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ORIGINAL PAPER

Distribution, sources, and risk assessment of polycyclic aromatic hydrocarbons (PAHs) in Kaokaowusu river sediments near a coal industrial zone

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Abstract This study systematically analyzed the contents, compositions, and sources of polycyclic aromatic hydrocarbons (PAHs) in river sediments near an important energy and chemical base in northwest China. In addition, their possible adverse effects on the ecology and human health were assessed. The PAH concentrations in this study area ranged from 2641.28 to 16783.72 (ng/g dw). PAHs of medium molecular weight (3-ring and 4-ring) showed the largest proportion, followed by PAHs of higher molecular weight (5-ring and 6-ring). The results of molecular diagnostic ratios and principal component analysis revealed that PAHs in the region have complex sources, with incomplete combustion of local fossil fuels and traffic exhaust factors being the main sources. The total toxic equivalent concentration of PAHs varied from 10.05 to 760.26 ng/g, and according to the sediment quality guidelines, PAHs have high potential ecological risk in the lower reaches of the river. The mean effect range-median quotient for the region was 0.46, and the combined ecological risk was at moderate to high levels (21% probability of toxicity). The lifetime carcinogenic risks for adults and children exposed to PAHs were 2.95×10^{-3} and 1.87×10^{-2}, respectively, which are much higher than the limit of 10^{-4}, indicating moderate to high potential cancer risks. Therefore, the local government should consider taking some environmental remediation measures. This study can provide theoretical support for pollution prevention measures and ecological restoration strategies for rivers in resource-rich areas.

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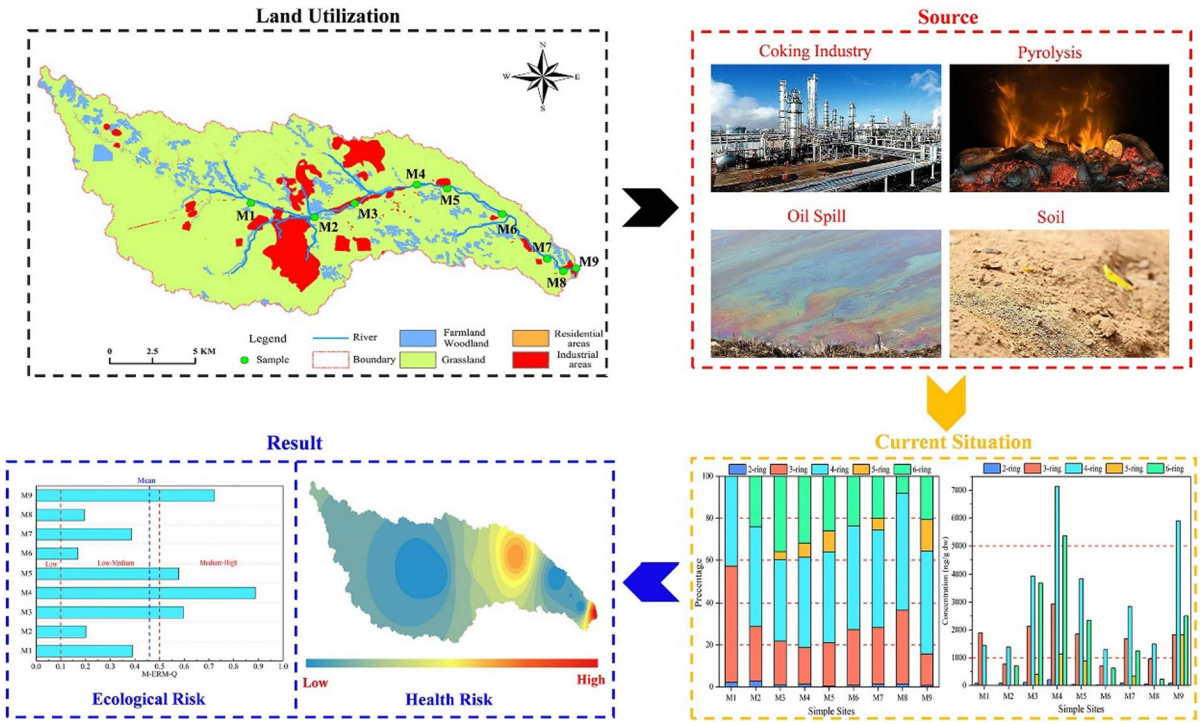
Graphical abstract



Keywords PAHs · Pollution characteristics · Source apportionment · Risk assessment · Industrial area · Sediment

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are hydrophobic aromatic compounds with two or more benzene rings (Pang et al., 2021), They have adverse effects on the human respiratory system and nervous system, and are teratogenic, carcinogenic, and mutagenic (Maleti et al., 2019; Ranjbar et al., 2019). Moreover, they can migrate over long distances, further increasing their potential risks. Therefore, the hazards of PAHs to the human body and environment attracted the wide attention of scholars around the world. In 1976, the U.S. Environmental Protection Agency selected 16 PAHs as precedence-controlled pollutants (Kieth & Telliard, 1979). PAHs have been found to be widespread in the natural environment, mainly originating from human activities, such as oil leakage, incomplete combustion of organic matter (coal, oil, wood), steel production, industrial emissions (Li et al., 2015, 2022; Suman et al., 2016). Owing to their hydrophobic and lipophilic nature (Zeng et al., 2018), PAHs in the atmosphere and rivers are prone to adsorption on suspended matter and ultimately settling in river sediments and soils. River sediments and soils serve as the source as well as the sinks of various pollutants, with low spatiotemporal variation (Li et al., 2015). The chemical composition and pollution level of sediments can accurately reflect the actual pollution status of the environment and are thus important indicators in the assessment of environmental pollution and ecological risk (Jing et al., 2018).

The Kuye River is a first-class tributary of the Yellow River in Northwest China, located in the transition zone between the Loess Plateau and the Mu Us Desert. The Kuye River basin has a large number of coal mines and coal processing heavy industrial areas, including the Shenfu coal field, which is one of the seven largest

coalfields in the world. Therefore, it is an important energy and chemical industry base in China. Under the situation of long-term coal mining, transportation, and processing, the ecological environment of the basin is very fragile, with complex pollutant sources. Moreover, the basin faces serious problems of soil erosion and desertification. The contradiction between water resources and water pollution is very strong. However, most studies on rivers in this region focused on water resources and sediment loss, paying little attention to water pollution, especially organic matter pollution. According to previous studies, the content, origin, and ecological risks of PAHs in river sediments are related to many factors, such as the levels of local economic development and industrialization, climatic factors, geographical conditions, physical and chemical properties of sediments, land use types, and human activities (Barhoumi et al., 2019; Lv et al., 2020; Van Metre et al., 2022). Zhang et al. (2022) discussed the impact of human lifestyle and land type on the PAH content in the soil of Beiluo River and discovered that the PAHs were mainly related to local energy production, with significantly higher contents in the dry season than in the wet season because of water dilution. Ma et al. (2022) investigated the cause of PAHs in the sediments of Dian Lake using molecular diagnostic ratio and principal component analysis (PCA), and found that the primary source of PAHs was the incomplete combustion of local coke and biomass, with high molecular weight PAHs in pore water reaching the levels of chronic or even acute toxicity. In the economically developed Pearl River Delta, industry and transportation development are relatively high, and frequent human activities have led to a medium risk level in the whole basin (Li et al., 2021).

In this study, nine river sediments samples (KaoKaowusu River) were collected from an industrial area and sixteen PAHs of serious concern were analyzed (Table S1). The objectives of this study were to: (1) explore the content and composition of PAHs in river sediments of industrial areas; (2) use molecular diagnostic ratio and PCA to qualitatively and quantitatively investigate the possible sources of PAHs; (3) assess the biological toxicity of PAHs, detrimental effects on local aquatic organisms and ecological risks based on the total equivalent concentration of BaP (TEQ_{BaP}), sediment quality guidelines (SQG), and mean effect range-median quotient (M-ERM-Q); (4) quantify of the exposure risk of sedimentary PAHs to adults and children using a lifetime carcinogenic risk model.

Materials and methods

Study area and sampling

This research area is located in the national heavy industry district of the Kuye River

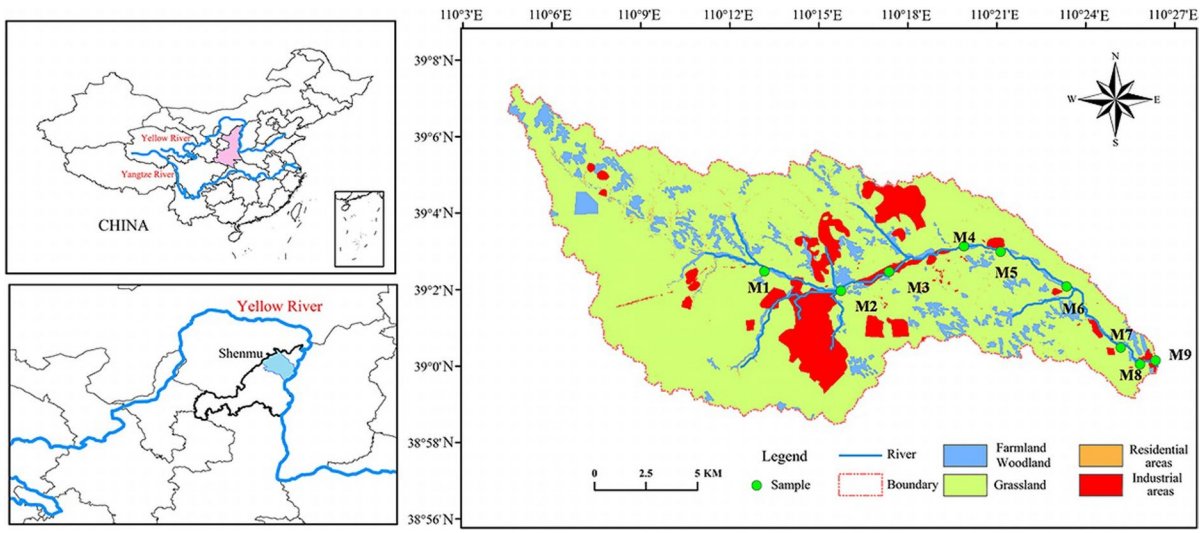


Fig. 1 Regional location of Kaokaowusu River and distribution of sampling points

Basin—Kaokaowusu River, which is a secondary branch of the Yellow River, with a total length of approximately 25.7 km and an area of approximately 252.2 km^2 (Fig. 1). The annual average temperature in this area is 8.9 °C, annual precipitation is 440.8 mm, and soil is dominated by aeolian sandy soil and loess. The main land types are industrial, residential, meadow, and woodland. The Ningtiaota Industrial Zone, which is based on coal mining and mainly produces gas and coal tar, is located upstream of the river, The industrial zone comprises industrial and mining enterprises of different scales. Woodland, and small farmlands distributed along both sides of the river. Downstream of the river is the residential area of Dianta Town, which also includes power plants and various mechanical repair factories.

In August 2021, a total of nine sediment samples were collected using clean stainless steel from a depth of 0–20 cm. A 1 m^2 sampling grid was set up in the river channel and a steel shovel was used to collect five sediment samples at the four corners and the center, after removing debris, such as stones and branches, the sediments were placed in polyethylene bags and mixed evenly. The location and number of the sampling points were then accurately recorded using a GPS locator. The specific coordinates are listed in Table S2.

Extraction and determination

Pretreatment: PAHs were extracted from sediments according to the method of Han et al. (2021). In brief, sediment samples of certain weight were weighed, and after freeze-drying for 24 h, the sediment samples were ground and filtered through a 100-mesh screen. Subsequently, 1.2 g of active copper powder and 25 mL of dichloromethane (CH_2Cl_2) and normal hexane (C_6H_{14}) (volume ratio of 1:1) were added, ultrasonic extraction was performed for 20 min, and the supernatant was extracted through centrifugation. The above steps were repeated four times in total. The mixture was stripped to 2 mL with nitrogen, further purified, and concentrated with a silica gel/alumina/anhydrous sodium sulfate column. The cylinder was then rinsed again with an admixture of 50 mL of dichloromethane and n-hexane. Finally, nitrogen was used to blow off the mixed liquid to almost dryness, and the volume was rinsed to 1.5 mL with n-hexane. Finally, a 10 μL standard solution of sixteen PAHs was added before the measurement.

Instrument and determination: The sixteen PAHs were analyzed using mature determination methods that have been studied by our research group (Yang et al., 2022). The detection was performed using an Agilent 7890 A gas chromatograph; the chromatographic column type was HP-5 ms, the carrier gas was super-purity nitrogen, and the injection volume was 1 μL. Firstly, the initial oven temperature was maintained at 80 °C for 4 min; Subsequently, it was increased to 170 °C at 8 °C/min, and maintained for 10 min; it was further increased to 250 °C at 8 °C/min, and continued for 10 min; Finally, the temperature was continuously to 300 °C at 8 °C/min, and held for 10 min. The inlet temperature was 280 °C, the FID detector temperature was 320 °C, and the splitting ratio was 10:1. The recoveries of the 16 PAHs were 75.68–105.14%, and the detection limits were 0.1–0.49 ng/g dw, A spectrogram of the standard sample is shown in Fig. S1.

Analysis method

Source analysis of PAHs

PAHs in river sediments primarily originate from two sources: natural and anthropogenic sources. In general, small molecular weight PAHs are believed to stem from soil parent material, petroleum, and pyrolysis (Laflamme & Hites, 1988), whereas macromolecular PAHs are believed to be products of combustion and pyrolysis (Li et al., 2018). Molecular diagnostic ratios, a simple source analysis method based on the physical and chemical properties of individual PAHs, have been diffusely utilized to qualitatively analyze PAH sources in water, atmosphere, and soil. In this study, four molecular ratios of Ant/(Ant + Phe), BaA/(BaA + Chr), Fla/(Fla + Pyr), and LMW/HMW were calculated to confirm the origin of PAHs in this area. Table S3 shows the possible source of PAHs corresponding to different molecular ratios.

PCA is a statistical method that can reduce the dimensionality of the data space. This method transforms a set of potentially correlated variables into a set of linearly independent variables through orthogonal transformation, and extracts factors with eigenvalues greater than 1, (Gdara et al., 2017; Wang et al., 2015). In this manner, the relationship and sources between PAHs can be analyzed. The method has been widely

used to determine how land use and the surrounding environment affect PAHs content in riverbed sediments. In this study, the PCA model of IBM SPSS Statistics 26.0 was used to quantitatively analyze the sources of PAHs.

Toxicity and ecological risk assessment

Equivalent concentration of BaP

BaP was the first discovered among the 16 PAHs analyzed in this study, and it is also the most toxic PAH (Aghadadashi et al., 2019). The concentration of PAHs at each site was converted into the equivalent concentration of BaP using Formula (1), and the calculation formula is as follows:

$$\mathrm{TEQ_{BaP}} = \sum_{i=1}^{16} C_i \times \mathrm{TEF} \tag{1}$$

where $\mathrm{TEQ_{BaP}}$ is the equivalent concentration of BaP; C_i is the measured content of various PAHs; TEF is the toxic equivalent factor of many types of PAHs. The specific data are presented in Table S4.

Sediment quality guidelines

SQG is an important method for assessing sediment pollution in river water bodies and soil (Long et al., 1995). It has been widely used in the environmental risk assessment of soil and estuary sediments, protection of aquatic benthic organisms, pollution control, and related research on water environment functions. In this study, the method was employed to evaluate the risk of sedimentary PAHs to benthic organisms and the ecological environment. In SQG, toxicity is measured in terms of the effects range low (ERL) and effects range-median (ERM). PAH contents below ERL indicate the absence or rare occurrence of negative biological effects; PAH contents between ERL and ERM indicate occasional occurrences of adverse biological effects; PAH contents above ERM indicate the frequent occurrence of adverse biological effects. The ERL and ERM of each PAH are listed in Table 3.

Mean effect range-median quotient

M-ERM-Q is suitable for calculating the comprehensive toxicity of PAHs in river water, suspended matter, sediment, and soil (Long et al., 1998; Yuan et al., 2021). It is calculated as follows:

$$M-\mathrm{ERM}-Q = \frac{\sum_{i=1}^{n} C_i/\mathrm{ERM}_i}{n} \tag{2}$$

In the formula, C_i is the content of a single PAH; ERM_i is the interval effect median concentration of the corresponding PAHs; n is the number of PAHs. Different risk levels are divided according to different M-ERM-Q values, the specific categories are listed in Table S5.

Human health risk assessment

Humans are exposed to the risks of PAHs in sediments mainly through oral intake, skin contact, and respiration. According to existing studies, the risk levels of these three pathways of exposure are as follows: oral cavity > skin > respiration (Froger et al., 2021). In this study, a lifetime carcinogenic risk model was selected to assess the carcinogenic potential of PAHs in adults and children in the region. The specific calculation methods of the three carcinogenic pathways are presented as formulas (3–5) (Kr et al., 2021). The specific meanings and values of the variables in the formulas are listed in Table S6, where $\mathrm{CS_{TEQ}}$ is the BaP equivalent concentration of the sediments at each site, and the calculation method is presented in Eq. 1.

$$\mathrm{ILCR_{ing}} = \frac{\mathrm{CS_{TEQ}}\times \mathrm{EF}\times \mathrm{ED}\times \mathrm{IR}}{\mathrm{BW}\times \mathrm{AT}\times 10^6} \times \mathrm{CSF}_o \tag{3}$$

$$\mathrm{ILCR_{der}} = \frac{\mathrm{CS_{TEQ}}\times \mathrm{EF}\times \mathrm{ED}\times \mathrm{SA}\times \mathrm{AF}\times \mathrm{ABS}}{\mathrm{BW}\times \mathrm{AT}\times 10^6} \times \frac{\mathrm{CSF}_o}{\mathrm{GIABS}} \tag{4}$$

$$\mathrm{ILCR_{inh}} = \frac{\mathrm{CS_{TEQ}}\times \mathrm{EF}\times \mathrm{ED}\times \mathrm{ET}\times \mathrm{IUR}}{\mathrm{AT}\times \mathrm{PEF}} \tag{5}$$

$$\mathrm{TLCR} = \mathrm{ILCR_{ing}} + \mathrm{ILCR_{der}} + \mathrm{ILCR_{inh}} \tag{6}$$

Results and discussion

Analysis of the levels of PAHs

Table 1 present the statistical characteristics of the concentrations of the 16 PAHs. Among them, the contents of BkF and BaP were both minor and below the limit of detection. The detection rates of DahA

Table 1 Descriptive statistics of the levels of PAH Congeners (ng/g dw) in the study area

PAH	Max	Median	Min	Mean	SD	CV (%)	Detection rate (%)
Nap	205.77	78.14	20.47	79.17	51.58	65.16	100
Acp	475.64	139.18	49.12	152.20	130.35	85.64	100
Ace	1653.42	751.73	242.82	751.79	491.70	65.40	100
Fle	1300.08	204.08	49.47	302.87	359.05	118.55	100
Phe	385.13	174.74	86.02	191.54	110.51	57.70	100
Ant	449.65	200.01	117.43	239.81	117.07	48.82	100
Fla	1451.68	386.09	72.14	437.19	437.60	100.09	100
Pyr	4979.64	2573.68	990.87	2382.78	1440.51	60.46	100
BaA	316.86	156.76	59.63	161.44	90.63	56.14	100
Chr	454.54	279.11	158.21	274.26	99.29	36.20	100
BbF	1268.09	368.60	ND	402.24	464.25	115.42	55.56
BkF	ND	ND	ND	ND	–	–	0
BaP	ND	ND	ND	ND	–	–	0
DahA	563.10	ND	ND	107.37	204.37	190.35	22.22
InP	634.32	ND	ND	140.29	262.46	187.09	22.22
BghiP	4723.47	1794.77	ND	1714.32	1471.22	85.82	88.89
∑PAH	16783.72	7578.13	2641.28	7337.26	4751.55	64.76	

ND, The content is below the monitoring limit; SD, standard deviation; CV, coefficient of variation; dw, dry weight

Table 2 Comparison of PAH levels (ng/g dw) in river sediments from the study area with those in other locations

Area	Country	PAHs compositions	Concentration range (Mean)	Degree of pollution
This research	China	16	2641.3–16783.7 (7337.3)	H–HV
Suzhou Industrial Park (Yuan et al., 2021)	China	15	180–81,000 (5700)	M–HV
Pearl River Delta (Yu et al., 2018)	China	15	55.7–381	L–M
Poyang Lake (Li et al., 2018)	China	16	73.2–367.2	L–M
Wangyang River(Zhang et al., 2015)	China	16	545.1–134054.7	M–HV
Liaohe River (He et al., 2016)	China	16	82.5–25374.4 (3149.2)	L–HV
Yangtze River (Chen et al., 2018)	China	16	312.3–9081.1 (2400.2)	HV
Wei River (Pang et al., 2022)	China	16	60.5–10241.1 (2250.4)	L–HV
Forcados River (Iwegbue et al., 2021)	Nigeria	16	1620–19,800	H–HV
Bahia Blanca Estuary (Oliva et al., 2015)	Argentina	17	19.7–30054.5	L–HV
Clyde River (Vane et al., 2019)	Britain	15	2.3–4226	L–H
St. Lawrence (Corminboeuf et al., 2022)	Canada	16	71–5672	L–HV

L Low Level, *M* Moderate Level, *H* High Level, *HV* Very High Level

and InP were low at 22.22%, while the detection rates of PAHs with low molecular weight (2- ring and 3-ring) were 100%, and the overall average detection rate was 74.31%. As shown in the table, the average concentration of Pyr (2382.78 ng/g dw) was the highest, followed by Bghip (1714.32 ng/g dw) and Ace (751.79 ng/g dw). The total content of PAHs in this area was 2641.28–16783.72 ng/g dw, the mean concentration was 7337.26 ng/g dw, and the coefficient of variation was between 36.20 and 190.35%. These values indicate the strong spatial variability of PAHs, and the higher coefficient of variation suggests that the content and pollution degree of surface sediments are related to industrial and human activities around

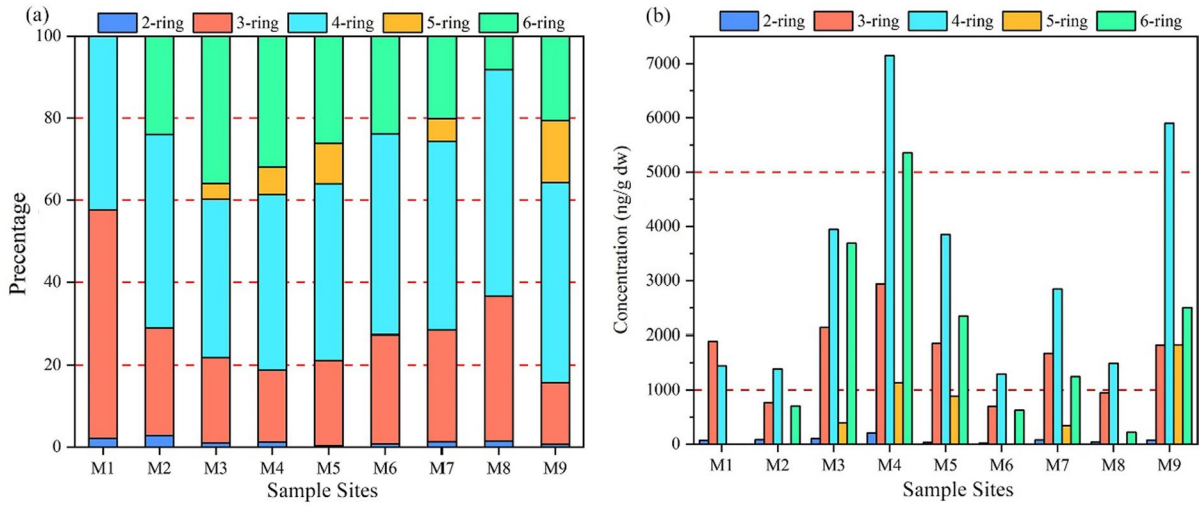


Fig. 2 Composition and concentration of five types of PAHs

the river. In addition, previous studies reported that the content of PAHs in soil is also affected by total organic carbon, sediment particle size, soil composition, and water flow status (Han et al., 2021; Nguyen et al., 2021; Seidl et al., 2022).

Referring to the content of PAHs in sediments, PAH pollution was divided into four levels (Han et al., 2021), as exhibited in Table S5. The average concentration of PAHs in Kaokaowusu river sediments was 7337.26 ng/g dw, and far exceeded 5000 ng/g dw at some sites, Therefore, the region is experiencing high and heavy PAH pollution. Table 2 lists the concentrations of PAHs in the sediments of the study area and river sediments in other areas. Comparatively, the study area has much higher levels of PAHs than the Pearl River Delta, Poyang Lake, Yangtze River, Clyde River, St. Lawrence River, and other areas; PAHs in the sediments of the industrial region were comparable to those in the sediments of rivers in Suzhou Industrial Park, Wangyang River, Liaohe River, Wei River, and Port Blanca estuary, and the PAH content in some of the above-mentioned rivers was much higher than that in the study. In general, the degree of contamination of the research area was

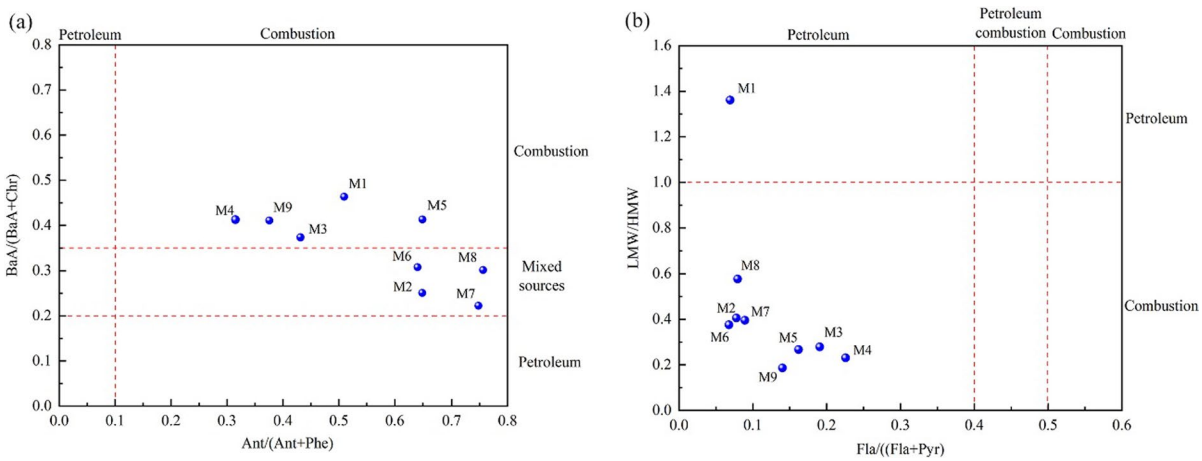


Fig. 3 Molecular diagnostic ratios of PAHs

at the same level. The PAH pollution in the above-mentioned areas may be attributable to the focus on industrial development, heavy transportation, and dense population flow. It is worth noting that the results of this comparative analysis should be treated with caution, because the land functions, sampling methods, and sampling numbers of the sampling areas are different, and the types of PAHs measured were inconsistent, resulting in large differences in the final overall content (Barhoumi et al., 2019).

Composition analysis of PAHs

The physical and chemical properties, sources, and biological toxicity widely vary among PAHs with different ring numbers and molecular weights (Han et al., 2021; Jing et al., 2018). PAHs with high molecular weights have much higher toxicity than those with low molecular weights. Based on the number of rings of PAHs, the 16 PAHs were separated into five categories in this study: 2-ring (Nap); 3-ring (Acp, Ace, Fle, Phe, Ant); 4-ring (Fla, Pyr, BaA, Chr); 5-ring (BbF, BkF, BaP, DahA); and 6-ring (InP, BghiP). The composition and content of the five different classes of PAHs are shown in Fig. 2. The components of PAHs slightly differed among different sites. The concentration of the 2-ring PAHs was 20.47–205.77 ng/g dw, accounting for 0.35–3.03%; the concentration of the 3-ring PAHs was 302.19–2939.82 ng/g dw, accounting for 14.5–58.18%; the concentration of the 4-ring PAHs was 1123.35–7148.21 ng/g dw, accounting for 39.0–54.37%; the concentration of the 5-ring PAHs was ND-1831.2 ng/g dw, accounting for 0–15.9%; the concentration of the 6-ring PAHs was ND-5357.79 ng/g dw, accounting for 0–36.49%. In general, the proportions of the 2-ring and 5-ring PAHs at each site were relatively low, while the 3-ring and 4-rings PAHs were the main components, and the 6-ring PAHs had different proportions at each site. The contributions of the various types of PAHs followed the order: 4-ring > 3-ring > 6-ring > 5-ring > 2-ring. Summarizing previous studies, low molecular weight PAHs in sediments mostly originate from diagenetic factors, oil transportation, leakage, atmospheric transportation, and dust deposition (Deng et al., 2013; Huang & Batterman, 2014; Pang et al., 2022). High molecular weight PAHs are generated by thermal decomposition (fossil fuels, biomass, grass, plastic, and transportation) (Chiu et al., 2018; Yunker et al., 2002). Accordingly, the PAH sources in this area are primarily thermal decomposition, oil spills, and dust deposition in local industrial areas. However, the specific source of PAHs cannot be effectively determined by simple composition comparison, and further analysis using new methods was required.

Source analysis of PAHs

Molecular diagnostic ratio of PAHs

Driven by natural and human factors, PAHs are globally widespread. The molecular diagnostic ratio is a simple approach for analyzing the sources of PAHs based on thermodynamic stability (Aghadadashi et al., 2019; Sereshk & Bakhtiari, 2014). This method has been utilized to determine the origin of PAHs (Huang et al., 2021). In this study, the sources of PAHs were determined using the molecular ratios of four PAH pairs: Ant/(Ant + Phe), BaA/(BaA + Chr), Fla/(Fla + Pyr), and LMW/HMW. The specific evaluation methods are presented in Table S3. As shown in Fig. 3, the molecular ratios of the above four pairs ranged from 0.32–0.76, 0.22–0.42, 0.06–0.23, and 0.18–1.36, respectively. From the ratio of Ant/(Ant + Phe), the PAHs in the study area can be attributed to the combustion of coal and biomass. The BaA/(BaA + Chr) values of M2, M6, M7, and M8 were between 0.2 and 0.35, indicating that PAHs at these four sites are attributable to geogenic sources, oil leakage, and material burning, while PAHs at other sites are attributable to coal burning. The Fla/(Fla + Pyr) values were below 0.4 at all sites, reflecting oil spill as the main source. Among all the sampling points, only M1 showed an LMW/HMW ratio greater than 1, reflecting the combustion of coal and petroleum as the main source. In terms of the current situation of land use (Fig. 1), the Ningtiaota Industrial Zone is located upstream, where coal washing plants and coal chemical companies are distributed; coal storage plants of various sizes are distributed in the middle reaches, with relatively heavy transportation activities, coal-burning power plants are distributed downstream. The above analysis results show that PAHs in the Kaokaowusu river have multiple sources rather than single sources, with the combustion of coal, biomass, and petroleum (such as diesel and gasoline) as the main source. Complete combustion, in addition to oil spills, may have resulted from oil and

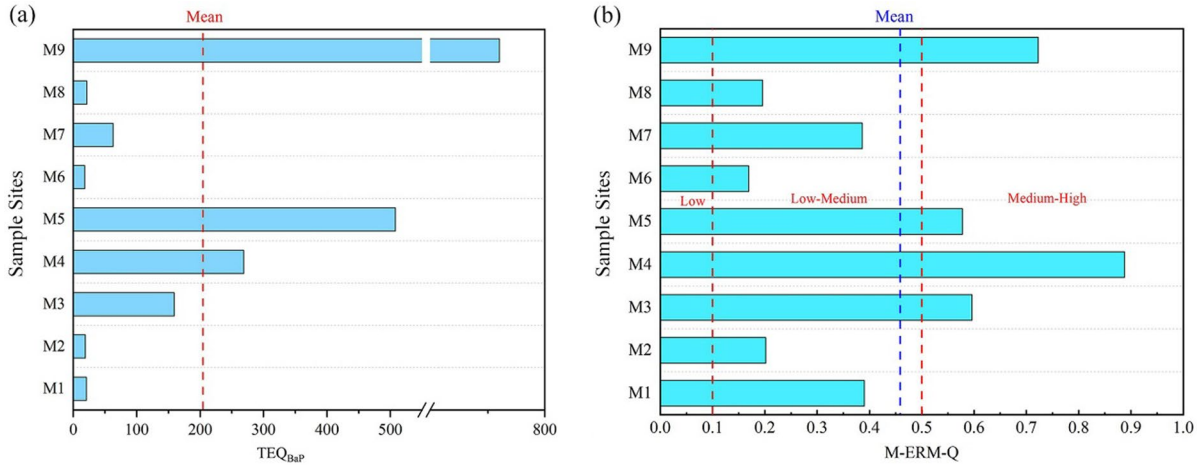


Fig. 4 Toxicity equivalent quotient (TEQ_{BaP}) and mean effect range-median quotient (M-ERM-Q) of PAH levels in sediments at each sampling Site

its by-products from local coke processing plants and vehicles.

Principal component analysis

Previous studies have shown that the photolysis of PAHs in natural environments may lead to deviations in the results of molecular diagnostic ratios (Gbeddy et al., 2021). To further explore the sources of PAHs under the influence of different human activities, the PCA method was applied to quantitatively confirm the origin of PAHs. PCA can be used to analyze and observe the relationship between variables by reducing the number of variables as much as possible and extracting the basic characteristic factors and then determining the possible sources of PAHs (Wang et al., 2015). Three factors were calculated through PCA, which together accounted for 91.03% of the total variance, as shown in Table S7 and Fig. S2. The first factor explained 63.9% of the total variance, and

Table 3 Biological toxicity assessment of PAHs in the sediment

PAH	ERL	ERM	Concentration (ng/g dw)	< ERL	ERL–ERM	> ERM
Nap	160	2100	20.47–205.77	8/9	1/9	0/9
Acp	44	640	49.12–475.64	0/9	9/9	0/9
Ace	16	500	242.82–1653.42	0/9	4/9	5/9
Fle	19	540	49.47–1300.08	0/9	8/9	1/9
Phe	240	1500	86.02–385.13	6/9	3/9	0/9
Ant	85.3	1100	117.43–449.65	0/9	9/9	0/9
Fla	600	5100	72.14–1451.62	6/9	3/9	0/9
Pyr	665	2600	990.87–4979.64	0/9	5/9	4/9
BaA	261	1600	59.63–316.86	7/9	2/9	0/9
Chr	384	2800	158.21–454.54	7/9	2/9	0/9
BbF	320	1880	ND-1268.09	4/9	5/9	0/9
BkF	280	1620	ND	9/9	0/9	0/9
BaP	430	1600	ND	9/9	0/9	0/9
DahA	63.4	260	ND-563.10	7/9	0/9	2/9
InP	1700	9600	ND-634.32	9/9	0/9	0/9
BghiP	430	1600	ND-4723.47	1/9	4/9	4/9

this factor had a strong load on PAHs with different molecular weights, such as Nap, Acp, Pyr, BbF, and BghiP. Nap, Acp, and Ace were considered to be representative molecules of coke oven combustion and are related to boiler emissions. In addition, studies have shown that Nap is related to oil leakage (Simcik et al., 1999; Zhang et al., 2015). Under the condition of incomplete combustion, higher contents of Phe and Fla have been found in soot and air fallout (Boonyatumanond et al., 2006; Gdara et al., 2017). Pyr, BaA, Chr, BbF, InP, and BghiP are regarded as tracers for diesel and gasoline-powered vehicle exhaust and natural gas (Chiu et al., 2018). The study area includes a large number of coal chemical bases, iron and steel plants, and coal processing enterprises. In the process of material transportation and production, dust and gas will cause sediment pollution through dry and wet deposition. On the whole, PC1 is related to the incomplete combustion of fossil fuels and traffic factors. Therefore, PC1 was attributed to the local heavy coking industry. The PC2 explained 17.16% of the total variance, with a strong correlation for DahA and a moderate correlation for Ant. DahA originates from the incomplete combustion of coal, such as heating, cooking, whereas Ant originates from the combustion and pyrolysis of wood and coal (Huang et al., 2022; Liu et al., 2009). Therefore, PC2 was attributed to pyrolysis. PC3, which explained 9.97% of the total variance, was only strongly correlated with Fle. Fle is a lightweight PAH related to petrogenesis, as well as oil leakage (Corminboeuf et al., 2021; Semenov et al., 2018). Combined with the relatively strong spatial variation of Fle content in this area, PC3 can be attributed to rock properties and oil spills, representing a mixed source.

Ecological risk assessment

Equivalent concentration of BaP

PAHs with different molecular weights pose different levels of toxicity and risk to the human body. Small molecular weight PAHs (2–3 ring) have acute toxicity, and large molecular weight PAHs (4–6 ring) have potential carcinogenic hazards (Witt, 1995). Among the 16 PAHs considered in this study, BaP is the most toxic and carcinogenic. The potential ecological environment risk posed by mixed PAHs was evaluated by quantifying the concentration of each PAH as the equivalent concentration of BaP through TEF. In this study, the equivalent concentration of BaP (TEQ_{Bap}) of the 14 PAHs detected in the sediments was calculated, and the potential consequences are presented in Table S8 and Fig. 4. The TEQ_{BaP} of the 14 PAHs ranged from 10.05 (M6) to 760.26 (M9) ng/g dw; the mean was 204.33 ng/g dw, and the proportion of the five strongly carcinogenic PAHs was 45.2–95.51%, with a wide range of variation. According to the human health standard provided by Environment Canada, 600 ng/g is a safe level of toxicity for PAHs (Canadian & Quality, 2007). At M9, TEQ_{BaP} was higher than 600 ng/g. M9 is located in Dianta Town, which includes thermal power plants and industrial zones of different scales. Dianta Town is also a transportation hub. Therefore, M9 having the highest concentration and largest proportion of carcinogenic PAHs can be attributed to industrial and

Table 4 Health risk of human exposure to PAHs in the sediment

Site	Adults				Children			
	$ILCR_{ing}$	$ILCR_{der}$	$ILCR_{inh}$	TLCR	$ILCR_{ing}$	$ILCR_{der}$	$ILCR_{inh}$	TLCR
M1	1.93E−04	1.06E−04	1.66E−08	3.00E−04	1.45E−03	4.47E−04	3.81E−09	1.90E−03
M2	1.78E−04	9.76E−05	1.52E−08	2.75E−04	1.33E−03	4.12E−04	3.51E−09	1.75E−03
M3	1.49E−03	8.16E−04	1.27E−07	2.30E−03	1.12E−02	3.44E−03	2.93E−08	1.46E−02
M4	2.51E−03	1.38E−03	2.15E−07	3.88E−03	1.88E−02	5.80E−03	4.94E−08	2.46E−02
M5	4.74E−03	2.60E−03	4.06E−07	7.34E−03	3.56E−02	1.10E−02	9.35E−08	4.65E−02
M6	1.68E−04	9.24E−05	1.44E−08	2.61E−04	1.26E−03	3.90E−04	3.32E−09	1.65E−03
M7	5.88E−04	3.22E−04	5.03E−08	9.10E−04	4.41E−03	1.36E−03	1.16E−08	5.77E−03
M8	2.03E−04	1.11E−04	1.74E−08	3.14E−04	1.52E−03	4.69E−04	4.00E−09	1.99E−03
M9	7.09E−03	3.89E−03	6.07E−07	1.10E−02	5.32E−02	1.64E−02	1.40E−07	6.96E−02

transportation factors, which led to the high carcinogenic and ecological risks at the site.

Sediment quality guidelines

As the types of PAHs in the sediments were much higher than the 16 species determined, the application of TEQ_{BaP} to evaluate the ecological risks of sediments introduced some uncertainties. Therefore, it was essential to further appraise the biological toxicity and ecological risk of PAHs in sediments (Nisbet, 1992). The SQG method has been widely used to determine the potential environmental risks and adverse biological effects of PAHs in sea estuary and river sediments. The 14 PAHs detected at 9 sites were compared with each threshold of SQG. Table 3 list the ERL, ERM, and concentration ranges of the 14 PAHs (It is noteworthy that the ERL and ERM values for InP were interpolated from the data of high molecular weight PAHs because they were lacking.). The contents of BkF, BaP, and InP were lower than ERL. The concentrations of NaP, Phe, Fla, BaA, Chr, and BbF were also lower than ERL at most sites, and they were between ERL and ERM only at few sites. This indicated that the above-mentioned types of PAHs do not or occasionally have adverse biological effects. It is worth noting that the concentrations of Ace, Fle, Pyr, DahA, and BghiP were higher than ERM at a considerable number of sites, which may reflect frequent adverse biological effects. In addition, the contents of Ace, Pyr, DahA, and BghiP were higher than ERM at M9. In general, the potential ecological risk of PAHs in this downstream region was already at a high level, which may pose serious risks to local aquatic organisms.

Mean effect range-median quotient

M-ERM-Q considers the comprehensive toxicity of multiple PAHs, and can quantitatively evaluate the ecological risks posed by PAHs at various locations (Meng et al., 2019). The M-ERM-Q values of the detected PAHs were calculated. As shown in Fig. 4, the M-ERM-Q values of nine samples varied from 0.17 to 0.89, with an average value of 0.46. The M-ERM-Q values of the sites were greater than 0.1, the range of 0.1–0.5 at five sites, in the range of 0.5–1.5 at four sites, and reached the maximum at M4. The ecological risk was at an upper-middle level, with an overall toxicity probability of 21%. Continued attention must be paid to the variation in the concentration of PAHs in this area, especially on the adverse effects of biological toxicity. Moreover, the input of PAHs should be reduced to mitigate the risks posed to aquatic organisms and humans.

Human health risk assessment

The total lifetime carcinogenic risk (TLCR) model was employed to evaluate the carcinogenic risk of PAHs in sediments for adults and children. TLCR values of less than 10^{-6}, 10^{-6}–10^{-4}, and greater than 10^{-4} are considered to be acceptable, moderate, and unacceptable, respectively (Gbeddy et al., 2020; Kr et al., 2021). The specific evaluation criteria are shown in Table S5. The specific evaluation criteria are exhibited in Table 4. The TLCR values were greater than 10^{-4} at all sites, revealing that the potential carcinogenic risk of the area is at a moderate level. The higher content and toxicity of high molecular weight PAHs (4–6 rings) were the main reasons for this result. Among the three carcinogenic pathways of oral intake, skin contact, and respiration, respiration presented a far lower carcinogenic risk than the other two, which agrees with the results of other scholars (Xu et al., 2022). Compared with adults, children are at higher risk, because of factors, such as weight and daily exposure. Therefore, attention should be paid to the carcinogenic hazards arising from oral intake and skin contact in this area, and exposure to river sediments should be reduced.

Conclusion

In this study, the concentration, composition, source, toxicity of PAHs in river sediments of a heavy industrial area were analyzed. In addition, their adverse impacts on the ecology and human health were assessed. The content of PAHs in the sediments of Kaokaowusu River ranged from 2641.28 to 16783.72 ng/g, and the average content was 7337.26 ng/g. Compared with other areas, the pollution level is higher, with 3-ring and 4-ring PAHs being the main components. According to the results of the molecular diagnosis ratio, PAHs in the study area originate from the incomplete combustion of coal and biomass. At the same time, the source of

PAHs in this area was quantitatively analyzed through PCA. According to the results, the higher concentration of PAHs could be attributed to the incomplete combustion of fossil fuels, local heavy traffic, pyrolysis sources, rock properties, and oil leakage. Based on toxicity calculations of TEQ_{BaP}, SQG, and M-ERM-Q, the PAHs pose high biotoxicity and ecological risk in the downstream area of the river, and they may adversely affect local aquatic organisms. In addition, the mean lifetime cancer risk for adults and children in this region was 2.95×10^{-3} and 1.87×10^{-2}, respectively, indicating that the potential cancer risk in this region is already at a high level. In general, the government should continue to pay attention to the changes in the content of PAHs in this area, as well as the ecological and carcinogenic risks of the PAHs, and maintain continuous monitoring.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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