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Sediment Heavy Metal Pollution Assessment in Changwang and Wuyuan Rivers in Hainan Island, China

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Abstract: Heavy metal pollution may pose a significant threat to aquatic ecosystems. To assess heavy metal pollution, sediment samples were collected from Changwang and Wuyuan Rivers between June and December 2019. An inductively coupled plasma mass spectrometer was used to analyze the concentrations of As, Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn. The results revealed that Changwang River had significantly high concentrations of Cr (240.70 mg kg^{-1}), Co (36.02 mg kg^{-1}), Ni (108.70 mg kg^{-1}), and Cu (36.61 mg kg^{-1}), whereas As (7.55 mg kg^{-1}) was elevated in Wuyuan River. In addition, Cd, As, Pb and Zn concentrations were below China's sediment quality standard limits (GB 3838-2002), but the Cr level exceeded the limit. However, Cr and Ni exceeded the probable effect concentrations (PEC) and thus may cause toxic effects. The contamination factor, geo-accumulation index, and modified degree of contamination revealed that Changwang experienced considerable to very high heavy metal pollution, while Wuyuan had low to moderate pollution. The pollution load index demonstrated that the rivers were polluted during all seasons. Additionally, the risk index showed considerable and moderate risks in Changwang and Wuyuan, respectively. The metal ecological risk was ranked as Cd > Ni > As > Co > Cu > Cr > Mn > Pb > Zn. Multivariate analyses categorized heavy metals into two groups based on their potential sources: group one included Cr, Co, Ni, Cu, Mn, and Zn, while group two contained As, Cd, and Pb. The study provides valuable data on heavy metal pollution, which needs improvement for the studied rivers. The data can be used to assess pollution risks and manage riverine sediment quality.

Keywords: heavy metals; sediment quality; ecological risk; anthropogenic activities

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1. Introduction

Heavy metal pollution of rivers can originate from diverse pathways, including wastes from anthropogenic activities or naturally from soil erosion and weathering of rocks [1,2]. Globally, industrial, agricultural, and urban waste discharge have consistently contributed significant amounts of heavy metals to river ecosystems [3]. From 1970 to 2017, Li et al. [4] investigated the global heavy metal pollution of rivers and lakes. They found higher heavy metal pollution of rivers in Asia than in Europe, noting increasing Cd, Mn, Cu, Fe, Ni, and Cr concentrations. In China, several rivers are severely polluted by heavy metals, resulting from increased industrialization and agricultural activities [5,6]. Lian & Lee [7] examined sediment heavy metal pollution in 102 Chinese rivers. They found high Cd, Zn, Cu, Co, Cr, Mn, Ni, As, Hg, and Pb concentrations, indicating significant impacts of anthropogenic activities on the riverine sediments. Additionally, sediment heavy metal pollution has been extensively studied in many rivers in China. For example, studies have been conducted in the Xiangjiang River [8,9], Houjing River [10], Wei River Basin [11], Yellow River [12], Pearl River Delta [9], Changhua River Estuary [13], etc.

Sediments are important components of aquatic ecosystems being pollutants sink and providing nutrients to benthic organisms [14]. Thus, heavy metals discharged into the water bodies often settle at the bottom sediments [15]. In addition, sediments can adsorb heavy metals from the water column. Therefore, heavy metal concentrations in sediments typically exceed that in surface water [16]. Furthermore, aquatic ecosystems are threatened by heavy metal pollution and bioaccumulation, causing toxic effects [17]. Notably, the accumulation of heavy metals in water bodies may endanger aquatic organisms and pose risks to human health [18]. Heavy metal accumulation in sediments may also deteriorate water quality through pollution effects. As a result, heavy metals are vital indicators of water quality assessment [19]. Therefore, examining sediment heavy metal pollution can help understand the anthropogenic influence on the river ecology.

Generally, several studies have investigated sediment-heavy metal pollution of Hainan Island rivers. For example, Zhao et al. [5] reported unpolluted to very high sediment heavy metal pollution in eight Hainan Island rivers. In addition, Xu et al. [6] investigated the sediment quality of twenty Hainan Island rivers. Their findings revealed significant heavy metal pollution, with high levels reported in Nandu, Changhua, and Wanquan Rivers. The present study was conducted in Hainan Island in the Changwang and Wuyuan Rivers. These rivers are vital to the region as they provide water for farmland irrigation, livestock farms, and aquaculture production. As a result, they are polluted by agricultural, livestock, domestic, industrial, and aquaculture effluents. However, despite studies assessing several rivers in Hainan Island, Changwang and Wuyuan have yet to be examined. To bridge the information gap, this study (1) analyzed concentrations of Cd, Cu, Zn, Mn, As, Pb, Co, Cr, and Ni in riverine sediments; (2) assessed heavy metal pollution using individual and synergistic indices; (3) investigated the possible ecological risks caused heavy metal pollution, and (4) determined relationships between heavy metals using multivariate techniques to give insight into their potential sources. In addition, we hypothesized spatial and seasonal variations in heavy metal pollution due to the anticipated changes and variations in pollution load. Combining individual and synergistic methods provides a more comprehensive assessment of heavy metal pollution. Therefore, this study's findings help assess and control heavy metal pollution.

2. Materials and Methods

2.1. Study Area

Two rivers selected for this study included Changwang and Wuyuan, found in Haikou City, Hainan Island, China (Figure 1). Haikou is the capital city of Hainan province. It is highly populated, with approximately 2.3 million residents. Its climate varies from humid subtropical to tropical monsoon with warm annual temperatures. January and February are the coldest months when the temperature drops to 16–21 °C, whereas July and August are the hottest, with a temperature range between 25 and 29 °C. The studied rivers are located in the urban/semi-urban areas of Haikou City. They are, therefore, significantly impacted by anthropogenic activities, such as domestic, municipal, industrial, and agricultural waste pollution. Changwang River is located in the Longhua district in the Southern part of Haikou. It is about 22 km long, flowing from Zuntan to Xinpo town. The river drains through areas characterized by various agricultural, aquaculture, and livestock activities. On the other hand, the Wuyuan River is situated in the Western part of Haikou. It is famous because of the establishment of the Wuyuan River Stadium. Wastes from urban and sub-urban runoffs significantly pollute the river. Therefore, thirteen (13) sampling sites were selected in Changwang and Wuyuan Rivers for this study based on the potential of being polluted by the nearby land use and anthropogenic activities (Figure 1).

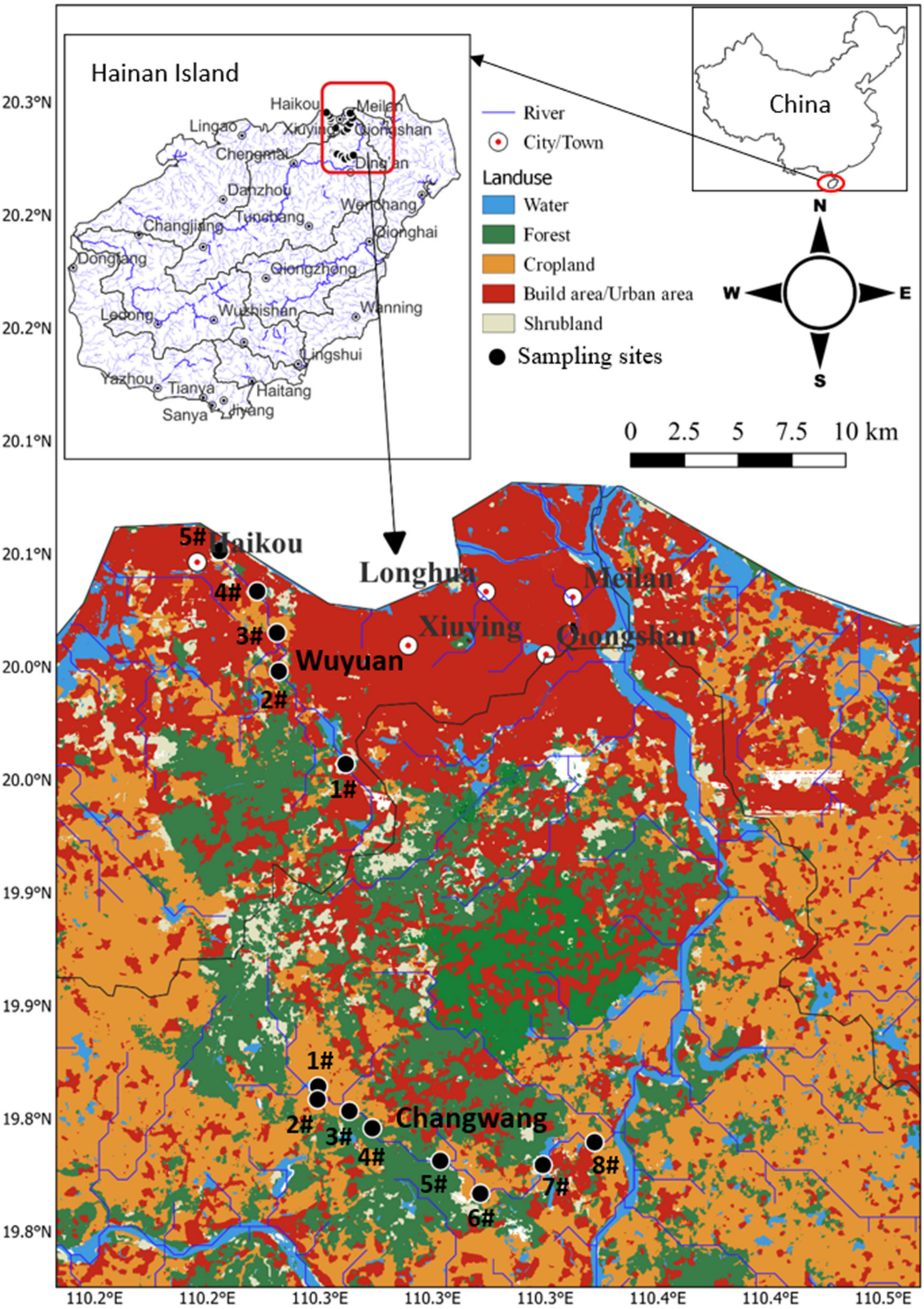

Figure 1. Study map showing sampling sites in Changwang and Wuyuan Rivers.

2.2. Samples Collection and Preparation

This study was conducted during June (Summer), September (Autumn), and December (Winter) 2019. A stainless-steel grab sampler was used to collect surface sediment samples (<3 cm). About 4–5 sub-samples were put in zip polyethylene and transported to

Hainan University, China, in an ice box at −20 °C. Then, the samples were dried at 70 °C in a hot air oven until a constant weight was achieved. After that, the samples were ground into powder using a mortar and sieved through a 0.15 mm stainless steel sieve.

2.3. Heavy Metals Analysis

An electric cooker was used to digest the prepared sediment samples. First, approximately 0.1 g of the sediment sample was dissolved in 5 mL nitric acid and heated at 180 °C for 3 h. Next, the dissolved sample was mixed with 2 mL each of nitric acid, hydrofluoric acid, and perchloric acid and heated at 180 °C for 5 h. Then, the mixture was acid-driven and reduced to about 1 mL. After that, 10 mL nitric acid (2%) was added, and the mixture was filtered through a 0.45 µm membrane before analysis. Finally, an inductively coupled plasma mass spectrometer (ICP-MS, Thermo scientific iCAP RQ) was used to analyze heavy metal concentrations, expressed as mg kg^{-1}, dry weight. Quality assurance and quality control (QA/QC) was based on the standard reference (GBW07307a). Additionally, standard internal solutions Rh and ln were employed for calibration. The precision of the analysis estimated by the coefficient of variation of three replicates was within 5%. The sample recovery rate for the analysis ranged from 98.2 to 103.7%.

2.4. Statistical Analysis

A two-way analysis of variance (ANOVA) was used to investigate the spatial and seasonal effects on heavy metal concentration. A simple main effect analysis was performed where there was a significant difference. In addition, the heavy metals' relationships were explored using Pearson's correlation coefficient analysis, cluster analysis, principal component analysis, and factor analysis. The analyses were done with SPSS version 28, R version 4.2.3, and Origin 2023.

3. Heavy Metal Pollution Assessment

3.1. Sediment Quality Assessments

Sediment quality guidelines (SQGs) help determine the toxic nature of heavy metals in aquatic ecosystems [20]. In addition, SQGs can be used to interpret sediment quality and identify pollutants of management concern. Therefore, consensus-based SQGs were utilized to analyze riverine sediment quality in this study [20]. The guideline comprises a threshold effect concentration (TEC) and a probable effect concentration (PEC). While values above PEC may indicate potential toxic effects, values below TEC suggest non-toxic effects. Therefore, the metals were evaluated based on the reference limits by calculating the sample percentage <TEC, TEC–PEC, and >PEC.

3.2. Geo-Accumulation Index and Contamination Factor

Individual indices, such as the geo-accumulation index (I_{geo}) and contamination factor (CF), are extensively employed to assess sediment heavy metal pollution [21]. The indices show the relationship between heavy metal in sediment and background values. Thus, they can reflect metal pollution from anthropogenic sources. The I_{geo} and CF are estimated using the sediment heavy metal concentration ratio to geochemical background reference values [22,23]. This study's heavy metals background values were obtained from Fu [24], who analyzed over 1000 deep soil samples from Hainan Island. The I_{geo} and CF values were estimated as follows.

$$I_{geo} = log_2\left(\frac{C_{si}}{1.5 \times C_{bi}}\right) \tag{1}$$

$$CF = \frac{C_{si}}{C_{bi}} \tag{2}$$

C_{si} is the measured heavy metal concentration in sediment, while C_{bi} is the background concentration of heavy metal, as presented in Table 1 [24].

Table 1. Spatio-temporal variation in mean (±SD) heavy metal concentration (mg kg^{-1}) in sediments and background values.

	Season	**Cr**	**Mn**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Pb**
Changwang	Autumn	255.93 ± 34.80	933.10 ± 230.01	40.70 ± 6.56	130.51 ± 24.60	43.27 ± 6.42	140.42 ± 31.70	3.48 ± 0.42	0.23 ± 0.04	19.18 ± 2.81
	Summer	252.17 ± 63.50	1221.37 ± 448.02	38.25 ± 11.10	107.34 ± 29.12	35.53 ± 8.72	97.52 ± 21.20	3.45 ± 0.80	0.14 ± 0.03	12.90 ± 3.23
	Winter	213.89 ± 46.30	764.84 ± 186.01	29.11 ± 6.97	88.36 ± 23.10	31.01 ± 5.99	88.00 ± 12.80	3.55 ± 0.78	0.18 ± 0.03	17.59 ± 4.20
	F-value	0.290	0.704	0.662	0.906	1.031	1.358	0.002	1.791	1.188
	p-value	0.750	0.502	0.522	0.414	0.368	0.271	0.998	0.183	0.317
Wuyuan	Autumn	75.02 ± 12.31	213.33 ± 42.20	8.05 ± 0.91	31.95 ± 3.27	17.40 ± 1.84	49.94 ± 6.05	6.20 ± 0.96	0.11 ± 0.02	16.51 ± 1.95
	Summer	114.08 ± 13.30	420.46 ± 142.02	12.90 ± 4.17	43.37 ± 11.90	24.67 ± 4.38	119.52 ± 51.70	12.89 ± 3.52	0.18 ± 0.05	22.02 ± 2.28
	Winter	160.80 ± 59.80	881.66 ± 413.01	23.29 ± 11.10	64.47 ± 27.90	26.40 ± 6.40	74.52 ± 18.40	3.57 ± 0.72	0.14 ± 0.04	13.55 ± 2.97
	F-value	0.618	0.966	0.673	0.346	0.383	1.355	1.712	0.650	1.286
	p-value	0.545	0.391	0.517	0.710	0.685	0.272	<0.001 *	0.529	0.290
Changwang	All seasons	240.70 ± 27.71	973.02 ± 176.01	36.02 ± 4.78	108.70 ± 14.60	36.61 ± 4.08	108.60 ± 13.71	3.49 ± 0.38	0.19 ± 0.02	16.55 ± 1.99
Wuyuan	All seasons	116.60 ± 21.40	505.04 ± 155.03	14.75 ± 4.06	46.62 ± 10.12	22.83 ± 2.67	81.31 ± 18.73	7.55 ± 1.55	0.14 ± 0.02	17.36 ± 1.60
	F-value	9.519	3.335	9.267	9.062	5.876	1.500	14.104	2.059	0.083
	p-value	0.004 *	0.077	0.005 *	0.005 *	0.021 *	0.229	<0.001 *	0.161	0.775
Background values [24]		27.52	163.52	4.03	7.24	6.10	44.43	1.34	0.04	24.36

Note(s): * Statistically significant difference at $p < 0.05$, ANOVA.

I_{geo} category: $I_{geo} < 0$ unpolluted, $0 \leq I_{geo} < 1$ unpolluted to moderately polluted, $1 \leq I_{geo} < 2$ moderately polluted, $2 \leq I_{geo} < 3$ moderately to heavily polluted, $3 \leq I_{geo} < 4$ heavily polluted, $4 \leq I_{geo} < 5$ heavily to extremely polluted, and $I_{geo} \geq 5$ extremely polluted. *CF* category: $CF < 1$ low pollution, $1 \leq CF < 3$ moderate pollution, $3 \leq CF < 6$ considerable pollution, and $CF \geq 6$ very high pollution.

3.3. Modified Degree of Contamination and Pollution Load Index

The modified degree of contamination (*mCd*) and the pollution load index (*PLI*) are synergistic indices for assessing heavy metal pollution. The *mCd* evaluates the contamination of sediments by multiple heavy metals [25]. In addition, *PLI* is a straightforward method for determining metal pollution levels [22]. Thus, these indices aggregate pollution caused by all the heavy metals. The *mCd* [26] and *PLI* [27] were determined as follows.

$$mCd = \frac{1}{n}\sum_{i=1}^{n} \times CF_i \tag{3}$$

$$PLI = (CF_1 \times CF_2 \times CF_2 \ldots\ldots CF_n)^{1/n} \tag{4}$$

where: *CF* is the contamination factor.

mCd category: $mCd < 1.5$ unpolluted, $1.5 \leq mCd < 2$ slightly polluted, $2 \leq mCd < 4$ moderately polluted, $4 \leq mCd < 8$ moderately to heavily polluted, $8 \leq mCd < 16$ heavily polluted, $16 \leq mCd < 32$ severely polluted, $mCd \geq 32$ extremely polluted. *PLI* category: $PLI < 1$ unpolluted and $PLI > 1$ polluted.

3.4. Potential Ecological Risk

The potential ecological risk index (*RI*) is typically employed to investigate heavy metals pollution risks to the aquatic environment [22,28]. Therefore, *RI* can help identify heavy metals of high ecological risk to inform management decisions [29]. The *RI* was computed using the following equations.

$$\mathrm{E_r^i} = \mathrm{T_r^i} \times \mathrm{C_f^i} = \mathrm{T_r^i} \times \frac{\mathrm{C_s^i}}{\mathrm{C_n^i}} \tag{5}$$

$$RI = \sum_{i=1}^{n} \mathrm{E_r^i} \tag{6}$$

where: $\mathrm{E_r^i}$ is the potential ecological risk factor of element *i*, $\mathrm{T_r^i}$ is the toxic response factor of element *i*. The $\mathrm{T_r^i}$ for Cd (30), Cr (2), Mn (1), Co (5), Ni (5), Cu (5), Zn (1), As (10), and Pb (5) were obtained from Zhang et al. [9]. *RI* category: $RI < 150$ low risks, $150 \leq RI < 300$ moderate risks, $300 \leq RI < 600$ considerable risks, and $RI \geq 600$ very high risks.

4. Results and Discussion

4.1. The Concentration of Heavy Metals

Figure 2 presents sediment heavy metals concentration for the sampling sites. Chan 2# and Chan 4# had the highest concentration of metals, whereas Chan 7# and Wuy 3# had the lowest levels. Mn was the dominant metal recording the highest concentration, while Cd concentration was the lowest. Two-way ANOVA indicated a significant spatial difference in heavy metal concentration ($F = 5.012$, $p < 0.001$) but not seasonally ($p > 0.05$). The results suggest a significant spatial influence on heavy metal pollution. There were significant spatial differences in Cr ($F = 9.52$, $p = 0.004$), Co ($F = 9.27$, $p = 0.005$), Ni ($F = 9.06$, $p = 0.005$), Cu ($F = 5.88$, $p = 0.021$), and As ($F = 14.10$, $p < 0.001$). As a result, Changwang River had significantly high concentrations of Cr (240.70 ± 27.71 mg $\mathrm{kg^{-1}}$), Co (36.02 ± 4.78 mg $\mathrm{kg^{-1}}$), Ni (108.70 ± 14.60 mg $\mathrm{kg^{-1}}$), and Cu (36.61 ± 4.08 mg $\mathrm{kg^{-1}}$), while As (7.55 ± 1.55 mg $\mathrm{kg^{-1}}$) was significantly high in Wuyuan River (Table 1).

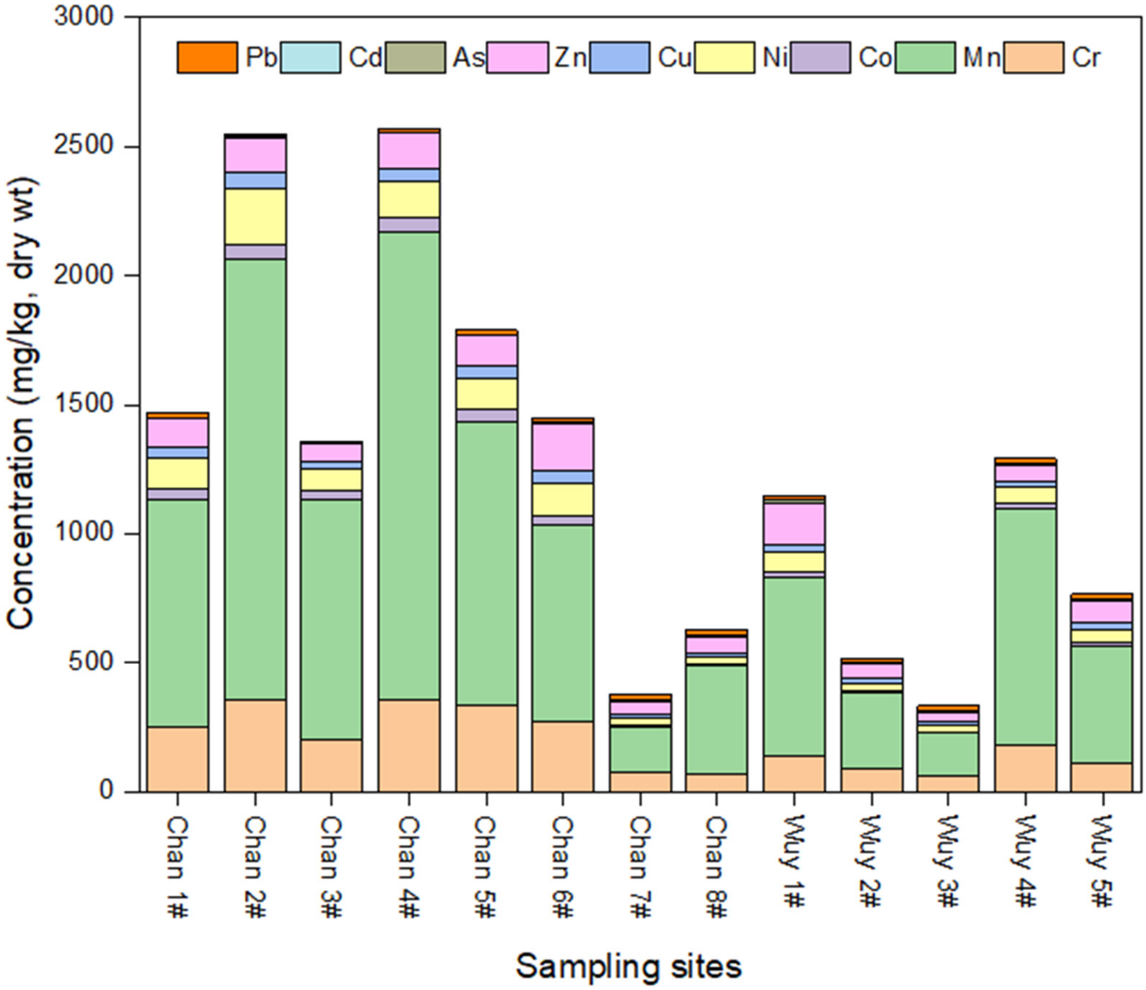

Figure 2. Heavy metals concentration in sediments. Chan (Changwang) and Wuy (Wuyuan).

According to the findings of this study, the elevated heavy metal concentration in Changwang River was primarily caused by fertilizer and chemical runoff from the nearby croplands. It can be noticed that the Changwang River drains through the agricultural areas (Figure 1). In addition, principal component analysis (PCA) explored the spatial and seasonal variation in heavy metal concentrations. The PCA separated the rivers (Figure 3a) but not the seasons (Figure 3b). Components 1 and 2 explained 56.9 and 25.1% of the total variations. Increasing Co, Mn, Cr, Ni, Cu, and Zn constrained Changwang River. In contrast, the Wuyuan River was influenced by increasing Pb, As, and Cd. Thus, the PCA results also revealed a stronger spatial effect than a seasonal effect on heavy metal pollution. Furthermore, the average concentration of all metals, except Pb, exceeded their background values (Table 1), suggesting anthropogenic input of heavy metals into the rivers. Compared with the SQGs, Cr, Ni, Cu, Zn, As, and Pb concentrations in 33.33%, 30.77%, 51.28%, 28.21%, 7.69%, and 5.13% of the samples were between TEC and PEC (Table 2). However, Cr and Ni concentrations in 58.97% and 51.28% of samples exceeded PEC, suggesting these metals may cause toxic effects. Meanwhile, As, Cd, Pb, and Zn concentrations were below China's sediment quality standard limit [30], whereas the Cr level was elevated. In addition, Cu concentration in Changwang River was slightly above China's sediment quality standard limit.

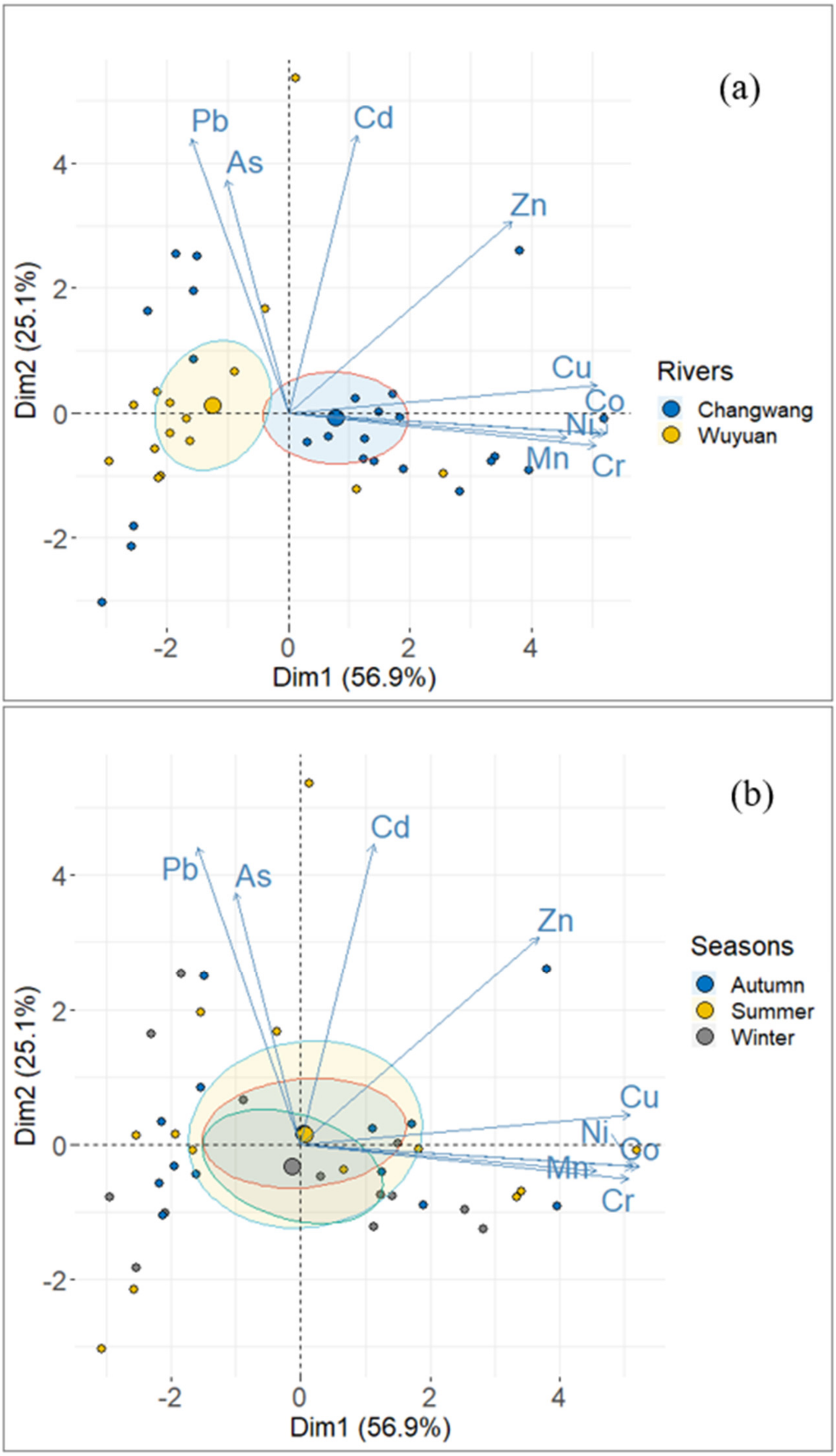

Figure 3. Principal component analysis of heavy metals based on: (**a**) rivers and (**b**) season.

Table 2. Sediment quality guidelines, China's primary standard, mean metal concentrations (mg kg^{-1}), and comparison with other rivers.

	Cr	**Mn**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Pb**	**Reference**
Changwang, Hainan Island	240.70	973.02	36.02	108.70	36.61	108.61	3.49	0.19	16.55	This study
Wuyuan, Hainan Island	116.60	505.04	14.75	46.62	22.83	81.31	7.55	0.14	17.36	This study
TEC	43.40	-	-	22.70	31.60	121.00	9.79	0.99	35.80	[20]
PEC	111.00	-	-	48.60	149.00	459.00	33.00	4.98	128.00	[20]
% of samples < TEC	7.69	-	-	17.95	48.72	71.79	92.31	100.00	94.87	This study
% of samples between TEC-PEC	33.33	-	_	30.77	51.28	28.21	7.69	0.00	5.13	This study
% of samples > PEC	58.97	-	-	51.28	0.00	0.00	0.00	0.00	0.00	This study
Primary standard, China	80.00	-	-	-	35.00	150.00	20.00	0.50	60.00	[30]
20 Hainan Island rivers	70.40	-	-	27.20	58.40	129.50	15.50	0.60	54.40	[6]
8 Hainan Island rivers	56.48	-	-	-	33.35	102.10	8.79	0.33	43.44	[5]
Changhua River Estuary	53.10	-	-	-	15.00	73.70	9.51	0.09	27.00	[13]
Pearl River Estuary	118.10	-	-	-	81.00	140.00	33.10	5.60	105.90	[3]
Xiangjiang River	20.44	1805.17	23.19	57.14	101.36	443.32	54.90	13.68	214.91	[8]
Yangtze River Estuary	52.10	-	-	-	28.00	78.00	11.60	0.20	21.90	[31]
Jialu River	60.80	-	-	42.44	39.22	107.58	6.31	2.93	29.35	[32]
Liaohe River	35.06	-	-	17.73	17.82	50.24	9.88	1.20	10.57	[33]
Bortala River	51.55	-	-	22.32	30.09	99.19	9.67	0.17	31.98	[34]
Zijiang River	67.51	1322.89	16.76	34.66	34.19	141.90	31.53	3.0	35.68	[9]

Note(s): TEC: a threshold effect concentration. PEC: a probable effect concentration. 8 Hainan rivers: Nandu, Wenlan, Beimen, Zhubi, Changhua, Ningyuan, Lingshui, and Wanquan [5]. 20 Hainan rivers: Wenjiao, Nandu, Wenlan, Beimen, Chun, Paipu, Zhubi, Changhua, Ganen, Nangen, Baisha, Wanglou, Ningyuan, Tengjiao, Shentian, Lingshiu, Longtou, Longguan, Wanquan, and Jiuqu [6].

This study's heavy metal concentration was compared with those from Hainan Island and other studies (Table 2). The concentration of Cd, Zn, As, Pb, and Cu were comparable with most previous studies but lower than those in 20 Hainan Island rivers [6], Xiangjiang River [8], Pearl River Estuary [3], and Zijiang River [9]. Moreover, Cu (36.61 mg kg^{-1}) and Zn (108.61 mg kg^{-1}) concentrations in Changwang River were comparable with 8 Hainan Island rivers [5] but higher than Changhua River Estuary [13], Yangtze River Estuary [31], Liaohe River [33], and Bortala River [34]. In contrast, Ni (108.70 mg kg^{-1}), Cr (240.70 mg kg^{-1}), and Co (36.02 mg kg^{-1}) concentrations in Changwang River exceeded other studied rivers (Table 2). Notable, Mn concentrations in Changwang River (973.02 mg kg^{-1}) and Wuyuan River (505.04 mg kg^{-1}) were significantly low compared to Zijiang River [9] and Xiangjiang River [8]. The varying anthropogenic impacts may be responsible for the spatial changes in heavy metal pollution.

4.2. Heavy Metals Pollution Assessment

4.2.1. Geoaccumulation Index and Contamination Factor

The I_{geo} values varied significantly in the studied rivers (Figure 4a). In Changwang River, the average I_{geo} values of Cr (2.02 ± 1.73), Ni (2.52 ± 2.60), and Co (1.72 ± 2.73) indicated moderate to heavy pollution ($I_{geo} < 3$), whereas Cd (1.23 ± 1.58), Cu (1.44 ± 2.02), and Mn (1.08 ± 2.72) indicated moderate pollution ($I_{geo} < 2$). Meanwhile, the average I_{geo} values of As (0.24 ± 2.36) and Zn (0.25 ± 1.68) reflected unpolluted to moderate pollution levels ($I_{geo} < 1$). In Wuyuan River, the average I_{geo} values of As (1.57 ± 1.04), Cd (1.04 ± 0.79), Cr (1.22 ± 0.91), Cu (1.15 ± 0.77), and Ni (1.71 ± 1.09) indicated moderate pollution, whereas Co (0.75 ± 1.20), Mn (0.37 ± 1.38), and Zn (−0.08 ± 1.02) indicated unpolluted to the moderate pollution level. Notably, Pb had unpolluted levels ($I_{geo} < 0$) with an average I_{geo} of −1.48 ± 1.24 and −1.17 ± 0.57 in Changwang and Wuyuan, respectively (Figure 4a).

Figure 4b presents *CF* results, which are consistent with those of I_{geo}. In Changwang River, the average *CF* values of Ni (15.02 ± 9.91), Co (8.94 ± 5.81), Cr (8.74 ± 4.93), and Cu (6.00 ± 3.28) reflected very high pollution levels ($CF \geq 6$), whereas Cd (4.64 ± 2.48) and Mn (5.95 ± 5.26) indicated considerable pollution ($CF < 6$). Meanwhile, average *CF* values of Zn (2.45 ± 1.51) and As (2.61 ± 1.39) indicated moderate pollution ($CF < 3$), while Pb (0.68 ± 0.40) showed a low pollution level ($CF < 1$). In Wuyuan River, the average *CF* value of Ni (6.44 ± 5.39) showed a very high pollution level, whereas Cr (4.24 ± 3.02), As (5.64 ± 4.49), Cu (3.74 ± 1.70), Cd (3.55 ± 2.05), Co (3.66 ± 3.90), and Mn (3.09 ± 3.66) reflected considerable pollution ($CF < 6$). In contrast, the *CF* value of Zn (1.83 ± 1.63) indicated moderate pollution, whereas Pb (0.71 ± 0.26) indicated a low pollution level. Generally, I_{geo} and *CF* results suggest that Changwang River experienced considerable to very high pollution, while Wuyuan River had low to moderate heavy metal pollution.

This study's results conform with the previous findings of Xu et al. [6], who found high As, Cu, and Cd pollution, moderate Ni pollution, and unpolluted Zn, Cr, and Pb levels in 20 Hainan Island rivers. Nevertheless, Zhao et al. [5] reported unpolluted to moderately polluted As, Cu, Cr, Cd, Zn, and Pb in 8 Hainan Island rivers. Meanwhile, Chai et al. [8] found severe Cd and moderate Mn, Cu, Pb, As, and Zn pollution in the Xiangjiang River, China. In addition, the Zijiang River in China witnessed severe Sb pollution and moderate to heavy Cd, Co, Mn, As, and Pb pollution [9]. However, the Lishui River in Southern China experienced moderate Mn and Pb pollution and unpolluted levels of Cr, Co, Ni, Cu, and Zn [35]. Furthermore, previous studies of riverine sediments also observed the unpolluted Pb, Ni, Cr, Cu, and Zn in Houjing River, China [10], Koshi River, Nepal [4], and Meghna River Estuary, Bangladesh [36].

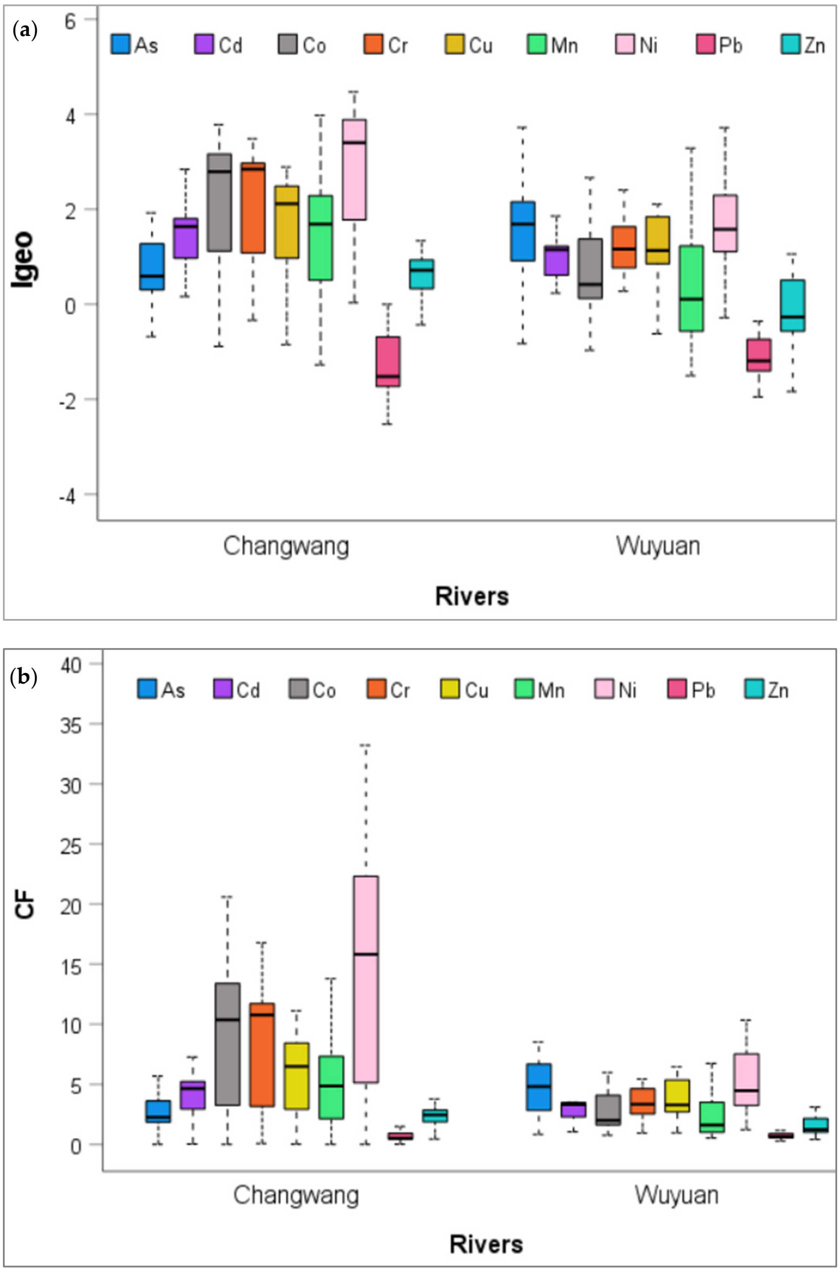

Figure 4. Heavy metal (**a**) geoaccumulation index (I_{geo}) and (**b**) contamination factor (*CF*) in riverine sediments.

4.2.2. Modified Degree of Contamination and Pollution Load Index

As shown in Figure 5, the average *mCd* varied from 2.22 to 9.68. It could be noted that most sites in the Changwang River had significantly higher *mCd* values than the Wuyuan River. The *mCd* results presented in Figure 5 indicated that 5 sampling sites were moderately polluted ($mCd < 4$), 6 sites were moderate to heavily polluted ($mCd < 8$), and 2 sites (e.g., Chan 2# and Chan 4#) were heavily polluted ($mCd > 8$). The observation concurs with heavy metal concentration results (Figure 2), as Chan 2# and Chan 4# recorded the highest metal concentration. Likewise, the pollution status demonstrated by *mCd* also conforms with those of I_{geo} and *CF* discussed above.

Figure 6 illustrates the seasonal *PLI* variation. The average *PLI* varied from 3.58 to 4.75 and 2.05 to 3.34 in Changwang and Wuyuan Rivers, respectively, indicating pollution during all seasons ($PLI > 1$). Notably, Changwang River recorded high *PLI* values during Autumn (4.75) and Summer (4.08). The findings agree with Ali et al. [17], who also reported high metal pollution in the Karnaphuli River, Bangladesh, during summer. Similarly, the results corroborate with Xu et al. [6], who found high metal pollution (average *PLI* = 6.3) in 20 Hainan Island rivers. However, Zhao et al. [5] found unpolluted to considerable pollution in sediments from the 8 Hainan Island rivers. In addition, Chai et al. [8] reported high metal pollution in the Xiangjiang River, China. Likewise, Hoang et al. [10] found high metal pollution in Houjing River, China (average *PLI* = 2.1). In contrast, the Lishui River, China (*PLI* = 0.48–1.68) experienced low metal pollution [35]. In addition, unpolluted levels were reported by Li et al. [37] in the Koshi River, Nepal (average *PLI* = 1.01), and Siddique et al. [36] in the Meghna River Estuary, Bangladesh (*PLI* = 0.09–0.45).

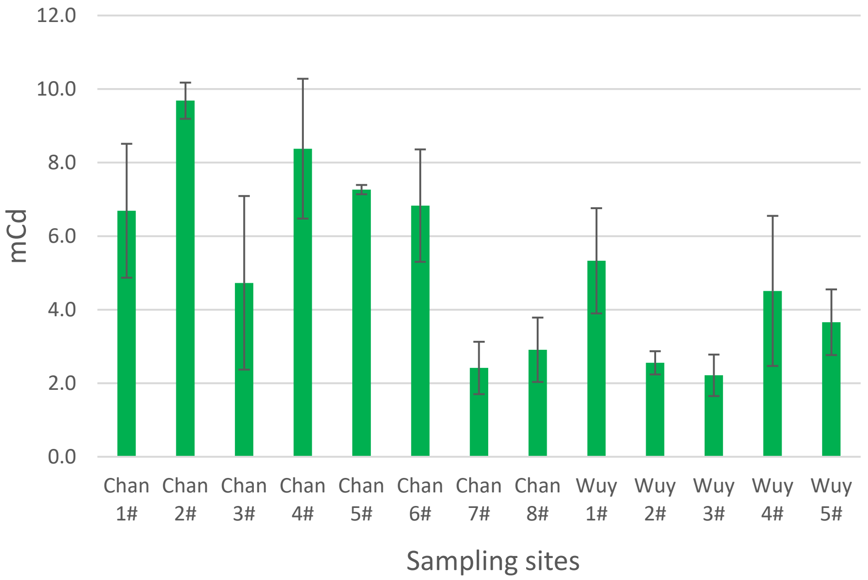

Figure 5. Modified degree of contamination (*mCd*) of heavy metals in sediments. Chan (Changwang) and Wuy (Wuyuan).

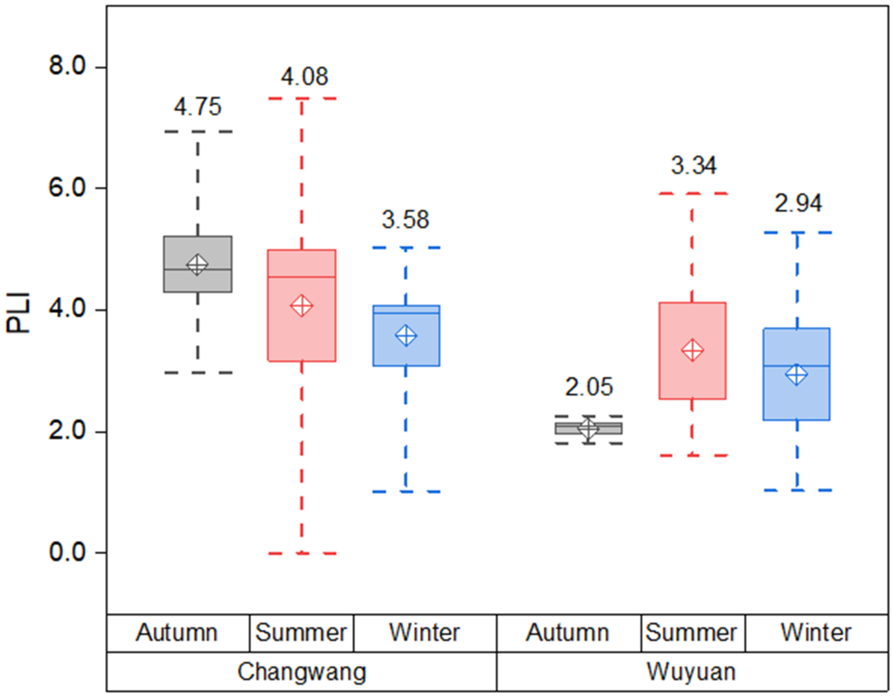

Figure 6. Seasonal pollution load index (*PLI*) variation in the riverine sediments.

4.3. Potential Ecological Risks

Figure 7 presents heavy metal potential ecological risks. Generally, *RI* varied from 158.78 to 429.92. Therefore, based on *RI* classifications, 6 and 7 sampling sites experienced considerable and moderate risks, respectively (Figure 7). Interestingly, the distribution of *RI* followed an almost similar trend with the *mCd* demonstrated in Figure 6. Therefore, most of the sampling sites in Changwang River, which had high *mCd* values, also experienced considerable risks. In contrast, all Wuyuan River sampling sites except Wuy 1# experienced moderate risk. These results suggest that the Changwang River exhibited high pollution risks than the Wuyuan River. Notably, the *RI*, I_{geo}, *CF*, *mCd*, and *PLI* values were generally higher in the Changwang River than in the Wuyuan River. Therefore, as discussed earlier, the significant heavy metal pollution in the Changwang River is mainly caused by fertilizer and chemical runoff from the nearby croplands. These findings could be supported by the high levels of heavy metals recorded here.

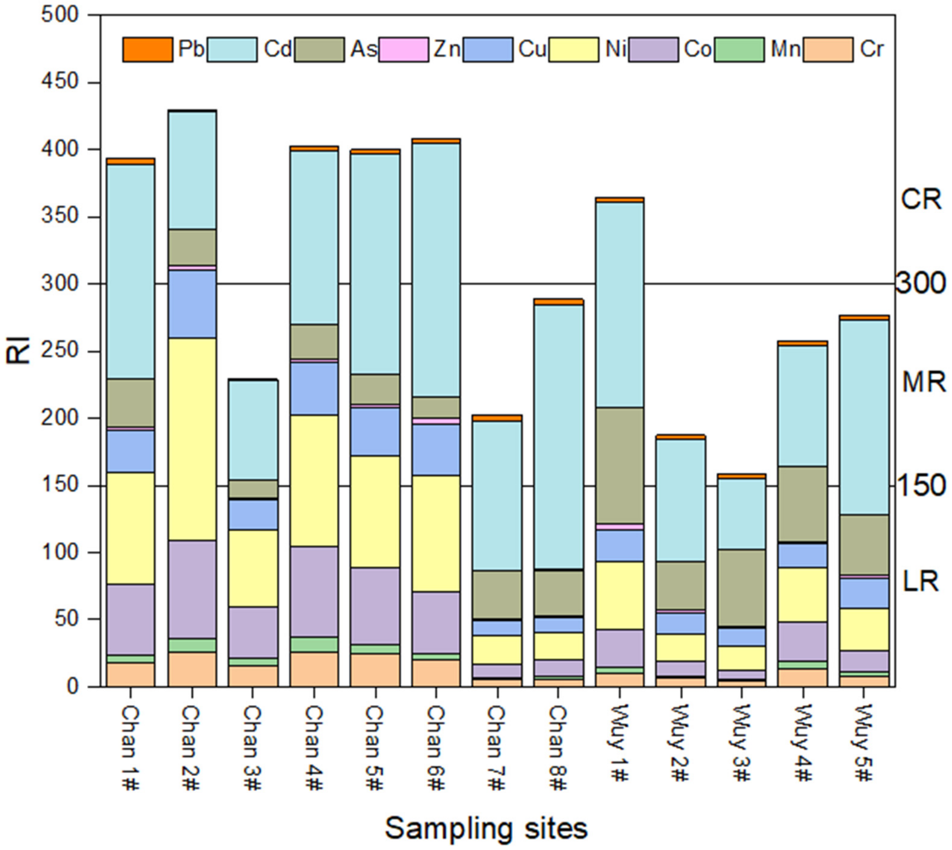

Figure 7. Spatial variation in the ecological risk index (*RI*) for heavy metals in sediments. CR (considerable risk), MR (moderate risk), LR (low risk), Chan (Changwang), and Wuy (Wuyuan).

Additionally, the heavy metals risk factors (E_r^i) followed the descending order Cd (126.51) > Ni (58.59) > As (37.72) > Co (34.54) > Cu (25.66) > Cr (14.02) > Mn (4.85) > Pb (3.46) > Zn (2.21). As shown in Figure 7, Cd posed the highest potential ecological risk, despite having the lowest concentration. In contrast, with extremely high concentrations, Mn posed no significant ecological risks. This observation is probably attributable to the faster dissolution and transportation of Cd [3]. Therefore, the results of potential ecological risk suggest controlling heavy metal pollution, especially for Cd. The findings of this study concur with previous studies. For example, Zhang et al. [9] reported that Cd and Sb posed significant ecological risks, while Mn had the lowest risk in the Zijiang River. In addition, Chai et al. [8] found that Cd caused severe ecological risk in the Xiangjiang River, while Mn induced low risk.

4.4. Heavy Metal Pollution Potential Sources

The potential sources usually influence the distribution and variation in sediment's heavy metal concentration [11,13,21]. Multivariate statistical methods, such as Pearson's correlation coefficient analysis, factor analysis, principal component analysis, and cluster analysis, are vital for assessing heavy metals and identifying pollution sources in the riverine environment [5,21,38]. In addition, multivariate approaches are useful for data reduction, investigating spatial and temporal changes, and clustering purposes [5,39].

Therefore, multivariate techniques were employed in this study to help give insight into heavy metal pollution sources. Figure 8 presents the results of the hierarchical cluster analysis. The sampling sites were categorized into two clusters (vertical dendrogram). Cluster 1 comprised sites in the Wuyuan River, except for Chan 7# and Chan 8#. These sites had similar pollution levels, characterized by high As, Cd, Pb, and Zn concentrations. In contrast, cluster 2 comprised the Changwang River sites, associated with high Cr, Co, Ni, Cu, Mn, Zn, Cd, and Pb concentrations. Additionally, the horizontal dendrogram classified heavy metals into two clusters. It could be seen that cluster 1 comprised Cr, Co, Ni, Cu, Mn, and Zn, while cluster 2 comprised As, Cd, and Pb. Interestingly, these findings agree with Pearson's correlation coefficient results presented in Figure 9. Cr significantly positively correlated with Mn (r = 0.80), Co (r = 0.96), Ni (r = 0.91), Cu (r = 0.90), and Zn (r = 0.54) at $p < 0.01$, reflecting common pollution sources. In contrast, Pb significantly positively correlated with As (r = 0.49) and Cd (r = 0.64) but correlated negatively with the above metals, suggesting that Pb, As, and Cd have a common pollution source.

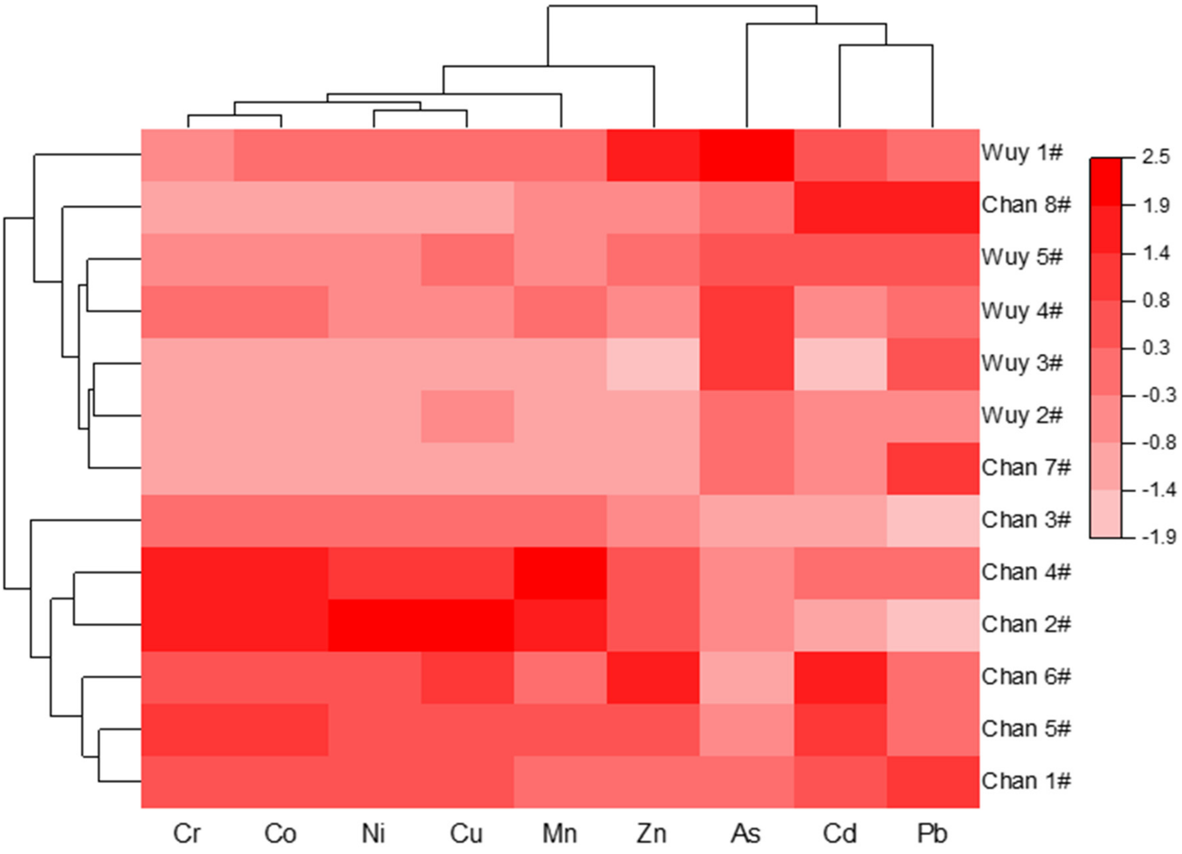

Figure 8. Dual hierarchical cluster analysis for the sampling sites and heavy metals.

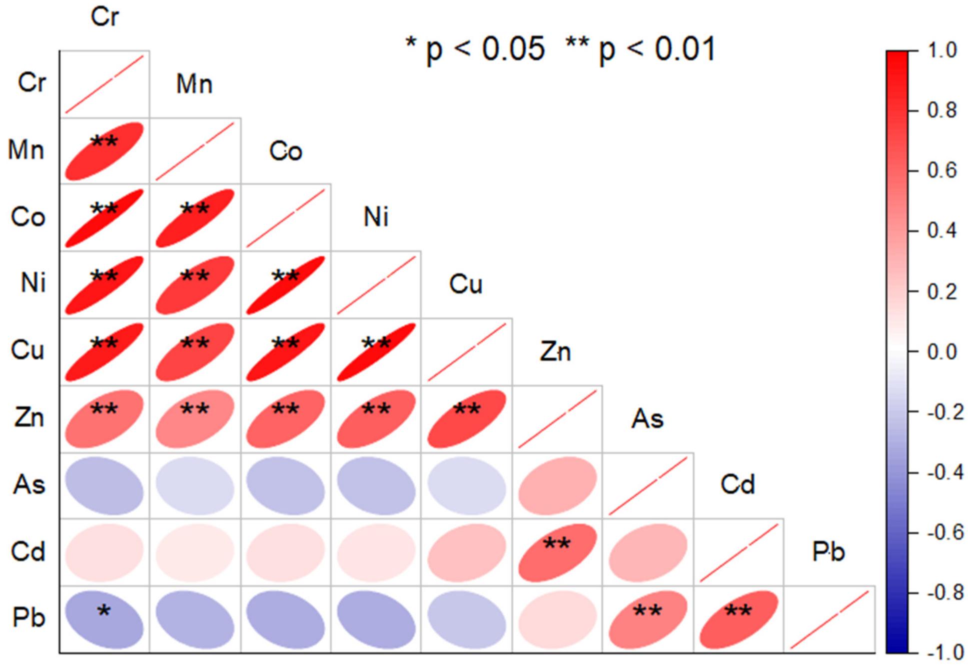

Figure 9. Heavy metals Pearson correlation coefficients analysis.

Furthermore, PCA/FA investigated the relationship between heavy metals. The Kaiser-Meyer-Olkin (KMO) value of 0.711 and Bartlett's test ($p < 0.001$; $\chi^2 = 420.186$) indicated the PCA validity. Table 3 summarizes the rotated PCA results, while Figure 10 illustrates the component plot. The analysis produced two components (eigenvalue >1.0), which explained 81.91% of the variation. Component 1, explaining 56.8% of the total variance, showed positive loading for Cr, Co, Ni, Cu, Mn, and Zn. In contrast, Component 2, responsible for 25.1% of the total variance, had positive loading for As, Cd, and Pb. These results also agree with cluster analysis and Pearson correlation results discussed above, confirming that these metals could be grouped into two categories based on their pollution sources. Anthropogenic activities, including agricultural fertilizers, electroplating wastes, and industrial and domestic discharge, are possible sources of heavy metals [38]. Generally, Cr, Cd, Pb, and Zn originate from agricultural runoff, fertilizers, and wastes from the tannery, textile, and chemical industries [40]. Therefore, this study's results might reflect pollution from diverse sources. As discussed earlier, pollutants greatly influence these rivers from industrial, agricultural, aquaculture, and livestock production.

Table 3. Rotated component matrix of heavy metals in riverine sediments.

Heavy Metal	Component 1	Component 2	Communalities
Cr	0.949	−0.128	0.917
Mn	0.858	−0.104	0.748
Co	0.982	−0.094	0.973
Ni	0.965	−0.093	0.940
Cu	0.960	0.053	0.924
Zn	0.710	0.555	0.812
As	−0.166	0.709	0.530
Cd	0.233	0.834	0.750
Pb	−0.271	0.840	0.779
Eigenvalues	5.112	2.259	
% Variance	56.804	25.105	
% Cumulative variance	56.804	81.909	

Note(s): Extraction Method: Principal Component Analysis. Rotation Method: Varimax with Kaiser Normalization. Rotation converged in 3 iterations.

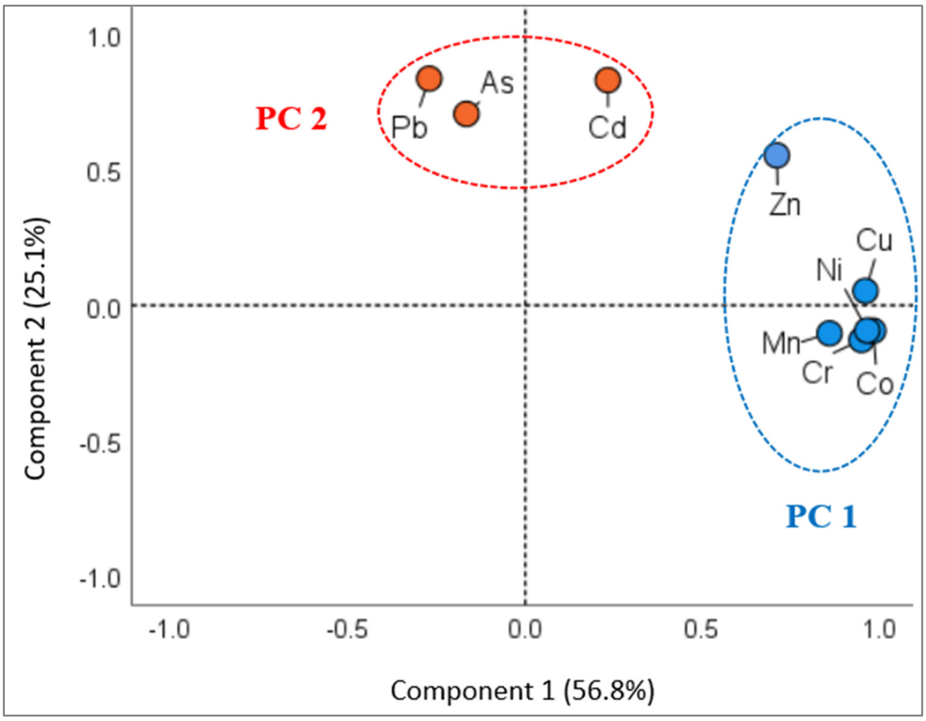

Figure 10. The plot of heavy metal component loadings in sediment. PC (Principal component).

Cd might originate from sources such as agriculture due to excessive fertilizer and pesticide application [34], thus accumulating in river sediments. Fertilizer applications also contribute to Cu enrichment in topsoil, which pollutes rivers through surface runoff. In addition, utilizing Co as a livestock feed additive could result in food chain Co accumulation, ultimately causing water pollution [41]. Meanwhile, Mn, As, and Pb are naturally occurring elements from the mining and weathering of rocks, soils, and sediments [42]. Previous work indicated that smelting and mining activities significantly contributed to As, Cd, and Pb pollution [9,43]. In summary, the multivariate techniques showed that Cr, Co, Ni, Cu, Mn, and Zn pollution could be attributable to anthropogenic sources such as industrial, domestic effluents, agricultural activities, etc. However, in addition to anthropogenic pathways, As, Cd, and Pb might have also originated from natural sources.

5. Conclusions

Sediment heavy metal pollution was examined using samples from the Changwang and Wuyuan Rivers in Hainan Island, China. Heavy metal pollution was assessed using different methods, such as the I_{geo}, *CF*, *mCd*, *PLI*, and *RI*. The Changwang River had elevated Cr, Co, Ni, and Cu levels, while the Wuyuan River had a high As level. The concentrations of Mn and Cd were the highest and lowest, respectively. According to I_{geo}, *CF*, and *mCd*, Changwang River experienced considerable to very high heavy metal pollution, while the Wuyuan River had low to moderate pollution. *PLI* showed that the rivers were polluted in all seasons. Sediment quality assessment revealed that Ni and Cr exceeded probable effect concentrations (PEC), indicating that the metals may cause toxic effects. The risk analysis revealed that Cd posed a significantly higher ecological risk than other metals. Additionally, the risk index showed that the Changwang River experienced considerable risk, while the Wuyuan River had moderate risk. All the multivariate analyses, including cluster analysis, Pearson's correlation coefficient, principal component, and factor analysis, unanimously categorized heavy metals into two groups based on their potential pollution sources. Group one comprised Cr, Co, Ni, Cu, Mn, and Zn, mostly from anthropogenic activities, while group two included As, Cd, and Pb originating from various pathways, including natural sources. This study provides a basis for strengthening the management measures to ensure riverine sediment quality. Generally, it is vital to ensure measures, such as efficient wastewater treatment, regulating excess fertilizer usage in agricultural farms, and restricting the discharge and dumping of wastes into the rivers. In addition, there is a need for further investigating the rivers' pollution to formulate appropriate control strategies.

Author Contributions: E.Y. and E.M. performed the experiment, wrote, and revised the main manuscript text. F.J. and D.W. collected and analyzed the samples. Z.G. and P.Z. conceptualized, designed, and supervised the study. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data for this study are available from the corresponding author upon reasonable request.

Conflicts of Interest: This manuscript is neither published nor under consideration for publication elsewhere. All the authors agree on submitting this manuscript, and they do not have any conflict of interest.

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