Heavy metals in soil, water sediment, and ambient PM2.5 across the Yangtze River economic belt: integrated environment risk identification and countermeasures

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ABSTRACT

To explore the integrated pollution features of heavy metals in environmental multi-media throughout the Yangtze River Economic Belt (YREB) is of significance for national/local decision-makers to establish a sustainable development strategy. Distributions, enrichments, and induced environmental risks of typical toxic metals in soil, water surface sediment, and PM2.5 from the 11 provinces/municipalities of the YREB were studied based on bibliometric analysis. Compared with the background values, Cu, Zn, Cd, Pb, Hg, and Ni were enriched in both soil and surface sediments to a certain extent. Also, the provincial contents of As and Cr were higher than the standard limit of PM2.5. Spatially, the heavy metal pollution in environmental multi-media was distributed primarily in the central and southern parts of the YREB, specifically in the Hunan, Guizhou, Hubei, and Jiangxi provinces. The noncarcinogenic risks of As in soil and PM2.5 were the main contributor to the total risk. In the surface sediment, the average potential ecological risk for Cd and Hg reached Level V (very high ecological risk) and Level IV (high ecological risk), respectively, which is much higher than that for the other metals (Level I, low potential ecological risk). Consequently, Cd, Hg, and As were considered as priority control metals, and targeted risk management policies were recommended.

Key words | heavy metals, risk management, sediment, soil, PM2.5, Yangtze River Economic Belt

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HIGHLIGHTS

- Heavy metals in environmental multi-media (surface water sediment, soil, and ambient PM2.5) were investigated across the Yangtze River Economic Belt.
- Integrated environment risk identification was carried out based on regional eco-risk and health risk assessment.
- Targeted countermeasures were recommended to efficiently manage the identified "hot" heavy metals/areas from an environmental multi-media perspective.

INTRODUCTION

Relying on the gold channel of the Yangtze River, the Yangtze River Economic Belt (YREB) stretches across eastern, central, and western China, which is a booming Chinese economic belt. The YREB covers an area of more than 2 million km², spanning nine provinces and two municipalities, namely, Anhui, Guizhou, Hunan, Hubei, Jiangxi, Zhejiang, Sichuan, Jiangsu, Yunnan, Chongqing, and Shanghai. As a dense urban zone in China, the YREB includes the Yangtze River Delta city group, the Triangle of Central China, and the Chengdu & Chongqing City Group. As a basin economic zone with the highest economic density in China, the YREB constitutes more than 40% of the annual national GDP, with a population of 580 million, lending great advantage in terms of regional development and capacity. In the past decade, rapid industrialization and urbanization have led to the establishment of a large number of enterprises in the steel metallurgy, automobile, electronics, petrochemical, and textile and clothing sectors, and strategic emerging industries. However, a lot of problems related to development have surfaced, such as unreasonable industrial planning and layout, unbalanced regional development, and ecological pollution (Zhu & Shao 2020). To meet the national green and sustainable development requirements, it is important to take the regional economy and its whole environment and resident health as the foundation for the purpose of a joint management decision-making. Recently, some researchers have focused on environmental issues in the YREB, mainly by identifying the relationship between urban land use and anthropogenic activities (Liu *et al.* 2018), exploring impacts of socioeconomic and natural factors on PM2.5 pollution (Liu *et al.* 2020), and identifying the problem of waste dumping surrounding the YREB (Kang *et al.* 2020). However, a representative study of heavy metal pollution across China is rarely done from the economic zone perspective. Elevated heavy metals will affect directly/indirectly environmental multi-media safety (mainly including soil, water, and air) and human health because of their toxicity, persistence, and bioaccumulation (Kim *et al.* 2019). In recent years, some researchers have reported elevated levels of heavy metals in soil (Zhou & Wang 2019), PM2.5 (Xie *et al.*

2020), sediment, and fishes (Yi *et al.* 2011) in the Yangtze River basin. Therefore, it is of significance to explore the environmental multi-media risk of typical heavy metals and develop a regionally targeted risk management policy from the economic belt perspective.

The three primary purposes of the present study were (1) to investigate the bibliometric contents of typical heavy metals in soil, PM2.5, and water surface sediment from the YREB on a provincial scale; (2) to carry out a bibliometric environmental pollution and risk evaluation of the studied heavy metals; and (3) to put forward integrated management countermeasures.

METHODS AND MATERIALS

Data collection

The bibliometric contents of eight typical heavy metals (Cd, Cr, Pb, Hg, Cu, Ni, As, and Zn) in environmental multi-media from the YREB were collected in the database of Web of Science (SCI and SSCI databases) and the China National Knowledge Infrastructure (CNKI). The key words of literature screening, in this study, were 'Yangtze Economic Belt', 'heavy metal', 'metals', 'soil', 'PM2.5', 'surface sediment', and so on. For the purpose of making the extracted data more effective, the sampling time of the relevant literature was limited to 2010–2018 (soil) and 2014–2018 (PM2.5 and water surface sediment). In addition, the collected literature met the following criteria: (i) the data were extracted from the monitoring study; (ii) the sampling sites and periods were clear; (iii) the heavy metal contents were accurately measured; and (iv) the extracted data were monitored in nine provinces and two municipalities in the YREB. The detailed extracted references are shown in Supplementary Tables S1, S2, and S3.

Geo-accumulation index

The geo-accumulation index, represented as I_{geo} (Equation (1)), is employed to quantitatively assess metals' enrichment

degree in soil or sediment (Muller 1969):

$$I_{\text{geo}} = \log_2\left[\frac{C_n}{1.5 \times B_n}\right] \tag{1}$$

where the studied metal content is represented by C_n (mg/kg), and the corresponding values of the referred metals are shown as B_n (mg/kg). The enrichment levels were divided into seven groups based on the values of I_{geo}: (i) $I_{\text{geo}} \leq 0$, uncontaminated; (ii) $0 < I_{\text{geo}} \leq 1$, uncontaminated to moderately contaminated; (iii) $1 < I_{\text{geo}} \leq 2$, moderately contaminated; (iv) $2 < I_{\text{geo}} \leq 3$, moderately contaminated to heavily contaminated; (v) $3 < I_{\text{geo}} \leq 4$, heavily contaminated; (vi) $4 < I_{\text{geo}} \leq 5$, heavily to extremely contaminated; and (vii) $I_{\text{geo}} > 5$, extremely contaminated.

Environmental risk assessment methodology

Health risk assessment

People generally faced the risk of exposure to heavy metals via three pathways – ingestion, inhalation, and dermal contact (Li *et al.* 2018). In the present study, the health risk of soil metals under all three exposure pathways was considered. However, the exposure risk of PM2.5-bound heavy metals only considered risk through the predominant inhalation exposure pathway. The evaluation model of health risk for possible receptors (children and adults) in this study was established by referring to the Dutch National Institute of Public Health Agency (Van den Berg 1994), U.S. Environmental Protection Agency (USEPA 1996, 2002), and Technical Guidelines for the Risk Assessment of Contaminated Site (HJ 25.3–2014) (MEPPRC 2014), and the calculation of three different exposure doses is shown in Equations (2)–(7).

$$\text{ADD}_{\text{ing}} = \frac{\text{CS} \times \text{IR} \times \text{CF} \times \text{FI} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \tag{2}$$

$$\text{ADD}_{\text{derm}} = \frac{\text{CS} \times \text{AF} \times \text{CF} \times \text{SA} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \tag{3}$$

$$\text{ADD}_{\text{inh}} = \frac{\text{CS} \times \text{PM10} \times \text{DAIR} \times \text{PLAF} \times \text{FSPO} \times \text{CF} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}}$$
(Soil)
$$\tag{4}$$

$$\text{ADD}_{\text{inh}} = \frac{\text{CS} \times \text{DAIR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (\text{PM2.5}) \tag{5}$$

where ADD_{ing}, ADD_{inh}, and ADD_{derm} are the average exposure doses per day via the three pathways (mg/kg·day). CS is equal to the content of studied metal in soil/sediment (mg/kg), or PM2.5 (ng/m^3). The values of the other parameters are shown in Table 1, which were obtained from the references (Van den Berg 1994; USEPA 1996, 2002; MEPPRC 2014), while the local values of SA, EF, ED, and AT of MEPPRC were applied preferentially.

Then, the noncarcinogenic risks of heavy metals in soil and PM2.5 were calculated as Eqs. (6) and (7), respectively.

$$\begin{aligned} \text{HI} &= \text{HQ}_{\text{ing}} + \text{HQ}_{\text{derm}} + \text{HQ}_{\text{inh}} \\ &= \left(\frac{\text{ADD}_{\text{ing}}}{\text{RfD}_{\text{ing}}}\right) + \left(\frac{\text{ADD}_{\text{derm}}}{\text{RfD}_{\text{derm}}}\right) + \left(\frac{\text{ADD}_{\text{inh}}}{\text{RfD}_{\text{inh}}}\right) \end{aligned} \tag{6}$$

$$\text{HQ}_{\text{inh}} = \frac{\text{ADD}_{\text{inh}}}{\text{RfD}_{\text{inh}}} \tag{7}$$

where HQ (hazard quotient) was divided into three categories (HQ_{ing}, HQ_{derm}, and HQ_{inh}), corresponding to the above three pathways. The hazard index, represented by

Table 1 │ The values of the receptor exposure parameters

	Definition	Unit	Values Child	Values Adult
IR	Ingestion rate	mg/day	200	100
CF	Conversion factor	kg/mg	10^{-6}	
FI	Fraction of ingestion	–	1.0	
EF	Exposure frequency	day/a	200	
ED	Exposure duration	a	6	74
BW	Body weight	kg	16	60
AT	Average contact time	day	EF × ED	
AF	Adherence factor	mg/day	0.2	0.07
SA	Exposure skin	cm^2	2,800	5,700
ABS	Dermal absorption factor	–	0.001	
PM10	Content of inhalable particulates in ambient air	mg/m^3	0.3	
DAIR	Daily air inhalation rate	m^3/day	7.6	20
PIAF	Retention fraction of inhaled particulates in body	–	0.75	
FSPO	Fraction of soil-borne particulates in outdoor air	–	0.5	

HI, was equal to the sum of HQ through different pathways for each studied metal. The acceptable risk limit is $HI = 1$, and the greater the HQ, the higher the corresponding risk. RfD (mg/(kg·d)) means the reference dose, and the specific value is shown in Table 2 (MEPPRC 2014). Among them, the RfD_{ing} value was given directly, while RfD_{derm} and RfD_{inh} were calculated via the following equation (MEPPRC 2014):

$$RfD_{inh} = \frac{RfC \times DAIR_{adult}}{BW_{adult}} \tag{8}$$

where the RfD_{ing}, RfD_{derm}, and RfD_{inh} mean the reference doses via three exposure routes, mg/(kg·d); ABS_{gi} is the absorption efficiency factor of the digestive tract, unitless. RfC means reference concentrations through inhalation, mg/m^3. In the HJ 25.3–2014 (MEPPRC 2014), $DAIR_{adult}$ and BW_{adult} were equal to 14.5 m^3/day and 56.9 kg, respectively.

Potential ecological risk index

To further study the ecological risk of the water surface sediment metals, the potential ecological risk index was developed (Chen *et al.* 2019). The specific calculation method is shown in the following formulas (Hakanson 1980):

$$E_r^i = T_r^i \times C_f^i = T_r^i \times C^i / C_i \tag{9}$$

$$RI = \sum_{i=1}^{n} E_r^i \tag{10}$$

where the potential risk for the studied metal is represented by E_r^i, and RI is equal to the sum of it. The values of the toxic-response factor (T_r^i) recommended by Hanson for metals are 5 (Cu), 2 (Cr), 30 (Cd), 1 (Zn), 5 (Pb), 40 (Hg), 10 (As), and 5 (Ni) (Xu *et al.* 2008). C_f^i means contamination factor. C^i is equal to metal content, and C_i is the corresponding value for reference, which is represented by the soil background values (CNEMC 1990). The different ecological risk levels are shown in Table 3.

RESULTS AND DISCUSSION

Environmental multi-media contents and spatial distributions of metals

Heavy metals in soil

The extracted content data of soil heavy metals in the YREB are shown in Supplementary Table S4. The mean contents

Table 2 | The specific value of RfD for the exposure pathway (mg/(kg·d))

RfD	RfD_{ing}	RfD_{derm}	RfC	RfD_{inh}
Cu	4.00×10^{-2}	1.20×10^{-2}	–	–
Cr	3.00×10^{-3}	6.00×10^{-5}	1.14×10^{-4}	2.90×10^{-5}
Cd	1.00×10^{-3}	1.00×10^{-5}	1.00×10^{-5}	2.55×10^{-6}
Zn	3.00×10^{-1}	6.00×10^{-2}	–	–
Pb	3.50×10^{-3}	5.25×10^{-4}	1.38×10^{-2}	3.52×10^{-3}
Hg	3.00×10^{-4}	2.10×10^{-5}	3.00×10^{-4}	7.66×10^{-5}
As	3.00×10^{-4}	1.23×10^{-4}	1.50×10^{-5}	3.83×10^{-6}
Ni	2.00×10^{-2}	5.40×10^{-3}	9.00×10^{-5}	2.30×10^{-5}

Table 3 | Ecological risk level criterion

Level	E_r^i value	Extent of ecological risk of single metals	RI	Extent of ecological risk of all metals
I	$E_r^i < 40$	Low potential ecological risk	RI < 150	Low potential ecological risk
II	$40 \leq E_r^i < 80$	Moderate ecological risk	$150 \leq RI < 300$	Moderate ecological risk
III	$80 \leq E_r^i < 160$	Considerable ecological risk	$300 \leq RI < 600$	High ecological risk
IV	$160 \leq E_r^i < 320$	High ecological risk	RI ≥ 600	Very high ecological risk
V	$E_r^i \geq 320$	Very high ecological risk	–	–

of each soil metal were the highest as follows: Cu (126.33 mg/kg), Ni (426.43 mg/kg), and Hg (0.83 mg/kg) in Anhui Province; Cr (97.45 mg/kg) and Pb (115.89 mg/kg) in Jiangxi Province; Cd (4.54 mg/kg), As (36.34 mg/kg), and Zn (241.62 mg/kg) in Hunan Province. Compared with the soil background contents, the contents of Cu, Ni, Hg, Pb, Zn, and Cd exceeded the background values in all provinces, indicating that soils in the YREB have suffered different degrees of such heavy metal pollution over the years. In addition, Anhui Province (Cu, Hg, and Ni), Jiangxi Province (Cr and Pb), and Hunan Province (Cd, Zn, and As) have two or more heavy metal contents reaching the highest value among all provinces, indicating that there was a relatively high heavy metal pollution.

Based on Supplementary Table S4, the provincial distributions of the eight soil metals from the YREB were obtained by using ArcGIS 10.0 and are shown in Figure 1. The high-value regions of Cu, Cr, Cd, Zn, and Pb were mainly distributed in the central YREB, specifically in the Hubei, Hunan, Anhui, and Jiangxi provinces. Metal
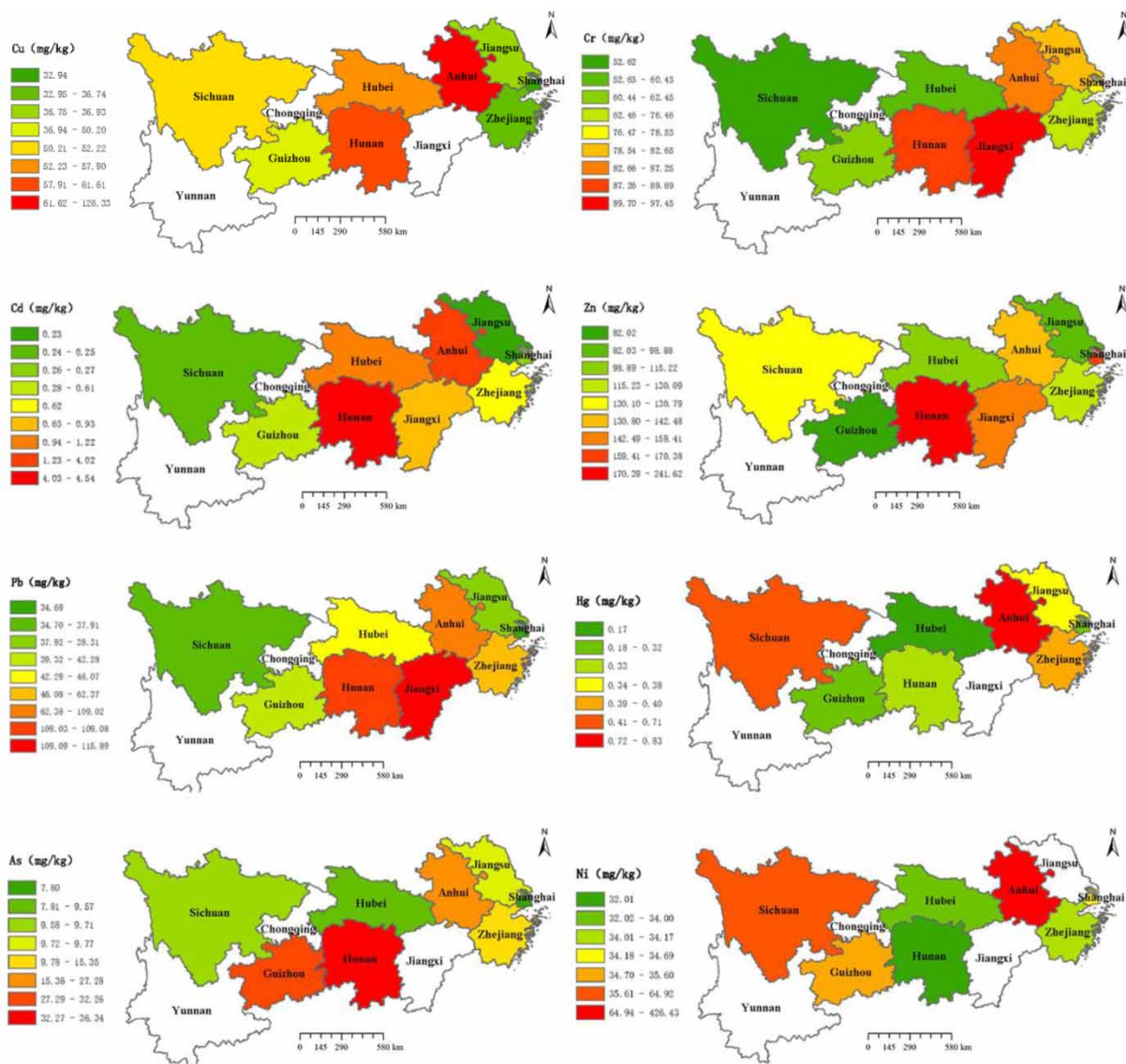

Figure 1 | The provincial distributions of the soil metals in the YREB.

contents were relatively low in the eastern and western parts of the YREB. To sum up, the distribution of metals basically tended to be high in the middle and low at both ends. However, the high-value regions of Hg, As, and Ni were scattered, specifically Hg in the Sichuan, Anhui, and Zhejiang provinces (As in the Guizhou, Hunan, and Anhui Provinces; Ni in the Sichuan and Anhui provinces). The results showed that, except for Hg and Ni, most of the heavy metal contents in the central part of the YREB were generally higher than those at both ends. For Hg and Ni, their provincial spatial distributions were similar, which were high at both ends and low at the middle.

To further quantitatively identify the soil metal enrichment level in the YREB, the geo-accumulation index was utilized, and the evaluation results are displayed in Table 4. Table 4 indicates that the general enrichment levels of Cu, Cr, Zn, Pb, and As in soil from the provinces were the moderately contaminated level or below it. Among them, the enrichment levels of Cr were the uncontaminated to moderately contaminated level or below it. The enrichment levels of Cd in Anhui Province ($I_{geo} =$ 4.79) and Hunan Province ($I_{geo} = 4.96$) fell into the category of Class VI, which was heavily to extremely contaminated, and these levels were the highest among all provinces. For Cd and Ni, the enrichment levels in Anhui Province belonged to Class V, which was heavily contaminated. To sum up, among all studied metals, the overall enrichment levels of Cd and Hg were higher than others, which deserved priority control. From a provincial perspective, the overall concentrations of studied metals

Table 4 │ Geo-accumulation index for soil metals in the YREB

Province	Cu	Cr	Cd	Zn	Pb	Hg	As	Ni
Anhui	2	0	5	1	2	4	1	4
Guizhou	1	0	3	0	1	2	1	0
Hubei	2	0	4	1	1	1	0	0
Hunan	1	0	5	2	2	2	2	0
Jiangsu	1	0	1	0	1	2	0	–
Jiangxi		1	3	1	2	–	–	–
Shanghai	0	0	1	1	0	2	0	0
Sichuan	1	0	1	1	0	3	0	1
Zhejiang	1	0	3	1	1	3	0	0

in the Anhui, Hubei, and Hunan provinces were higher than those of the others, specifically Cd, Hg, and Ni in Anhui Province, and Cd in the Hunan and Hubei provinces.

PM2.5-bound heavy metals

The extracted concentration data of PM2.5 heavy metals in the YREB are shown in Supplementary Table S5. The mean concentrations of each metal in PM2.5 were the highest as follows: Cu (353.82 ng/m^3), Cr (47.26 ng/m^3), and Hg (1.48 ng/m^3) in Hunan Province, Ni (51.20 ng/m^3), Zn (1,141.10 ng/m^3), Pb (468.70 ng/m^3), and Cd (21.50 ng/m^3) in Jiangxi Province, and As (105.00 ng/m^3) in Yunnan Province. Among them, the Jiangxi and Hunan provinces had four and three kinds of heavy metals with the highest average contents, indicating the serious nature of enrichment in these two provinces. To further assess the enrichment characteristics of heavy metals, the National Ambient Air Quality (GB3095-2012), the Air Quality Guidelines for Europe, and the Ambient Air Quality and Cleaner Air for Europe were used to compare the average contents of PM2.5-bound heavy metals. The results revealed that the average content of Pb in the Yunnan and Jiangxi provinces and that of Cd in Hubei, Hunan, Jiangxi, and Yunnan exceeded the limits. The average content of Ni in the Hunan, Jiangxi, and Yunnan provinces exceeded the Air Quality Guidelines for Europe. Particularly, the average concentrations of As and Cr were higher than their limits in all the studied provinces. In short, the Jiangxi, Yunnan, and Hunan provinces had a higher enrichment degree of PM2.5 heavy metals than the others.

According to the provincial distributions of PM2.5-bound metals in the YREB, as shown in Figure 2, the high-value regions of Cu, Cr, and Cd were generally distributed in the five provinces of Sichuan, Yunnan, Guizhou, Hunan, and Jiangxi. The content of Cr in Jiangsu Province was also relatively high. Zn, Pb, and As were distributed primarily in the eastern and southern parts of the YREB, specifically the Sichuan, Yunnan, Huanan, and Jiangxi provinces. The high-value regions of Ni are mostly concentrated in the southern part of the YREB. Similar to the soil, PM2.5-bound Hg contents were relatively high at both ends and low at the middle.
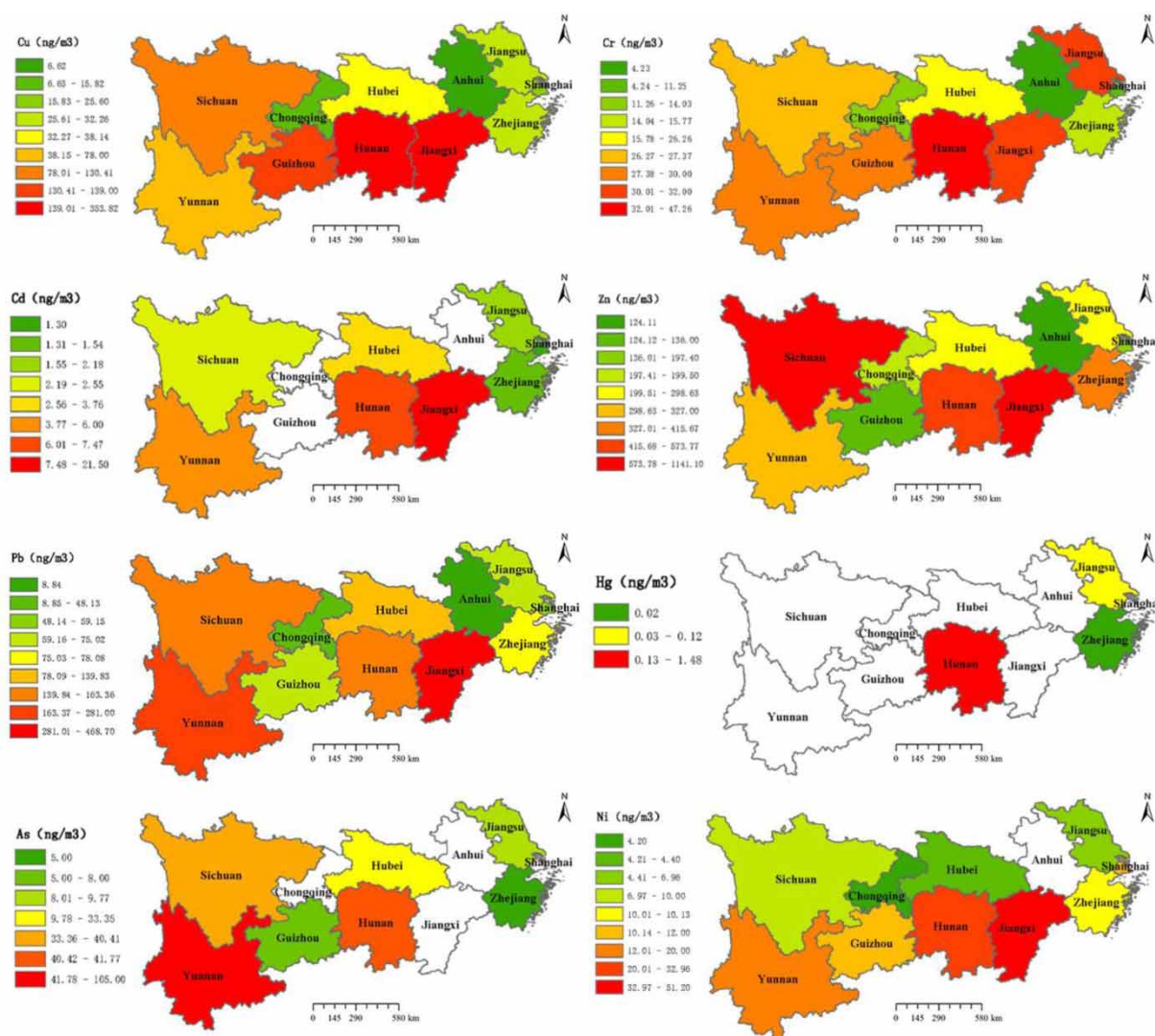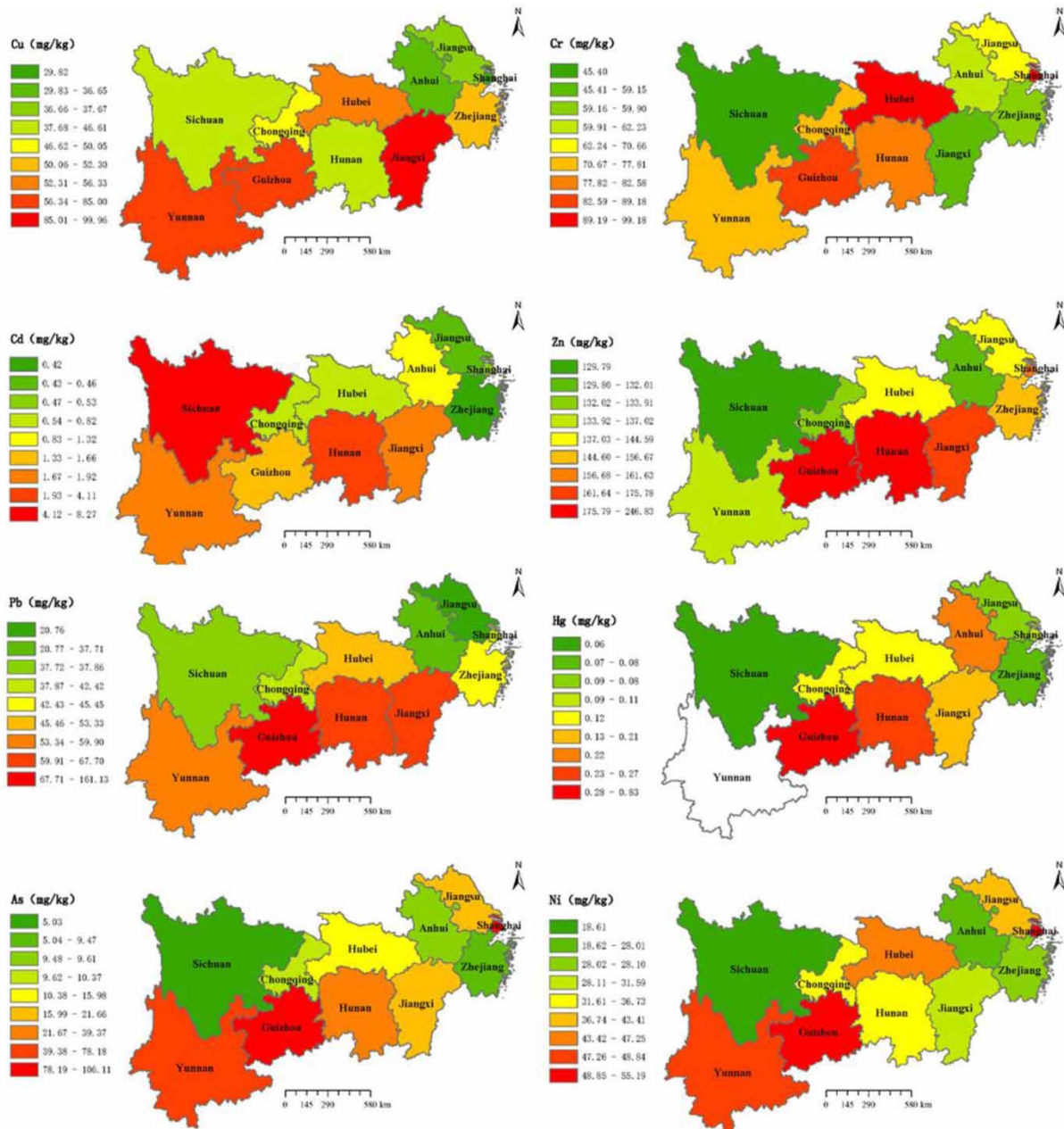
Figure 2 │ The provincial distributions of the PM2.5-bound metals in the YREB.

Heavy metals in the surface sediment

The extracted content data of the metals in the surface sediment of the YREB are shown in Supplementary Table S6. The average concentrations of each heavy metal were the highest as follows: Pb (161.13 mg/kg), Zn (246.83 mg/kg), Ni (55.19 mg/kg), and Hg (0.83 mg/kg) in Guizhou Province, Cr (99.18 mg/kg) and As (106.11 mg/kg) in Shanghai, Cu (99.96 mg/kg) in Jiangxi Province, and Cd (8.27 mg/kg) in Sichuan Province. Compared with the soil background values, Cu, Zn, Cd, Pb, Hg, and Ni were enriched in the water surface sediment of the YREB in most of the studied provinces. Moreover, the metal contents in the sediment in Guizhou, Hubei, Hunan, Shanghai, and Chongqing all exceeded the background values, among which the contents of five metals (Zn, Pb, Hg, As, and Ni) from Guizhou Province reached the highest levels, which warrants more attention.

As drawn in Figure 3, the provincial distributions of surface sediment metals in the YREB showed that the high-value regions of Pb, Cd, and As were mostly concentrated in the southern parts (Yunnan, Guizhou, and Hunan) of the YREB, while Cr, Hg, and Zn were primarily distributed in the middle YREB. For Cu and Ni, they

Figure 3 | The provincial distributions of the surface sediment metals in the YREB.

were generally enriched in the Yunnan, Guizhou, and Hubei provinces. In short, the heavy metals in the water surface sediment have a higher content in the middle YREB.

According to the geo-accumulation index, the enrichment levels of heavy metals in the water surface sediment are displayed in Table 5. The enrichment

levels of Cu and Zn were all Class III or below, while Cr and Ni were all in the uncontaminated to moderately contaminated level or in the uncontaminated level. Furthermore, the enrichment level of Cd in the Anhui, Guizhou, Hunan, Jiangxi, Sichuan, and Yunnan provinces was all in the heavily contaminated level or above, especially in the Sichuan (extremely contaminated) and

Table 5 | Geo-accumulation index for water surface sediment metals

Province	Cu	Pb	Zn	Cr	Ni	Cd	Hg	As
Anhui	1	0	1	0	0	4	2	0
Guizhou	2	3	2	0	1	4	4	3
Hubei	1	1	1	1	1	3	1	0
Jiangsu	1	0	1	0	1	2	0	1
Hunan	1	1	1	0	0	5	2	2
Zhejiang	1	1	1	0	0	2	0	0
Jiangxi	2	1	1	0	0	4	2	1
Sichuan	1	0	1	0	0	6	0	0
Shanghai	0	1	1	1	1	2	1	3
Chongqing	1	1	1	0	0	3	1	0
Yunnan	2	1	1	0	1	4		3

Hunan provinces (heavily to extremely contaminated). Additionally, the enrichment level of Hg in Guizhou Province fell into Class V, which was heavily contaminated. From a provincial perspective, the overall enrichment levels of the surface sediment metals in the Guizhou, Hunan, and Yunnan provinces were relatively high. To sum up, Cd and Hg generally had higher enrichment levels than the other metals.

Environmental risk assessment

Health risk assessment for the metals in soil and PM2.5

The average daily intake through the three pathways of soil and from the inhalation of PM2.5 in the YREB is shown in Supplementary Tables S7 and S8. The results showed that the daily intake and the noncarcinogenic risk of all metals for child receptors were greater than those for adults, which were consistent with the findings of the previous studies (Zhao *et al.* 2017; Aendo *et al.* 2019). Also, the results proved that children were more sensitive receptors than adults due to their exposure characteristics. The average daily intake of soil metals decreased as ingestion > inhalation > dermal contact, with ingestion having the maximal exposure risk. The noncarcinogenic risk of soil heavy metals is shown in Table 6. The HIs of the studied soil metals for adults in the studied provinces were lower than 1, indicating that the adults in the YREB faced the corresponding little noncarcinogenic risk. However, the HIs of As for children in the Guizhou, Hunan, and Anhui provinces, and those of Ni in Anhui Province all exceeded 1, which showed that children faced the noncarcinogenic risk of As and Ni in soil from the above provinces. For

Table 6 | Noncarcinogenic risk of soil heavy metals in the YREB

HI	Receptor	Shanghai	Zhejiang	Guizhou	Hubei	Anhui	Jiangsu	Sichuan	Hunan	Jiangxi
Cu	Adult	1.39×10^{-3}	1.55×10^{-3}	2.12×10^{-3}	2.44×10^{-3}	5.33×10^{-3}	1.56×10^{-3}	2.20×10^{-3}	2.60×10^{-3}	–
	Child	1.04×10^{-2}	1.16×10^{-2}	1.58×10^{-2}	1.83×10^{-2}	3.98×10^{-2}	1.16×10^{-2}	1.65×10^{-2}	1.94×10^{-2}	–
Cr	Adult	1.54×10^{-1}	1.50×10^{-1}	1.22×10^{-1}	1.18×10^{-1}	1.71×10^{-1}	1.62×10^{-1}	1.03×10^{-1}	1.76×10^{-1}	1.91×10^{-1}
	Child	5.18×10^{-1}	5.04×10^{-1}	4.12×10^{-1}	3.98×10^{-1}	5.75×10^{-1}	5.45×10^{-1}	3.47×10^{-1}	5.91×10^{-1}	6.42×10^{-1}
Cd	Adult	4.51×10^{-3}	1.06×10^{-2}	1.03×10^{-2}	2.07×10^{-2}	6.84×10^{-2}	3.91×10^{-3}	4.30×10^{-3}	7.73×10^{-2}	1.58×10^{-2}
	Child	9.79×10^{-3}	2.30×10^{-2}	2.24×10^{-2}	4.49×10^{-2}	1.48×10^{-1}	8.49×10^{-3}	9.33×10^{-3}	1.68×10^{-1}	3.43×10^{-2}
Zn	Adult	9.65×10^{-4}	7.37×10^{-4}	4.65×10^{-4}	6.53×10^{-4}	8.07×10^{-4}	5.60×10^{-4}	7.41×10^{-4}	1.37×10^{-3}	9.03×10^{-4}
	Child	7.20×10^{-3}	5.50×10^{-3}	3.47×10^{-3}	4.87×10^{-3}	6.02×10^{-3}	4.18×10^{-3}	5.53×10^{-3}	1.02×10^{-2}	6.73×10^{-3}
Pb	Adult	1.73×10^{-2}	3.12×10^{-2}	2.11×10^{-2}	2.30×10^{-2}	5.45×10^{-2}	1.96×10^{-2}	1.89×10^{-2}	5.45×10^{-2}	5.79×10^{-2}
	Child	1.27×10^{-1}	2.28×10^{-1}	1.54×10^{-1}	1.68×10^{-1}	3.98×10^{-1}	1.44×10^{-1}	1.39×10^{-1}	3.99×10^{-1}	4.23×10^{-1}
Hg	Adult	2.00×10^{-3}	2.52×10^{-3}	2.00×10^{-3}	1.05×10^{-3}	5.28×10^{-3}	2.42×10^{-3}	4.49×10^{-3}	2.10×10^{-3}	–
	Child	1.39×10^{-2}	1.74×10^{-2}	1.39×10^{-2}	7.27×10^{-3}	3.65×10^{-2}	1.67×10^{-2}	3.10×10^{-2}	1.45×10^{-2}	–
As	Adult	1.20×10^{-1}	2.36×10^{-1}	4.97×10^{-1}	1.47×10^{-1}	4.20×10^{-1}	1.50×10^{-1}	1.50×10^{-1}	5.60×10^{-1}	–
	Child	4.36×10^{-1}	8.58×10^{-1}	1.80×10	5.35×10^{-1}	1.52×10	5.46×10^{-1}	5.43×10^{-1}	2.03×10	–
Ni	Adult	5.96×10^{-2}	5.87×10^{-2}	6.11×10^{-2}	5.84×10^{-2}	7.32×10^{-1}	–	1.11×10^{-1}	5.49×10^{-2}	–
	Child	1.03×10^{-1}	1.01×10^{-1}	1.05×10^{-1}	1.01×10^{-1}	1.26×10	–	1.92×10^{-1}	9.46×10^{-2}	–

children and adults, the HIs of each soil heavy metal ranked as follows: As > Cr > Ni > Pb > Cd > Hg > Cu > Zn. The noncarcinogenic risk of As and Cr was higher for the possible receptors than the other metals, while the noncarcinogenic risk of Ni was generally low in the provinces.

The noncarcinogenic risk of PM2.5-bound heavy metals (Table 7) illustrated that the HQs of Cr, Pb, and Hg via the inhalation pathway were lower than 1. Among them, the HQs of Pb and Hg were much lower than 1, showing the acceptable noncarcinogenic risk. For adults, Cr in Jiangxi Province (2.81) and As in the provinces of Hubei (2.90), Sichuan (3.52), Yunnan (9.14), and Hunan (3.64) caused the obvious noncarcinogenic risk to possible receptors. For children, the noncarcinogenic hazard quotients (HQs) of Cd in Jiangxi, Yunnan, and Hunan, Ni in Jiangxi, and As in Hubei, Sichuan, Yunnan, Hunan, and Jiangsu were all higher than 1, implying that children faced a noncarcinogenic health risk in these provinces. Similar to soil metals, the HQs of each metal of PM2.5 for children and adults

ranked as follows: As > Cd > Cr > Ni > Pb > Hg. People faced the noncarcinogenic risk of As in most of the studied provinces, and the overall level of the noncarcinogenic risk of Cr was relatively high but did not exceed the limits, which was similar to soil metals. Differences between the metal risk in soil and PM2.5 were that the noncarcinogenic risk of Cd in PM2.5 was generally high, with the noncarcinogenic risk existing in some provinces.

Potential ecological risk assessment

As shown in Table 8, the results of potential ecological risk of the metals in the water surface sediment were calculated. The average potential ecological risk for each metal was decreased in the following order: Cd (603.32) > Hg (120.45) > As (33.69) > Cu (12.37) > Pb (11.03) > Ni (7.19) > Cr (2.45) > Zn (2.17). Generally, among the studied metals, Cd was in Level V (very high ecological risk), Hg belonged to Level IV (high ecological risk), and the others were in Level I (low potential ecological risk). The

Table 7 | The noncarcinogenic hazard quotient of PM2.5-bound metals in the YREB

HQ	Receptor	Anhui	Chongqing	Jiangxi	Shanghai	Guizhou	Hubei	Sichuan	Yunnan	Hunan	Jiangsu	Zhejiang
Cr	Adult	4.86×10^{-2}	1.61×10^{-1}	3.68×10^{-1}	1.29×10^{-1}	3.45×10^{-1}	3.02×10^{-1}	3.15×10^{-1}	3.45×10^{-1}	5.43×10^{-1}	3.66×10^{-1}	1.81×10^{-1}
	Child	6.93×10^{-2}	2.30×10^{-1}	5.24×10^{-1}	1.84×10^{-1}	4.91×10^{-1}	4.30×10^{-1}	4.48×10^{-1}	4.91×10^{-1}	7.74×10^{-1}	5.21×10^{-1}	2.58×10^{-1}
Cd	Adult	–	–	2.81×10	1.70×10^{-1}	–	4.92×10^{-1}	3.34×10^{-1}	7.83×10^{-1}	9.75×10^{-1}	2.85×10^{-1}	2.00×10^{-1}
	Child	–	–	4.00×10	2.42×10^{-1}	–	7.01×10^{-1}	4.75×10^{-1}	1.12×10	1.39×10	4.06×10^{-1}	2.86×10^{-1}
Pb	Adult	8.37×10^{-4}	4.56×10^{-3}	4.44×10^{-2}	5.60×10^{-3}	7.01×10^{-3}	1.32×10^{-2}	1.41×10^{-2}	2.66×10^{-2}	1.55×10^{-2}	7.10×10^{-3}	7.39×10^{-3}
	Child	1.19×10^{-3}	6.50×10^{-3}	6.32×10^{-2}	7.98×10^{-3}	9.99×10^{-3}	1.89×10^{-2}	2.01×10^{-2}	3.79×10^{-2}	2.20×10^{-2}	1.01×10^{-2}	1.05×10^{-2}
Hg	Adult	–	–	–	–	–	–	–	–	6.44×10^{-3}	5.22×10^{-4}	8.70×10^{-5}
	Child	–	–	–	–	–	–	–	–	9.18×10^{-3}	7.44×10^{-4}	1.24×10^{-4}
As	Adult	–	–	–	–	6.96×10^{-1}	2.90×10	3.52×10	9.14×10	3.64×10	8.51×10^{-1}	4.35×10^{-1}
	Child	–	–	–	–	9.92×10^{-1}	4.14×10	5.01×10	$1.30E+01$	5.18×10	1.21×10	6.20×10^{-1}
Ni	Adult	–	6.09×10^{-2}	7.43×10^{-1}	1.65×10^{-1}	1.74×10^{-1}	6.39×10^{-2}	1.45×10^{-1}	2.90×10^{-1}	4.78×10^{-1}	1.01×10^{-1}	1.47×10^{-1}
	Child	–	8.68×10^{-2}	1.06×10	2.36×10^{-1}	2.48×10^{-1}	9.11×10^{-2}	2.07×10^{-1}	4.13×10^{-1}	6.81×10^{-1}	1.44×10^{-1}	2.09×10^{-1}

Table 8 | The potential ecological risk for each metal in the water surface sediment

Province	Cu	Pb	Zn	Cr	Ni	Cd	Hg	As
Anhui	8.11	7.25	1.78	2.04	5.21	396.25	125.14	8.58
Guizhou	16.71	30.99	3.33	2.92	10.26	498.50	476.57	88.97
Hubei	12.46	10.26	1.95	3.20	8.78	245.93	68.57	14.27
Jiangsu	8.33	3.99	1.94	2.32	8.07	136.67	47.35	17.58
Hunan	9.96	12.33	2.86	2.71	6.83	1,234.25	156.91	35.15
Zhejiang	11.57	8.74	2.11	1.96	5.22	124.80	44.19	8.45
Jiangxi	22.11	13.02	2.37	1.94	5.87	577.00	120.00	19.34
Sichuan	10.31	7.28	1.75	1.49	3.46	2,481.00	34.29	4.49
Shanghai	6.60	7.80	2.18	3.25	9.76	158.00	62.86	94.74
Chongqing	11.07	8.16	1.80	2.51	6.56	232.44	68.57	9.26
Yunan	18.81	11.52	1.85	2.55	9.08	551.70	–	69.80
Minimum	6.60	3.99	1.75	1.49	3.46	124.80	34.29	4.49
Mean	12.37	11.03	2.17	2.45	7.19	603.32	120.45	33.69
Maximum	22.11	30.99	3.33	3.25	10.26	2,481.00	476.57	94.74

ecological risk of Cd and Hg was much higher than that of others. From a provincial perspective, except for Cd, the risk of each sediment heavy metal in Sichuan Province was generally low, with the potential ecological risk of Zn, Cr, Ni, As, and Hg being the lowest in Sichuan Province. Additionally, five kinds of heavy metals (Zn, Pb, Hg, As, and Ni) in Guizhou Province had the highest risk values. The ecological risk of Cd and Hg in Hunan Province was also relatively high, which warrants attention.

Risk management and control measures

With a comprehensive analysis of the above evaluation results of the heavy metal pollution in soil, PM2.5 and water surface sediment, some pertinent countermeasures and suggestions for risk management of environmental multi-media of the Yangtze Economic Belt were put forward as follows:

(1) Priority metals should be given priority control. The preliminary heavy metal pollution assessment results revealed that the pollution levels of Cd and Hg in the soil and water surface sediment were higher than those of the other metals, and the enrichment degree of As, Cr, and Cd in PM2.5 was a little bit heavier

than that of other metals. Moreover, the environmental risk assessment results concluded that the noncarcinogenic risk contributions of As accounted for the highest proportion in both soil and PM2.5. In the surface sediment, the average potential ecological risk (Table 8) for Cd and Hg reached Level V (very high ecological risk) and Level IV (high ecological risk), respectively. To sum up, Cd, Hg, and As were identified as the priority control metals in environmental multi-media of the YREB, which deserved special attention to reduce the relevant human health risk as much as possible.

(2) A joint source control mechanism of environmental multi-media heavy metal pollution in the central and southern parts of the YREB is recommended. The previous results indicated that heavy metal pollution in soil, PM2.5, and surface sediment is distributed mainly in the central and southern parts of the YREB, specifically in the Hunan, Guizhou, Hubei, and Jiangxi provinces. Through the literature review, as pollution sources, Cd and Hg mainly emanated from industrial emissions, especially nonferrous metal smelters (Lv et al. 2016), and As and Hg generally originated from coal combustion (Li et al. 2015; Ma et al. 2020). Hence, extensive industrial activities, nonferrous metal smelters, and coal combustion were probably the main

reasons for the obvious heavy metal enrichment in the three environmental media in these provinces. As we know, Hunan Province was famously known as the 'Hometown of Nonferrous Metals' and Guizhou Province was known as the 'Mercury Capital', due to its rich mercury resources. Additionally, extensive mining activities mainly contributed to heavy metal pollution in Jiangxi Province (Zhang *et al.* 2020). Also, developed heavy industries in Hubei Province have posed a relatively high health risk for possible receptors (Zhang *et al.* 2019). Therefore, due to the regional characteristics of heavy metal pollution (Li *et al.* 2020), effective communication and cooperation among each related city are of importance. So, it is recommended for the above provinces to build a joint source control mechanism of heavy metal to enhance risk management and control.

(3) The relevant education and publicity on environmental health need to be given enough attention, especially for children. The results of health risk assessment for the metals in soil and PM2.5 concluded that child receptors were more likely to be exposed to relatively high doses than adult receptors. So, it is highly important to pay sufficient attention to children and provide relevant environmental health knowledge to them. For this purpose, it is recommended to provide the relevant compulsory courses in kindergartens or primary schools.

CONCLUSIONS

In accordance with the growing green and sustainable development requirements in the YREB, an overall evaluation on the distribution, enrichment degree, and environmental risk of provincial heavy metal pollution in the three environmental media (soil, PM2.5, and water surface sediment) is of significance for both national and local decision-makers. The pollution assessment showed that heavy metal pollution, mainly Cd and Hg, was enriched in the Hunan, Jiangxi, and Anhui provinces. Spatially, the pollution in the three environmental media showed the obvious regional characteristics and is distributed mainly in the central and

southern parts of the YREB. Environmental risk assessment showed that children were more likely to be exposed to greater risk than adults, and the average daily intake of soil metals was the highest by ingestion. Moreover, As contributed the highest noncarcinogenic risk in soil and PM2.5. The potential ecological risk assessment concluded that the risk levels of the two metals, Cd (Level V) and Hg (Level IV), in the water surface sediment were the highest among all metals, and that the Guizhou and Hunan provinces had much higher ecological risk than others. Finally, three recommended risk management countermeasures for heavy metal pollution in the YREB were: (i) to give priority control to the priority metals (Cd, Hg, and As); (ii) to build a joint source control mechanism of heavy metal pollution in the Hunan, Guizhou, Hubei, and Jiangxi provinces; and (iii) to accord more importance to provide relevant education and publicity on environmental health to people, especially children.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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