Distributions, compositions, and ecological risk assessment of typical pollutants in surface sediment of Xihe River, China

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Abstract

Distributions and compositions of six phthalic acid esters (PAEs), eight phenol compounds and seven heavy metals in riverbed sediments of the Xihe River in Shenyang, China. The ecological risks of these typical pollutants were investigated and evaluated based on the risk quotient (RQ) and potential ecological risk index (PERI) methods. The concentrations of total $\sum_6$ PAEs, $\sum_8$ phenols and $\sum_7$ heavy metals in sediments varied from 92.83 to 293.66 $\mu\text{g/g dw}$, 8.87 to 83.73 $\mu\text{g/g dw}$ and 0.46 to 419 $\mu\text{g/g dw}$. The main pollutants in Xihe River sediments are DEHP, DIBP, phenol, P-methylphenol and Cd. More than half target PAEs and phenol compounds in sediment of the Xihe River exhibited medium or high ecological risk. Cd poses a very high ecological risk to the Xihe River Basin. It is imperative to take some effective measures to reduce the pollution of those contaminants.

1. Introduction

Due to Chinese rapid urbanization and industrialization, rivers are increasingly threatened by all kinds of pollutants (Zhao et al., 2018). Riverbed sediments as important sinks and sources for pollutants, the concentration of pollutants in it has a great impact on aquatic organisms and the water environment, even influence the entire ecosystem (Mi et al., 2019; Hu et al., 2020).

Phthalic acid esters (PAEs) are typical plasticizer, whose environmental behavior has attracted considerable attention (Yang et al., 2019; Khalid Muzamil Gani et al., 2016). The US Environmental Protection Agency has identified six congeners, including di-methyl phthalate (DMP), di-ethyl phthalate (DEP), di-n-butyl phthalate (DnBP), di-2-ethylhexyl phthalate (DEHP), di-n-octyl phthalate (DnOP) and butylbenzyl phthalate (BBP), as priority pollutants (Gao et al., 2018; Alkan Nigar et al., 2021). Phenol compounds are important chemical raw materials and are used in large quantities to make phenolic resin, epoxy resin and synthetic fiber (Wang et al., 2020). Phenol compounds have a strong destructive effect on the human body, and can inhibit the central nervous system and have strong toxic, persistent and bio-accumulative effects (Li et al., 2015). Due to the potential biological toxicity of heavy metals, they may cause further adverse effects on human beings through bioaccumulation (Xu et al., 2017). Because of potential risks to humans and the environment of these typical pollutants, the comprehensive evaluations of ecological risks are required.

The Xihe River is 78 kilometers long and flows through the Tiexi District, the most important industrial estate in Shenyang (Liu., 2016). Due to the complete variety and large number of factories around the basin, various types of wastewater are one of the main sources of water for the Xihe River. A variety of pollutants in the basin such as ammonia nitrogen, PCDD and dibenzofurans have been investigated and studied by many people (Ke et al., 2018; Lin et al., 2013; Ke et al., 2019). It is still lack of information on the pollution level of common PAEs, phenol compounds and heavy metals in the river sediment, limited work has reported on the potential risk of these pollutants in the Xihe River.

In this study, we selected some representative sites as the study area to conduct a risk assessment of heavy metals, PAEs and phenol compounds in Xihe River. The purpose of this study is: (1) to study the distribution and compositions of heavy metals, PAEs, phenols and heavy metals in surface sediments of Xihe River; (2) To evaluate the ecological risk of heavy metals, PAEs and phenol compounds in sediments.

2. Materials And Methods

2.1. Sample collection

Figure 1 provides sampling locations for this study. Sediment samples were collected from 11 sampling points in Xihe River. The riverine sediments were collected by use of a grab bucket (depth of approximately 0.1 m) and then put in a clean stainless steel lunch box. In each sampling point, three duplicate samples were collected and mixed evenly, then transported back to the laboratory and all samples were stored in an icebox stored at 4°C.

2.2. Chemical analysis

Use a centrifuge for 30 minutes (4°C; 9000r/min) to extract interstitial water, and store it in a 4°C walk-in cooler before collecting samples for chemical analysis. The concentrations of phthalates (PAEs) and phenols were analyzed by Agilent 6890 – 5973 GC/MS. The sediment samples (0.5g) were digested in 20mL of a 1:1:2 guaranteed reagent $\text{HNO}_3 + \text{HClO}_4 + \text{HF}$ for 10h, the concentrations of heavy metals were determined by Inductively coupled Plasma Atomic Emission Spectrometry (ICP-AES, Thermo).

2.3. Risk Assessment

2.3.1. Risk quotient

The risk quotient (RQ) method was used to assess the potential environmental risk of the PAEs and phenols in this work. According to the European technical guidance document (TGD) on risk assessment (EC, 2003; Li et al., 2017; Li et al., 2016*; Zhang et al., 2018; Li et al., 2016**), RQ for PAEs and phenols congeners were calculated from the measured environmental concentration (MEC) divided by the predicted no effect concentration (PNEC), which is described as follows ;

$$\text{RQ} = \frac{\text{MEC}}{\text{PNEC}}$$

$$\text{PNEC}_{\text{sed}} = \frac{K_{\text{susp-water}}}{\text{RHO}_{\text{susp}}} \times \text{PENC}_{\text{water}} \times 1000$$

$$K_{\text{susp-water}} = F_{\text{water-susp}} + F_{\text{solif-susp}} \times F_{\text{oc-susp}} \times \frac{K_{\text{oc}}}{1000} \times \text{RHO}_{\text{solid}}$$

where MEC is the measured environmental concentration and PNEC is the predicted no effect on concentration. where $PNEC_{\text{water}}$ values were calculated from the lowest acute toxicity data, RHO_{susp} is the density of wet suspended matter, which is 1150 kg/m^3 ; RHO_{solid} is the density of the solid phase, which is 2500 kg/m^3 ; $F_{\text{water-susp}}$ is the volume fraction of water in suspension, which is defined as $0.9 \text{ m}^3/\text{m}^3$; $F_{\text{solid-susp}}$ is the volume fraction of solid in suspension, which is defined as $0.1 \text{ m}^3/\text{m}^3$; $F_{\text{oc-susp}}$ is the mass fraction of organic carbon in suspension, which is assigned as 0.1 kg/kg ; K_{oc} is the partition coefficient of organic carbon-water (L/kg).

The PERI (Ke et al.,2017; Ben Mna Haïfa et al.,2021; Islam Md Saiful et al.,2018; Shi et al.,2016; Chai et al.,2017) was used to assess the comprehensive potential ecological risk of heavy metals in sediment and was initially introduced by Hakanson (1980). The potential ecological risk factor of a given metal (E_r^i) is defined as :

$$E_r^i = T_r^i \times \frac{C_i}{C_0}$$

$$RI = \sum_{i=1}^n T_r^i \times \frac{C_i}{C_0}$$

where C_i is the concentration of metal i in sediment, C_0 is the concentration of the same element in background sediment, T_r^i is the biological toxicity factor of an individual element, which was determined for $\text{Cu} = \text{Pb} = 5$, $\text{Zn} = 1$, $\text{Hg} = 40$, $\text{As} = 10$, $\text{Cr} = 2$ and $\text{Cd} = 30$ (Suresh et al., 2011), E_r^i is the potential ecological risk factor of a single metal, and RI is the comprehensive potential ecological risk index of the metals. The PERI of heavy metals was categorized according to five levels, as shown in Table.1.

2.4. Quality assurance and Quality control (QA/QC)

Quality assurance and quality control were assessed in duplicates, with method blanks and standard reference materials. The content of pollutants was determined by three replicates. No target pollutants were detected in the blank control groups. Quantitative analysis of all target compounds was corrected by data of blank test and recovery. The deviations were less than 10%. The quality of the analytical procedures was tested by the recovery measurements on the Chinese national geo-standard (GBW-07333 and GBW-07314). All of the reagents were guaranteed analytical grade or higher. The lab glassware (bottles, tubes, etc.) were pre-cleaned by soaking in 10% HNO_3 (w/w) for at least 2 days, followed by soaking and rinsing with de-ionized water prior to use (Zhuang et al., 2015)

3. Results And Discussion

3.1. Distribution of PAEs in the Xihe River

The concentration of $\sum_6$ PAEs was shown in Table. 2. In the Xihe River, the mean concentrations of each PAEs is: DEHP (200.96 $\mu\text{g/g dw}$) > DNOP 5.88 $\mu\text{g/g dw}$) > DiBP (2.67 $\mu\text{g/g dw}$) > DBP (1.46 $\mu\text{g/g dw}$) > DMP (0.20 $\mu\text{g/g dw}$) > DEP (0.15 $\mu\text{g/g dw}$). DEHP and DNOP were the dominant pollutants in sediments. The maximum concentrations of DEHP and DNOP appear in source of Xihe River (St2; 284.95 $\mu\text{g/g dw}$) and Ganguan (St3; 7.90 $\mu\text{g/g dw}$). The proportion of $\sum_6$ PAEs at each sampling point is shown in Figure. 2. According to the chart, it can be seen that the upstream of the fine river is greatly affected by the pollution of phthalates, with the detected content of six phthalates accounting for about 75–92% of the whole river. Due to the Xihe River mainly flows through Tiexi District, the largest industrial area in Shenyang, all kinds of factories around the river basin are complete, and the Xiannvhe sewage treatment plant, the largest sewage treatment plant in Shenyang, is near its upstream. Therefore, the Xihe River became the largest recipient of industrial effluents and domestic wastewater from Shenyang. In the Dawula (St.9), the concentration of $\sum_6$ PAEs has dropped significantly, among which the main pollutants DEHP and DNOP in the sediments gradually decreased to 87.94 $\mu\text{g/g dw}$ and 2.93 $\mu\text{g/g dw}$, which were about one third of the upstream concentration. This may have been caused by the constant dilution of the river

3.2. Distribution of phenols in the Xihe River

The concentrations of phenol compounds ($\sum_8$ phenols) in sediment from the Xihe River were given in Table. 3. The concentration of $\sum_8$ phenols in sediments varied from 8.86 to 83.72 $\mu\text{g/g dw}$ in the Xihe River. The main pollutants are phenol and p-methylphenol. The proportion of $\sum_8$ phenols was shown in Figure.3. On the upstream of Xihe River, the content of phenol and p-methylphenol is 71.5% and 80.2% of the overall sampling point. In addition, the contents of 2, 4-dimethylphenol and 2, 4-di-tert-butylphenol in the upstream area were second only to phenol and p-methyl phenol, accounting for 89.3% and 81.3% of the total sampling points, respectively., and the remaining phenols were less than 0.5 $\mu\text{g/g dw}$. Very similar spatial distribution has been found between the pollution levels of PAEs and phenolic compounds, indicating that both pollutants might be subjected to the same contamination sources. In general, the pollution level in the upstream of Xihe River is much greater than that in the downstream.

3.3. Distribution of heavy metals in the Xihe River

The concentration of $\sum_7$ heavy metals was shown in Table. 3. In the Xihe River, the mean concentrations of each heavy metals is: Zn (333 $\mu\text{g/g dw}$) > Cr (91.68 $\mu\text{g/g dw}$) > Cu (82.43 $\mu\text{g/g dw}$) > Pb (61.66 $\mu\text{g/g dw}$) > As (19.80 $\mu\text{g/g dw}$) > Cd (18.44 $\mu\text{g/g dw}$) > Hg(0.68 $\mu\text{g/g dw}$). From the perspective of pollution, the order of the sampling points is St.4 ($\sum_7$ heavy metals = 760.63 $\mu\text{g/g}$) > St.10 ($\sum_7$ heavy metals = 636.80 $\mu\text{g/g}$) > St.8 ($\sum_7$ heavy metals = 577.86 $\mu\text{g/g}$) > St.7 ($\sum_7$ heavy metals = 559.69 $\mu\text{g/g}$) > St.6 ($\sum_7$ heavy metals = 421.02 $\mu\text{g/g}$). The distribution ratio of the seven heavy metals at each sampling point is shown in Fig. 4. It can be found that the high proportion of metals in St.4 and st. 10 is due to the presence of many factories for the manufacture of machinery and automobile parts, located along the river and not just in upstream of Xihe River. Zn is the most important pollutant in Xihe River, followed by Cr and Cu, which also confirms the reason for the high Content of Zn and Cu in the sediment of this basin from the side.

3.4. Ecological risk assessment

The ecological risk assessment of $\sum_6$ PAEs and $\sum_8$ phenols in sediments was carried out based on the calculated RQ values. In Figs. 5 and 6, among which DEHP and phenol are high-risk in all sampling sites, p-methyl phenol and DIBP are high-risk in three of the four sampling sites. Half of the target PAEs and phenolic compounds in the river have a moderate or high ecological risk to organisms, which means that some effective measures must be taken to reduce the pollution of these pollutants. Among the seven types of heavy metals, Cd is high-risk at all sampling sites, and the other heavy metals are low-risk. Explain that Cd is the main pollutant in this area. As a whole, because the potential ecological risk factor of Cd is too high, the comprehensive potential ecological risk index in the region presents a high risk. Therefore, it is necessary to strengthen the detection of Cd in wastewater and reduce the concentration of Cd in wastewater to reduce the potential ecological risk of heavy metals in the region as a whole. However, Due to the lack of specific biotoxicity number of phenol in sediment and the simple evaluation method in this paper, further research might be needed to establish the real risks of phenolic compounds to benthic organisms.

4. Conclusion

The distributions of PAEs, phenol compounds and heavy metals in the Xihe River were investigated and the ecological risk was evaluated in this study. Screen out several pollutants with potential risks to the environment. The results revealed that six PAEs and eight phenols, DEHP, DIBP, phenol and p-methyl phenol were the most abundant pollutants in the Xihe River. The pollution level in the upstream of Xihe River is much higher than that in the downstream. The ecological risk assessment showed that DBP, DEHP, phenol, p-methyl phenol and Cd in the Xihe River have a high ecological risk to relevant organisms. Thus, only by precisely controlling the above pollutants can the overall ecological risk in the basin be effectively reduced it is imperative to control the discharge of toxic sewage into the rivers so as to reduce the potential risks in the Xihe River.

Declarations

Ethical Approval

Before the journal editing process is completed, the paper will not be submitted elsewhere

Consent to Participate

Xin Ke, Xun Sun and Zheng Yan all agreed to participate.

Consent to Publish

Xin Ke, Xun Sun and Zheng Yan all agreed to publish.

Authors Contributions

Xin Ke and Xun Sun conceived and designed experiments. Xun Sun performed experiments, statistical analysis and chemical analysis. Xun Sun wrote manuscript. Xin Ke and Zheng Yan provided technical and editorial assistance.

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-Competing Interests

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.

-Availability of data and materials

No additional unpublished data and materials are available.

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Tables

Table.1. Classification of PERI

Assessment criterion	PERI				
	Low	Moderate	Considerable	High	Very high
	≤40	40—80	80-160	160-320	≥320
RI	≤150	150—300	300-600	≥600	

Table.2. Concentrations of ∑6PAEs and ∑8phenol compounds in sediment of the Xihe River (μg/g dw)

	range	mean	SD
DMP	0.159-0.227	0.198	0.025
DEP	0.085-0.188	0.148	0.040
DBP	0.792-2.132	1.461	0.475
DiBP	0.872-3.746	2.67	1.085
DnOP	2.929-7.903	5.877	1.923
DEHP	87.94-284.945	200.961	83.105
$\sum_6$ PAES	92.83-293.662	210.577	84.457
phenol	3.938-28.858	14.88	8.944
p-methylphenol	3.548-51.835	19.679	18.860
2,3-dimethylpheno	0.049-0.088	0.064	0.014
2,4-dimethylphenol	0.300-1.815	0.702	0.643
3,5-dimethylphenol	0.156-0.213	0.184	0.027
2,4,6-trimethylphenol	0.050-0.086	0.073	0.014
2,4-di-tert-butylphenol	0.565-0.875	0.755	0.117
2,6-di-tert-butyl-4-ethylphenol	0.014-0.021	0.017	0.003
$\sum_8$ Phenols	8.870-83.726	36.353	28.273

Table.3. Concentrations of $\sum_7$ heavy metals in sediment of the Xihe River ($\mu\text{g/g dw}$)

	range	mean	SD	TEC	PEC
Cu	39-118.5	82.43	29.33	31.6	149
Cd	9.2-27.6	18.44	6.69	0.99	4.98
Pb	12.8-126.8	61.66	46.2	35.8	128
Cr	73-101.6	91.68	10.27	43.4	111
Hg	0.46-0.83	0.68	0.13	0.18	1.06
As	12.9-31.2	19.8	6.74	9.79	33
Zn	201-419	333	83.34	121	459.8

TEC = threshold effect concentration, PEC = possible effect concentration.

Table.4. Risk quotient (RQ) of $\sum_6$ PAEs and $\sum_8$ phenol compounds

	RQ(St1)	RQ(St2)	RQ(St3)	RQ(St9)
DMP	0.77	0.80	0.88	0.62
DEP	0.52	0.24	0.49	0.40
DBP	21.47	10.98	4.93	2.93
DiBP	29.50	25.14	22.60	6.86
DnOP	13.64	8.88	18.29	6.78
DEHP	41.17	42.47	23.07	13.11
phenol	2885.77	1374.53	393.81	1297.87
p-methylphenol	95.99	21.03	6.57	22.18
2,3-dimethylpheno	0.12	0.06	0.08	0.08
2,4-dimethylphenol	0.85	0.18	0.15	0.14
3,5-dimethylphenol	0.10	0.14	0.10	0.13
2,4,6-trimethylphenol	1.00	1.07	0.95	0.62
2,4-di-tert-butylphenol	-	-	-	-
2,6-di-tert-butyl-4-ethylphenol	-	-	-	-

Table.5. Potential ecological risk factor (E_{tr}^i) of $\sum_7$ heavy metals

		(St4)	(St6)	(St7)	(St8)	(St10)
Cu	5	29.92	19.04	22.40	9.85	24.44
Cd	30	7527.27	5318.18	2509.09	6300.00	3490.91
Pb	5	25.12	7.64	6.66	2.99	29.63
Cr	2	3.33	2.52	3.06	3.51	3.41
Hg	40	0.83	0.72	0.79	0.46	0.60
As	10	25.8	21.25	15.34	14.66	35.45
Zn	1	6.09	3.17	6.60	6.11	4.25

Figures

Figure 1

Sampling sites in Xihe River, China

St.1. Gili Bridge, St.2.source of Xihe River, St.3. Ganguan, St.4. Fuguan, St.5. Aijin,
St.6. Xiaoyu, St.7.Dapan, St.8.Zhangyi, St.9. Dawula, St.10. Qianyu, and St.11. Tutaizi.

Figure 2

Distributions and compositions of $\sum_6$ PAEs in sediment of the Xihe River ($\mu\text{g/g dw}$).

DMP = Dimethyl phthalate, DEP = Diethyl phthalate, DBP = Dibutyl phosphate, DEHP = Di-2-ethylhexyl phthalate, DIBP = Di-iso-butyl phthalate, DNOP = Di-n-octyl phthalate.

Figure 3

Proportion of $\sum_8$ phenols compounds in each sampling dots of the Xihe River sediments ($\mu\text{g/g dw}$)

p-p= p-methylphenol, 2,3-p= 2,3-dimethylphenol, 2,4-p= 2,4-dimethylphenol,

3,5-p= 3,5-dimethylphenol, 2,4,6-p= 2,4,6-trimethylphenol,

2,4-dp= 2,4-di-tert-butylphenol, 2,6-dp= 2,6-di-tert-butyl-4-ethylphenol.

Figure 4

Proportion of $\sum_7$ heavy metals in sediment of the Xihe River ($\mu\text{g/g dw}$)

Figure 5

Ecological risk assessment of PAES and phenols in surface sediments of Xihe River

Figure 6

Ecological risk assessment of heavy metals in surface sediments of Xihe River