



# Distribution, source and pollution level of heavy metals in river sediments from South China

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## ARTICLE INFO

### Keywords:

Sediment pollution  
South China  
Geoaccumulation index  
Enrichment factor  
Pearson's correlation matrix  
Principal component analysis

## ABSTRACT

The sediment pollution caused by heavy metals has attracted a great deal of attention due to its persistence, bioaccumulation and toxicity. This research was the first to consider the whole of South China to obtain an overall profile of heavy metal spatial distribution, possible sources and pollution levels in river systems. For these data, 14 selected heavy metals were analysed in river sediments collected from sampling sites in Guangdong, Fujian, Guangxi and Hainan Provinces. The geoaccumulation index and enrichment factor revealed that river systems in South China were universally contaminated by Cd, As and Sn, which might be distributed by anthropogenic activities. Moreover, Guangdong Province, a relatively developed area in South China, was relatively polluted by certain heavy metals such as Ni, Cu, Zn and Mn. Multivariate statistical analyses such as Pearson's correlation matrix and a principal component analysis determined that several of the heavy metals might be derived from similar anthropogenic activities such as industrial effluents and domestic sewage discharge. In terms of heavy metal contamination in South China, necessary measures should be undertaken to protect rivers in South China.

## 1. Introduction

It is well known that rivers not only produce life but also multiply and generate human culture (Bhardwaj et al., 2017). Rivers perform a suite of ecological functions, such as water transport, aquaculture, habitat and shielding effects (Liao et al., 2016a). However, due to rapid industrialization, heavy metals have been discharged into rivers without effective purification (Zhang et al., 2017). Heavy metals have attracted much more attention on account of their inherent toxicity, vast sources, non-degradability, bioaccumulation and persistence in the aquatic environment (Gao et al., 2016; Paramasivam et al., 2015). Following the discharge of heavy metals into rivers, contamination can be distributed between different components of these aquatic systems, such as water, sediments and biota (Ali et al., 2016; Maanan et al., 2015). Consequently, only a small amount of the heavy metals remains in the water column, and the majority is deposited in the sediments (Malvandi, 2017). More specifically, heavy metals are bound to sediments through multiple mechanisms, including particle surface adsorption, ion exchange, co-precipitation and complexation with organic matter (Dong et al., 2014; Passos et al., 2010; Peng et al., 2009). River sediments serve as a reservoir or sink of heavy metals for aquatic

organisms (Chapman et al., 1998; Sundelin and Eriksson, 2001). Additionally, several of the sediment-bound metals can be released into the water column through sediment resuspension, desorption reactions, reduction or oxidation reactions (Dong et al., 2012; Feng et al., 2007; Zhao et al., 2013), and this release may be more hazardous to animal and human life via the food chain. Thus, sediments in the aquatic environment can play a significant role in the deposition and transmission of heavy metals.

Heavy metals in sediments can originate from both natural sources (e.g., geological weathering, atmospheric precipitation and erosion from wind, waves, storms and bioturbation) and anthropogenic activities (e.g., industrial discharge, mining, transportation, and agricultural and urban activities) (Feng et al., 2011; Keshavarzi et al., 2015; Sun et al., 2015). Thus, sediment quality serves as a useful parameter for characterizing the influence of natural sources and anthropogenic activities; furthermore, sediment quality can provide evidence of anthropogenic effects on ecosystems and direct the policy and management of the surrounding areas (Wang et al., 2014; Xu et al., 2014).

In many countries, such as the Member States of the European Union and Japan, effective management and restoration technologies have been established to protect local ecological environments and

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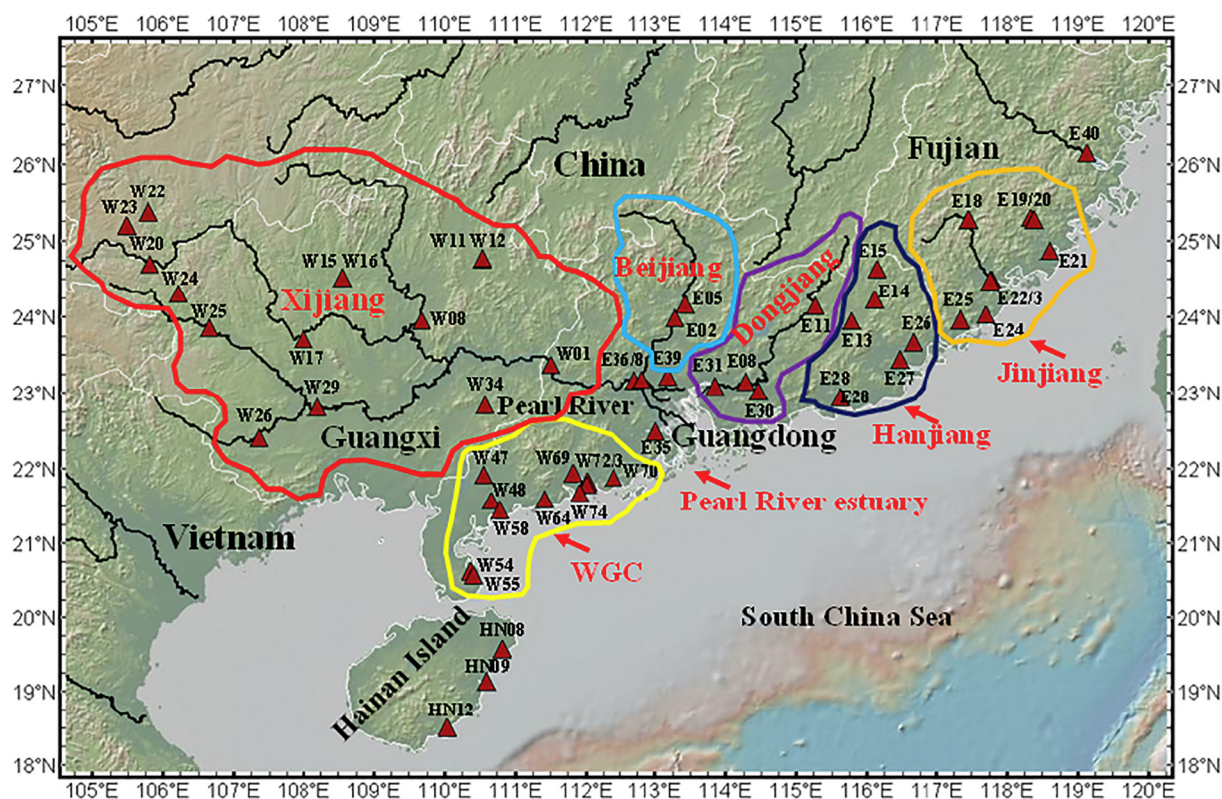


Fig. 1. Location of the study area and river sampling sites in Guangdong Province, Fujian Province and Guangxi Province, South China.

drinking water (Gu et al., 2012). Compared with other developing countries, China is currently suffering from great challenges with regard to heavy metal contamination due to rapid economic growth and intense industrialization (Wang et al., 2013; Wu et al., 2016). South China in particular, as one of the most rapidly developing regions in China, has undergone considerable environmental changes since the reform and opening-up policy of 1978, especially in local electronics industries that have generated a considerable amount of heavy metal contamination (Zhang et al., 2017). Thus, there is an urgent need to conduct research regarding heavy metal contamination in South China.

In this context, we selected several representative regions of South China as study areas, including Guangdong, Guangxi, Fujian and Hainan Provinces, which are shown in Fig. 1. Guangdong Province is a traditional industrial zone, while Guangxi, Fujian and Hainan Provinces are traditional agricultural regions. Studies on heavy metal pollution have demonstrated that rapid economic development will significantly affect the water quality of rivers (Wan et al., 2008; Zhou et al., 2007). Guangdong is the province with the most highly developed economy and the highest aquaculture production rate in China. The well-known Pearl River Delta (PRD), located in Guangdong Province (102°14'–115°53'E, 21°31'–26°49'N), is one of the most important regions for waterborne commerce (Yang et al., 2012). At present, numerous studies have focused primarily on the total content of heavy metals and pollution levels in the Pearl River, such as in the eight estuaries of the Pearl River (Ip et al., 2007; Wang et al., 2008; Yu et al., 2010), the Pearl River itself in the Guangzhou region (Li et al., 2009; Min et al., 2000) and so on; however, little information is available on the overall spatial distribution of heavy metal pollution in river systems over the entirety of Guangdong Province. Moreover, the rapid development of industries in Guangxi, Fujian and Hainan Provinces might also contribute to the degradation of the quality of their river systems, threatening the survival of aquatic life. Therefore, it is essential to determine the concentration of heavy metals and their potential hazardous risks in these river systems, on which little work has been

completed. Additionally, to gain a more comprehensive understanding, a profile of these three provinces that shows an overall spatial distribution of heavy metals should be developed.

In summary, few studies have considered South China as a whole to obtain an overall heavy metal spatial distribution in the river systems. Therefore, this study aims to address the research gap to provide worthwhile information on the spatial distribution and possible sources and pollution levels of heavy metals. In recent decades, a considerable number of indexes have been developed to assess heavy metal pollution levels, spatial distribution and source apportionment, such as the geoaccumulation index ( $I_{geo}$ ), enrichment factor (EF) and so on (Feng et al., 2011; Müller, 1979, 1981). The goals of this paper are as follows: (1) examine the spatial distribution of heavy metals in the rivers of South China; (2) assess heavy metal contamination using the  $I_{geo}$  and EF methods; and (3) identify the possible sources of these metals in river sediments from South China.

## 2. Sampling and methods

### 2.1. Study areas

The present study primarily examines the concentrations of heavy metals in South China, a region characterized by an uncoordinated economy; thus, we will discuss each area separately. The river study sites were distributed throughout South China and mainly comprised Guangdong, Guangxi, Fujian and Hainan Provinces (Fig. 1).

The river system in Guangdong Province generally comprises the Pearl River Basin, Hanjiang River Basin and coastal rivers of western and eastern Guangdong. As the second largest river in China, the Pearl River is a collection of all rivers and streams in the PRD, comprising three tributaries: the Dongjiang River, Beijiang River and Xijiang River. These three rivers comprise 89.6% of the total flow of the Pearl River. The Xijiang River is the main tributary that accounts for approximately 70.8% of the total annual flow. The Xijiang River flows throughout

Yunnan, Guizhou, Guangxi and Guangdong Provinces, finally terminating at the South China Sea near Macau. The Dongjiang River, originating in Jiangxi Province and primarily situated in Guangdong Province, is the major source of drinking water for large cities such as Hong Kong, Shenzhen, Guangzhou and so on. The Beijiang River flows from north to south through cities such as Shaoguan (a city well known for the large-scale production of nonferrous metals), Yinde, Qingyuan, Foshan and so on in the PRD in Guangdong Province. The Hanjiang River, the second largest river in Guangdong Province, covers a basin area of 30,112 km<sup>2</sup> with approximately 200 million residents. This river has also supported cultural heritage in the Chaoshan region, which comprises the cities of Chaozhou (a town with a lengthy history), Shantou (one of China's special economic zones) and Jieyang. The dense river network in these two basins provides favourable conditions for economic growth and social development in Guangdong Province, and this also applies to the rivers surrounding the western and eastern Guangdong coast.

The Xijiang River and drainage area in Guangxi Province, which form a critical channel connecting the southwestern PRD Economic Zone, comprise the Nanning–Liuzhou national priority development region. Moreover, the Xijiang River Economic Belt in Guangxi Province, which is located in the upstream section of the Xijiang River, serves as the critical ecological barrier for the downstream section of the Pearl River Delta. Thus, the ideal development of the Xijiang River Economic Belt occurs under these unique conditions.

The Jinjiang River is the third largest river in Fujian Province, with a total length of 182 km, a total drainage area of 5629 km<sup>2</sup> and a mean annual runoff of  $55.02 \times 10^8$  m<sup>3</sup>. The Dongxi and Xixi rivers are the two major tributaries of the Jinjiang River. As the primary drinking water source for 8 million residents in the region, the Jinjiang River flows through many counties and cities before finally reaching Quanzhou Bay, the famous starting point for the Maritime Silk Road of ancient China. Quanzhou Bay is adjacent to the intensely industrialized cities of Quanzhou, Jinjiang, Shishi and Hui'an, which comprise one of the fastest economically developing areas in Fujian Province.

Hainan Island, located in the northern Southern Sea, is the second largest tropical island in China. Hainan is one of the most famous tourist destinations due to the island's tropical beaches and lush forests. The island receives over 1.6 m of rainfall annually and is drained by mountainous rivers that are predominantly < 100 km in length. The Wanning River, located in the southern part of the island, is the largest river (350 km in length), while the Nandu River in the northern part of the island is 314 km long. There are relatively few factories in Hainan Province, and tourism plays a significant role in its economy.

The river system in South China is the main water source for industrial, agricultural and domestic supplies, and this system plays a significant role in the sustainable development of cities. Coinciding with the rapid development of industrialization and urbanization, there are increasing heavy metal contamination problems in South China. Therefore, based on the unique benefits for the health of local residents and drinking water quality, much greater efforts should be directed towards evaluating the pollution levels in the rivers of South China. To achieve this objective, there were 54 river sampling sites in this study; these sites are labelled with different letters and numbers to distinguish among them (Fig. 1). Table 1 shows the location and the description of each sampling site. Moreover, Fig. 2a, b and c show the upstream sections of the rivers in South China, while Fig. 2d shows the midstream sections of the rivers. The mangrove ecosystem in South China is presented in Fig. 2f.

## 2.2. Sediment sampling and pretreatment

Fifty-four surface sediment (0–4 cm) samples were collected using a box corer in the rivers of South China. All sub-samples were sealed separately in clean polyethylene bags, placed in a cooler at 4 °C, and transported to the Key Laboratory of Radiological and Interdisciplinary

Sciences in Soochow University.

The pretreatment involved converting the sediments into samples for analysis through the procedures of drying, grinding and digesting. Specifically, after freeze-drying, each of the sediment samples was heated in an oven at  $105 \pm 2$  °C to a constant weight. All the data in this paper were based on dry weights for further analyses. For the determination of total heavy metals, a 0.1-g dry weight sediment sample was first weighed on a mass balance and digested by an HCl–HNO<sub>3</sub>–HF–HClO<sub>4</sub> mixture in a Teflon vessel on a hot plate. The liquid samples were then evaporated to a near-dry state on the hot plate. Finally, the samples were adjusted to a suitable centrifuge tube with double deionized water and filtered with a membrane (0.45 μm) for testing (Liao et al., 2016b).

## 2.3. Analytical methods

The river sediments were ashed in a muffle furnace at 450 °C for 4 h to estimate the organic matter content (OM) (Bacardit et al., 2012). Furthermore, the total concentrations of the heavy metals vanadium (V), chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), gallium (Ga), arsenic (As), cadmium (Cd), antimony (Sb), lead (Pb), manganese (Mn), tin (Sn) and thallium (Tl) were determined by sector field inductively coupled plasma mass spectrometry (SF-ICP-MS, Finnigan Element 2, Bremen, Germany). All samples were analysed in triplicate to demonstrate reproducibility of the equipment. All standards were prepared from the dilution of 1000 mg L<sup>-1</sup> stock standard solutions in 5% (v/v) ultrapure nitric acid. The analytical procedure was checked with certified standards (GBW07314, GBW07437, GBW(E) 130286-89). The results were within the uncertainty interval specified by the certified values. The analytical blanks always produced values < 3% of the measured contents.

## 2.4. Calculations

The contamination levels of heavy metals in the sediment were assessed with  $I_{geo}$ , which was originally introduced by Müller (1969). The  $I_{geo}$  value was defined by the following equation:

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

where  $C_n$  is the measured concentration of metals (n) in the sediment, and  $B_n$  is the geological background (BG) concentrations of metals (n) in different provinces. To determine  $B_n$ , we consulted the yearbook BG values for the soil environment in China. The BG values (mg/kg) of select heavy metals in South China are summarized in Table 2. The constant term 1.5 is the background matrix correction factor that involves potential lithological variations in the background value. The  $I_{geo}$  value for each metal can be estimated and classified into seven classes: Class 0 ( $I_{geo} \leq 0$ ), uncontaminated; Class 1 ( $0 < I_{geo} \leq 1$ ), uncontaminated to moderately contaminated; Class 2 ( $1 < I_{geo} \leq 2$ ), moderately contaminated; Class 3 ( $2 < I_{geo} \leq 3$ ), moderately to heavily contaminated; Class 4 ( $3 < I_{geo} \leq 4$ ), heavily contaminated; Class 5 ( $4 < I_{geo} \leq 5$ ), heavily to extremely contaminated; and Class 6 ( $I_{geo} \geq 5$ ), extremely contaminated (Chowdhury et al., 2015; Varol, 2011).

The EF, which normalizes the concentration of a trace metal to that of a conservative element, has been used to further assess anthropogenic influences (Rahn and McCaffrey, 1979). Because the anthropogenic sources of Titanium (Ti) can be ignored and its behaviour is conservative (Bacardit et al., 2012), we selected Ti as the normalizing element. The EF is described with the following equation:  $EF = (C_M/Ti)_{sample} / (C_M/Ti)_{background}$ , where  $(C_M/Ti)_{sample}$  and  $(C_M/Ti)_{background}$  are the ratios of a given metal to Ti in the sediment sample and the background sample, respectively. The value of  $EF = 1.5$  was adopted as an assessment criterion. Generally, if the value of an EF is between 0.5 and 1.5, this suggests that heavy metals may be completely derived

Table 1 Heavy metal (mg/kg) concentrations and organic matter in sediments from all study sites in South China. WGC refers to rivers on the western Guangdong coast.

Table with 18 columns: Site, Longitude, Latitude, Province, Region, OM, V, Cr, Co, Ni, Cu, Zn, Ga, As, Cd, Sn, Sb, Tl, Pb, Mn. Rows list 42 study sites (E02 to HN12) with their respective metal concentrations and organic matter percentages.





Fig. 2. Photographs of the sampling sites in different areas of the river basin and the estuary.

from crustal material or natural weathering processes, and if an EF is  $> 1.5$ , this suggests that a significant portion of the heavy metal may originate from non-crustal material, such as that produced by anthropogenic activity.

### 2.5. Statistical analysis

To interpret mutual correlations among the internal metal concentrations in South China, a bivariate correlation analysis characterized by Pearson's correlation matrix (PCM) was performed with the statistical software SPSS 17.0. Additionally, to distinguish the hypothetical sources of heavy metals, a principal component analysis (PCA) was conducted using factor extraction with an eigenvalue  $\geq 1$  (SPSS 17.0) (Zhu et al., 2012).

## 3. Results and discussion

### 3.1. Total concentrations and spatial distribution of heavy metals

The concentrations of heavy metals at different sites in South China are summarized in Table 1. The metals in the sediments from Guangdong Province, the province with the most highly developed economy, ranged from 7.195–86.485 for V, 5.869–66.559 for Cr, 0.578–19.687 for Co, 2.364–77.787 for Ni, 3.955–35.320 for Cu, 22.658–178.903 for Zn, 1.149–27.316 for Ga, 5.830–133.623 for As, 0.012–2.420 for Cd, 1.193–109.294 for Sn, 0.147–2.929 for Sb, 0.062–0.580 for Tl, 9.093–83.827 for Pb, and 73.218–954.330 mg/kg for Mn. Metals in sediments from the whole of Guangdong Province were compared to other studies and BG values (Table 2). Manganese had the highest mean values in the study area for all sampling sites in Guangdong Province,

and it reached  $325.781 \pm 1.408$  mg/kg, whereas Cd had the lowest mean concentration ( $0.258 \pm 0.014$  mg/kg) throughout Guangdong, which was similar to the concentration of Cd in Lingdingyang (0.29 mg/kg) (Li et al., 2000). Moreover, the mean concentration of As was approximately 3 times higher than that of Zhanjiang Harbour (Guo and Huang, 2006). Lastly, the mean concentrations of Ni, Zn, Sb, Sn and Mn were 1.110, 1.272, 1.489, 3.171 and 1.168 times greater than the local soil BG value, respectively. These results indicated that the rivers throughout Guangdong Province had obtained relatively high heavy metal concentrations. As shown in Table 1, the concentrations of V, Cr, Co, Ni, Cu, Zn, Ga, As, Cd, Sn, Sb, Tl, Pb and Mn in Fujian Province ranged between 11.837 and 35.373, 4.322–18.122, 0.493–6.020, 1.169–4.610, 1.792–14.593, 17.182–111.131, 1.224–12.804, 16.535–112.825, 0.028–0.123, 7.652–39.994, 0.173–0.339, 0.122–0.343, 8.719–74.180 and 93.301–832.840 mg/kg, respectively. We observed that all heavy metal levels were within the range of BG values for Fujian Province, except for the mean concentrations of As and Sn, which were 7.8 and 3.3 times greater than the BG values, respectively. As described in Table 1, the concentrations of V, Cr, Co, Ni, Cu, Zn, Ga, As, Cd, Sn, Sb, Tl, Pb and Mn in Guangxi Province were 42.088–192.849, 11.932–40.470, 2.159–14.932, 7.290–38.542, 5.980–36.101, 37.742–109.380, 1.731–10.530, 26.010–163.306, 0.074–1.282, 7.320–33.630, 0.900–13.753, 0.140–0.879, 20.869–93.844 and 194.636–882.690 mg/kg, respectively. Compared with the BG values, low concentrations of investigated heavy metals, with the exception of Zn, As, Cd, Sn, Pb and Mn, were detected at sites in Guangxi Province. Due to limited data, the concentrations at the three sites (HN08, HN09 and HN12) in Hainan Province were only listed here without further analysis.

Additionally, a profile (Fig. 3) regarding the spatial distribution of

**Table 2**  
Background values (mg/kg) of selected heavy metals in South China.

Location	V	Cr	Co	Ni	Cu	Zn	Ga	As	Cd	Sn	Sb	Tl	Pb	Mn	Reference
Guangdong Province	65.3	50.5	7	14.4	17	47.3	13.9	8.9	0.056	5.8	0.54	0.682	36	279	MEP (2009)
Fujian Province	79.5	44	8.8	18.2	22.8	86.1	19.1	6.3	0.074	4.4	0.65	0.821	41.3	301	MEP (2009)
Guangxi Province	129.9	82.1	10.4	26.6	27.8	75.6	15.2	20.5	0.2670	3.30	2.93	0.7820	24.0	446	MEP (2009)



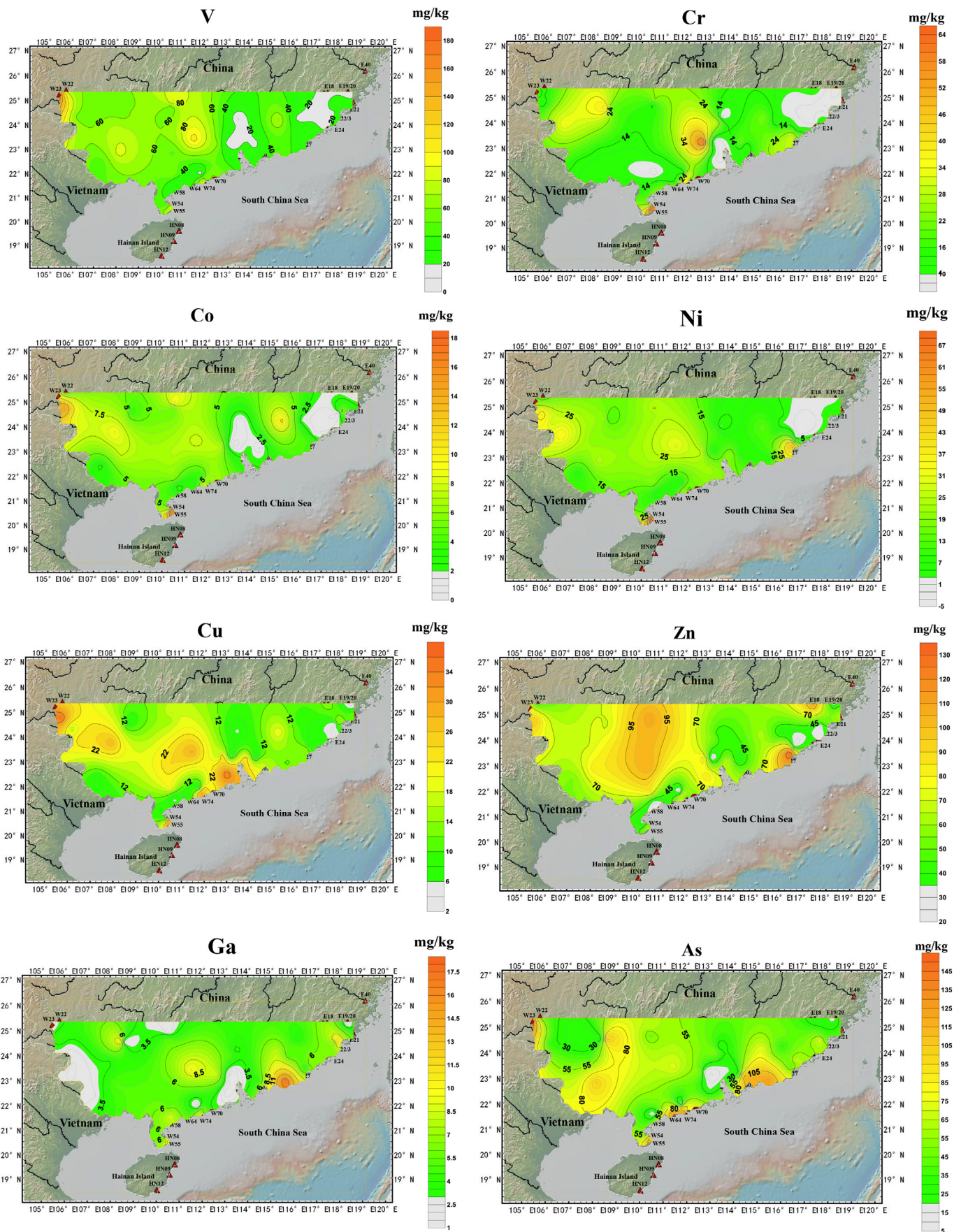


Fig. 3. Distribution of the concentrations of selected heavy metals in South China.



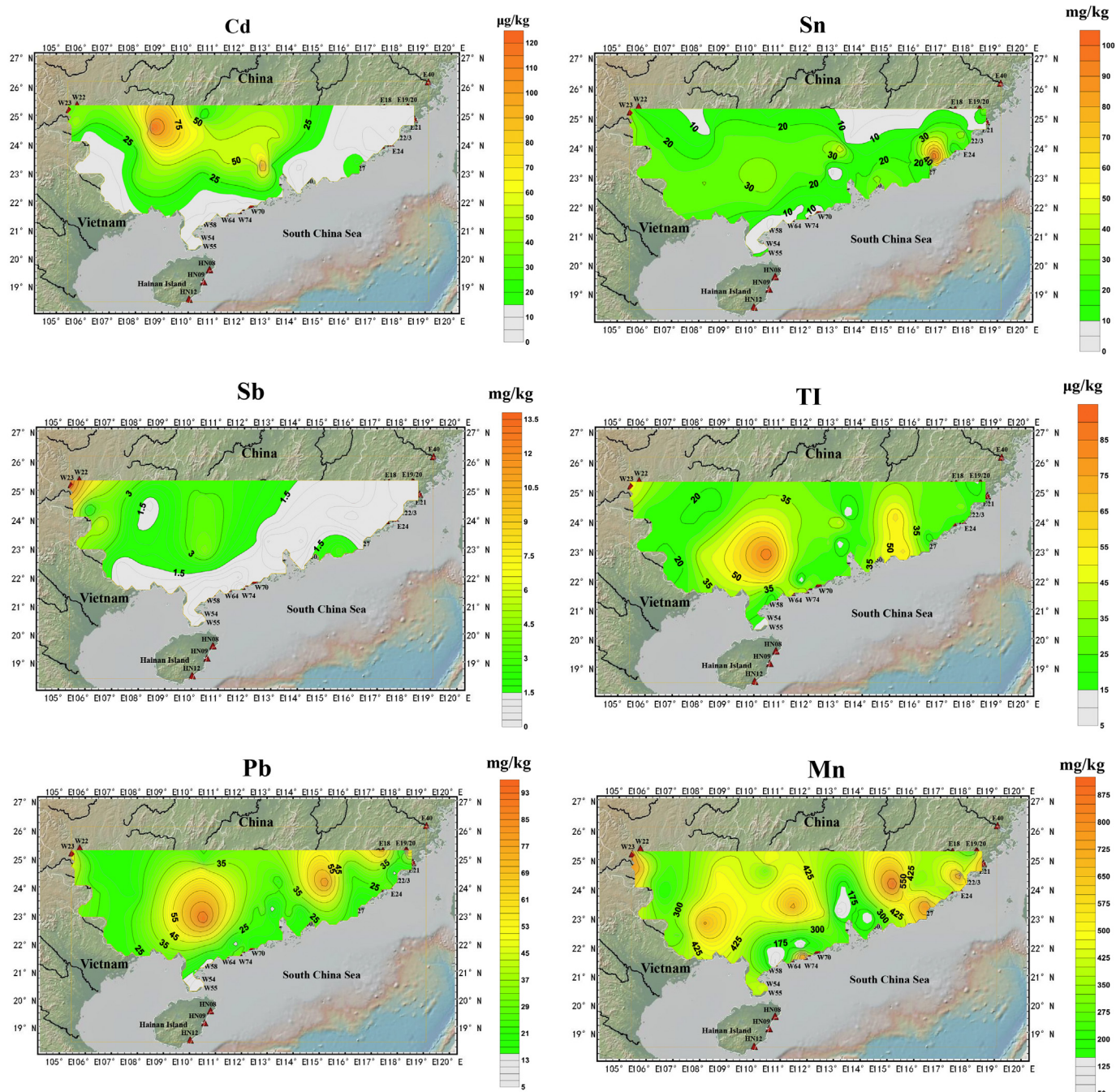


Fig. 3. (continued)

heavy metals was created to show the overall contamination levels in South China. We observed that several heavy metals, such as As, Zn, Cd, Sn and Mn, had accumulated in high concentrations in South China compared with the respective background values, which was consistent with the results discussed above. Moreover, each individual heavy metal exhibits a significant spatial variation among different provinces as is likely influenced by natural weathering and anthropogenic input, which is further explained by the contamination assessment.

### 3.2. Contamination assessment

$I_{geo}$  was selected to further explain the pollution levels in South China in comparison with the associated BG values. As shown in Fig. 4a, few of the  $I_{geo}$  values for V, Cr, Co, Cu, Ga, Tl and Pb were greater than zero for Guangdong Province, indicating uncontaminated status for V,

Cr, Co, Cu, Ga, Tl and Pb. Moreover, Cd had the highest  $I_{geo}$  value (3.86) of all target metals, while As had the highest mean  $I_{geo}$  value (1.7). Additionally, the resulting  $I_{geo}$  values for the Sn samples at 17 sites were greater than zero, indicating the presence of Sn contamination. Although the average  $I_{geo}$  values for Zn and Sb were less than zero, 9 and 11 sites had positive values, respectively, indicating minor to moderate metal contamination. Lastly, we observed that both Ni and Mn had multiple positive values, indicating that several sediments were moderately polluted by Ni and Mn for Guangdong Province. These results showed that Cd, As, Sn, Zn, Sb, Ni and Mn contamination at different levels was universally present in the Pearl River Basin, Hanjiang River Basin and in coastal rivers of western Guangdong. For Fujian Province, the  $I_{geo}$  values for Jinjiang River are presented in Fig. 4b. The majority of the  $I_{geo}$  values for V, Cr, Co, Ni, Cu, Zn, Ga, Cd, Sb, Pb, Tl and Mn were less than or approached zero, indicating that these sites in

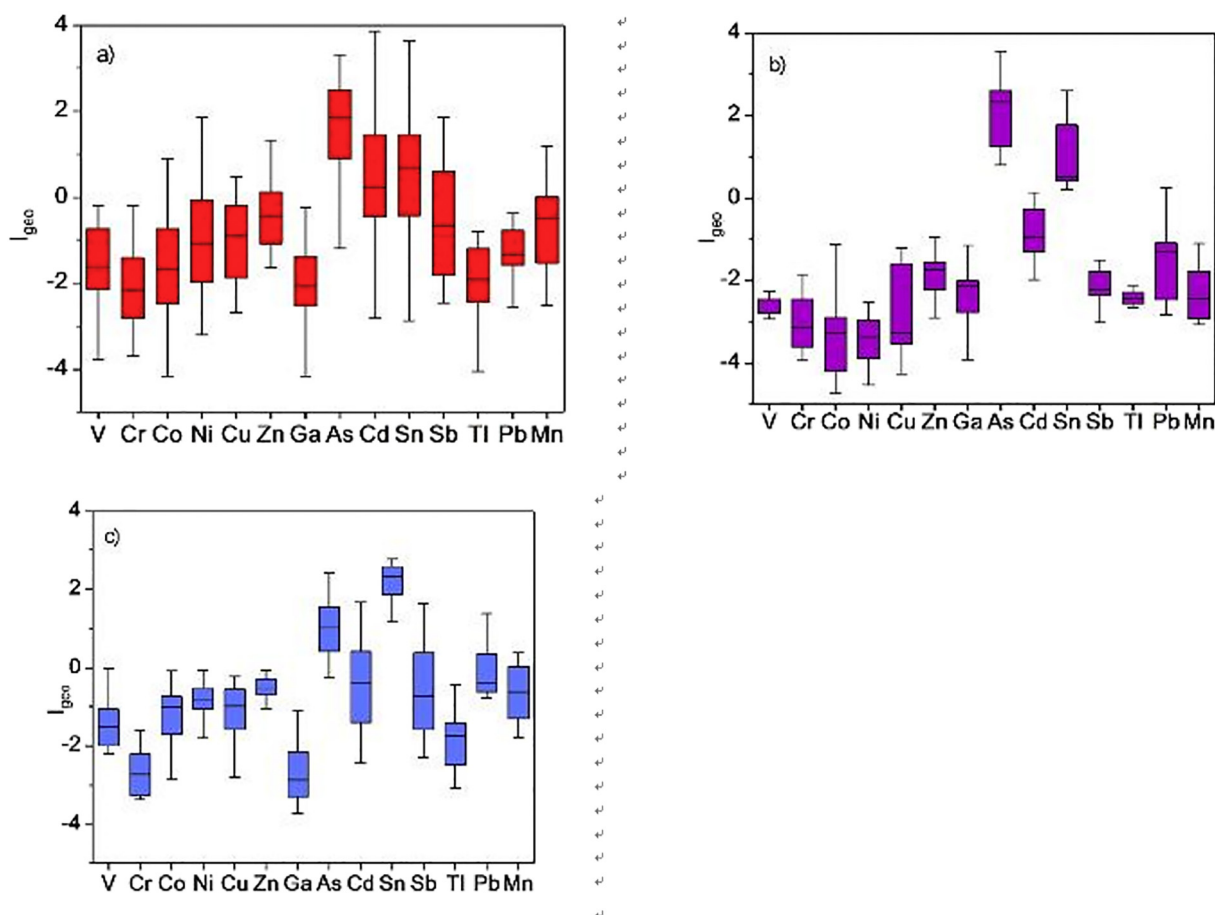


Fig. 4. Geo-accumulation indexes ( $I_{geo}$ ) of selected heavy metals in surface sediments from rivers in South China: a) Guangdong Province; b) Fujian Province; c) Guangxi Province.

Fujian Province were not polluted. The  $I_{geo}$  values for As and Sn ranged from 0.81 to 3.57 and from 0.21 to 2.59, respectively, in the sediment from all sites, indicating a minor to moderate pollution level of these two heavy metals for Fujian Province. More specifically, both As and Sn had high  $I_{geo}$  values at sites E21, E22, E23, E24 and E25, which were located in the downstream stretch of the Jinjiang River, indicating that the As and Sn in these sites might have been deposited in sediment by an upstream point source. For Guangxi Province, the  $I_{geo}$  values were calculated for the Xijiang River, and the results are summarized in Fig. 4c. The concentrations of V, Cr, Co, Ni, Cu, Zn, Ga, Sb, Tl and Mn can be classified as unpolluted because few of the associated  $I_{geo}$  values were higher than zero; however, most of the  $I_{geo}$  values for Sn were 1–2, indicating moderately polluted sediment. Moreover, 13 sites indicated  $I_{geo}$  values for As ranging from 0.21 to 1.79, suggesting the unpolluted to moderately polluted class. For Guangxi Province, five sites had positive  $I_{geo}$  values for Cd and Pb, indicating potential contamination of Cd and Pb. Considering that the Xijiang River flows through both Guangxi and Guangdong Provinces, we might observe that the Cd, As and Sn contamination was carried by the Xijiang River.

In the present study, the calculated EF values in the sediments from all study sites in South China are summarized in Table 3. For Guangdong Province, the mean EF values for V, Cr, Co, Ga and Tl in river sediments were  $< 1.5$ , suggesting that they were entirely from crustal material or natural origin; meanwhile, the relatively high EF values for As, Cd and Sn were  $\geq 5$ , indicating an anthropogenic input on all sites in Guangdong Province, which was consistent with their high  $I_{geo}$  values. Moreover, several EF values for Ni, Cu, Zn, Pb, Sb and Mn slightly exceeded 1.5, demonstrating that anthropogenic activities contributed to them partially. For Fujian Province, the EF values of the river

sediments for V, Cr, Co, Ni, Cu, Zn, Ga, Sb and Tl were  $< 1.5$ , indicating that these metals originated primarily from natural sources. Moreover, the relatively high EFs for As, Cd, Sn, Pb and Mn might be derived from anthropogenic activities. For Guangxi Province, we also calculated the EF values for sediments and determined that only As, Sn and Pb in the majority of the sites were associated with non-crustal material, whereas V, Cr, Co, Cu, Ga and Tl were characterized by  $EF < 1.5$ , indicating natural sources. The rest of the heavy metals were characterized by an  $EF < 1.5$  or an  $EF \geq 1.5$ , demonstrating that they were either from anthropogenic activities or natural sources.

In summary, the calculations discussed above revealed that the three studied areas, Guangdong, Fujian and Guangxi Provinces in South China, had common As, Cd and Sn contamination, which might be released by anthropogenic activities. Our research also presented other heavy metals that might be from either natural sources or anthropogenic input. Additionally, it may be determined that a number of sites in these three areas were influenced by different levels of heavy metal pollution, such as Ni, Cu, Zn and Mn, which should receive greater attention. Lastly, the EF values for multiple sites with heavy metal contamination were higher than 1.5, whereas the  $I_{geo}$  values were less than zero, indicating that these sites were influenced by anthropogenic sources; however, these sites still maintained uncontaminated status owing to bioturbation in the upper mixed layers or dilution by coarse sediments (Zhao et al., 2016).

### 3.3. Identification of contamination sources

To determine a likely common metal source for the rivers in South China, PCM was applied to identify the degree of correlation among the



**Table 3**  
Calculated EF values in sediments from all study sites in South China.

Site	V	Cr	Co	Ni	Cu	Zn	Ga	As	Cd	Sn	Sb	Tl	Pb	Mn
E02	0.62	0.93	0.36	1.36	0.72	1.85	0.59	9.96	12.30	13.99	4.19	1.08	1.28	0.62
E05	0.93	1.13	0.99	1.39	2.65	7.18	1.96	23.24	78.91	7.02	21.22	1.13	4.81	0.93
E08	0.39	0.77	0.15	0.93	0.42	1.24	0.57	5.19	0.54	4.69	0.59	0.61	0.85	0.39
E11	0.82	0.31	1.11	0.74	0.86	0.97	0.42	6.08	1.76	2.15	0.67	0.61	1.68	0.82
E13	0.79	0.26	0.98	0.70	0.93	1.32	0.44	6.65	1.61	3.80	0.66	0.80	1.44	0.79
E14	0.79	0.62	0.81	0.55	2.01	2.28	1.25	11.20	4.02	16.27	1.14	1.35	2.37	0.79
E15	0.62	0.28	0.73	0.57	0.79	1.66	0.40	8.45	1.87	2.45	3.61	0.69	1.07	0.62
E26	0.53	0.28	0.28	0.22	0.43	0.68	0.57	9.06	1.24	23.39	0.84	0.52	0.69	0.53
E27	0.69	1.27	1.18	7.50	1.82	6.14	0.57	10.09	8.55	4.75	2.01	0.47	1.05	0.69
E28	1.08	0.82	0.95	0.91	1.07	2.57	2.04	21.58	3.57	6.60	8.56	1.31	1.32	1.08
E30	1.11	0.52	0.61	1.31	2.14	2.10	0.69	30.14	5.65	13.10	5.06	1.39	1.29	1.11
E31	0.79	0.48	0.73	2.92	3.15	3.09	0.42	5.31	3.59	7.65	1.96	0.81	1.69	0.79
E35	0.89	0.28	0.94	2.40	3.19	2.82	0.18	3.41	8.35	4.79	2.24	0.52	0.99	0.89
E36	0.64	0.95	0.74	1.30	0.70	1.19	0.45	4.04	5.58	3.36	1.87	0.27	0.43	0.64
E38	1.19	1.84	1.45	3.26	5.37	6.54	1.49	3.63	72.50	0.68	1.30	1.63	3.11	1.19
E39	0.91	0.56	0.42	2.19	1.24	2.31	0.44	5.46	20.12	5.13	2.89	0.71	1.52	0.91
W01	0.88	0.48	0.92	2.00	1.08	2.87	1.49	6.18	32.77	3.07	3.46	0.52	1.75	0.88
W47	1.00	0.14	0.91	1.32	1.30	1.93	0.42	5.53	1.94	4.46	1.17	0.84	1.05	1.00
W48	1.35	1.41	1.44	1.71	1.62	3.58	2.76	2.84	8.89	1.46	1.76	1.28	2.50	1.35
W54	1.38	0.97	1.56	2.15	0.98	1.32	0.73	12.83	1.15	1.42	0.55	0.51	0.51	1.38
W55	0.58	0.58	1.23	2.37	0.88	0.61	0.31	5.77	0.13	0.56	0.73	0.06	0.11	0.58
W58	1.03	1.04	0.90	0.99	0.90	1.52	1.64	15.09	0.63	3.06	1.35	0.79	1.09	1.03
W64	1.41	0.73	0.92	1.24	1.29	2.33	0.94	35.04	3.22	3.70	1.06	1.75	1.61	1.41
W69	0.86	1.42	2.76	2.07	1.99	2.97	0.49	7.07	7.10	7.58	2.02	0.54	1.52	0.86
W70	1.32	0.48	1.33	1.62	1.61	1.66	0.34	11.83	1.97	1.68	3.15	0.53	0.71	1.32
W72	1.33	1.12	1.51	1.99	2.43	2.20	0.76	16.30	2.48	3.41	2.25	0.90	1.21	1.33
W73	3.18	1.65	1.77	1.53	6.08	3.57	1.75	20.09	2.97	1.49	2.83	1.81	3.29	3.18
W74	1.40	0.85	1.34	1.80	2.20	1.79	0.71	15.87	1.80	2.45	3.69	0.65	0.99	1.40
E18	0.38	0.47	0.35	0.25	1.04	2.24	0.64	5.70	1.05	3.01	0.76	0.53	2.92	2.04
E19	0.99	0.56	0.40	0.53	0.66	1.64	0.32	13.27	4.44	10.45	0.95	0.75	1.07	3.04
E20	0.83	0.55	0.31	0.85	0.44	2.51	0.55	19.91	2.18	44.38	1.65	1.42	9.99	8.48
E21	0.72	0.41	0.57	0.58	1.14	1.78	0.50	13.69	3.68	4.63	0.73	0.60	1.42	5.05
E22	0.64	0.18	0.99	0.32	0.93	0.73	0.97	10.94	1.82	3.49	0.43	0.44	0.38	1.95
E23	0.83	0.66	0.31	0.39	0.60	1.20	1.39	45.12	6.37	34.82	1.95	1.60	1.84	10.60
E24	0.58	0.58	0.42	0.40	0.28	0.77	0.69	19.09	1.62	10.77	0.70	0.50	1.48	3.43
E25	0.61	0.45	0.44	0.18	0.85	1.25	0.96	22.99	1.06	5.39	1.46	0.77	1.67	3.03
E40	0.41	0.71	0.16	0.16	0.22	0.34	1.13	30.87	1.28	3.69	0.46	0.59	0.47	0.53
W08	0.86	0.34	0.78	1.23	1.07	1.12	0.66	3.67	2.07	7.97	0.86	0.49	2.28	1.63
W11	0.71	0.30	0.87	1.54	0.95	2.73	0.63	4.01	3.91	12.25	2.42	0.83	3.66	1.12
W12	0.73	0.30	0.93	1.01	0.69	1.48	0.13	3.16	0.58	4.87	0.99	0.47	1.12	1.08
W15	2.50	1.39	1.71	2.36	1.19	4.61	0.84	28.69	26.46	40.73	2.16	0.99	6.28	6.03
W16	0.74	0.97	1.04	1.52	0.93	1.94	1.36	2.50	4.89	4.37	0.61	0.48	2.27	1.89
W17	0.72	0.38	1.09	0.90	1.24	1.13	0.27	2.35	0.79	6.75	0.62	0.33	1.07	1.06
W20	0.85	0.14	1.36	1.37	1.23	1.18	0.29	4.16	1.64	8.45	2.62	0.46	1.44	1.46
W22	0.83	0.22	0.45	0.42	1.24	0.73	0.29	3.50	1.93	4.92	5.40	0.82	1.27	0.81
W23	1.28	0.18	0.81	0.62	0.92	0.94	0.10	6.89	0.86	8.70	4.06	0.55	2.01	1.71
W24	0.81	0.57	1.46	1.81	1.01	1.70	0.29	3.05	0.60	12.62	0.88	0.49	1.69	1.16
W25	0.79	0.70	1.32	2.28	1.46	1.54	0.23	3.41	0.78	12.84	3.29	0.46	1.74	0.73
W26	0.61	0.30	0.39	0.52	0.49	1.36	0.29	6.74	0.52	15.39	0.62	0.43	1.69	1.15
W29	0.92	0.21	1.02	1.27	0.84	1.53	0.47	6.97	1.84	12.88	0.92	0.78	1.71	2.18
W34	0.73	0.38	1.37	1.78	1.74	2.78	0.72	6.01	1.42	20.42	2.83	2.25	7.84	1.92

heavy metals and OM. According to the Pearson correlation coefficients, a significant correlation was detected between multiple heavy metals and OM (Table 4). Specifically, a significant correlation was present between Co, Ni, Cu, Zn, Mn and OM, indicating that the complexation and chelation with these heavy metals might occur due to the OM. The V, Co, Ni, Cu and Zn concentrations were significantly correlated with each other, indicating that these five elements had similar sources. Cr and Mn displayed a close relationship with V, Co, Ni, Cu and Zn. Additionally, Ga displayed a close relationship with Cr, Zn, As, Cd, Tl and Pb. Similarly, a close relationship was detected between As and V, Co, Cu, Sb, Tl and Mn. Moreover, there was a positive correlation between Sb and V, Co, Cu, Zn, Tl and Mn and for Cd with Zn and Pb. Lastly, Tl showed a correlation with V, Co, Cu, Zn, Pb and Mn, as did Pb with V, Co, Cu, Zn and Mn. These results suggested a common potential source of these heavy metals. In contrast, no correlations occurred between Sn and other heavy metals, suggesting that Sn pollution might originate from a separate source compared with the other heavy metals.

PCA was performed on the normalized data to determine the

influencing factors and potential sources of the pollutants in the sediments from South China. The corresponding eigenvalue, principal components (PCs), percentage of variance by different components extracted and the factor loadings of different variables are presented in Table 5. The results of the PCA identified four PCs with eigenvalues > 1 that accounted for approximately 72.52% of the total variance in the sediment data set, suggesting that different controlling factors or sources were responsible for the heavy metals in the sediments from South China. PC1 accounted for 38.67% of the total variance and was explained by the high loading for V (0.856), Co (0.839), Ni (0.739), Cu (0.815), Zn (0.789) and Mn (0.612), which was consistent with the high correlation among these metals discussed previously. Moreover, Sb, Tl and Pb were also added into PC1 considering their close relationship with V, Co, Ni, Cu and Zn. Previous studies had shown Ni, Zn and Mn contamination to be present in Guangdong Province in South China and derived from industrial sectors such as electroplating factories or metal industries (Lee et al., 2017; Liu et al., 2014). Hence, it could be concluded that the Ni, Zn and Mn contamination of several sites in

**Table 4**  
Pearson correlation coefficients between heavy metals and organic matter.

	TOC	V	Cr	Co	Ni	Cu	Zn	Ga	As	Cd	Sn	Sb	Tl	Pb	Mn
TOC	1.000														
V	0.317*	1.000													
Cr	0.174	0.319*	1.000												
Co	0.380**	0.772**	0.520**	1.000											
Ni	0.482**	0.543**	0.670**	0.805**	1.000										
Cu	0.575**	0.704**	0.375**	0.765**	0.673**	1.000									
Zn	0.532**	0.470**	0.293*	0.519**	0.623**	0.619**	1.000								
Ga	−0.058	0.078	0.288*	0.199	0.153	0.107	0.343*	1.000							
As	0.092	0.600**	0.245	0.423**	0.253	0.288*	0.130	0.282*	1.000						
Cd	0.052	0.217	0.182	0.150	0.255	0.229	0.582**	0.482**	0.032	1.000					
Sn	−0.136	0.132	0.020	−0.022	0.022	−0.025	0.020	−0.041	0.186	0.016	1.000				
Sb	0.272*	0.738**	0.041	0.397**	0.262	0.470**	0.282*	−0.097	0.380**	0.157	0.151	1.000			
Tl	0.212	0.412**	−0.037	0.286*	0.079	0.404**	0.418**	0.304*	0.442**	0.142	0.248	0.383**	1.000		
Pb	0.204	0.332*	−0.034	0.261	0.146	0.333*	0.591**	0.271*	0.105	0.368**	0.193	0.264	0.647**	1.000	
Mn	0.360**	0.541**	0.149	0.462**	0.321*	0.379**	0.397**	0.136	0.442**	0.096	0.190	0.274*	0.331*	0.406**	1.000

\*\* Correlation is significant at the 0.01 level (2-tailed).

\* Correlation is significant at the 0.05 level (2-tailed).

**Table 5**  
Principle component analysis for selected heavy metals in river sediments from South China.

Components	Initial eigenvalues			Heavy metals	Component matrix			
	Total	Variance %	Cumulative %		PC1	PC2	PC3	PC4
1	5.414	38.669	38.669	V	0.856	−0.015	−0.365	−0.053
2	1.861	13.293	51.963	Cr	0.481	−0.612	0.156	0.354
3	1.725	12.318	64.280	Co	0.839	−0.356	−0.134	−0.019
4	1.154	8.241	72.521	Ni	0.739	−0.545	0.068	−0.047
5	0.921	6.578	79.099	Cu	0.815	−0.196	−0.079	−0.283
6	0.808	5.769	84.868	Zn	0.759	0.048	0.433	−0.277
7	0.648	4.626	89.494	Ga	0.347	0.114	0.635	0.522
8	0.361	2.576	92.070	As	0.554	0.131	−0.359	0.583
9	0.305	2.175	94.245	Cd	0.416	0.113	0.670	−0.083
10	0.252	1.800	96.045	Sn	0.150	0.401	−0.227	0.314
11	0.236	1.688	97.732	Sb	0.583	0.194	−0.447	−0.271
12	0.147	1.052	98.784	Tl	0.568	0.629	−0.020	0.069
13	0.122	0.871	99.655	Pb	0.548	0.590	0.286	−0.221
14	0.048	0.345	100.000	Mn	0.612	0.194	−0.188	0.146
				Eigenvalues	5.414	1.861	1.725	1.154
				% total variance	38.669	13.293	12.318	8.241
				% cumulative	38.669	51.693	64.280	72.521

Guangdong Province originated from industrial sectors. Moreover, the uncontaminated status for V, Co, Ni, Cu, Zn, Mn, Sb, Tl and Pb at multiple sites in South China likely originated from natural sources. PC2 accounted for 13.29% of the total variance associated with Cr, Ni, Tl, Pb and Sn in which the loadings of Cr and Ni were negative. Considering the high  $I_{geo}$  value for Sn in most of the sites in South China, it could be inferred that Sn originated from a different anthropogenic source than Cr and Ni (smelting and mining) (Li et al., 2000; Liu et al., 2003) and Tl and Pb (Industrial effluents) (Liu et al., 2014). PC3, which comprised 12.32% of the contribution, had a loading of Ga (0.635) and Cd (0.670). Considering that Cd, the most toxic heavy metal with a very high ecological risk, was correlated with Ga, we suggest that these two heavy metals were likely sourced from industrial sewage from the electroplating and electronic industries and runoff from agricultural sites in South China (Cheung et al., 2003). PC4 was dominated by As (loading 0.583) and Ga (loading 0.522) with a contribution rate of 8.241%. As plays an important role in insecticides, herbicides, fungicides, desiccants, defoliant, and animal feed additives (Matschullat, 2000; Smedley and Kinniburgh, 2002), while GaAs is an example of a semiconductor, a type of material used in virtually all modern electronics (Tran et al., 2018). Thus, it could be inferred that PC4 might represent an agricultural source or modern electronics in South China.

#### 4. Conclusion

Helpful tools and methods such as  $I_{geo}$ , EF, PCM and PCA were used to assess heavy metal pollution and identify the possible sources of heavy metal contamination in South China. The field study showed that heavy metals such as As, Cd, and Sn have accumulated at significantly high levels in South China when compared with the respective background values, and this finding was consistent with their elevated  $I_{geo}$  levels. The concentrations of heavy metals in the downstream stretch of the Xijiang River in Guangdong Province were higher than those in the upstream section of the Xijiang River in Guangxi Province, suggesting that a more developed region might sustain a greater level of pollution. Moreover, from the perspective of pollution distribution, we can observe that each individual heavy metal exhibited a significant spatial variation among the different provinces that might be derived from natural weathering and anthropogenic input. The results of the PCM and PCA showed that contaminated river sediments in South China were influenced by anthropogenic activities such as industrial effluents and domestic sewage discharge, whereas the uncontaminated sediments were affected by natural sources such as natural lithogenic actions or riverbank erosion. Lastly, the non-point sources of heavy metal pollution, such as agriculture, urban surface runoff and soil erosion, should not be ignored due to their complexity and difficult analysis. Thus, effective management and restoration work should be conducted



in contaminated areas, and additional investigations and risk assessments should be undertaken in uncontaminated areas in South China.

## Acknowledgements

This work was supported jointly by grants from the National Science Foundation of China (41773004, 41676031, 41306047), the Guangdong Natural Science Foundation (2017A030313252), the Youth Innovation Promotion Association of CAS (2017395), the Jiangsu Provincial Key Laboratory of Radiation Medicine and Protection, the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD), the General Financial Grant from the China Postdoctoral Science Foundation (2016M590494) and the Jiangsu Postdoctoral Science Foundation (1501057C).

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