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Impact of intensive land use on heavy metal concentrations and ecological risks in an urbanized river network of Shanghai

Yongsheng Zeng[a], Chunjuan Bi[a,b,*], Jinpu Jia[a], Lin Deng[a], Zhenlou Chen[a,*]

[a] Key Laboratory of Geographic Information Science of Ministry of Education, School of Geographic Sciences, East China Normal University, Shanghai, China
[b] Institute of Eco-Chongming (IEC), East China Normal University, Shanghai, China

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ABSTRACT

Heavy metal pollution characteristics in the urbanized river network are closely related to urban land use. The existing studies mainly focused on the pollution characteristics of heavy metals in single rivers, but fewer studies reported the impact of land use on the concentrations and ecological risks of heavy metals in surface water and sediments of the urban river network. This study combined land use types with heavy metal pollution in rivers, and explored the specific impact of intensive land use on the characteristics of heavy metal pollution in rivers, which provided scientific basis for the study of heavy metal pollution in plain river networks. The concentrations, spatial variations and potential ecological risk levels of cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), lead (Pb) and zinc (Zn) in surface water and sediments from five different intensive land use areas of Shanghai river network (industrial areas, agricultural areas, central city, suburban towns and Chongming Island) were analyzed. Results showed the concentrations of Cd, Cr, Cu, Pb and Zn in surface water of the Shanghai river network were relatively low, but Hg pollution existed in some rivers. Influenced by intensive land use, heavy metal concentrations in the central city were generally higher than those in other areas. Heavy metals in sediments had considerable ecological risk, in which Hg and Cd were the main contributors, and showed the highest potential ecological risk in central city. Acid volatile sulfides (AVS) and total organic carbon (TOC) in river sediments affected the migration and transformation of Cd, Hg and Zn. Industrial activities, traffic emissions and coal-burning emissions were the main sources of heavy metals. Intensive land use caused by anthropogenic activities led to the weakening of hydrodynamic conditions and the discharge of a large number of pollutants into the river network. In the central city with intensive anthropogenic activities, heavy metals in the river network showed higher levels than those in other areas. This study found that intensive land use and high-intensity anthropogenic activities led to the high pollution level of heavy metals in the plain urbanized river networks. Quantitative analyses of the effects of intensive land use on heavy metal pollution in urbanized river networks were suggested to conduct further to protect the urban river water quality.

1. Introduction

Heavy metal pollution is a common environmental problem in many river systems of the world because of the rapid population growth, urbanization and industrialization (Staley et al., 2015; Nguyen et al., 2016; Zhao et al., 2018). Heavy metal contaminated sediments have seriously threatened Chinese aquatic ecosystems (Deng et al., 2020). Having the characteristics of high enrichment and low migration, heavy metals cannot be degraded and tend to be reserved in sediments associated with organic matter or other compounds, which seriously threaten the natural ecosystem and human health (Yan et al., 2016) due to their toxicity, bioaccumulation and persistence level in the environment (Lu and Yu, 2018). Some heavy metals, such as As, Cd, Hg and Pb, could impair the human endocrine system and were screened as environmental endocrine disruptors by the US Environmental Protection Agency (EPA) (Ren and Jun, 2004).

Land use changes will become more frequent and intense because of anthropogenic activities, which significantly influence the ecosystem services (Huang et al., 2019; Talukdar et al., 2020). Due to rapid urbanization, high-intensity human activities and land use have caused a series of extreme events such as droughts, floods, and landslides in China in the past two decades (Ali et al., 2020). Heavy metals show imparity distribution under different land uses (Liu et al., 2019). The intensive anthropogenic activities promote heavy metal pollution in

* Corresponding authors at: Key Laboratory of Geographic Information Science of Ministry of Education, School of Geographic Sciences, East China Normal University, Shanghai, China (C. Bi, Z. Chen).
E-mail addresses: cjbi@geo.ecnu.edu.cn (C. Bi), zlchen@geo.ecnu.edu.cn (Z. Chen).

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rivers (Song et al., 2010). Affected by urbanization, the riparian areas are prone to heavy metal pollution (Liu et al., 2019). Due to long-term industrialization and anthropogenic activities, urban rivers were occupied or straightened and a large number of pollutants were discharged into the river, which endangered the aquatic biota and deteriorated the river water quality (Staley et al., 2015; Yan et al., 2016; Paul, 2017). The tidal river network hydrodynamic poses a severe challenge to the protection of rivers (Yao et al., 2006). Ali et al. (2019) found that the dam affected the flow pattern of the middle and upper reaches of the Yangtze River. River hydrology is an important factor in the migration and transformation of heavy metals (Shafer et al., 1997; Garneau et al., 2015). Zhang et al. (2001) found that the accumulation of heavy metals in the river was related to river hydrodynamics. However, in the plain river network area, rivers are connected with each other, and river water flows backward during the course of high tide, resulting in poor fluidity and weak hydrodynamic conditions. The river water quality is affected by natural processes such as precipitation and weathering, as well as anthropogenic activities such as urbanization and industrial production (Carpenter et al., 1998; Singh et al., 2005). Agricultural and industrial activities can cause river pollution in the area (Bu et al., 2014).

Shanghai is located at the confluence of the Yangtze River and Qiantang River with an urbanized river network. There is a total of 43,253 rivers with a total length of 28,714 km, an area of 497.53 km^2, and the density of the river network is 4.53 km/km^2, from the 2018 Shanghai Statistical Yearbook of Shanghai Municipal Statistics Bureau (SMSB) (SMSB, 2018). The risk assessment and influencing factors of heavy metals in several river sediments of Shanghai and the spatial distribution and sources of heavy metals in sediments of the Yangtze Estuary were reported (Han et al., 2017; Liang et al., 2017). But the river networks in coastal areas are more vulnerable to anthropogenic activities such as intensive urban land use and industrial production (Bi et al., 2018). Liu et al. (2016) and Liu et al. (2019) reported the effects of land-use variations on the distribution of heavy metals in riparian soils. Anthropogenic emission of pollutants was considered as an important cause of water pollution (Alizadeh et al., 2018). Heavy metals can settle from surface water to sediment (Deng et al., 2020). There is a lack of study on the impact of intensive land use on heavy metals in surface water and sediments of the river network. Heavy metal pollution in plain river network was different from a single river and more influenced by different intensive anthropogenic activities. Based on a large number of previous studies, this study further refined the research ideas and mainly focused on the characteristics and ecological risks of heavy metals in different areas influenced by different land use types, such as industrial areas use for industrial activities, agricultural areas using for farming production, traffic and human active area in the urban central city, all those activities would discharge heavy metals into the river. The river network distributes widely in urban area, industrial area, agricultural areas and suburban towns. Although connected with each other, river flow is very slow in most of the river, which make it influenced more by land use types and can be used as a good case study. This study can provide a scientific basis for the prevention and control of heavy metals in other urban areas with different anthropogenic activities, plain river network areas and poor hydrodynamics.

The difficulties of this study were to combined land use types with the pollution characteristics of heavy metals in surface water and sediments of river networks, and scientifically explored the impact of intensive land use on the spatial distribution and ecological risks of heavy metals in rivers. The challenge was that it requires a lot of detailed and up-front work in determining the main types of land represented by the sampling sites.

This paper is based on the idea of studying the distribution and ecological risks of heavy metals in urban river network under the influence of different intensive land use. The methods of sample collection, sample analysis and data analysis were first explained. Next, from the aspects of heavy metal concentrations and spatial distribution, the variations of heavy metals in surface water and sediments of river network under different land use types were analyzed. Based on above studies, the influencing factors, ecological risks and sources of heavy metals in sediments were discussed and the main results were concluded finally.

2. Materials and methods

2.1. Study area and sample collection

The study area of Shanghai is located in the eastern Yangtze River Delta (30°40′ - 31°53′N, 120°51′ - 122°12′E), and the total area is 6340.5 km^2, which is one of the largest cities and also the center of finance, trade and shipping in China. The mean altitude of Shanghai is about 4 m, and the general trend of land topography is inclined slightly from east to west. The soil in the southwest of Shanghai is yellow–-brown soil, and the marsh plains and coastal plains are paddy soils, gray fluvo-aquic soils, and delta plains and tidal flats have coastal saline soils (SMSB, 2018). The upper soil in Shanghai is clayey. Next is a phreatic aquifer (sand layer) 0.5–2.5 m below the surface (Shen et al., 2014).

The area of industrial and warehousing, agricultural planting, central city and suburban towns in shanghai is 839, 2794, 660 and 660 km^2, respectively, from Xiao et al. (2017), SMSB (2018) and Shanghai City Master Plan (2017–2035) (SCMP) (SCMP, 2018). The total area of rivers and lakes in Shanghai is 620.98 km^2, amounting to 9.79% of the area of Shanghai (SMSB, 2018).

The climate in Shanghai is subtropical monsoon climate, mild and humid, with abundant rainfall. The mean precipitation is 1258 mm, and the annual precipitation in each district is 1066–1462 mm, with an annual rainy day of 124 days. In 2017, the annual precipitation in Shanghai was 1388.8 mm, and the annual runoff in the city was 437.8 mm. The annual sunshine hours are 1876 h. The mean temperature in Shanghai is 17.5 °C, of which 18.2 °C is in the central city and 17.5 °C in the suburbs. The urban climate is characterized by heat islands and rain islands (SMSB, 2018). In 2017, Shanghai's population density was 3814 persons/km^2, which reached a maximum of 34058 persons/km^2 in the central city (SMSB, 2018). The rivers and creeks in Shanghai are interconnected with each other, forming a river network. Except for the Huangpu River, most of the rivers in the river network are cut and controlled by flood gates, which weakens the hydrodynamic conditions of the river network (Bi et al., 2018), and further affects the water physic-chemical parameters of the rivers (Alizadeh et al., 2018). Since the 1980s, Shanghai has experienced rapid urbanization and industrialization, resulting in more serious environmental changes (Wang et al., 2008). Located in China's highly urbanized, industrialized and densely populated area, the Shanghai river network is a typical river network with weak hydrodynamic conditions (Bi et al., 2018).

A total of 53 sampling sites from 44 rivers in Shanghai were selected to collect surface water and sediment samples respectively. The sampling sites were laid out by the grid layout method and were positioned by a global positioning system (GPS) (Fig. 1). In order to analyze the distribution characteristics of heavy metals in different functional areas, the whole Shanghai municipality was divided into industrial areas (R1, n = 11), agricultural areas (R2, n = 14), central city (R3, n = 11), suburban towns (R4, n = 11) and Chongming Island (R5, n = 6) according to the function of land use in each area. According to the buffer area represented by the sampling points, combined with the division of land use in Shanghai in the study of Yu et al. (2013), the impact of local land use and land cover types on the regional division was avoided. About 2 L surface water (5–15 cm depth) samples (two copies) were collected by using a plexiglass sampler at each sampling point and stored in 1 L clean polyethylene plastic bottles. Sediments (0–15 cm depth) of the river network were sampled by using the Ekman-Birge bottom sampler (HYDRO-BIOS, Germany) and stored in sealed polyethylene bags. Dissolved oxygen (DO), temperature and pH

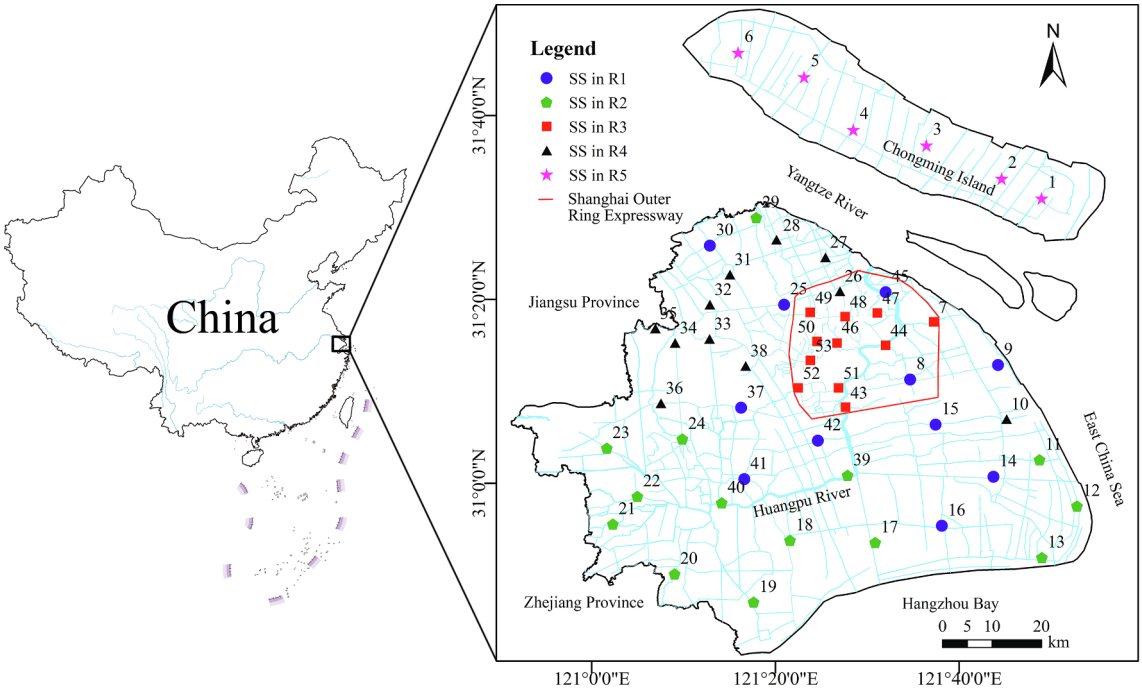

Fig. 1. Map showing sampling sites in the Shanghai river network.

of surface water were measured in situ by using portable DO meter (YSI55, USA) and pH meter (IQ150, USA), respectively. The samples of surface water and sediment were quickly brought back to the laboratory and stored at 4 °C and −18 °C, respectively.

2.2. Sample preparation and analysis

Surface water samples were filtered through a 0.45 μm microporous membrane, which was dried at high temperature before use. Sediment samples were dried in an electric heating constant temperature air drying oven (40 °C), ground and mixed evenly, and then passed through 60 and 230 mesh nylon sieves, which were used for the determination of TOC and heavy metals, respectively.

Take about 0.1 g of unscreened sediment sample in a 50 ml beaker, add 10 ml of 0.05 mol/L $(NaPO_4)_6$ solution, stir well with a glass rod overnight, and then ultrasonically disperse for 10 min. Then used a laser diffraction particle analyzer (LS13320, Beckman Coulter, USA) to measure the particle size (Ding et al., 1998). Take about 0.3 g sample of undried sediment was taken and analyzed for AVS contents by methylene blue spectrophotometry (GB 17378.5-2007). The total organic carbon (TOC) in sediments was measured using the potassium dichromate oxidation-external heating method (Nelson and Sommers, 1982).

Take 10 ml of surface water sample into a 25 ml colorimetric tube, add 1 ml of BrCl solution and let it stand for 2 h. Hydroxylamine hydrochloride was dripped into the colorimetric tube to discolor the solution, and then the volume was adjusted to 25 ml with ultrapure water. After being thoroughly mixed, it was used to test the Hg concentration in surface water sample. Take 0.2000 g of a sediment sample into a 50 ml colorimetric tube, add 5 ml ultrapure water and 5 ml aqua regia. After thorough mixing, let stand overnight and perform cold digestion. The next day, after shaking well, the colorimetric tube was placed in a thermostatic water bath and heated in a water bath at 95 °C for 2 h, and thoroughly mixed every half hour. After cooling, makeup to 50 ml with ultrapure water, shake and let stand overnight to make the solution clear, it was used to test the Hg concentration in sediments sample. Take 0.1000 g of a sediment sample into polytetrafluoroethylene digestion tank, add 3 ml HNO_3, 3 ml HF and 1 ml $HClO_4$ in order, and place in an oven (180 °C) for 2 h. After cooling, open the cover and place it on a hot plate (140 °C) to catch acid to the last drop, and then make up to 50 ml with 2% HNO_3. After standing, take the supernatant to test the Cd, Cr, Cu, Pb and Zn Concentrations in sediments sample.

The concentration of Hg in surface water and sediment sample were measured by atomic fluorescence spectrometry (AFS) (AFS-9230, Beijing Jitian, China). AFS has been widely used to measure Hg in water and sediments due to its reliability in measuring Hg concentrations (Zhang et al., 2016; Liang et al., 2017; Qu et al., 2018). Compared with other methods, AFS has the advantage of lower detection limit and higher accuracy when measuring Hg concentrations in water and sediments. The concentrations of Cd, Cr, Cu, Pb and Zn in surface water were measured by inductively coupled plasma mass spectrometry (ICP-MS) (ELEMENT 2, Germany). The concentrations of heavy metals in river water is low, so instruments with low detection limits are required for measurement. ICP-MS is becoming a workhorse for Cd, Cr, Cu, Pb and Zn analysis in water (Nguyen et al., 2013; Islam et al., 2015; Xu et al., 2017), not only because it's excellent lower detection limits, but also have the advantages of wide dynamic range and rapid semi-quantitative analysis. The concentrations of Cd, Cr, Cu, Pb and Zn in sediments were measured by the atomic absorption spectrometer (AAS) (AANALYST800, Perkin-Elmer, USA). Compared with other measurement methods, AAS has the advantages of low cost and high efficiency. AAS was widely used to accurately measure the concentrations of heavy metals in sediments (Goher et al., 2014; Nguyen et al., 2016).

2.3. Quality assurance and quality control (QA/QC)

In order to ensure the accuracy of heavy metal determination, in every batch digestion procedure, GSB04-1714-2004 and GBW07309 (GSD-9), from the National Standard Research Center of China, were used as quality control standards for surface water and sediment respectively. The recoveries of heavy metals ranged from 86% to 128%. In every batch of digestion and analytical procedures, three reagent blanks and triplicate samples were processed. The relative standard deviations were less than 10%. In order to reduce the pollution, the experimental containers were soaked in 10% HNO_3 and stored more than 24 h, then rinsed with tap water and ultra-pure water sequentially.

2.4. Data analysis

2.4.1. Geo-accumulation index method (I_{geo})

The geo-accumulation index method (I_{geo}) proposed by Muller (1969) was used to analyze heavy metal pollution in sediments of the Shanghai river network. The degree of heavy metal pollution of heavy metals could be assessed by I_{geo} (Islam et al., 2015). The I_{geo} has been widely used to assess the pollution levels of heavy metals in sediments (Liang et al., 2017; Han et al., 2017). There are other pollution assessment methods, such as comparing with environmental quality standards to analyze the pollution levels. But I_{geo} can quantify the pollution levels of heavy metals, and can clearly compare the impact of different land use types on heavy metal pollution in river networks. The calculation formula is as follows:

$$I_{geo} = Log_2\,[C_n/(1.5B_n)] \tag{1}$$

where C_n is the concentrations of element n in sediments; B_n is the geochemical background of element n, the geochemical background values of Cd, Cr, Cu, Hg, Pb and Zn in Shanghai are 0.13, 75.0, 28.6, 0.1, 25.5 and 86.1 μg/g, respectively (Wang et al., 1992); the constant *1.5* is used to account the possible variations in the background values (Islam et al., 2015). The geo-accumulation index consists of seven classes or grades: Level 0 (practically unpolluted): $I_{geo} < 0$; Level 1 (unpolluted to moderately polluted): $0 < I_{geo} < 1$; Level 2 (moderately polluted): $1 < I_{geo} < 2$; Level 3 (moderately to heavily polluted): $2 < I_{geo} < 3$; Level 4 (heavily polluted): $3 < I_{geo} < 4$; Level 5 (heavily to extremely polluted): $4 < I_{geo} < 5$; Level 6 (extremely polluted): $5 < I_{geo}$.

2.4.2. Potential ecological risk index method (RI)

The potential ecological risk index method (*RI*) (Hakanson, 1980) was used to evaluate the potential ecological risk posed by heavy metals in sediments (Maanan et al., 2015; Zhang et al., 2016). This method can not only analyze the pollution degree of a single pollution element, but also determine the comprehensive ecological risk of pollutants (Han et al., 2017). *RI* is a very useful, accurate and comprehensive method to evaluate the combined pollution risk of heavy metals in an aquatic system (Xu et al., 2017). The calculation formula is as follows:

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} \left(T_i \times \frac{C_i}{C_o}\right) \tag{2}$$

where n is the number of heavy metals; RI is the potential ecological risk index; E_r^i is the Potential ecological risk coefficient for a single element; T_i is the toxicological response coefficient for heavy metals; The toxicity coefficients of Cd, Cr, Cu, Hg, Pb and Zn based on the results from previous studies were determined to be 30, 2, 5, 40, 5 and 1, respectively (Zhang et al., 2016); C_i is the measured concentration of heavy metals in sediments; C_o is the background value of heavy metals in sediments. In this study, the background value of the soil environment in Shanghai is selected as the background value. The background values of Cd, Cr, Cu, Hg, Pb and Zn in Shanghai are 0.13, 75.0, 28.6, 0.1, 25.5 and 86.1 μg/g, respectively (Wang et al., 1992), respectively. The potential ecological risk coefficient and potential ecological risk index classification and pollution degree are shown in Table 1.

Table 1
The classification of potential ecological risk of heavy metals.

Potential ecological risk coefficient	Ecological risk	Potential ecological risk index	Ecological risk
$E_r^i < 40$	low	$RI < 150$	low
$40 \le E_r^i < 80$	moderate	$150 \le RI < 300$	moderate
$80 \le E_r^i < 160$	considerable	$300 \le RI < 600$	considerable
$160 \le E_r^i < 320$	high	$RI \ge 600$	very high
$E_r^i \ge 320$	very high		

2.4.3. Statistical analysis

Analysis of Variances (ANOVA) ($p < 0.05$) of heavy metal concentrations in different areas was performed by using a one-way ANOVA test. One-way ANOVA can test whether there is a significant difference between the mean values of dependent variables of sample multiple groups affected by a single factor. Bu et al. (2014) used one-way ANOVA to compare river water quality in different land-use areas. Pearson correlation analysis and principal component analysis (PCA) analysis was used to analyze the influencing factors and source apportionment of heavy metals by using SPSS 23. The significant level was set at $p < 0.05$ (two-tailed). Pearson correlation analysis and PCA is suitable for check the influence factors and identify the sources of heavy metals in water and sediments, and is widely used in many reports (Maanan et al., 2015; Zhang et al., 2016; Xu et al., 2017). Compared with other methods, the result of PCA can reflect the source of all heavy metals in the sample (Nguyen et al., 2013). Statistical figures were drawn by origin 9.1.

3. Results and discussion

3.1. Physic-chemical parameters and sediments particle sizes analysis of Shanghai river network

Physic-chemical parameters such as temperature (T), pH, DO, salinity, conductivity (EC), total suspended solids (TSS) and total phosphorus (TP) in surface water, AVS and TOC in sediments are important factors affecting the concentrations, migration and transformation of heavy metals in rivers (Zhang et al., 2014; Garneau et al., 2015; Han et al., 2017; Liang et al., 2017). In the development and improvement of water quality prediction models, water quality indicators such as salinity and sediment load are important considerations (Chen and Chau, 2019; Shamshirband et al., 2019). As shown in Table 2, the lowest values of pH and DO in surface water appeared in the central city, followed by suburban towns and industrial areas. Central city had the highest TP value of 0.92 mg/L, followed by suburban towns, agricultural areas and Chongming Island. Intensive land use and agricultural activities resulted in increased TP contents in rivers (Bu et al., 2014). Areas with higher urbanization had worse surface water quality (Wang et al., 2008). There was no significant difference in temperature and TSS contents in surface water among different areas.

The contents of AVS and TOC in sediments of the central city were the highest among all areas, followed by agricultural areas and suburban towns. The growth of population, the increasing discharge of urban sewage and agricultural activities caused organic matter pollution and consumed DO in the river (Bu et al., 2014; Zhang et al., 2014). Sulfur could be released during organic matter decomposition (Du Laing et al., 2009).

The proportion of clay contents in the river network sediments from the central city was higher than those from other areas, indicating more weak hydrodynamic conditions in central cities (Fig. 2). The silt was the most widely distributed particles and accounted for 55.3%-81.14% of the total sediments. The proportion of sand in the river sediment of Chongming Island was much higher than those of other areas influenced by the least human disturbance and strong hydrodynamic conditions, which made less fine particles to remain in the river.

3.2. Heavy metal levels in river surface water and sediments

The concentrations of Cd, Cr, Cu, Hg, Pb and Zn from surface water of the Shanghai river network varied between 0.04 and 0.25, 0.21–3.44, 1.0–12.9, 0.01–5.42, 0.05–3.39 and 6.62–95.8 μg/L, respectively. Compared with other rivers (Table 3), the concentrations of Cd, Cr, Cu, Pb and Zn in surface water were lower than those from other rivers such as the Wenrui Tang River (Qu et al., 2018), the rivers near Laizhou Bay (Xu et al., 2017) in China, and also lower than those of the Mississippi River in the USA except for Cd (Bussan et al., 2017), the

Table 2
Mean physic-chemical parameters in surface water and sediments from different areas of Shanghai river networks.

Areas	*Surface water*							*Sediments*	
	T (°C)	pH	DO (mg/L)	Salinity (ng/L)	EC (μS/cm)	TSS (mg/L)	TP (mg/L)	AVS (μg/g)	TOC (mg/g)
Industrial areas	21.22	7.63	4.42	0.35	739.9	358.6	0.43	335.7	5.92
Agricultural areas	21.79	7.88	6.78	0.42	809.2	362.6	0.44	669.9	9.98
Central city	20.61	7.50	2.44	0.43	854.5	309.0	0.92	1244.7	12.43
Suburban towns	20.75	7.69	2.98	0.52	964.3	308.1	0.63	448.9	7.97
Chongming Island	19.62	8.05	7.28	0.33	599.5	333.0	0.24	97.8	6.87
Mean	21.00	7.74	4.74	0.42	811.8	337.0	0.54	608.0	8.82

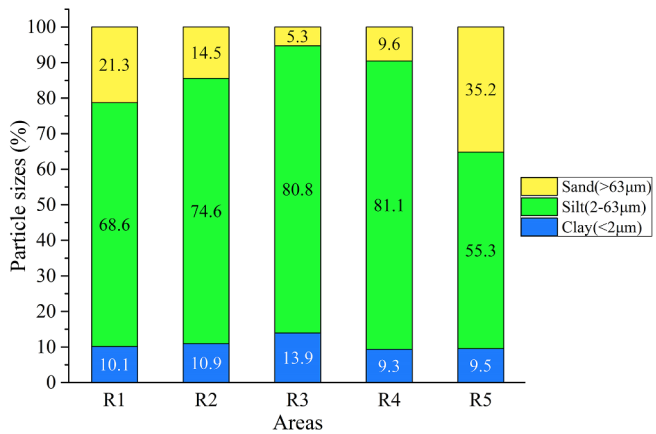

Fig. 2. Particle composition in sediments of the Shanghai river network.

Korotoa River in Bangladesh except for Zn (Islam et al., 2015), the Ismailia Canal in Egypt except for Cr and Zn (Goher et al., 2014), the To Lich River in Vietnam (Nguyen et al., 2013) and the Brahmani River in Indian (Swain and Sahoo, 2017) except for Cd and Zn, but higher than those from the Pardo River in Brazil except for Pb (Alves et al., 2014). Hg concentrations was higher than that of the Wenrui Tang River (Qu et al., 2018). Overall, the concentrations of heavy metals in surface water of the Shanghai river network were at a low level.

The concentrations of Cd, Cr, Cu, Hg, Pb and Zn in sediments of the Shanghai river network varied between 0.08 and 5.62, 38.8–549.1, 20.1–521.6, 0.06–3.28, 10.4–115.5 and 80.0–1404.8 μg/g, respectively. Compared with other rivers (Table 3), the concentrations of Cd, Cr, Cu, Pb and Zn in sediments were higher than those from other rivers such as the Yangtze Estuary (Wang et al., 2015), the Bortala River (Zhang et al., 2016), the Songhua River except for Cr (Li et al., 2017) and the Pearl River Delta except for Cd and Pb (Zhao et al., 2018) in China, and also higher than those of the Credit River watershed in Canada (Cowden et al., 2015), the Pardo River in Brazil (Alves et al., 2014), the Mvudi River in South Africa except for Cd (Edokpayi et al., 2016), the Gironde Estuary in France (Larrose et al., 2010) and the Red River in Vietnam (Nguyen et al., 2016) except for Pb, but the concentrations of Pb and Zn were lower than the Dill River in Germany (Martin, 2015). Hg concentrations was higher than those of the Bortala River and the Pearl River in China, the Gironde Estuary in France and the Credit River watershed in Canada, but lower than those of the Songhua River in China and the Feni river estuary in Bangladesh (Islam et al., 2018). In general, the concentrations of heavy metals in sediments of the Shanghai river network were at a relatively high level compared with other rivers. Anthropogenic activities were the main reason for heavy metal pollution in rivers (Edokpayi et al., 2016). Shanghai is a large city with a high degree of intensive urban land use (Bi et al., 2018). Long-term urbanization and land use have caused heavy metal pollution in river network sediments.

3.3. Spatial variations of heavy metals influenced by intensive land use

3.3.1. Spatial variations of heavy metals

Cd, Cr, Cu, Hg, Pb and Zn concentrations in river surface water varied little among different areas ($P > 0.05$), which might be due to the fluidity of surface water in the river network. The flow of river

Table 3
Comparison of heavy metal concentrations in surface water (μg/L) and sediment (μg/g) of rivers in different areas.

	Country	Sites	Cd	Cr	Cu	Hg	Pb	Zn	References
Surface water (μg/L)	China	Study Area	0.08	0.6	3.5	0.79	0.45	25.8	This study
	China	the Wen-Rui Tang River	0.98	5.3	20.9	0.03	4.2	72.1	(Qu et al., 2018)
	China	the rivers Laizhou Bay	0.49	3.5	114		1.51	113	(Xu et al., 2017)
	American	the Mississippi River	0.57	0.2	2.1		0.31	1.6	(Bussan et al., 2017)
	Indian	the Brahmani River	5.6	24.7	7.6		10.8	22.7	(Swain and Sahoo, 2017)
	Brazil	the Pardo River	0.05	0.5	3.3		1.8	13.3	(Alves et al., 2014)
	Bangladesh	the Korotoa River	8	73	61		27		(Islam et al., 2015)
	Vietnam	the To Lich River		2.9	4.5		8.1	51.1	(Nguyen et al., 2013)
	Egypt	the Ismailia Canal	0.45		7		18	15	(Goher et al., 2014)
Sediments (μg/g)	China	Study Area	0.53	106.3	87.5	0.42	35.2	296.8	This study
	China	the Yangtze Estuary	0.19	79.1	24.7		23.8	82.9	(Wang et al., 2015)
	China	the Songhua River	0.27	121.4	13.3	0.56	18.8	92.5	(Li et al., 2017)
	China	the Bortala River	0.17	51.6	30.1	0.02	32	99.2	(Zhang et al., 2016)
	China	the Pearl River Delta	0.84	55.2	42.9	0.1	44.6	135.9	(Zhao et al., 2018)
	Germany	the Dill River			83.4		107.9	323.9	(Martin, 2015)
	France	the Gironde Estuary	0.48	78.4	24.5	0.16	46.8	168	(Larrose et al., 2010)
	Canada	the Credit river watershed	0.28	7.4	12.6	0.07	8.9	63.6	(Cowden et al., 2015)
	Bangladesh	the Feni river estuary		35.3		0.71	6.5		(Islam et al., 2018)
	South Africa	the Mvudi River		51.6	26.8		3.9	24	(Edokpayi et al., 2016)
	Brazil	the Pardo River	0.06	25.5	21		9.7	40.3	(Alves et al., 2014)
	Vietnam	the Red River	0.35	85.7	83		66	127	(Nguyen et al., 2016)

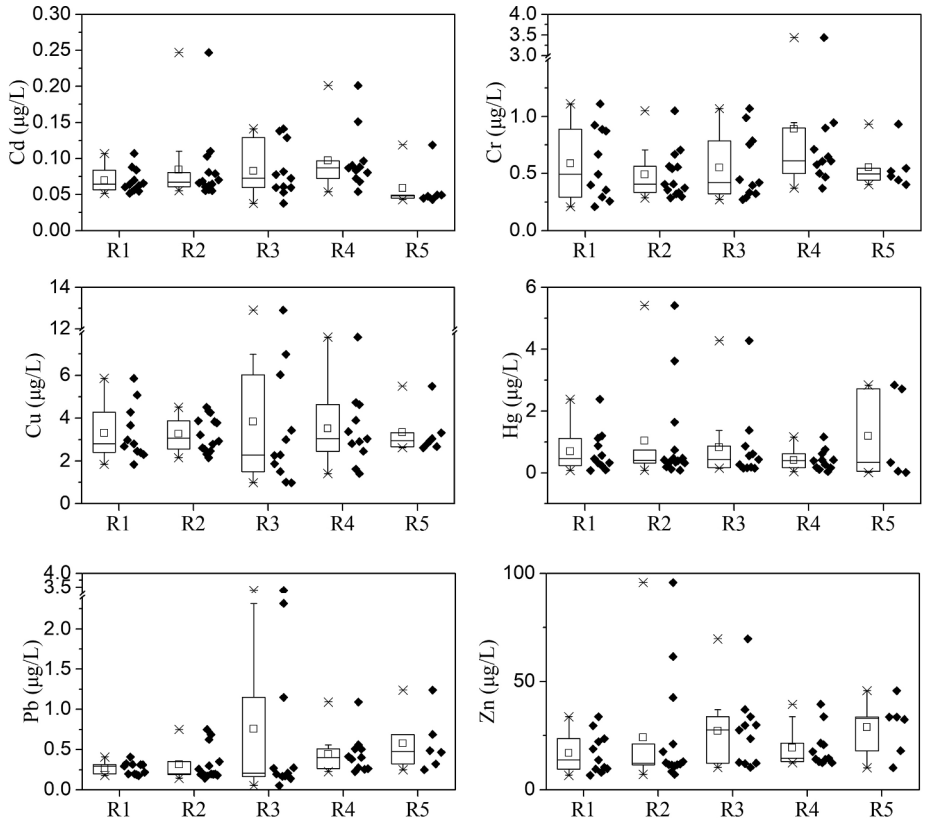

Fig. 3. Spatial variation of heavy metals in surface water of the Shanghai river network.

water affects the stagnation and dissolution of heavy metals in surface water, resulting in different concentrations of heavy metals in sediment and water (Nguyen et al., 2013). Compared with sediments, surface water has greater fluidity and mixing, which makes the concentrations of dissolved heavy metals in surface water have no significant difference in different areas. The highest concentrations of Cd, Hg and Zn were found in agricultural areas (Fig. 3), which was related to the application of chemical fertilizers in agricultural activities (Naveedullah et al., 2013). Chemicals such as herbicides used in agricultural activities also contain a large amount of Hg (Marrugo-Negrete et al., 2017). The highest concentrations of Cu and Pb found in the central city were 12.9 and 3.39 μg/L at site 51 and 47, respectively, which were from the black and odorous river. The discharge of urban domestic sewage will increase the concentrations of Cu in the river (Islam et al., 2015), and Pb enriched in black and smelly rivers may be derived from the use of gasoline and the discharge of wastewater from industrial production (Mohiuddin et al., 2012; Islam et al., 2015).

There was no significant difference of Cd, Cr, Cu and Zn concentrations in the river sediments among different areas ($P > 0.05$), but Hg and Pb concentrations in sediments varied significantly ($P < 0.05$) in different areas. Concentrations and types of heavy metals in rivers varied by land use type (Bussan et al., 2017). The mean concentrations of Hg and Pb in sediments of the central city were the highest among the five areas (Fig. 4). Land intensive use and anthropogenic activities in the central city of Shanghai were significantly higher than those in other areas (Bi et al., 2018), which will increase the concentrations of heavy metals in urban river sediments. A similar conclusion was reported by Lundy et al. (2017). Although there was no significant difference of Cd, Cr, Cu and Zn concentrations among different areas, the highest concentration of Cd was found at sites 51 and the highest concentrations of Cr, Cu and Zn were found at sites 28. Both sites were located in black and odorous river sections. Rivers with black odors were often accompanied by high concentrations of heavy metals (Islam et al., 2015).

Overall, the concentrations of heavy metals in the river network were significantly higher in the areas with intensive land use and more anthropogenic activities were concentrated, such as central city and suburban towns. In areas of lower anthropogenic activities and land use, such as Chongming Island, the concentrations of heavy metals in river networks were significantly lower. The amount and intensity of pollutants emission in the areas with intensive land use and high-intensity anthropogenic activities were significantly higher than those in other areas, which led to the relatively high pollution of heavy metals (Song et al., 2010; Edokpayi et al., 2016). Paul (2017) also found that land-intensive urban areas were an important source of heavy metals in the Ganges.

3.3.2. Factors influencing spatial variations of heavy metals in sediments by land use

Hg concentrations in surface water of Shanghai river network showed a very significant positive correlation with pH ($r = 0.470$, $p < 0.01$) and DO ($r = 0.455$, $p < 0.01$), indicating that pH and DO had a certain impact on the migration and conversion of Hg in surface water. The pH value of river water was between 7.12 and 9.29, with a mean value of 7.74, which was slightly alkaline. High pH value promoted the adsorption and precipitation of heavy metals in river water (Zhang et al., 2014). Heavy metals deposited in sediments could release into the water due to the changes of pH and other properties of the river (Miao et al., 2006). The poor correlation between temperature, salinity, TSS and TP with heavy metal concentrations in surface water indicated that those physic-chemical parameters had little effect on heavy metal pollution in the Shanghai river network. Similar result was also found by Xu et al. (2017).

Heavy metals such as Cd and Zn are abundantly present in sulfides in AVS-rich deposits. The sulfide exhibits a weak solubility in the reduced state of the deposit, and thus a large amount of AVS is combined with the cationic metals (Zhang et al., 2014). As showed in Fig. 5, AVS contents were significantly positively correlated with Cd, Hg and Zn in

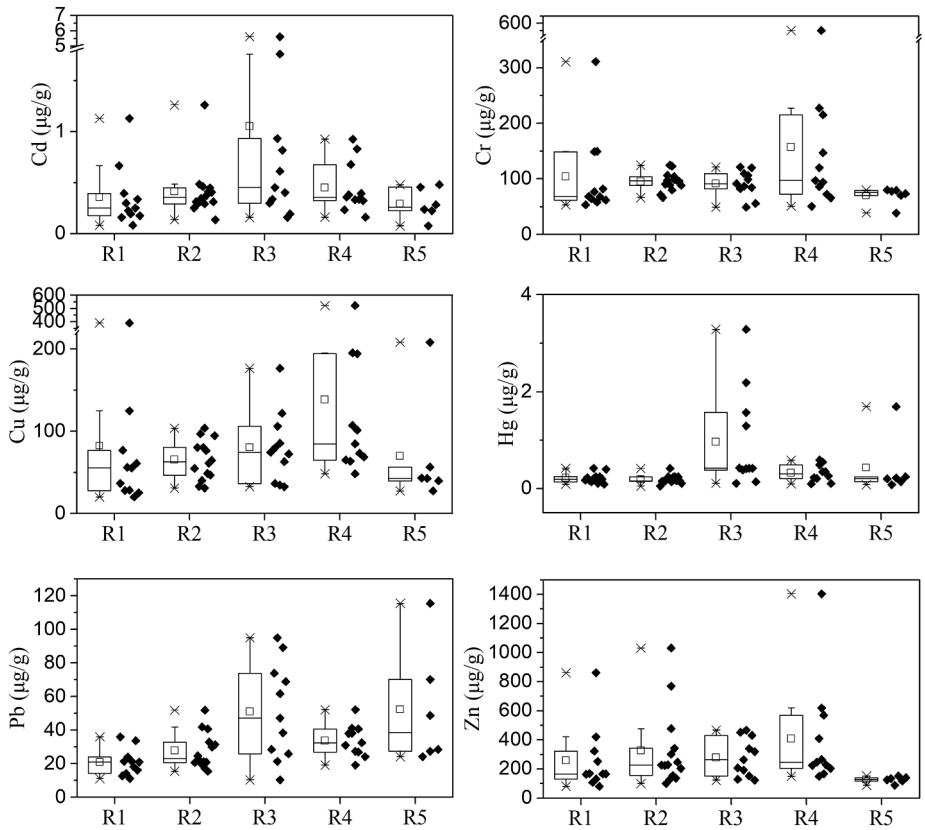

Fig. 4. Spatial variation of heavy metals in sediments of the Shanghai river network.

sediments of Shanghai river network ($P < 0.01$), and TOC contents were very significantly positively correlated with Cd, Hg and Zn ($P < 0.01$), indicating that AVS and TOC affected the migration and transformation of Cd, Hg and Zn in sediments. AVS was an important heavy metal-binding phase in sediments. Zhao et al. (2018) found that organic compounds could affect the concentrations and distribution of heavy metals in sediments by adsorbing small particles and then absorbing large amounts of heavy metals. The part of low to medium molecular weight in the organic matter could promote the dissolution of heavy metals (Du Laing et al., 2009; Zhang et al., 2014). The concentrations of heavy metals in sediments of the Shanghai river network showed no correlation with particle sizes, indicating that the particle composition did not affect heavy metals, which was consistent with the conclusions reported by Mwanamoki et al. (2015) and Wang et al. (2016).

It can be seen from the above analysis, that most physic-chemical

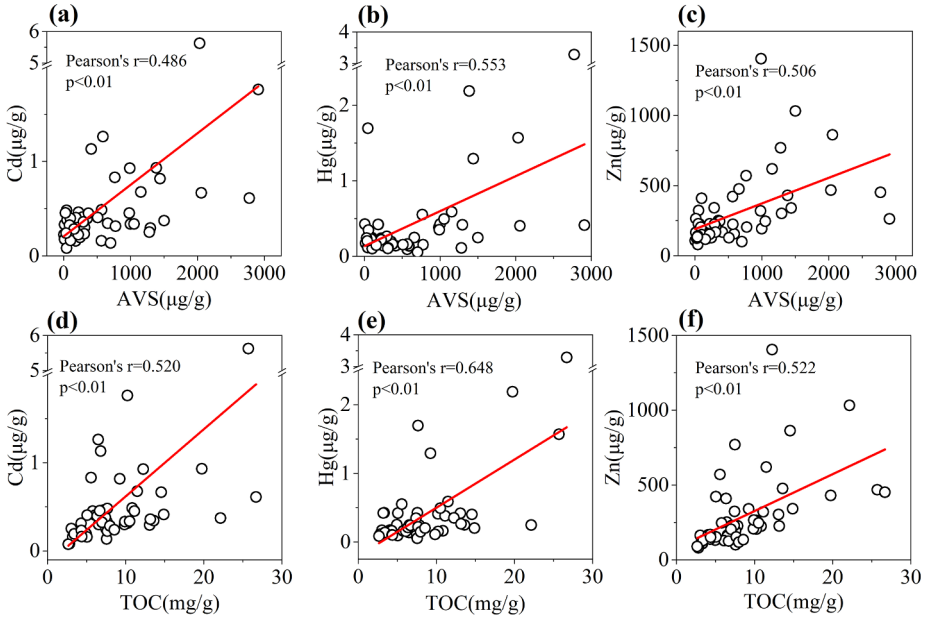

Fig. 5. Correlations between heavy metal concentrations and AVS or TOC contents in sediments of the Shanghai river network.

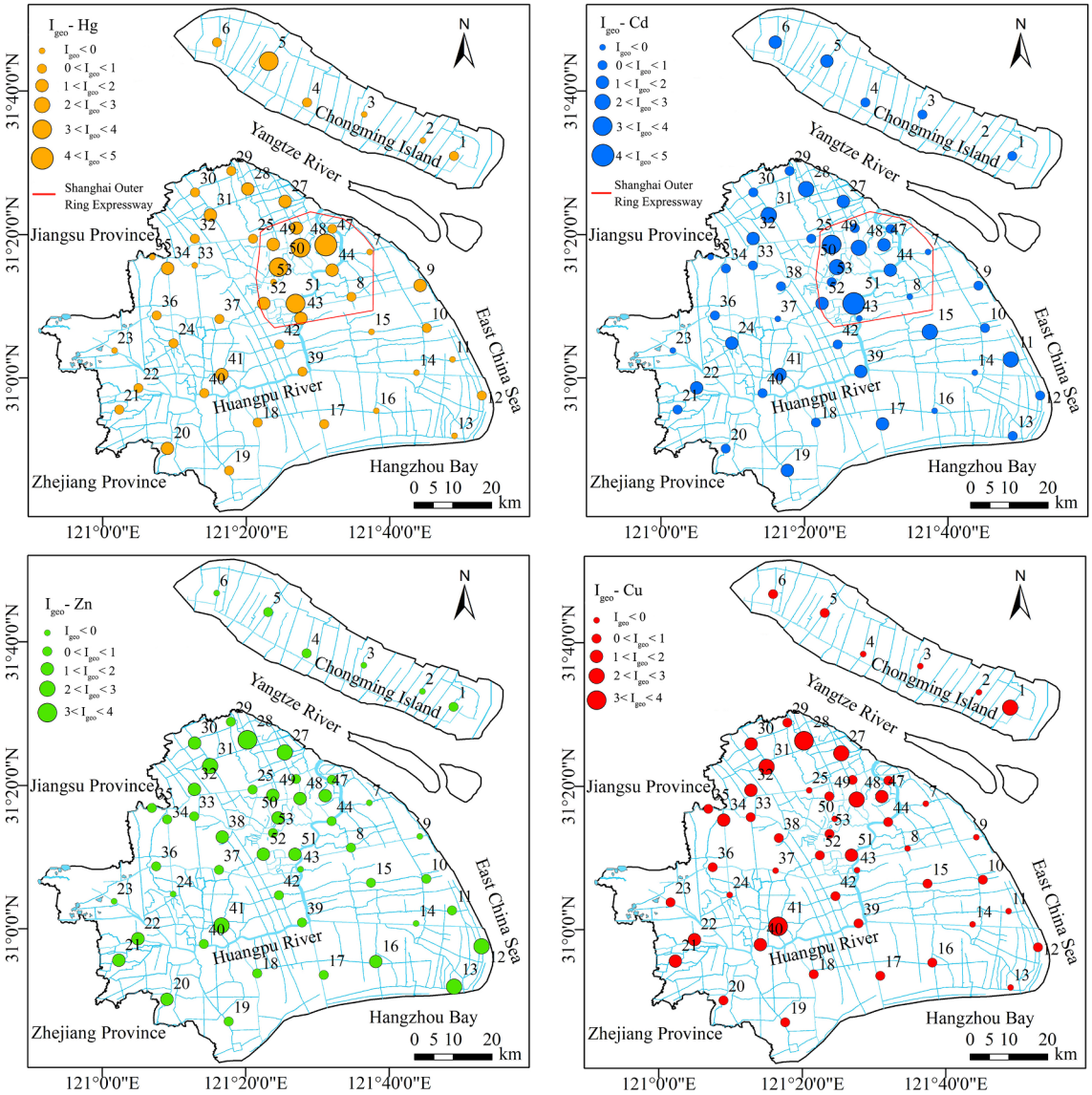

Fig. 6. The I_{geo} of heavy metals in sediments of the Shanghai river network.

parameters have no correlation with heavy metal concentrations, only the pH and DO with Hg concentrations in surface water, the AVS and TOC contents with Cd, Hg and Zn concentrations in sediments, which with the positive correlation, indicating that the rivers in plain river network, heavy metal concentrations is more affected by different land use types, and different land use types will affect the concentrations and distribution of heavy metals by changing the contents of AVS, TOC and DO in the river.

3.4. Ecological risk assessment of heavy metals in sediments

The I_{geo} method was used to evaluate the pollution level of heavy metals in sediments of the Shanghai river network. The mean I_{geo} of Hg, Cd, Zn and Cu were 1.45, 1.42, 1.20 and 1.03, respectively, which was moderately polluted. The mean I_{geo} of Pb and Cr were less than 0, which was practically unpolluted. Therefore, Shanghai river network sediments were mainly polluted by Hg, Cd, Zn and Cu. The pollution level of Hg and Cd in Shanghai rivers were mainly unpolluted to moderately polluted, moderately polluted and moderately to heavily polluted (Fig. 6). The I_{geo} of Cd and Hg in sediments from the central city were significantly higher than those from other areas. The central city within the outer ring expressway of Shanghai has the highest land utilization intensity (Bi et al., 2018). This indicated that intensive land use contributes significantly to the accumulation of heavy metals in sediments of the Shanghai river network in this area. The pollution levels of Zn and Cu in Shanghai rivers were mainly from unpolluted to moderately polluted and moderately polluted (Fig. 6). Moderately polluted and heavily polluted of Pb and Cd were also found in the Pearl River sediments of South China (Zhao et al., 2018) and the Korotoa River sediments of Bangladesh (Islam et al., 2015). The pollution extent of Cd and Hg were moderately to heavily polluted and heavily polluted at most sampling sites in Shanghai river sediments of China (Jia et al., 2013).

The mean potential ecological risk index (*RI*) of heavy metals in sediments of Shanghai river network was 309.8 (Table 4), which was higher than that in the Bortala River (Zhang et al., 2016) and the Yangtze River Estuary (Han et al., 2017), suggesting a considerable ecological risk. The highest *RI* (651.8) was found in the central city, suggesting a very high ecological risk. The *RI* of heavy metals in industrial areas, agricultural areas, Chongming Island and suburban towns varied between 150 and 300, suggesting the moderate ecological risk.

Cd and Hg were the two major contributors to *RI*, with a contribution rate of 53.0% and 38.8%, respectively. The mean potential

Table 4
Potential ecological risk assessment of heavy metals in sediments of the Shanghai river network.

Areas	E_r^i						*RI*
	Cd	Cr	Cu	Hg	Pb	Zn	
Industrial areas	80.9	2.8	14.4	87.7	4.1	3.0	192.8
Agricultural areas	94.1	2.5	11.4	76.0	5.5	3.8	193.3
Central city	239.5	2.4	14.0	382.6	10.0	3.2	651.8
Suburban towns	102.4	4.2	24.2	128.1	6.6	4.7	258.6
Chongming Island	66.6	1.9	12.2	170.7	10.3	1.5	263.1
Mean	120.1	2.8	15.3	164.3	6.9	3.4	309.8

ecological risk coefficient (E_r^i) of Hg in sediments of the Shanghai river network was 164.3, which showed a very high ecological risk, in which 30.2% of the sampling sites were at a considerable ecological risk level and 26% were up the high ecological risk. The mean E_r^i of Cd was 120.1, which was at considerable ecological risk, in which 47.2% of the samples reached a level of considerable ecological risk. In the central city. The mean E_r^i of Cd and Hg showed high risk and very high risk, respectively. And in the suburban towns, the E_r^i of Cd and Hg varied between 80 and 120, suggesting a considerable ecological risk. Land intensive use and anthropogenic activities in central city and suburban towns of Shanghai were significantly higher than those in other areas (Bi et al., 2018). The mean E_r^i of Cu was 15.3, which showed a low ecological risk. For Cr, Pb and Zn, the E_r^i values were below 40 and all samples were at a low ecological risk level. Zhang et al. (2016) found that Cu and Zn in sediments of Bortala River were at low ecological risk, and Cd, Cr and Hg were at moderate ecological risk.

Based on the above analysis, the hazard ranking of various heavy metals in sediments of the Shanghai river network were Hg > Cd > Cu > Pb > Zn > Cr. The ecological risks of Hg and Cd were the highest, contributing 91.8% of the total *RI* in sediments. The geo-accumulation index of Hg and Cd showed a high pollution level too. Deng et al. (2020) also found that Cd and Hg contributed 90% to *RI* in the water reservoir of Irrigation-T. Cd was the major contributor (57.17% contribution) to *RI* in sediments of the Yangtze River Estuary (Han et al., 2017). The I_{geo} value of Cd was the highest in sediments of the Korotoa River (Islam et al., 2015). Cd and Hg pollution levels were the highest in sediments of the Bortala River (Zhang et al., 2016). The other heavy metals in all areas were at low ecological risk. The *RI* of heavy metals in sediments of the river network in the Central city with the strongest intensive land use was the highest, followed by Chongming Island, suburban towns, agricultural areas and industrial areas, successively. When considering the treatment of heavy metal pollution in urban rivers, emphasis should be placed on the treatment of rivers in the areas with intensive land use, and heavy metals with high ecological risks such as Hg and Cd should be paid more attention.

3.5. Source apportionment of heavy metals in surface water and sediments

The results of the correlation analysis showed that Cu had a very significant positive correlation with Cr ($r = 0.45$, $p < 0.01$) and Pb ($r = 0.64$, $p < 0.01$). And Zn had a very significant positive correlation with Pb ($r = 0.434$, $p < 0.01$) and Cd ($r = 0.416$, $r < 0.01$). That correlation suggested the same sources or similar migration and transformation behavior of these heavy metals.

PCA is a commonly used method in heavy metal source analysis (Li et al., 2013). According to the eigenvalue > 0.7, four principal components (PC) of heavy metals in surface water were extracted, and the cumulative variance contribution rate was 83.1% (Table 5). Cr and Cu had higher loads in PC 1 with a contribution rate of 30.9%. Cd and Pb showed higher loads in PC 2 and PC 3, respectively, with their respective contribution rate of 19.7% and 17.3%. Hg had a higher load in PC 4, suggesting that Hg in the river mainly comes from coal-burning

Table 5
Factor loading and accumulated variance of heavy metals in surface water and sediments of the Shanghai river network.

		PC 1	PC 2	PC 3	PC 4
Surface water	Cd	0.11	**0.92**	0.05	0.10
	Cr	**0.89**	0.12	−0.15	−0.08
	Cu	**0.76**	−0.20	0.35	0.05
	Hg	−0.03	0.01	−0.05	**0.98**
	Pb	0.05	−0.04	**0.96**	−0.06
	Zn	0.40	−0.60	0.28	0.22
	Eigenvalue	1.85	1.18	1.04	0.91
	Variance contribution rate %	30.9	19.7	17.3	15.2
	Cumulative contribution rate %	30.9	50.6	68.0	83.1
Sediments	Cd	0.12	**0.87**	−0.01	
	Cr	**0.96**	−0.02	0.04	
	Cu	**0.93**	0.09	0.13	
	Hg	0.03	**0.75**	0.31	
	Pb	0.09	0.19	**0.96**	
	Zn	**0.89**	0.16	0.01	
	Eigenvalue	2.79	1.48	0.76	
	Variance contribution rate %	46.6	24.7	12.6	
	Cumulative contribution rate %	46.6	71.3	83.9	

(Song et al., 2011). Shanghai consumed 45.8 million tons of coal in 2017 (SMSB, 2018). A large amount of Hg was emitted into the environment because of coal-burning (Song et al., 2011).

According to the eigenvalue > 0.7, three principal components of heavy metals in sediments were extracted, and the cumulative variance contribution rate was 83.9% (Table 5). Cr, Cu and Zn had higher loads in PC 1 with a contribution rate of 46.6%, which were mainly from industrial activities. Zhao et al. (2018) found that the increasing concentrations of Cr, Cu and Zn in river water might be caused by industrial electroplating wastewater. In industrial production activities of electroplating and galvanizing, discharged wastewater contained a large number of heavy metals such as Cr, Cu and Zn (Meybeck et al., 2007; Qu et al., 2018). Cd and Hg had higher loads in PC 2 with a contribution rate of 24.7%, which were mainly from complex pollution of traffic and anthropogenic activities. Road traffic was an important factor affecting the increase of Cd concentrations in river waters (Njenga et al., 2009). Coal-burning caused a large amount of Hg to be released into the environment (Song et al., 2011). Cd and Hg were found to have the highest concentrations in the central city with heavy traffic, there was a very significant positive correlation between Cd and Hg. Pb showed a higher load in PC 3. Coal and oil burning were the major sources of anthropogenic Pb (Chen et al., 2014). In 2017, coal and oil consumed in Shanghai were 45.778 and 37.749 million tons, respectively (SMSB, 2018).

4. Conclusions

The concentrations, distribution characteristics, sources and potential ecological risk of heavy metals in surface water and sediments of the Shanghai river network were analyzed. Compared with other rivers, heavy metal concentrations in surface water were at low levels, while heavy metal concentrations in sediments were obviously higher than those in other rivers. The mean concentrations of Cd, Cr, Cu, Hg, Pb and Zn in sediments were 0.53, 106.3, 87.5, 0.42 35.2 and 296.8 μg/g, respectively. Hg and Pb concentrations in sediments varied significantly in different areas. The highest mean values of 1.02 and 53.43 μg/g were found in the central city with the strongest intensive land use and high intensity of anthropogenic activities. AVS and TOC contents in sediments influenced the migration and transformation of Cd, Hg and Zn. Source analysis showed that Cd and Hg were mainly from complex pollution of traffic and coal combustion. Intensive land use contributes significantly to the accumulation of heavy metals in sediments of the

Shanghai river network. Heavy metals in the river sediments showed a considerable ecological risk. Cd and Hg were the two major contributors to *RI*, with the contribution rate of 53.0% and 38.8%, respectively. Central city has the highest *RI* of 651.8, suggesting a very high ecological risk. In order to quantitatively analyze the effects of intensive land use on heavy metal distribution in urbanized river networks, more detailed information such as population density, historical changes of land use, traffic flow and a number of thermal power plants and industrial plants should be considered in the future research.

Credit authorship contribution statement

Yongsheng Zeng: Formal analysis, Writing - original draft, Writing - review & editing. **Chunjuan Bi:** Conceptualization, Writing - review & editing, Supervision, Project administration. **Jinpu Jia:** Writing - review & editing. **Lin Deng:** Resources. **Zhenlou Chen:** Supervision, Funding acquisition.

Declaration of Competing 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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