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Environmental Background Values and Ecological Risk Assessment of Heavy Metals in Watershed Sediments: A Comparison of Assessment Methods

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Abstract: The distribution and assessment of heavy metal pollution in sediments have been extensively studied worldwide. Risk assessment methods based on total content, background values, and sediment quality guidelines are widely applied but have never been compared. We systematically sorted out these evaluation methods, obtained evaluation results using actual monitoring data, and compared their applicability. The results showed that the background values of different metals are significantly different, which may depend on their mobility. Geoaccumulation index (I_{geo}) and enrichment factor (*EF*) values invariably decreased with the increase of background values for individual heavy metal enrichment risk assessment. Compared with EF, I_{geo} also showed a significant positive linear correlation with heavy metal content. Pollution load index (*PLI*), modified contamination degree (mC_d), and potential ecological risk index (*RI*) showed significant differences in response to background values and evaluation levels for the comprehensive risk of heavy metal enrichment, but their distribution trends along with the sampling points were basically identical. Toxic risk index (*TRI*), mean ERM quotient (*mERMQ*), and contamination severity index (*CSI*) were used to evaluate the damage degree of complex heavy metals to aquatic organisms and shared a similar whole-process distribution trend. The modified hazard quotient (*mHQ*), which is used to evaluate the toxicity of a single heavy metal to aquatic organisms, showed a significant positive linear correlation with the total content of each heavy metal, indicating that the toxic effect on organisms can be predicted through the direct monitoring. The results of this study have important guiding significance for the selection of evaluation methods for heavy metal pollution in sediments.

Keywords: risk assessment; method comparison; different types of background values; sediment quality guidelines; heavy metals

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1. Introduction

As one of the main pollutants in the water environment, heavy metals have raised concern regarding their effect on water ecosystem safety and human health. Heavy metals in water can migrate into sediments through physical, chemical, and biological reactions [1,2]. Meanwhile, heavy metals in sediments will be released upward with the change of environmental conditions, causing secondary pollution of the water environment [3]. The content of heavy metals in sediments is usually three to six orders of magnitude higher than that in water, which implies that sediment is the main storage reservoir of heavy metals in water environment and plays an important indicator role for water pollution [3,4]. Currently, various indexes have been developed to assess environmental risks for heavy metals in sediments based on their total contents, bioavailability, and toxicity [5–7]. Although many researches have highlighted that the morphological content can well reveal the migration and toxicity of heavy metals in sediments, total content can directly reflect the degree and source of contamination [8,9]. Therefore, risk assessment based on total concentration calculation remains an indispensable method in the study of heavy metal pollution in water environment and a key pathway to identify pollution sources.

Heavy metal risk assessment methods based on total concentration calculation have been widely proposed [10,11], mainly including two categories: one is related to background values, the other is related to sediment quality guidelines. Background values mainly include three types: soil background value in each investigated area (B_v), control value of surface sediment (C_{vs}, samples collected from the study area that are uncontaminated), and control value of deep sediment (C_{vd}, samples collected from the bottom of sedimentary column). However, the selection of background values in previous studies was not uniform, resulting in large differences in evaluation results and low comparability.

Sediment quality guidelines (SQGs) are the actual allowable values of a specific chemical substance in sediments that does not harm benthic aquatic organisms or other relevant water functions [7,12]. Although there is still no internationally agreed methodology, the SQGs have been useful in numerous applications, such as designing monitoring programs, interpreting historical data, ecological risk assessment, and developing sediment quality remediation objectives [13]. The current SQGs are mainly derived from various states in North America for both freshwater and marine ecosystems and are composed of a variety of calculations [14]. However, few studies have concentrated on the comparison of different methods, which may lead to the possible confusion of these evaluation indicators.

The pollution assessment method compares the concentration of heavy metals with background values and SQGs, which helps to evaluate the accumulation of heavy metals in sediments. Thus, it is essential to consider the intensity of metal pollution by inventorying the concentrations and their distribution in a riverine ecosystem. In addition, the various bases of assessment indices predict inconsistent risks of heavy metals in sediments of different regions [5]. In order to avoid biases in the risk assessment of heavy metals in sediments, a combination of different indicators should be used. However, the existing researches mainly focus on the superposition application of each single method [7,10] and thus lack mutual comparison, leading to confusion, especially when the effects of background values and SQGs have not been highlighted.

Therefore, we assume that each assessment method is sufficiently independent, and that the distribution and migration of heavy metals are mainly affected by pollution sources and environmental factors, so the risk of heavy metals depends on the content of heavy metals and background values. This study comprehensively compared and analyzed various evaluation methods based on the calculation of actual detection data, aiming to reveal the importance of the selection of evaluation methods, background values and SQGs. The main objectives were (a) to analyze the vertical distribution of heavy metals in sediments and to obtain three types of background values, (b) to compare the results of selected evaluation methods with the application of background values, and (c) to reveal the relationship between evaluation methods and background values and SQGs.

2. Materials and Methods

2.1. Study Area and Data Source

The Beijiang River is situated in Guangdong Province, China, and flows through Shaoguan City, Yingde City, Qingyuan City, and Foshan City from upstream to downstream, and then converges in the Pearl River Estuary. It is a major source of drinking water and an ecological safety barrier in Guangdong Province. With the adjustment of Guangdong's industrial layout, a large number of small- and medium-sized enterprises, taking advantage of the "golden waterway", move to the cities along the Beijiang River. The development of cement, ceramics, smelting, and other manufacturing industries has led to the continuous deterioration of water quality of the Beijiang River, which poses a serious threat to regional ecological and environmental security [15,16]. The data of this study were derived from the sampling survey of sediments throughout the Beijiang River. In April 2018, 23 columnar sediment samples were collected by drilling. The depth of each sample column was 3 m and was divided into 9 sections: 0–0.2 m, 0.2–0.4 m, 0.4–0.6 m, 0.6–0.8 m, 0.8–1.0 m, 1.0–1.5 m, 1.5–2.0 m, 2.0–2.5 m, and 2.5–3.0 m, respectively. Each part of the intercepted sediment sample was thoroughly mixed, and three small portions were taken out for pretreatment,

such as drying, grinding, and digestion. After volumetric determination, heavy metals, Cr, Ni, Cu, Zn, As, Cd, Tl, Pb, and Hg, as well as common metallic element Fe were measured by atomic absorption spectrometry. The detailed detection methods and quality assurance were shown in a previous study [17]. Moreover, in order to obtain control values of surface sediments suitable to the actual environment, three samples were collected from tributaries nearby that were not affected by point sources.

In particular, the data here were used only to analyze vertical distribution trends, with the purpose of obtaining control values for deep sediments, rather than analyzing pollution mechanisms. On the basis of obtaining background values, we uniformly used the total concentration data of heavy metals in the surface layer of 0–0.2 m to conduct pollution assessment and focused on analyzing the differences in the results obtained by different types of assessment methods.

2.2. Assessment Methods

The selected evaluation methods are all calculated based on total concentrations of heavy metals, background values and SQGs, which are introduced as follows.

2.2.1. Geoaccumulation Index (I_{geo})

The geoaccumulation index (I_{geo}) has been widely used in the assessment of heavy metal pollution and reveals the relationship between heavy metals in sediments and geochemical background values, which was originally proposed by Muller [18]. The I_{geo} value is defined as

$$I_{geo} = \log_2\left[\frac{C_n}{1.5B_n}\right] \quad (1)$$

where C_n represents the measured concentration of metal (n) (mg/kg), B_n represents the geochemical background value of metal (n) (mg/kg), and *1.5* is a factor used to minimize the impact of background value caused by lithological variation [19]. The I_{geo} consists of 7 levels, as shown in Table S1.

2.2.2. Enrichment Factor (*EF*)

Enrichment factor (*EF*) is considered to be an effective tool to evaluate the enrichment of pollutants in the environment. To ascertain the impact of anthropogenic activities on sediment, the measured heavy metal concentrations are compared with conservative elements (such as Al and Fe) that are not affected by weathering [20,21]. Here, Fe is selected as the conservative metal, and the *EF* value is defined as

$$EF = \frac{(C_n/C_{Fe})_{sample}}{(B_n/B_{Fe})_{background}} \quad (2)$$

where $(C_n/C_{Fe})_{sample}$ represents the ratio between the measured heavy metal concentration and Fe concentration in the contaminated sediment sample, and $(B_n/B_{Fe})_{background}$ represents the ratio between the measured heavy metal concentration and Fe concentration in the background sediment sample. $EF > 1.5$ indicates that heavy metals are derived from anthropogenic origin, while $EF < 1.5$ indicates that heavy metals are completely derived from natural weathering [22]. The *EF* consists of 7 levels, as shown in Table S2.

2.2.3. Contamination Factor (C_f) and Pollution Load Index (*PLI*)

Contamination factor (C_f) is used to show the contamination degree with a single metal and is the ratio of the content of each metal to its background value. Pollution load index (*PLI*) is an empirical index which provides a simple and comparative means for assessing metal pollution levels and is a geometric evaluation of the individual C_f [23]. The calculation methods of C_f and *PLI* are as follows:

$$C_f = \frac{C_n}{B_n} \tag{3}$$

$$PLI = \sqrt[n]{{C_f}^1 \times {C_f}^2 \times {C_f}^3 \times \ldots {C_f}^n} \tag{4}$$

where C_n is the measured concentration of the target heavy metal (mg/kg), B_n is the selected background concentration of the target heavy metal (mg/kg), and n is the quantity of the target heavy metal. The recommended pollution levels for *PLI* classification are as follows: no pollution ($PLI \leq 1$) and polluted ($PLI > 1$) [24].

2.2.4. Modified Contamination Degree (mC_d)

Contamination degree (C_d) is defined as the sum of all contamination factors (C_f). As it is not always feasible to analyze all the components used in this indicator, Abrahim and Parker proposed an improved method, which was defined as modified contamination degree (mC_d) [25], and it was calculated as

$$mC_d = \frac{\sum_{i=1}^{n} {C_f}^i}{n} \tag{5}$$

where ${C_f}^i$ represents the contamination factor, and n represents the number of target heavy metals. The classifications of contamination levels are shown in Table S3.

2.2.5. Potential Ecological Risk Index (*RI*)

Potential ecological risk of individual factor (${E_r}^i$) and potential ecological risk index (*RI*) were proposed by Hakanson to determine the ecological impact and potential risk of heavy metals exposure [23]. It illustrates ecological sensitivity and vulnerability towards toxic heavy metals and evaluates the comprehensive ecological risk. Four factors are considered: the type of target heavy metals, measured concentration, toxicity coefficient, and sensitivity of water body to heavy metals. The equation is

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \times {C_f}^i \tag{6}$$

where ${C_f}^i$ represents the contamination factor, and n represents the number of target heavy metals. ${T_r}^i$ represents the toxicity coefficient of a single heavy metal [6], and the toxicity coefficients of target heavy metals are shown in Table S4. The classifications of ecological risk levels are shown in Table S5.

2.2.6. Toxic Risk Index (*TRI*)

Toxic risk index (TRI_i) is known as a newly validated method for evaluating the ecotoxicity of system in view of the *TEL* (threshold effect level) and *PEL* (probable effect level) effects, which is applied to normalize the toxicities caused by different heavy metals and then facilitated the comparison of their relative effects [26]. The *TEL* and *PEL* values of target heavy metals are shown in Table S4. The potential acute toxicity of the heavy metals in a sediment sample can be assessed as the sum of TRI_i. The TRI_i and *TRI* can be calculated as

$$TRI_i = \sqrt{\frac{(C_i/TEL_i)^2 + (C_i/PEL_i)^2}{2}} \tag{7}$$

$$TRI = \sum_{i=1}^{n} TRI_i \tag{8}$$

where C_i represents the measured concentration of heavy metal i, and n represents the number of target heavy metals, TEL_i is the *TEL* value of the target heavy metal i, and PEL_i

is the *PEL* value of the target heavy metal *i*. The pollution levels of the *TRI* are classified as shown in Table S6.

2.2.7. Modified Hazard Quotient (*mHQ*)

The modified hazard quotient (*mHQ*) is used to evaluate sediment contamination by comparing heavy metal contents in the sediment with effect level standards (*PEL*, *SEL* (severe effect level), and *TEL*) [27]. It is considered a significant tool because it exemplifies the extent of risk each heavy metal poses to the biota and aquatic habitat [7]. *TEL*, *PEL*, and *SEL* values of target heavy metals are shown in Table S4. The *mHQ* is evaluated following the mathematical expression below:

$$mHQ = \sqrt{\frac{C_i}{TEL_i} + \frac{C_i}{PEL_i} + \frac{C_i}{SEL_i}} \quad (9)$$

where C_i represents the measured concentration of heavy metal *i*, TEL_i is the *TEL* value of the target heavy metal *i*, PEL_i is the *PEL* value of the target heavy metal *i*, and SEL_i is the *SEL* value of the target heavy metal *i*. The contamination levels of the *mHQ* are classified as shown in Table S7.

2.2.8. Mean ERM Quotient (*mERMQ*)

Mean ERM quotient (*mERMQ*) is proposed for assessing the potential effects of multiple heavy metal contamination in sediment. The sediment quality guidelines were developed from biological toxicity test of the benthic environment and classified into three levels by *ERL* (effect range low) and *ERM* (effect range medium) as rarely (<*ERL*), occasionally (*ERL-ERM*), or frequently (≥*ERM*) associated with adverse biological effects [28,29]. *ERL* and *ERM* values of target heavy metals are shown in Table S4. The *mERMQ* is calculated as follows:

$$ERMQ_i = \frac{C_i}{ERM_i} \quad (10)$$

$$mERMQ = \frac{\sum_{i=1}^{n} ERMQ_i}{n} \quad (11)$$

where *mERMQ* is the effect-range median quotient of multiple metal contamination, $ERMQ_i$ is the effect-range quotient of heavy metal *i*, C_i is the measured content of the target heavy metal *i*, ERM_i is the *ERM* value of the target heavy metal *i*, and *n* is the number of metals. The contamination levels of *mERMQ* are classified as shown in Table S8.

2.2.9. Contamination Severity Index (*CSI*)

The contamination severity index (*CSI*) is a new index based on *ERL* and *ERM* values to study the severity of heavy metal contamination in sediments, which was first proposed by Pejman [30], for the toxicity boundaries and adverse effect on the biota as well as weighted values for each heavy metal attributed by the ratio of the PCA/FA as site-specific factor [6]. The *CSI* is calculated as follows:

$$CSI = \sum_{i=1}^{n} w_i [(\frac{C_i}{ERL_i})^{1/2} + (\frac{C_i}{ERM_i})^2] \quad (12)$$

where w_i is the weight of the heavy metal *i*, C_i is the measured content of the target heavy metal *i*, ERL_i is the *ERL* value of the target heavy metal *i*, ERM_i is the *ERM* value of the target heavy metal *i*, and *n* is the number of selected metals. The pollution levels of the *CSI* are classified as shown in Table S9.

The ratio PCA/FA is used to obtain the weight (w_i) of each heavy metal. This method only considered the factors with human influence to calculate the weighted value. The weight of each heavy metal is calculated as follows:

$$w_i = \frac{(Loading\ value_i \times eigen\ value)}{\sum_{i}^{n} (Loading\ value_i \times eigen\ value)} \quad (13)$$

The *loading value*, *eigen value*, and w_i are shown in Table S10.

2.3. Statistical Analysis

All statistical analyses were performed using SPSS 19.0 software packages. Results were expressed as mean ± standard deviation. Differences were considered significant when $p < 0.05$. Principal component analysis (PCA) and factor analysis (FA) were performed to analyze the occurrence relationship between heavy metals to obtain the weight value w_i.

3. Results and Discussion

3.1. Vertical Distribution and Background Values of Heavy Metals

Figure 1 illustrates the concentration distribution of Cr, Ni, Cu, Zn, As, Cd, Tl, Pb, and Hg with depth in sediment cores from 23 sampling sites along the Beijiang River. Except for Ni and Cu, all other heavy metals fluctuated in the depth range of 0–0.6 m and showed a slightly decreasing trend. The results are comparable to those of previous studies, suggesting that the large fluctuation of heavy metals in surface sediments is mainly caused by human activities and hydraulic disturbance [31–33]. Given the prevalence of diagenesis, heavy metal concentrations declined rapidly in the subsequent depth of 0.6–2.5 m, indicating that heavy metals were gradually deposited into deep sediments under the action of sediment adsorption and gravity [34,35]. However, the deposition rate decreased with the increase of depth, and the rate of different metals varied slightly. After 2.5 m, heavy metal concentrations basically remained stable, indicating that sediments beyond this depth were barely disturbed by human activities and the diagenesis also decreased, which can be verified by isotope dating [36,37]. Therefore, the heavy metal concentrations in subsequent deep sediments can be defined as background values, which are defined as the deep control values (C_{vd}).

As a conservative metal element, the content of Fe is not affected by weathering and is significantly related to the distribution of heavy metals, which can effectively reflect the accumulation of heavy metals in sediments caused by human activities [4,7]. As shown in Figure S1, the distribution trend of Fe was basically consistent with that of the target heavy metals, which can be used to interpret the enrichment of heavy metals. In addition, we collected three surface sediment samples from tributaries not affected by point sources near the Beijiang River and detected the contents of target heavy metals and Fe, the results are presented in Table S11. Given the difference of geological conditions and the inevitable influence of anthropogenic sources, noticeable concentration gradients could be observed in these samples. Therefore, average values could be calculated separately and defined as the control values of surface sediments (C_{vs}).

Synthetically, the three types of background values were compared, and the results are shown in Table 1, where B_v represents the native soil background value [38]. The B_v, C_{vs}, and C_{vd} values of Cr, Ni, Cu, Zn, As, and Pb showed a decreasing trend of $B_v > C_{vs} > C_{vd}$ with the maximum multiple reached 3.53, indicating that the accumulation of these heavy metals in soils was significantly higher than that in the unpolluted sediments. By contrast, B_v values of Cd, Hg, and Fe were significantly lower than those of C_{vs} and C_{vd}, which might be attributed to their rapid mobility under the influence of environment [39–41]. Therefore, there are significant differences among the three kinds of background values of each heavy metal and their variation trends are completely different, which inevitably leads to great differences in the risk assessment results. In this light, it is particularly important to select the appropriate background value when conducting risk assessment.

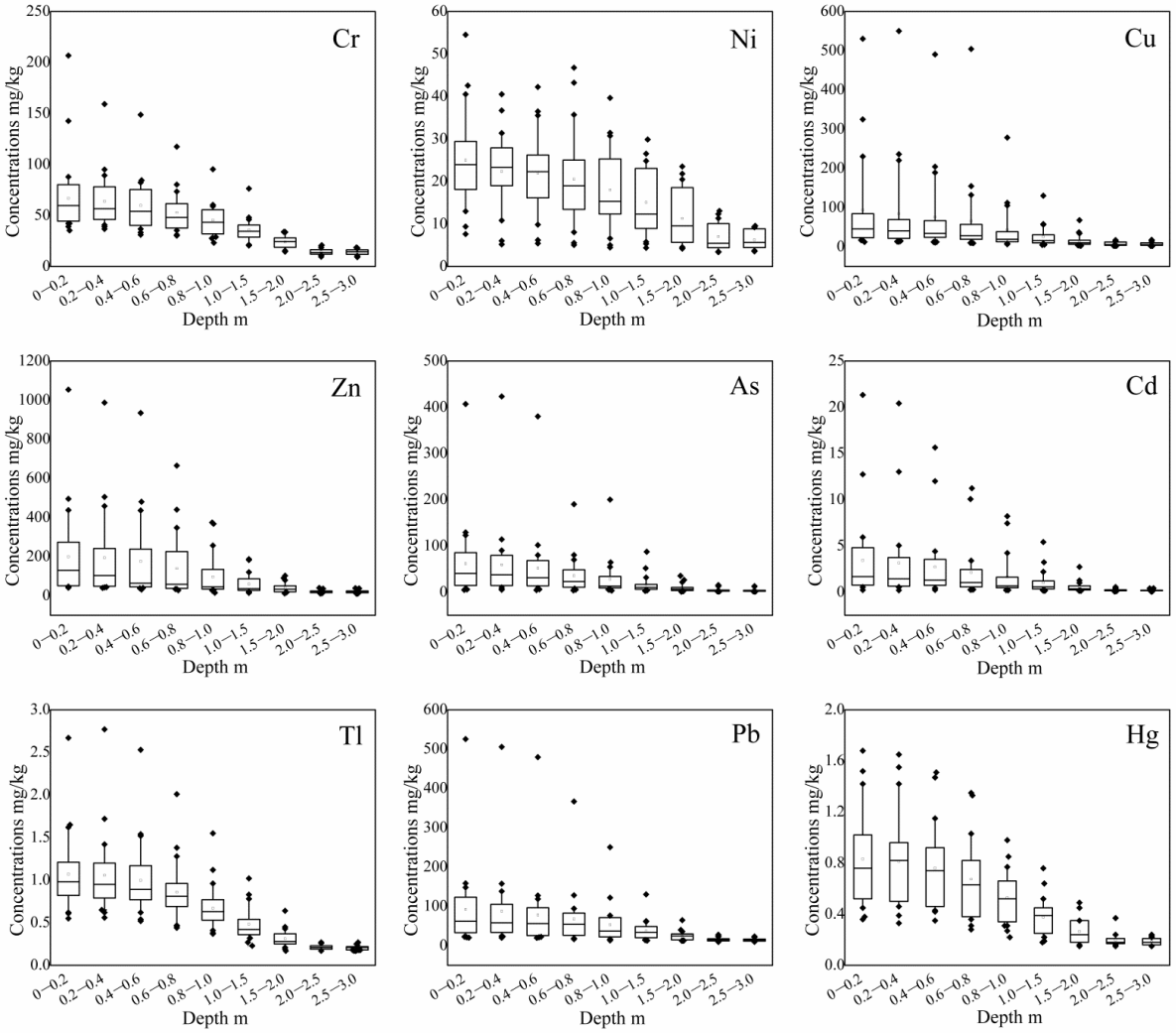

Figure 1. Vertical variations of heavy metals in sediments, all data in mean concentrations, dry weight, mg/kg. The boxes represent 25th and 75th percentiles, the middle horizontal lines represent the 50th percentile, the vertical line ends represent 1st and 99th percentiles, the small squares in the middle represent the mean value, and the diamond black dots represent outliers.

Table 1. Comparison of three types of background values of heavy metals and Fe in sediments, including soil background value (B_v), control value of surface sediment (C_{vs}), and control value of deep sediment (C_{vd}) in Guangdong Province, dry weight, mg/kg.

Metals	Cr	Ni	Cu	Zn	As	Cd	Tl	Pb	Hg	Fe
B_v	50.53	14.4	17.65	47.71	8.9	0.094	0.55	35.78	0.078	26,400
C_{vs}	31.16	8.7	13.22	31.15	5.56	0.22	0.56	21.79	0.25	33,846
C_{vd}	14.31	6.64	7.49	21.44	4.39	0.22	0.21	15.58	0.19	31,065

3.2. Effect of Background Values on Risk Assessment

I_{geo} and EF are risk assessment indexes based on the total concentration and the background value of a single heavy metal, which plays a crucial role in indicating the pollution degree of heavy metal enrichment caused by anthropogenic sources. As shown in Figure 2, it was surprising that the I_{geo} value of each target heavy metal increased toward its total concentration with a significant decrease was observable toward the background value. Given that the background values of Cr, Ni, Cu, Zn, As, and Pb are in the order of $B_v > C_{vs} > C_{vd}$, the calculated I_{geo} values showed an opposite decreasing trend. As an example, the range of I_{geo} values for Cr based on B_v, C_{vs} and C_{vd} were −1.09 to 1.45, −0.40 to 2.14,

and 0.72–3.27, respectively, and the corresponding evaluation levels also changed significantly, with the highest level showed moderately polluted (B_v) to heavily polluted (C_{vd}). These results indicate that I_{geo} values are only related to the total concentration of heavy metals based on selected background values and did not be affected by sediment properties [42]. However, sediment properties and composition can influence the availability of heavy metals in sediments and thus affect the risk assessment [43]. In addition, the risk values of heavy metals with the same content may differ due to the different background values for each type [44]. In contrast, given the *EF* value is not only affected by total concentrations and background values of heavy metals, also related to the distribution of conservative elements in the matrix. Figure 3 revealed that there was hardly any correlation between *EF* values of target heavy metals and total heavy metal contents, especially Tl and Hg, indicating that the *EF* value is susceptible to environmental geological factors [44,45]. This kind of comparison has not been mentioned in previous studies, but it is certain that comparability between these indicators clearly exists.

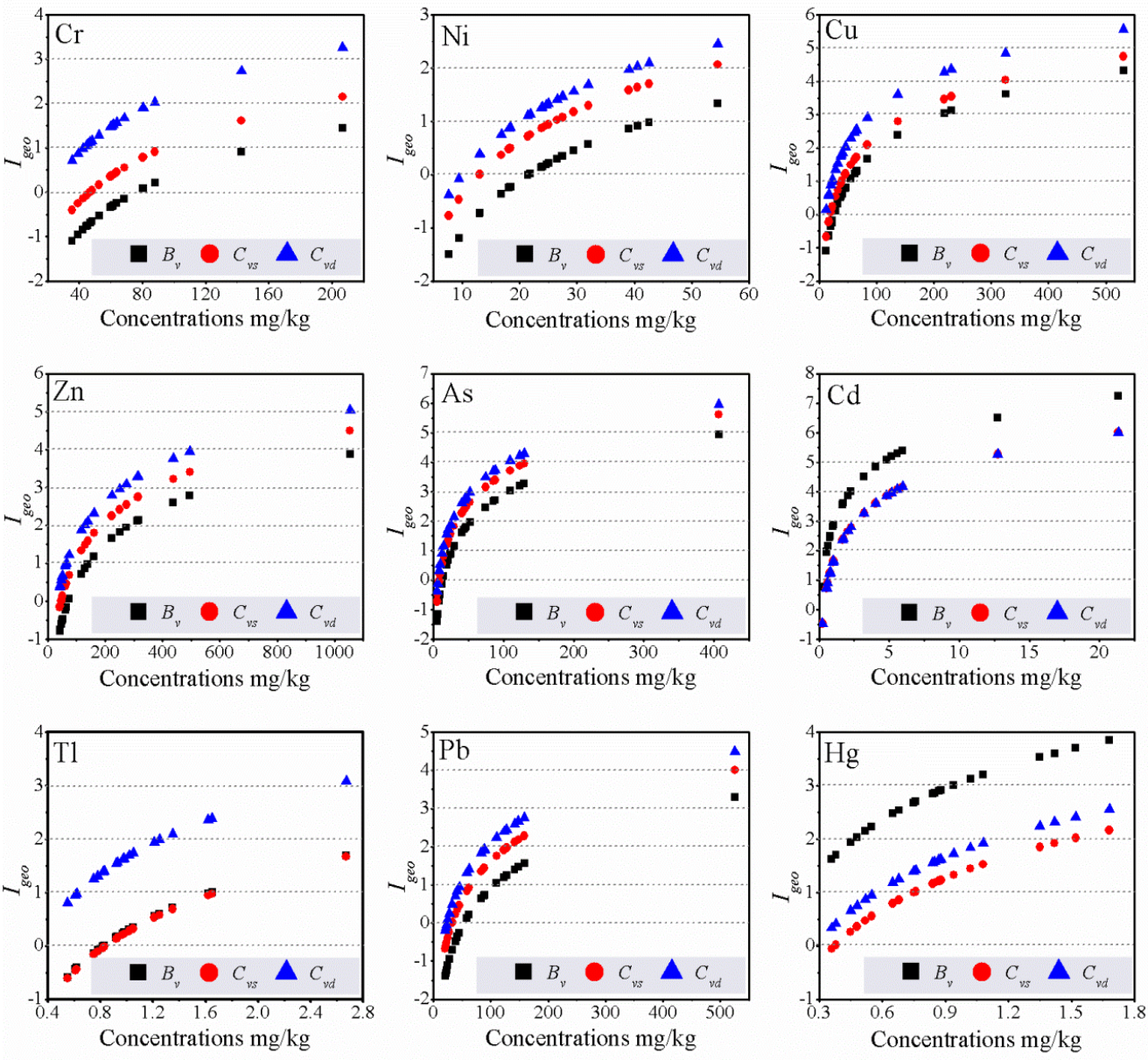

Figure 2. The I_{geo} values of Cr, Ni, Cu, Zn, As, Cd, Tl, Pb, and Hg in sediments were calculated based on different types of background values and total concentrations.

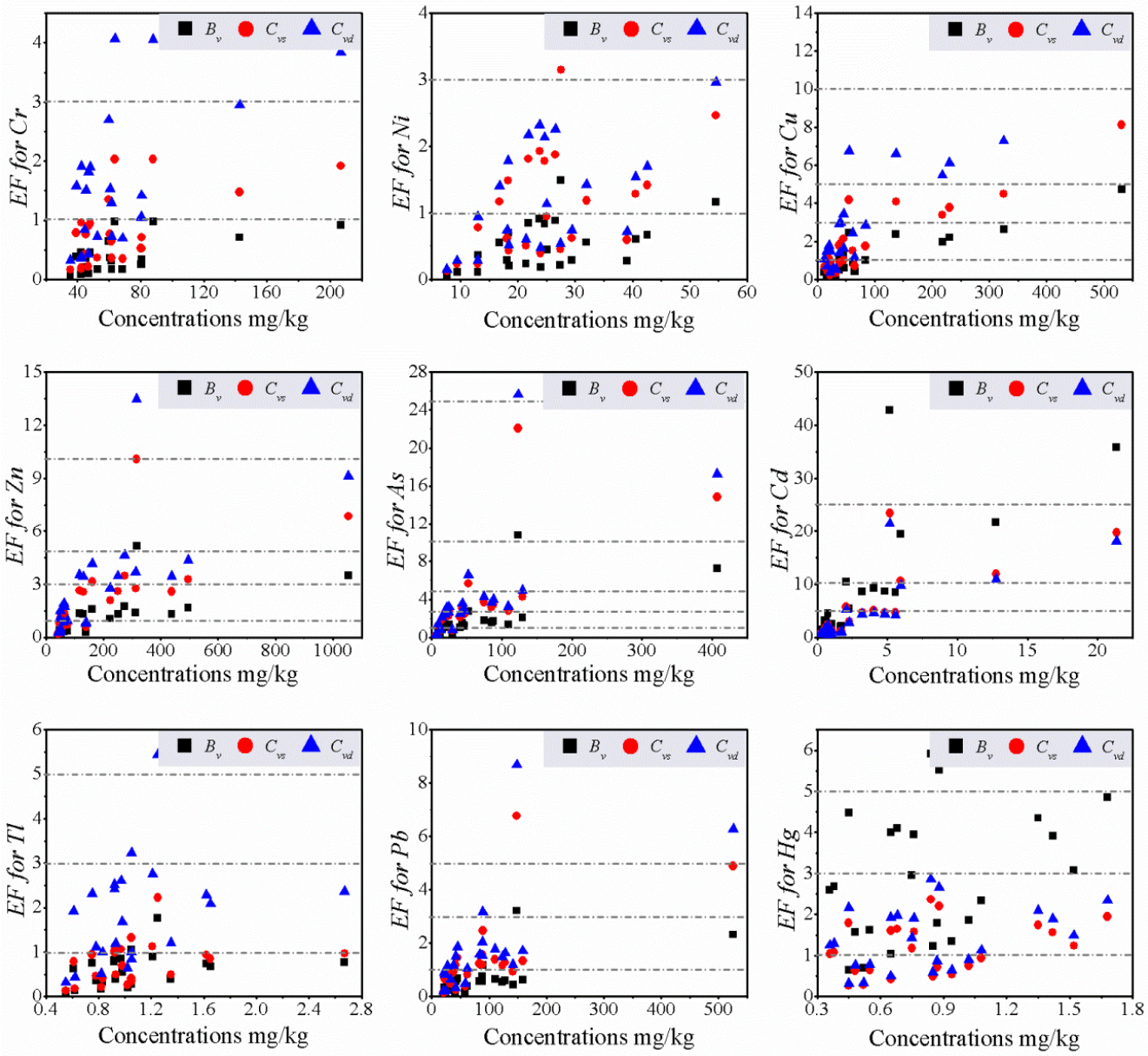

Figure 3. The *EF* values of Cr, Ni, Cu, Zn, As, Cd, Tl, Pb, and Hg in sediments were calculated based on different types of background values and total concentrations.

Traditionally, I_{geo} and *EF* values have been compared on the basis of different sampling sites with the purpose of identifying hazardous areas and sources of heavy metals. Figures S2 and S3 visually indicated the differences in I_{geo} and *EF* values among sampling sites. In view of the influence of pollution sources and human activities, heavy metal risks invariably fluctuated dynamically, e.g., the I_{geo} and *EF* of Cd fluctuated significantly in the whole investigated region, indicating a higher risk at the Maba confluence, especially with an *EF* value greater than 20 times, which is consistent with our previous findings [2,17]. Given the presence of $B_v < C_{vs} = C_{vd}$, I_{geo} curves based on the latter two background values were coincident along the path, while *EF* values were somewhat atypical. In addition, consistent results showed that both the I_{geo} and *EF* of a single sample decrease with the increase of background values. These results are expressed in a manner similar to those reported before and are indispensable information for the identification of significant pollution areas and sources in the watershed [10,46].

Synthetically, the selection of background value has significant effect on the enrichment risk of individual sample and individual heavy metal. A more interesting and widely

applicable finding is that I_{geo} values seem to be independent of geological background differences, which can compare the risk of the same heavy metal in different regions, whereas *EF* values are recommended to be more suitable for comparison of different heavy metals in the same sampling region.

In contrast, *PLI*, mC_d, and *RI* are indicators to evaluate polymetallic composite pollution based on background values, which can reflect the comprehensive risk of heavy metal enrichment in each sampling area. As shown in Figure 4, although the three indicators are all calculated according to C_f value, their subsequent change trends varied greatly under different background values. *PLI* showed an increasing trend of $B_v < C_{vs} < C_{vd}$, and mC_d followed an order of $C_{vd} > B_v > C_{vs}$, while *RI* was in the order of $B_v > C_{vd} > C_{vs}$. These differences have rarely been addressed in previous studies [5]. The main reason may be that the three kinds of background values of each heavy metal are always different, and the total concentration of heavy metals at each sampling site also varies significantly, which in turn varies with the heavy metals [31,47]. What counts is that such a great variation in toxicity coefficients inevitably lead to the reverse effect of individual heavy metal risks on the combined risk [6]. Given the difference of total concentrations of Cd reached a maximum of 88.9 times, and the B_v value of Cd was smaller than that of C_{vs} and C_{vd}, the mC_d and *RI* values were comparable to or significantly higher than those of the other two. Therefore, it can be inferred that the difference of background value and high risk of a single heavy metal has relatively little influence on *PLI*, while *RI*, influenced by the combination of background value and toxicity coefficient, should be closer to the real risk effect.

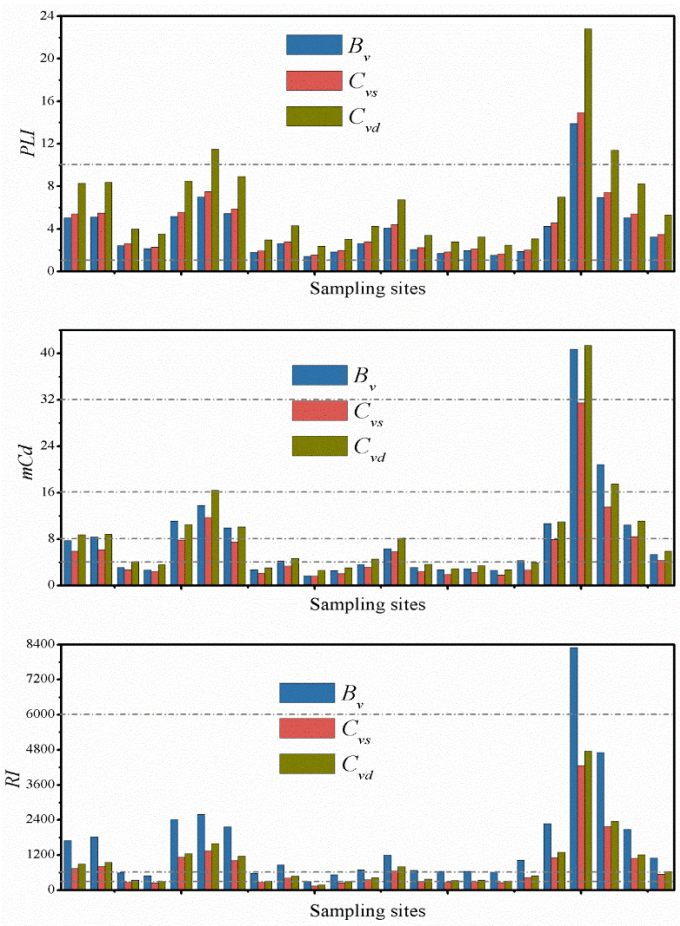

Figure 4. The distribution of the *PLI, mCd, RI* values for Cr, Ni, Cu, Zn, As, Cd, Tl, Pb, and Hg in sediments in the whole investigated region based on different types of background values.

From the perspective of comprehensive risk assessment in the whole investigated region, the distribution trends observed of *PLI*, mC_d, and *RI* were basically similar, which was

consistent with the results of previous studies [7,48]. The three indicators comprehensively reflected the regional differences in levels of heavy metal pollutants, pollution sources, and metal mobility [49], but there were some subtle differences in the corresponding evaluation levels. The relationship between the distribution of *PLI*, mC_d, *RI* and the individual heavy metal concentration was further analyzed, and the results are shown in Figures S4–S6. Incredibly, the effects of individual heavy metal concentrations on the comprehensive risk assessment indicators were almost identical. However, the absence of a significant linear correlation between any of the two indicated that the factors influencing these indicators are not unique. This is consistent with the previous results, indicated that the total content does not include information of sediment composition and availability of heavy metals [49], which affects the risk distribution to a certain extent.

In general, the application of *PLI*, mC_d, and *RI* can reflect the comprehensive risk of heavy metal enrichment in the investigated region and provide similar essential information to indicate the heavily polluted area, but they are not affected by the background value to the same extent. Given the comprehensive consideration of background value and toxicity coefficient, *RI* seems to be a better choice in the comprehensive risk assessment of heavy metals in sediments.

3.3. Effect of SQG Values on Risk Assessment

The SQGs are derived from empirically toxic experiments, giving a toxicity indicator for specific aquatic amphipods without considering sediment properties and heavy metal background values [13,27]. *TRI*, *mERMQ*, and *CSI* are comprehensive ecological risk assessment methods of heavy metals in sediments based on SQGs and total contents, which can provide biotoxicity levels of combined heavy metal pollution on aquatic organisms. As shown in Figures 5–7, although these three indicators were calculated based on different SQGs [6], the influence of single heavy metal concentration on *TRI*, *mERMQ*, and *CSI* was completely the same as observed from the comprehensive distribution trend. Inevitably, there were subtle differences between the target heavy metals. *TRI*, *mERMQ* and *CSI* were not in conformity with the total contents of Cr, Ni, and Hg, while a significant linear correlation could be observed with the contents of Cu, Zn, As, Cd and Pb. However, the relationship shown does not completely increase in the direction of increasing concentrations. The reason may be that Cu, Zn, As, Cd, and Pb mainly come from anthropogenic sources, such as the discharge of industrial wastewater and domestic sewage, which leads to serious heavy metal pollution with extremely high toxicity to local organisms [2,50]. On the contrary, Cr, Ni, and Hg mainly come from natural sources, and the influence of sediment characteristics cannot be ignored [43,51]. Meanwhile, the results also indicated that the three indicators have similar effects. Regardless of the fact that the results of different indicators for matching objects may be mutually complementary, it seems that choosing one of them is sufficient.

Figure S7 showed the distribution of *TRI*, *mERMQ*, and *CSI* along each sampling site in the whole investigated region, and it could be observed that the trend was almost identical, which further confirms the conclusion as indicated earlier. However, they do not indicate the same level of risk. In this light, *TRI* indicated that 43.48% of sampling sites had a very high toxicity risk (*TRI* > 20). *mERMQ* showed that 4.35% of samples were greater than 1.5 and 39.13% of samples were between 0.5 to 1.5, indicating that 43.48% of samples had a 49% probability of toxicity. Except for the *CSI* value of one sampling site being as high as 11.14 (ultra-high severity), the other values were all below 3, indicating that 88.86% of sampling areas were below medium-high severity [6,7]. Given the use of different SQGs, although it is impossible to get exactly the same results by applying these three indicators, the evaluation results for the same object are surprisingly consistent.

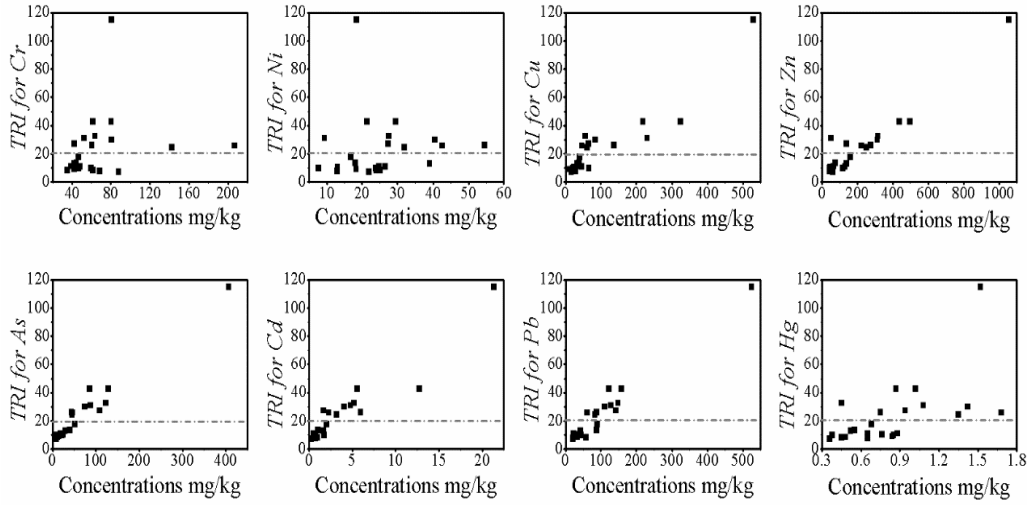

Figure 5. The *TRI* values of Cr, Ni, Cu, Zn, As, Cd, Pb, and Hg in sediments based on SQG values (*TEL*, *PEL*) and total heavy metal concentrations.

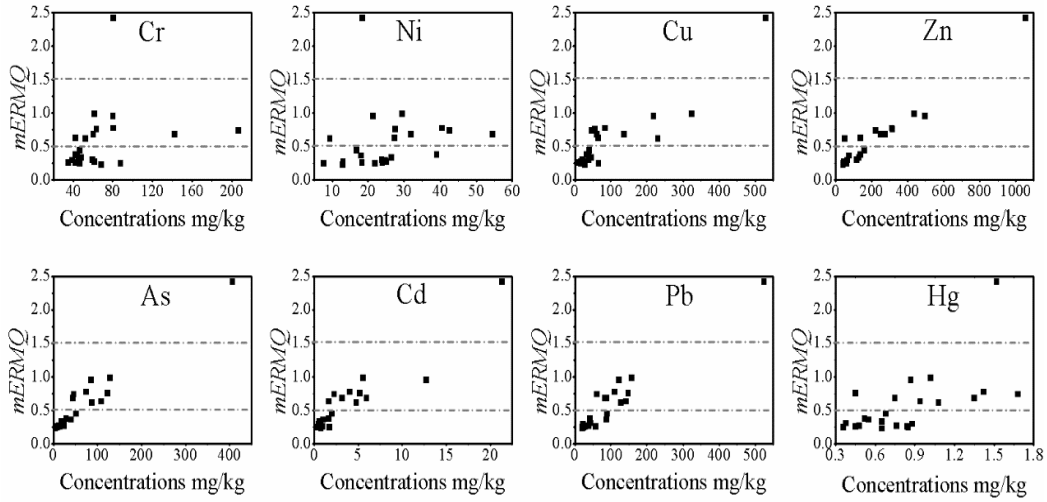

Figure 6. The *mERMQ* values of Cr, Ni, Cu, Zn, As, Cd, Pb and Hg in sediments based on SQG values (*ERM*) and total heavy metal concentrations.

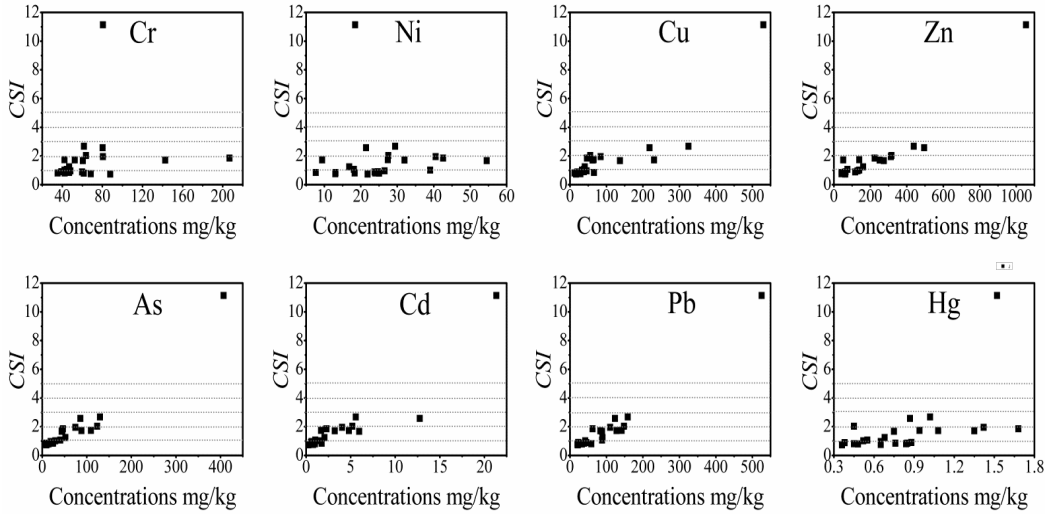

Figure 7. The *CSI* values of Cr, Ni, Cu, Zn, As, Cd, Pb, and Hg in sediments based on SQG values (*ERL*, *ERM*) and total heavy metal concentrations.

The *mHQ* assesses levels of contamination by describing each metal concentration observed in sediments with SQGs. The evaluation of *mHQ* is of utmost importance since it evaluates the risk of individual metal to the biota and the aquatic environment [52]. With respect to the *mHQ* values obtained, it was completely unexpected that they showed a significant positive linear relationship with individual heavy metal content, and increased towards the increasing of the content of heavy metals (Figure 8), indicating that the *mHQ* value is only related to the total concentration of heavy metals and has nothing to do with sediment properties [7]. Moreover, Figure 8 also showed the content distribution and corresponding risk of each target heavy metal in the investigated area. Therefore, the corresponding risk can be effectively predicted by measuring the content of heavy metals.

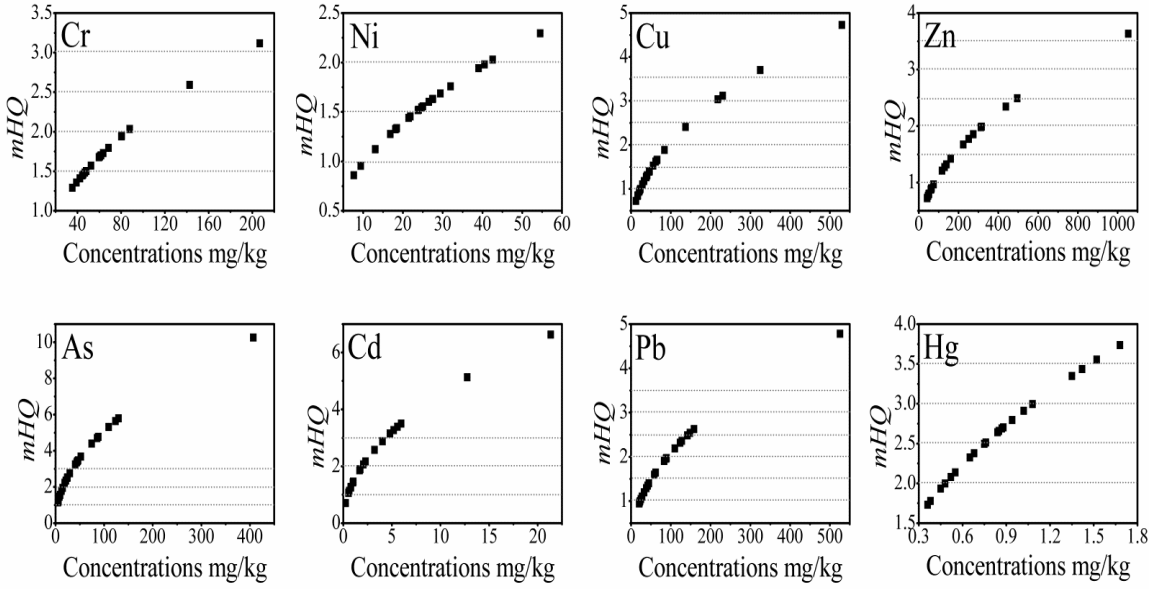

Figure 8. The *mHQ* values of Cr, Ni, Cu, Zn, As, Cd, Pb, and Hg in sediments based on SQG values (*TEL*, *PEL*, *SEL*) and total heavy metal concentrations.

By contrast, it can be obviously observed from Figure S8 that the *mHQ* distribution of each heavy metal varies with the sampling sites, and it is particularly different from the trend shown by the above comprehensive risk indicators. In this light, no significant anthropogenic pollution sources of Cr, Ni, and Hg were observed, while Cu, Zn, As, Cd, and Pb showed extreme severity of contamination [50]. This firmly confirms that anthropogenic inputs had a key contribution for the enrichment of heavy metals in surface sediments [53–55]. Further, more attention should be paid to As, Cd, and Pb at the entrance of the Maba River because of their high contributing ratio to the *mHQ* values. Undeniably, this representation with the distribution of sampling points is widely adopted, which can be better used to reveal the major pollution sources and the locations of serious pollution in the investigated area.

Synthetically, *TRI*, *mERMQ*, and *CSI* are mainly used for comprehensive ecological risk assessment of composite heavy metal pollution. Although the evaluation degrees of these three indicators are not completely consistent, their effects are basically the same, indicating that any of them can be utilized to meet the requirements rather than all of them. *mHQ* is used to evaluate the toxicity degree of each heavy metal to aquatic organisms, showing a significant positive linear correlation with the total content of each heavy metal, indicating that this index is not affected by sediment properties and has a wide range of applications.

4. Conclusions

Herein, risk assessment methods of sediment heavy metals calculated based on total content, background values, and SQGs were compared from the perspective of application

by substituted of actual monitoring data. The results showed that both I_{geo} and *EF* values for the risk assessment of single heavy metal enrichment decreased with the increase of background values. Compared with *EF*, I_{geo} also showed a significant positive linear correlation with heavy metal content, indicating that I_{geo} is not affected by geological factors and is suitable for the comparison of the same heavy metal in different regions. Considering the influence of sediment texture, *EF* is more suitable for the comparison of different heavy metals in the same sampling interval. *PLI*, mC_d, and *RI* showed significant differences in response to background values for the comprehensive risk of heavy metal enrichment, and the trends were $B_v < C_{vs} < C_{vd}$, $C_{vd} > B_v > C_{vs}$ and $B_v > C_{vd} > Cvs$, respectively. Although the evaluation levels of these factors were not identical, their distribution trends were basically the same along with the sampling points, indicating that they have equal evaluation effects. *TRI*, *mERMQ*, and *CSI* showed similar overall distribution trends, but their evaluation levels were not exactly the same, indicating that choosing any one of them can better reflect the toxicity of complex heavy metal pollution to aquatic organisms. Similar to I_{geo}, *mHQ* showed a significant positive linear correlation with the content of each heavy metal, indicating that it should not be affected by sediment properties and can be widely used to determine the toxicity of single heavy metals to aquatic organisms. The results of this study have important guiding significance for the selection of evaluation methods for heavy metal pollution in sediments. Further studies regarding the comparison with the method based on speciation content are needed to comprehensively identify the pollution status of heavy metals in sediments.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/w14010051/s1, Table S1. Classifications for index of geoaccumulation (I_{geo}). Table S2. Classifications for enrichment factor (*EF*). Table S3. Classifications for modified contamination degree (mC_d). Table S4. Sediment quality guidelines for metals in freshwater ecosystems that reflect TECs (below which harmful effects are unlikely to be observed) and PECs (above which harmful effects are likely to be observed), and toxicity coefficients (Tr_i) of heavy metals. Table S5. Classifications for potential ecological risk index (*RI*). Table S6. Classifications for toxic risk index (*TRI*). Table S7. Classifications for modified hazard quotient (*mHQ*). Table S8. Classifications for mean ERM quotient (*mERMQ*). Table S9. Classifications for contamination severity index (*CSI*). Table S10. The loading value, eigen value and w_i based on principal component analysis and factor analysis. Table S11. Heavy metal concentrations in surface sediments used as control values, all data in mean concentrations, dry weight, mg/kg. Figure S1. Vertical variations of Fe in sediments, all data in mean concentrations, dry weight, mg/kg. The boxes represent 25th and 75th percentiles, the middle horizontal lines represent the 50th percentile, the vertical line ends represent 1th and 99th percentiles, the small squares in the middle represent the mean value, and the diamond black dots represent outliers. Figure S2. The distribution of I_{geo} values for Cr, Ni, Cu, Zn, As, Cd, Tl, Pb and Hg in the whole investigated region based on different types of background values. Figure S3. The distribution of *EF* values for Cr, Ni, Cu, Zn, As, Cd, Tl, Pb and Hg in the whole investigated region based on different types of background values. Figure S4. The *PLI* values of Cr, Ni, Cu, Zn, As, Cd, Tl, Pb and Hg in sediments were calculated based on different types of background values and total concentrations. Figure S5. The mC_d values of Cr, Ni, Cu, Zn, As, Cd, Tl, Pb and Hg in sediments were calculated based on different types of background values and total concentrations. Figure S6. The *RI* values of Cr, Ni, Cu, Zn, As, Cd, Pb and Hg in sediments were calculated based on different types of background values and total concentrations. Figure S7. The distribution of the *TRI*, *mERMQ* and *CSI* values for Cr, Ni, Cu, Zn, As, Cd, Pb and Hg in sediments in the whole investigated region based on different types of SQG values. Figure S8. The distribution of the *mHQ* values for Cr, Ni, Cu, Zn, As, Cd, Pb and Hg in sediments in the whole investigated region based on different types of SQG values.

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