments. The contamination of PAHs in lake bay sediments had become more severe in some highly industrializing areas. The distribution of PAHs concentration in sediments was more dependent on specific pollutant sources than lake-zone type or other environmental factors. The combined combustion sources and petroleum contamination contribute to PAHs contamination of sediments. The 2-ring PAH compound (Nap) in sediments might pose negative effects for exposed receptors according to the ecological risk assessment. The findings of this study should help in understanding the levels, sources, and ecological risks of PAHs in the lake bay and adjoining rivers ecosystems, and to provide valuable information for implementing pollution reduction policies and protecting water resources and human health in these areas.

Author Contributions: Conceptualization, Y.D., H.W., and Z.Y.; methodology, Y.D.; validation, G.Z.; formal analysis, Y.D.; investigation, Y.D., M.Y., and H.Z.; data curation, H.W. and Y.D.; writing—original draft preparation, Y.D.; visualization, H.W.; writing—review and editing, Z.Y. and H.W.; project administration, Z.Y. and H.W. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by grants from National Natural Science Foundation of China (41671496 and 41977361), the National Science and Technology Major Project (2017ZX07204005), and Major Program of Natural Science Foundation of Jiangsu Province (BE2016703).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data sharing not applicable.

Acknowledgments: Not applicable.

Conflicts of Interest: There are no conflicts of interest to declare.

References

1. Yan, Z.; Song, N.; Wang, C.; Jiang, H. Functional potential and assembly of microbes from sediments in a lake bay and adjoining river ecosystem for polycyclic aromatic hydrocarbon biodegradation. *Environ. Microbiol.* **2020**, doi:10.1111/1462-2920.15104.
2. Yan, Z.; Hao, Z.; Wu, H.; Jiang, H.; Yang, M.; Wang, C. Co-occurrence patterns of the microbial community in polycyclic aromatic hydrocarbon-contaminated riverine sediments. *J. Hazard. Mater.* **2019**, *367*, 99–108.
3. Yan, Z.; He, Y.; Cai, H.; Van Nostrand, J.D.; He, Z.; Zhou, J.; Krumholz, L.R.; Jiang, H.-L. Interconnection of key microbial functional genes for enhanced benzo [a] pyrene biodegradation in sediments by microbial electrochemistry. *Environ. Sci. Technol.* **2017**, *51*, 8519–8529.
4. Nowacka, A.; Włodarczyk-Makuła, M. Monitoring of polycyclic aromatic hydrocarbons in water during preparation processes. *Polycycl. Aromat. Compd.* **2013**, *33*, 430–450.
5. DeBruyn, J.M.; Mead, T.J.; Wilhelm, S.W.; Sayler, G.S. PAH biodegradative genotypes in Lake Erie sediments: Evidence for broad geographical distribution of pyrene-degrading mycobacteria. *Environ. Sci. Technol.* **2009**, *43*, 3467–3473.
6. Jia, H.; Zhao, S.; Shi, Y.; Zhu, L.; Wang, C.; Sharma, V.K. Transformation of polycyclic aromatic hydrocarbons and formation of environmentally persistent free radicals on modified montmorillonite: The role of surface metal ions and polycyclic aromatic hydrocarbon molecular properties. *Environ. Sci. Technol.* **2018**, *52*, 5725–5733.

7. Haritash, A.; Kaushik, C. Biodegradation aspects of polycyclic aromatic hydrocarbons (PAHs): A review. *J. Hazard. Mater.* **2009**, *169*, 1–15.
8. Zhao, Z.; Zhang, L.; Wu, J. Polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) in sediments from lakes along the middle-lower reaches of the Yangtze River and the Huaihe River of China. *Limnol. Oceanogr.* **2016**, *61*, 47–60, doi:10.1002/lno.10197.
9. Wang, J.; Xia, K.; Waigi, M.G.; Gao, Y.; Odinga, E.S.; Ling, W.; Liu, J. Application of biochar to soils may result in plant contamination and human cancer risk due to exposure of polycyclic aromatic hydrocarbons. *Environ. Int.* **2018**, *121*, 169–177.
10. Hao, Z.; Wang, Q.; Yan, Z.; Jiang, H. Novel magnetic loofah sponge biochar enhancing microbial responses for the remediation of polycyclic aromatic hydrocarbons-contaminated sediment. *J. Hazard. Mater.* **2021**, *401*, 123859, doi:10.1016/j.jhazmat.2020.123859.
11. Han, J.; Liang, Y.; Zhao, B.; Wang, Y.; Xing, F.; Qin, L. Polycyclic aromatic hydrocarbon (PAHs) geographical distribution in China and their source, risk assessment analysis. *Environ. Pollut.* **2019**, *251*, 312–327.
12. Yan, Z.; Jiang, H.; Li, X.; Shi, Y. Accelerated removal of pyrene and benzo[a]pyrene in freshwater sediments with amendment of cyanobacteria-derived organic matter. *J. Hazard. Mater.* **2014**, *272*, 66–74.
13. Song, N.; Xu, H.; Yan, Z.; Yang, T.; Wang, C.; Jiang, H.-L. Improved lignin degradation through distinct microbial community in subsurface sediments of one eutrophic lake. *Renew. Energy* **2019**, *138*, 861–869.
14. Song, N.; Yan, Z.-S.; Cai, H.-Y.; Jiang, H.-L. Effect of temperature on submerged macrophyte litter decomposition within sediments from a large shallow and subtropical freshwater lake. *Hydrobiol.* **2013**, *714*, 131–144.
15. Tao, Y.; Li, W.; Xue, B.; Zhong, J.; Yao, S.; Wu, Q. Different effects of copper (II), cadmium (II) and phosphate on the sorption of phenanthrene on the biomass of cyanobacteria. *J. Hazard. Mater.* **2013**, *261*, 21–28.
16. Dachs, J.; Eisenreich, S.J.; Hoff, R.M. Influence of eutrophication on air–water exchange, vertical fluxes, and phytoplankton concentrations of persistent organic pollutants. *Environ. Sci. Technol.* **2000**, *34*, 1095–1102.
17. Jouanneau, Y.; Willison, J.C.; Meyer, C.; Krivobok, S.; Chevron, N.; Besombes, J.-L.; Blake, G. Stimulation of pyrene mineralization in freshwater sediments by bacterial and plant bioaugmentation. *Environ. Sci. Technol.* **2005**, *39*, 5729–5735.
18. Yan, Z.; Jiang, H.; Cai, H.; Zhou, Y.; Krumholz, L.R. Complex interactions between the macrophyte *Acorus calamus* and microbial fuel cells during pyrene and benzo[a]pyrene degradation in sediments. *Sci. Rep.* **2015**, *5*, 10709, doi:10.1038/srep10709.
19. Yan, Z.; Zhang, H.; Wu, H.; Yang, M.; Wang, S. Occurrence and removal of polycyclic aromatic hydrocarbons in real textile dyeing wastewater treatment process. *Desalin. Water Treat.* **2016**, *57*, 22564–22572.
20. Włodarczyk-Makula, M. The loads of PAHs in wastewater and sewage sludge of municipal treatment plant. *Polycycl. Aromat. Compd.* **2005**, *25*, 183–194.
21. Meng, Y.; Liu, X.; Lu, S.; Zhang, T.; Jin, B.; Wang, Q.; Tang, Z.; Liu, Y.; Guo, X.; Zhou, J. A review on occurrence and risk of polycyclic aromatic hydrocarbons (PAHs) in lakes of China. *Sci. Total Environ.* **2019**, *651*, 2497–2506.
22. Tao, Y.; Yao, S.; Xue, B.; Deng, J.; Wang, X.; Feng, M.; Hu, W. Polycyclic aromatic hydrocarbons in surface sediments from drinking water sources of Taihu Lake, China: Sources, partitioning and toxicological risk. *J. Environ. Monit.* **2010**, *12*, 2282–2289.
23. Qian, X.; Liang, B.; Liu, X.; Wang, J.; Liu, F.; Cui, B. Distribution, sources, and ecological risk assessment of polycyclic aromatic hydrocarbons in surface sediments from the Haihe River, a typical polluted urban river in Northern China. *Environ. Sci. Pollut. Res.* **2017**, *24*, 17153–17165.
24. Li, C.; Huo, S.; Yu, Z.; Xi, B.; Zeng, X.; Wu, F. Spatial distribution, potential risk assessment, and source apportionment of polycyclic aromatic hydrocarbons (PAHs) in sediments of Lake Chaohu, China. *Environ. Sci. Pollut. Res.* **2014**, *21*, 12028–12039.
25. Zhang, Y.; Guo, C.-S.; Xu, J.; Tian, Y.-Z.; Shi, G.-L.; Feng, Y.-C. Potential source contributions and risk assessment of PAHs in sediments from Taihu Lake, China: Comparison of three receptor models. *Water Res.* **2012**, *46*, 3065–3073.
26. Zhao, Z.; Zhang, L.; Deng, J.; Wu, J. The potential effects of phytoplankton on the occurrence of organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbons (PAHs) in water from Lake Taihu, China. *Environ. Sci.: Processes Impacts* **2015**, *17*, 1150–1156.
27. Tao, Y.; Yu, J.; Lei, G.; Xue, B.; Zhang, F.; Yao, S. Indirect influence of eutrophication on air–water exchange fluxes, sinking fluxes, and occurrence of polycyclic aromatic hydrocarbons. *Water Res.* **2017**, *122*, 512–525.
28. Yan, Z.; Zhang, Y.; Wu, H.; Yang, M.; Zhang, H.; Hao, Z.; Jiang, H. Isolation and characterization of a bacterial strain *Hydrogenophaga* sp. PYR1 for anaerobic pyrene and benzo [a] pyrene biodegradation. *RSC Adv.* **2017**, *7*, 46690–46698.
29. Zhao, S.; Wang, B.; Wang, D.; Li, X.; Huang, B.; Hu, P.; Zhang, L.; Pan, X. Environmental Behavior of PAHs in Dianchi Lake Distributions, Sources and Risk Assessment of Polycyclic Aromatic Hydrocarbons in Surface Sediments from Dianchi Lake, China. *Int. J. Environ. Res.* **2014**, *8*, 317–328.
30. Ren, C.; Wu, Y.; Zhang, S.; Wu, L.-L.; Liang, X.-G.; Chen, T.-H.; Zhu, C.-Z.; Sojinu, S.O.; Wang, J.-Z. PAHs in sediment cores at main river estuaries of Chaohu Lake: Implication for the change of local anthropogenic activities. *Environ. Sci. Pollut. Res.* **2015**, *22*, 1687–1696.
31. Li, H.-L.; Gao, H.; Zhu, C.; Li, G.-G.; Yang, F.; Gong, Z.-Y.; Lian, J. Spatial and temporal distribution of polycyclic aromatic hydrocarbons (PAHs) in sediments of the Nansi Lake, China. *Environ. Monit. Assess.* **2009**, *154*, 469, doi:10.1007/s10661-009-0752-9.
32. White, J.; Triplett, T. Polycyclic aromatic hydrocarbons (PAHs) in the sediments and fish of the Mill River, New Haven, Connecticut, USA. *Bull. Environ. Contam. Toxicol.* **2002**, *68*, 104–110.

33. Zakaria, M.P.; Takada, H.; Tsutsumi, S.; Ohno, K.; Yamada, J.; Kouno, E.; Kumata, H. Distribution of polycyclic aromatic hydrocarbons (PAHs) in rivers and estuaries in Malaysia: A widespread input of petrogenic PAHs. *Environ. Sci. Technol.* **2002**, *36*, 1907–1918.
34. Liu, F.; Liu, J.; Chen, Q.; Wang, B.; Cao, Z. Pollution characteristics and ecological risk of polycyclic aromatic hydrocarbons (PAHs) in surface sediments of the southern part of the Haihe River system in China. *Chin. Sci. Bull.* **2013**, *58*, 3348–3356.
35. Yuan, H.; Zhang, E.; Lin, Q.; Wang, R.; Liu, E. Sources appointment and ecological risk assessment of polycyclic aromatic hydrocarbons (PAHs) in sediments of Erhai Lake, a low-latitude and high-altitude lake in southwest China. *Environ. Sci. Pollut. Res.* **2016**, *23*, 4430–4441.
36. Bai, Y.; Meng, W.; Xu, J.; Zhang, Y.; Guo, C.; Lv, J.; Wan, J. Occurrence, distribution, environmental risk assessment and source apportionment of polycyclic aromatic hydrocarbons (PAHs) in water and sediments of the Liaohe River Basin, China. *Bull. Environ. Contam. Toxicol.* **2014**, *93*, 744–751.
37. Hu, G.; Luo, X.; Li, F.; Dai, J.; Guo, J.; Chen, S.; Hong, C.; Mai, B.; Xu, M. Organochlorine compounds and polycyclic aromatic hydrocarbons in surface sediment from Baiyangdian Lake, North China: Concentrations, sources profiles and potential risk. *Journal of Environmental Sciences* **2010**, *22*, 176–183.
38. Ko, F.-C.; Baker, J.; Fang, M.-D.; Lee, C.-L. Composition and distribution of polycyclic aromatic hydrocarbons in the surface sediments from the Susquehanna River. *Chemosphere* **2007**, *66*, 277–285.
39. Hale, S.E.; Meynet, P.; Davenport, R.J.; Jones, D.M.; Werner, D. Changes in polycyclic aromatic hydrocarbon availability in River Tyne sediment following bioremediation treatments or activated carbon amendment. *Water Res.* **2010**, *44*, 4529–4536.
40. Barakat, A.O.; Mostafa, A.; El-Sayed, N.B.; Wade, T.L.; Sweet, S.T. Polycyclic aromatic hydrocarbons (PAHs) in surface sediments of Lake Manzala, Egypt. *Soil Sediment. Contam.: An. Int. J.* **2013**, *22*, 315–331.
41. Oyo-Ita, O.E.; Offem, J.O.; Ekpo, B.O.; Adie, P.A. Anthropogenic PAHs in mangrove sediments of the Calabar River, SE Niger Delta, Nigeria. *Appl. Geochem.* **2013**, *28*, 212–219.
42. Pozo, K.; Perra, G.; Menchi, V.; Urrutia, R.; Parra, O.; Rudolph, A.; Focardi, S. Levels and spatial distribution of polycyclic aromatic hydrocarbons (PAHs) in sediments from Lengua Estuary, central Chile. *Mar. Pollut. Bull.* **2011**, *62*, 1572–1576.
43. Arias, A.H.; Vazquez-Botello, A.; Tombesi, N.; Ponce-Vélez, G.; Freije, H.; Marcovecchio, J. Presence, distribution, and origins of polycyclic aromatic hydrocarbons (PAHs) in sediments from Bahía Blanca estuary, Argentina. *Environ. Monit. Assess.* **2010**, *160*, 301, doi:10.1007/s10661-008-0696-5.
44. Baumard, P.; Budzinski, H.; Garrigues, P. Polycyclic aromatic hydrocarbons in sediments and mussels of the western Mediterranean Sea. *Environ. Toxicol. and Chem.: An. Int. J.* **1998**, *17*, 765–776.
45. Sanders, M.; Sivertsen, S.; Scott, G. Origin and distribution of polycyclic aromatic hydrocarbons in surficial sediments from the Savannah River. *Arch. Environ. Contam. Toxicol.* **2002**, *43*, 438–448.
46. Qian, X.; Liang, B.; Fu, W.; Liu, X.; Cui, B. Polycyclic aromatic hydrocarbons (PAHs) in surface sediments from the intertidal zone of Bohai Bay, Northeast China: Spatial distribution, composition, sources and ecological risk assessment. *Mar. Pollut. Bull.* **2016**, *112*, 349–358.
47. Deng, W.; Li, X.G.; Li, S.Y.; Ma, Y.Y. Source apportionment of polycyclic aromatic hydrocarbons in surface sediment of mud areas in the East China Sea using diagnostic ratios and factor analysis. *Mar. Pollut. Bull.* **2013**, *70*, 266–273.
48. Wang, L.; Yang, Z.; Niu, J.; Wang, J. Characterization, ecological risk assessment and source diagnostics of polycyclic aromatic hydrocarbons in water column of the Yellow River Delta, one of the most plenty biodiversity zones in the world. *J. Hazard. Mater.* **2009**, *169*, 460–465.
49. Tobiszewski, M.; Namieśnik, J. PAH diagnostic ratios for the identification of pollution emission sources. *Environ. Pollut.* **2012**, *162*, 110–119.
50. Zhao, Z.; Jiang, Y.; Li, Q.; Cai, Y.; Yin, H.; Zhang, L.; Zhang, J. Spatial correlation analysis of polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (OCPs) in sediments between Taihu Lake and its tributary rivers. *Ecotoxicol. Environ. Saf.* **2017**, *142*, 117–128.
51. Iqbal, J.; Overton, E.B.; Gisclair, D. Sources of polycyclic aromatic hydrocarbons in Louisiana Rivers and coastal environments: Principal components analysis. *Environ. Forensics* **2008**, *9*, 310–319.
52. Zhang, K.; Wang, J.-Z.; Liang, B.; Zeng, E.Y. Occurrence of polycyclic aromatic hydrocarbons in surface sediments of a highly urbanized river system with special reference to energy consumption patterns. *Environ. Pollut.* **2011**, *159*, 1510–1515.
53. Wang, Y.; Shen, C.; Shen, Z.; Zhang, D.; Crittenden, J.C. Spatial variation and sources of polycyclic aromatic hydrocarbons (PAHs) in surface sediments from the Yangtze Estuary, China. *Environ. Sci.: Process. Impacts* **2015**, *17*, 1340–1347.
54. Ribeiro, J.; Silva, T.; Mendonça Filho, J.G.; Flores, D. Polycyclic aromatic hydrocarbons (PAHs) in burning and non-burning coal waste piles. *J. Hazard. Mater.* **2012**, *199*, 105–110.
55. Santos, F.R.; Martins, D.A.; Morais, P.C.; Oliveira, A.H.; Gama, A.F.; Nascimento, R.F.; Choi-Lima, K.F.; Moreira, L.B.; Abessa, D.M.; Nelson, R.K. Influence of anthropogenic activities and risk assessment on protected mangrove forest using traditional and emerging molecular markers (Ceará coast, northeastern Brazil). *Sci. Total Environ.* **2019**, *656*, 877–888.
56. Long, E.; Ingersoll, C.; MacDonald, D. Calculation and uses of mean sediment quality guideline quotients: A critical review. *Environ. Sci. Technol.* **2006**, *40*, 1726–1736, doi: 10.1021/es058012d.
57. Li, Y.; Liu, X.; Liu, M.; Li, X.; Wang, Q.; Zhu, J.; Qadeer, A. Distribution, sources and ecological risk of polycyclic aromatic hydrocarbons in the estuarine-coastal sediments in the East China Sea. *Environ. Sci.: Process. Impacts* **2017**, *19*, 561–569.

-
58. Long, E.R.; MacDonald, D.D.; Smith, S.L.; Calder, F.D. Incidence of adverse biological effects within ranges of chemical concentrations in marine and estuarine sediments. *Environ. Manage.* **1995**, *19*, 81–97.
 59. Lin, B.-L.; Meng, Y.; Kamo, M.; Naito, W. An all-in-one tool for multipurpose ecological risk assessment and management (MeRAM) of chemical substances in aquatic environment. *Chemosphere* **2020**, 128826, doi:10.1016/j.chemosphere.2020.128826.
 60. Ancione, G.; Lisi, R.; Milazzo, M.F. Human health risk associated with emissions of volatile organic compounds due to the ship-loading of hydrocarbons in refineries. *Atmos. Pollut. Res.* **2020**. doi:10.1016/j.apr.2020.12.004.
 61. Neff, J.M.; Stout, S.A.; Gunster, D.G. Ecological risk assessment of polycyclic aromatic hydrocarbons in sediments: Identifying sources and ecological hazard. *Integrated Environ. Assess. Manag.: Int. J.* **2005**, *1*, 22–33.