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Comprehensive ecological risk assessment for heavy metal pollutions in three phases in rivers

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Abstract: Literature lacked in providing a comprehensive research on heavy metal detection in aquatic, biological and sedimentary states of rivers. The present study was imparted with all these three components of the river. Heavy metal toxicity or pollution index was used as a tool for ecological risk assessment by considering the single state studies conducted by many researchers. An intensive ecological risk assessment model was constructed and heavy metals were indicated as a serious threat to the environment. The model was applied to determining five toxic heavy metals in three states of the Songhua River. According to the ecological risk index, heavy metal pollution in three phases was categorized as aquatic>biological>sedimentary, while the overall descending order of heavy metal ecological risk index was as Cd>Hg>As>Pb>Cr. Cd and Hg were selected as the priority pollutants of Songhua River.

Key words: comprehensive ecological risk assessment; priority pollutants selection; heavy metal; Songhua River

1 Introduction

An increase in industrial production and urbanization was believed responsible for increasing concentration of heavy metals into the natural environment, not only posing serious threat to individuals and species but also causing a negative effect on the ecological system. Excessive amount of heavy metals resulted in severe impacts on human body, such as acute and chronic intoxication, cancer, teratogenesis, and mutations [1]. Currently, heavy metal contamination got great attention by researchers due to its potential hazard to environmental pollution locally and globally [2]. Different methods were adopted and implemented to calculate an ecological risk assessment of heavy metals in Xiawan Port sediments like potential ecological risk index (RI), risk assessment code (RAC) by modifying an index, modified potential ecological risk index (MRI) [3]. Four different methods, named mineralogical analysis, three-stage BCR sequential extraction procedure, dynamic leaching test and Hakanson potential ecological risk index method were used to evaluate the zinc residual leaching and its potential ecological risks on environment [4]. Ten fish species were collected from Bangshi River at Savar in Bangladesh in two different seasons to measure eight heavy metals (Pb, Cd, Ni, Cr, Cu, Zn, Mn, and As) [5]. The water quality index was implemented for heavy metals risk assessment and water quality characterization of River Soan, Pakistan [6]. However, most of the researchers detected heavy metals in single medium only. No intensive research on heavy metal pollution in all aquatic, biological and sedimentary phases of the river was reported. Although many studies investigated screening methods for priority pollutants in aqueous environments, most of them focused on heavy metal contamination of organic matters causing great threat to human body [7]. This was a problem to formulate a scientific evaluation model for determination of heavy metals as priority pollutants for intensive research on target rivers due to the demand of high standard heavy metal detection devices and operators. By considering heavy metal pollution assessment models [8−10] and priority pollutant screening methods [11−13] adopted and observed in different researches, the present study opted relevant indexes to construct a comprehensive ecological risk assessment model to detect heavy metals in aqueous, biological and

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sedimentary phases. Heavy metals with higher ecological risks were selected as the priority pollutants to control heavy metal pollution in rivers.

2 Migration and transmission of heavy metals among three phases in rivers

Heavy metals after entering in any environmental component were easily transmitted and accumulated in food chains [14], same in the case of water bodies, heavy metals eventually entered in human bodies by edible marine food like fish [15]. This process not only poisoned the fish, but also caused the alarming situation towards humans. Moreover, heavy metals in water bodies showed a tendency to react with organic polymers, forming a complex or chelate by adhering to surface of clay minerals. These pollutants were ultimately settled and accumulated in sediments and acted as secondary pollutants in water bodies by undergoing a series of physical, chemical and biological processes [16]. The migration and transmission process of heavy metals in river ecosystem is shown in Fig. 1.

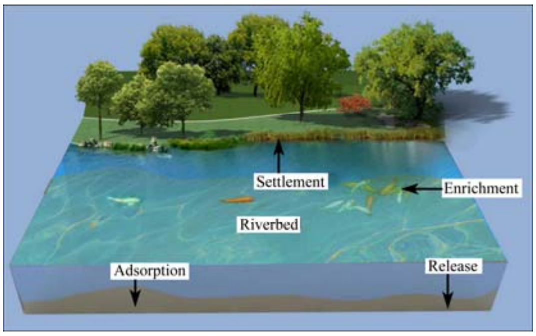

Fig. 1 Simulation of migration and transmission process of heavy metals in river ecosystem

3 Construction of risk evaluation model for heavy metals in three phases in rivers

3.1 Construction of index system

Table 1 shows the major assessment models for heavy metal pollutions locally and globally. Heavy metals pollution in rivers were mainly influenced by biological toxicity, absorption and contamination extent. By keeping in view the model construction concepts in literature, in the current study, toxicity coefficient, pollution index, and detection rate were selected as the key factors to construct the risk assessment model to detect heavy metals in rivers.

3.1.1 Toxicity coefficient of heavy metals (T_r^i)

Toxicity coefficient T_r^i represented the toxicity level and the biological sensitivity for heavy metals. Toxicity coefficients commonly used are shown in Table 2.

3.1.2 Pollution index of heavy metals (C_f^i)

Pollution index C_f^i represented the richness and pollution degree of a single heavy metal, which was denoted in Eq. (1):

$$C_f^i = C^i / C_n^i \tag{1}$$

where C^i is the detected value of a single heavy metal (mg/kg); C_n^i is the background value (mg/kg).

3.1.3 Detection rate of heavy metals (F_s^i)

Detection rate F_s^i symbolized the pollution scale and

Table 1 Main models of heavy metal pollution assessment

Model name	Formula	Indication
Index of geo-accumulation [8]	$I_{geo} = \log_2 \frac{C_n}{kB_n}$	C_n represents the content of element n in sediments; B_n refers to the background value of n in sedimentary rock (regular rock).
Pollution load index [17]	$CF_i = \frac{C_i}{C_{oi}}$ $PLI = \sqrt[n]{CF_1 \times CF_2 \times \cdots \times CF_n}$ $PLI_{zone} = \sqrt[n]{PLI_1 \times PLI_2 \times \cdots \times PLI_n}$	C_i is the detected content of element i; C_{oi} is the background value of i; PLI is the pollution bearing coefficient of a certain point; PLI_{zone} is the pollution bearing coefficient of a certain region.
Potential ecological risk index [9]	$RI = \sum_i^m T_r^i \times \frac{C_d^i}{C_r^i}$	C_d^i is the detected concentration of pollutants in sediments; C_r^i is the background value of pollutants in sediments; T_r^i refers to the toxicity coefficient of single factor pollutant.
Excess after regression analysis [10]	$ERA(B) = [E_s - (\beta C_{Mg} + \alpha)] / E_D$, $ERA(A) = (E_s - E_D) / E_D$	E_s is the total concentration of heavy metals; E_D is the background concentration; C_{Mg} refers to the Mg concentration; β represents the regression slope between heavy metals and Mg; α is the regression intercept between heavy metals and Mg.
Sediment enrichment factor [18]	$K_{SEF} = (s_E / s_{Al} - a_E / a_{Al}) / (a_E / a_{Al})$	S_E indicates the heavy metal content in sediments; S_{Al} is the Al content in sediments; a_E is the content of heavy metals in unpolluted sediments; a_{Al} is Al content in unpolluted sediments.

Table 2 Toxicity coefficients of different heavy metals

Heavy metal	T_r^i
Hg	40
Cd	30
Cr	2
As	10
Pb	5
Cu	5
Zn	1
Ni	2
Mn	2

detection rate of a single heavy metal, which was shown in Eq. (2):

$$F_s^i = S^i / S_t^i \tag{2}$$

where S^i is the number of sections that detected a single heavy metal; S_t^i indicates the total number of detection sections.

3.2 Weight of indexes

3.2.1 Weight determination

The weight determination method of heavy metals (ζ, η, θ) in the three phases was as follows: 1) Heavy metals weight determination coefficients in aquatic, biological and sedimentary phases were x, y and z, respectively; 2) Dividing water environment according to water qualities into I−V categories, and weight coefficients for these categories were α, β, γ, δ and ε, respectively; 3) Heavy metal weight coefficients x, y and z were multiplied with α, β, γ, δ and ε to obtain the weight coefficient of each heavy metal in every selected phase of water bodies, α_i, β_i, γ_i, δ_i and ε_i; 4) Determination of the number of different water quality categories A, B, C, D and E; 5) Calculation of the mean arithmetic weight of each heavy metal in various water components a, b and c to measure the heavy metal weight in aquatic, biological and sedimentary phases ζ, η, θ, respectively (Eq. (3)). Table 3 shows the heavy metals weights in three phases in rivers.

$$\zeta = \frac{a}{a+b+c};\ \eta = \frac{a}{a+b+c};\ \theta = \frac{a}{a+b+c} \tag{3}$$

where

$$a = \frac{\alpha_1 A + \beta_1 B + \gamma_1 C + \delta_1 D + \varepsilon_1 E}{A+B+C+D+E},$$

$$b = \frac{\alpha_2 A + \beta_2 B + \gamma_2 C + \delta_2 D + \varepsilon_2 E}{A+B+C+D+E},$$

$$c = \frac{\alpha_3 A + \beta_3 B + \gamma_3 C + \delta_3 D + \varepsilon_3 E}{A+B+C+D+E}.$$

3.2.2 Weight value determination

The analytic hierarchy process was adopted to determine the weight coefficient. This section referred to the concentration comparison method for a single heavy metal in different water environments to construct a verification matrix as described in Surface Water Quality Assessment Standard GB3838—2002. Six experts were invited to construct the verification matrix for I−V water quality in aquatic, biological and sedimentary phases. The common conclusion of their researches was adopted to construct the weight coefficient matrix, as shown in Table 4.

3.3 Comprehensive ecological risk index in three-phases in rivers

By considering the evaluation indexes in literature, the ecological risk indexes were established to detect heavy metals in aquatic, biological, and sedimentary phases (represented by W, F and S, Eq. (4)). Heavy metal impact on human was varied depending upon their

Table 3 Weight distribution of heavy metals in three phases in rivers

Three phase	Water quality category										Weight value
	I(α)		II(β)		III(γ)		IV(δ)		V(ε)		
	Weight index	No.	Weight index	No.	Weight index	No.	Weight index	No.	Weight index	No.	
Aquatic (x)	α_1	A	β_1	B	γ_1	C	δ_1	D	ε_1	E	ζ
Biological (y)	α_2	A	β_2	B	γ_2	C	δ_2	D	ε_2	E	η
Sedimentary (z)	α_3	A	β_3	B	γ_3	C	δ_3	D	ε_3	E	θ

Table 4 Weight coefficient of different heavy metals in rivers

Three phase	Weight coefficients in different water quality environments				
	α (0.363)	β (0.276)	γ (0.182)	δ (0.117)	ε (0.062)
Aquatic (0.625)	0.227	0.173	0.114	0.073	0.039
Biological (0.239)	0.087	0.066	0.043	0.028	0.015
Sedimentary (0.137)	0.050	0.038	0.025	0.016	0.008

accumulation in different phases, consequently, different risk weights were assigned to the heavy metals in different phases. Comprehensive ecological risk index R was obtained in Eq. (5) and heavy metals with the higher R values were selected as the priority pollutants in rivers.

$$W(F \text{ or } S) = T_{\mathrm{r}}^{i} \times C_{\mathrm{f}}^{i} \times F_{\mathrm{s}}^{i} \tag{4}$$

$$R = \zeta W + \eta F + \theta S \tag{5}$$

4 Model application and verification

4.1 Model application

4.1.1 Research region selection and data collection

The model was applied to the third largest river in China named Songhua River. Songhua River was connected to Nen River on the north and the Second Songhua River on the south. The two branches were merged into the mainstream of Songhua River at Sancha River and joined Heilongjiang River at Tongjiang City, having great impacts on the water quality of Heilongjiang River. Five main control pollutants Hg, Cd, Cr, As and Pb were selected as research objects and the samples were collected from the Second Songhua River and trunk stream of Songhua River from May 2011 to May 2012.

The water samples were collected from 10 sections for 8 times. Overall 88 fish samples were collected from 5 sections for once, including catfish (sarcophagous), carp and crucian (omnivory), chub (herbivore). One sediment sample was collected from each of 8 selected sections. The background information of fish samples was assembled from Ref. [19], 16 water samples and 23 sediment samples were collected from six rivers in May 2011 (Fig. 2). All samples were detected by ICP-MS.

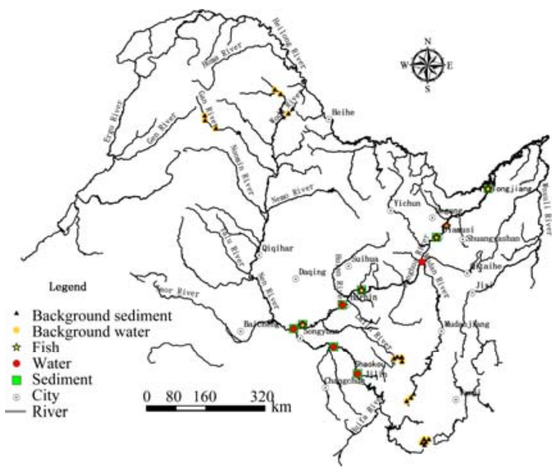

Fig. 2 Spatial distribution of sampling sections

4.1.2 Model calculation and results analysis

The target river was segmented according to water quality classification standards prescribed in Jilin Province Surface Water Function Regions DB22/388—2004 and Heilongjiang Province Surface Water Environment Regionalization and Water Quality Supplementary Standards DB23/485—1998. Shaokou to Tongjiang section was selected as the research target and it was divided into 11 water sampling areas, whereas, 1 section was followed under category II, 7 in category III and 3 in category IV. The calculation process of weight values was as follows, such as Hg.

$$a = \frac{0.173 \times 1 + 0.114 \times 7 + 0.073 \times 3}{1 + 7 + 3} = 0.108$$

$$b = \frac{0.066 \times 1 + 0.043 \times 7 + 0.028 \times 3}{1 + 7 + 3} = 0.041$$

$$c = \frac{0.038 \times 1 + 0.025 \times 7 + 0.016 \times 3}{1 + 7 + 3} = 0.024$$

$$\zeta = \frac{0.108}{0.108 + 0.041 + 0.024} = 0.624$$

$$\eta = \frac{0.041}{0.108 + 0.041 + 0.024} = 0.237$$

$$\theta = \frac{0.024}{0.108 + 0.041 + 0.024} = 0.139$$

The results of ecological risk assessment of heavy metals in Songhua River are show in Table 5 and Fig. 3. Table 5 and Fig. 3 explained that: 1) The biological toxicity of the five types of heavy metals in Songhua River were different with toxicity coefficient in a descending order of Hg>Cd>As>Pb>Cr, C_{f}^{i} showed that the richness of heavy metals in the three phases was as

Table 5 Comprehensive ecological risk assessment of heavy metals in Songhua River

Three phase	Heavy metal	T_{r}^{i}	C_{f}^{i}	F_{s}^{i}	W, F, S	ζ, η, θ	R
Water			1.40	0.34	19.04	0.62	
Fish	Hg	40	1.30	1	52.00	0.24	28.48
Sediment			0.75	1	30.00	0.14	
Water			1.30	1	39.00	0.62	
Fish	Cd	30	1.17	1	35.10	0.24	37.64
Sediment			1.20	1	36.00	0.14	
Water			5.11	0.76	7.77	0.62	
Fish	Cr	2	1.13	1	2.26	0.24	5.67
Sediment			1.10	1	2.20	0.14	
Water			2.34	1	23.40	0.62	
Fish	As	10	1.33	1	13.30	0.24	19.14
Sediment			1.03	1	10.30	0.14	
Water			3.37	0.50	8.43	0.62	
Fish	Pb	5	1.25	1	6.25	0.24	7.41
Sediment			0.98	1	4.90	0.14	

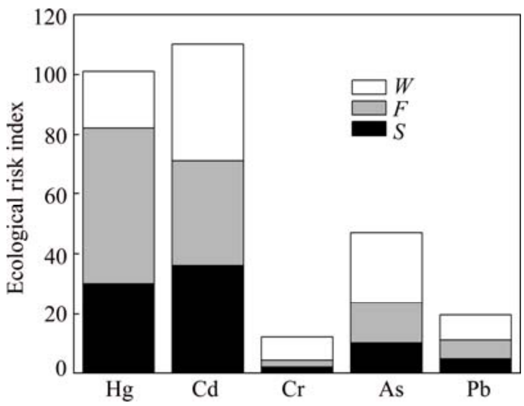

Fig. 3 Ecological risk index of heavy metals in three phases in Songhua River

aquatic>biological>sedimentary, F_s^i represented a higher detection rate of heavy metals; 2) The detection rate of Hg, Cr, and Pb in aquatic phase was less than 1, but the pollution index was marked up to 1.40, 5.11 and 3.37, respectively, this phenomenon indicated the uneven spatial distribution of these three kinds of heavy metals, mainly higher values were detected in aquatic phase, the pollution index of Hg and Pb in sediments was smaller than 1, but the detection rate was 100%, this was due to their high background values; 3) The ecological risk index of Hg in biological phase was the highest (52), greater than the sum of risk indexes in water and solid phases (49), the ecological risk of Cd was similar in three phases, while the other 3 heavy metals in an descending order was water>biological>solid; 4) The comprehensive ecological risk index (R) descending order of five heavy metals was Cd(37.64)>Hg(28.48)> As(19.14)>Pb(7.41)> Cr(5.67), and Cd and Hg were selected as priority pollutants in Songhua River.

4.2 Model verification

ZHU and WANG [20] reviewed about 51 papers consisted of 34478 samples from seven major Chinese water systems at different time. According to the heavy metal characteristics analysis, Songhua River was ranked third on ecological risk, following Pearl River and Haihe River. The ecological risk of Hg was higher than that of Cd. SUN et al [21] reported higher concentrations of Cd in fish of the Second Songhua River than that of the trunk stream Songhua River, while Hg concentration in fish was found higher in the trunk stream of the Songhua River. LU et al [22] investigated the higher ecological risk index of Hg and Cd in sediment samples collected from middle and lower stream of the Second Songhua River. The research evidenced the Hg and Cd as the greatest ecological risk to water, fish and sediments of the Songhua River. These researches were consistent with the results of the present study, which demonstrated the significance of this model.

4.3 Model discussion

Toxicity coefficient and heavy metal concentration were the most important indexes for most models (Table 1). By analyzing the model construction and selection methods of priority pollutants in foreign studies, the comprehensive ecological risk assessment model was constructed. The results illustrated that heavy metals with higher toxicity coefficient have higher comprehensive ecological risk index (R and T_r have significant correlation), but lacked in direct correlation due to the influence of pollutant concentration, background value and detection rate. For example, T_r(Cd)<T_r(Hg), but R(Cd)>R(Hg). The low detection rate of Hg in aquatic phase also decreased the ecological risk index of Hg in biological and sedimentary phases, while an ecological risk index of the Cd was calculated higher. Therefore, the detection rate was considered as an important influencing factor for comprehensive effects. This model was designed to screen out the priority heavy metal pollutants from rivers by adopting comprehensive ecological risk indexes. It was not only time and cost efficient, but also helpful for actual heavy metal assessment and transformation in the rivers. In addition, this work probed the integrated ecological risk of five types of toxic heavy metals, and was only applied in the case study of Songhua River, but further researches on more heavy metals and rivers were needed to explain the specifications of the application.

5 Conclusions and prospects

1) Heavy metal toxicity coefficient, pollution index and detection rate were selected as the key factors to calculate the ecological risk index in aquatic, biological and sedimentary phases. Different parameters such as sampling numbers, water quality and pollutant concentrations were selected for absolute study of three phases. Finally, the comprehensive ecological risk assessment model was constructed and heavy metals of higher risk indexes were concluded as priority pollutants.

2) The model was applied to assessing five types of toxic heavy metals in the Songhua River. The results concluded higher detection rate of heavy metals and descending order of their biological toxicity was as Hg>Cd>As>Pb>Cr, while the richness in phases was categorized as aquatic> biological> sedimentary.

3) The ecological risk index of five heavy metals in the single medium was as aquatic>biological> sedimentary, while the comprehensive ecological risk index in three phases was Cd>Hg>As>Pb>Cr. Therefore, Cd and Hg were selected as the priority pollutants of the Songhua River.

4) The team decided further study to mitigate the model limits as this was only applied for heavy metal detection in the Songhua River. Parameter selection and weight determination still needed intensive studies. Moreover, the research on the risk hierarchy was acting as a milestone in future studies.

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河流三相空间重金属污染综合生态风险评价

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摘　要：当前，有关重金属在河流水相、生物相和固相三相空间的综合污染效应研究尚未见报道，因此主要围绕河流三相空间重金属风险评估展开研究。大量学者在开展单一介质空间生态风险评估时，主要考虑重金属毒性系数和污染指数。本文构建的综合生态风险评价模型表明重金属对环境安全具有严重威胁。该模型应用于松花江 5 种有毒重金属污染综合效应评价，结果表明，5 种有毒重金属在单一介质中生态风险指数均表现为水相>生物相>固相，三相空间综合生态风险指数由高到低排序为 Cd>Hg>As>Pb>Cr。在此基础上，将 Cd 和 Hg 筛选为松花江重金属优控污染物。

关键词：综合生态风险评价；优控污染物筛选；重金属；松花江

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