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Underestimated heavy metal pollution of the Minjiang River, SE China: Evidence from spatial and seasonal monitoring of suspended-load sediments

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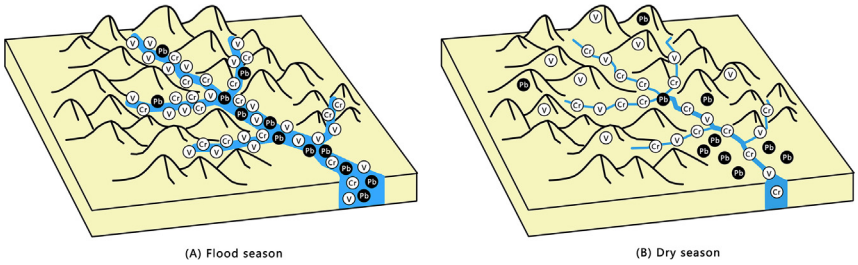
HIGHLIGHTS

- Suspended particulate matter (SPM) samples have higher heavy metal concentrations.
- Spatial and seasonal variations of particulate heavy metals are evident.
- Natural-anthropogenic control on transport and discharge of particulate heavy metals
- The Minjiang River has worse heavy metal pollution than previously known.
- High spatiotemporal resolution investigations on SPM samples are highly recommended.

GRAPHICAL ABSTRACT



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ABSTRACT

Previous assessments on rivers in SE China with highly developed economy and enormous population indicate diverse and relatively low particulate heavy metal pollution levels. However, the controlling mechanisms for heavy metal enrichment and transport remain enigmatic. Here, we target a mesoscale mountainous river, the Minjiang River, and obtain grain size, mineralogical and heavy metal concentration (Pb, Cd, Cr, Mn, Mo, Zn, V, Co, Ni, Cu) data from seasonal suspended particulate matter (SPM) near the river mouth, riverbed sediments and SPM samples from mainstream and major tributaries of the river. The results indicate that SPM samples have higher particulate heavy metal concentrations than riverbed sediments collected in pairs. Heavy metal concentrations of Cd, Zn, Cr, V, Co, Ni and Cu are higher in upstream SPM samples than those in downstream regions, whereas Pb, Mn and Mo concentrations don't show this spatial variation. Most heavy metals (e.g., Pb and Zn) show high concentrations in flood seasons and relatively low concentrations in dry seasons, revealing a hydrologic control. However, Cr and Mn show high concentrations in some dry season samples, suggesting incidental anthropogenic input events. The SPM-based pollution assessments using enrichment factor, geoaccumulation index and potential ecological risk index demonstrate that the Minjiang River is moderately to strongly polluted by particulate Pb, Cd, Mo and Zn contaminations and most particulate heavy metals have moderate to considerable potential ecological risks. We contend that transport and discharge of particulate heavy metals by the Minjiang River are controlled by both natural and anthropogenic forcings and the pollution levels are worse than previously known. Our findings suggest that particulate heavy metal discharge by subtropical mountainous rivers is related to sediment types and hydrologic characteristics. Therefore, high-spatiotemporal-resolution investigations on river SPM samples are highly recommended to better evaluate particulate heavy metal pollution levels and aquatic environmental conditions.

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1. Introduction

High concentrations of heavy metals (such as Pb, Zn and Cr) in aquatic environments are harmful for ecological systems (Ye et al., 2012; Bolan et al., 2013; Geng et al., 2015; Xu et al., 2018a). Heavy metals are usually hard-degradable and can cumulate in food chains by converting into more toxic metallo-organic compounds and eventually do harm to human health (Salah et al., 2015; Geng et al., 2015; Vareda et al., 2016; Vareda et al., 2019; Kumar et al., 2019). It is well known that more than 90% heavy metal load in aquatic systems is carried by suspended particulate matter (SPM) and sediments (Sinclair et al., 1989; Zheng et al., 2008; Sheng, 2017). How heavy metals concentrate in particulate matter during transport remains complex and enigmatic but is crucial to better control and prevention of heavy metal pollution in aquatic environments (Saeedi et al., 2004; Yuan et al., 2014; Wang et al., 2017a). Rivers serve as the major carriers and provide pathways for particulate matter to final sediment sinks, such as lakes, estuaries and global oceans (Zhang, 1999; Song et al., 2010; Hu et al., 2015). As a result, particulate heavy metal pollution is a hot topic in river environmental assessment of river systems and also is a worldwide environmental problem. (Zhang et al., 2007; Bi et al., 2017).

A great number of investigations have focused on pollution levels and sources of particulate heavy metals in worldwide rivers for decades (e.g., Hung and Hsu, 2004; Marchand et al., 2006; Salah et al., 2012; Fu et al., 2012; Mwanamoki et al., 2014). Results up to now reveal quite different pollution levels in different rivers for most heavy metals. For example, particulate Pb in the highly industrialized Rhine River (Western Europe) shows strongly polluted to extremely polluted (Gu et al., 2013; Hahn et al., 2018) and Pb in the sparsely populated Lena River (Russia) also shows strongly polluted (Rachold et al., 1996), while Pb in the lowly industrialized Nile River (Africa) displays moderately polluted (Gu et al., 2013). Most high pollution levels are commonly thought to be associated with industrial activities (e.g., ore mining and smelting) (Aleksander-Kwaterczak and Helios-Rybicka, 2009; Varol, 2011; Pheiffer et al., 2014) and domestic emissions (Gu et al., 2013; Mwanamoki et al., 2014), which are significantly influenced by urban development (Song et al., 2010; Xia et al., 2012; Gu et al., 2013). It is interesting that the rivers in South and East Asia, which account for the largest proportion of sediment discharges on the global scale (Milliman and Farnsworth, 2011), are not the worst polluted by heavy metals based on previously reported evaluations, although these regions have the largest populations and most industries. China in this region, a populous and rapidly developing country, doesn't indicate extreme river heavy metal pollution.

There are many rivers running into sea in southeast China (Fig. 1), where the economy is highly developed and the population is enormous. Previous heavy metal pollution assessments have been made for the Yangtze River (Feng et al., 2004; Zhang et al., 2009; Yuan et al., 2014; Guo and Yang, 2016), the Hangzhou Bay (Fang et al., 2009), the Minjiang River (Xu et al., 2014b), the Quanzhou Bay (Yan et al., 2020), the Xiamen Bay (Zhang et al., 2007) and the Pearl River (Li et al., 2000; Qi et al., 2010; Yang et al., 2012) (Fig. 1). Note that most these evaluations are based on the riverine surface sediments. The surface bulk sediment, with a certain thickness, probably covers mixed heavy metal information for many years and cannot represent instantaneous river pollution levels (Zhang et al., 2009; Wang et al., 2012; Maghrebi et al., 2018). In addition, environmental changes in post-deposition processes can more or less influence species and concentrations of heavy metals (Huang et al., 2010; Qin et al., 2012; Sun and Hu, 2014). Suspended particulate matter, instead, records real-time solid materials delivered by rivers and is commonly combined with many kinds of heavy metals (Sinclair et al., 1989; Zhang, 1999; Cui, 2000). Therefore, high temporal resolution SPM record allows to characterize river particulate heavy metal pollution status and to study pollution changes over a short-term time scale.

The Minjiang River is the largest river in Fujian Province (Xu et al., 2014b; Jian et al., 2020b). As a subtropical mountainous river with the strong monsoon climate, it experiences abundant rainfall and has a relatively smaller population density than other plain-dominated rivers in China. Previous studies on sediment heavy metal pollution of the Minjiang River mainly focused on estuary and surface bulk sediments (Hong et al., 2003; Xu et al., 2014b; Chen et al., 2014; Bi et al., 2017). The published data show high Pb pollution levels and overwhelmingly low-level pollutions for most other heavy metals. These studies favor that high concentrations of heavy metals are associated with geological background of the Minjiang River basin and anthropogenic activities (e.g., mining, coal combustion and wastewater discharge). The low pollution is due to the point distribution of heavy metal pollution and the dilution and self-purification effects of the Minjiang River.

In this study, we collect SPM samples within a one-year hydrological cycle (from October 2016 to October 2017) at a fixed location in the lower reach of the Minjiang River and also collect riverbed sediment and SPM samples from the mainstream and major tributaries (Fig. 2B). We present grain size, mineralogical, heavy metal geochemical and environmental assessment results and corresponding interpretations for these SPM and riverbed sediment samples. The aims are to: (1) evaluate spatial and seasonal variations in heavy metals of the Minjiang River-delivered sediments and (2) unravel potential pollution sources of the heavy metals and (3) decipher heavy metal enrichment mechanisms during sediment transport by this mountainous river.

2. Background of the Minjiang River

The Minjiang River is located in the regions of 25–29° north latitude and 116–120° east longitude and flows east into the Taiwan strait (Fig. 2). The river catchment involves many tributaries, including Shaxi, Futunxi, Jianxi in upper reaches, Youxi, Gutianxi in middle reaches and Meixi, Dazhangxi in lower reaches (Fig. 2B). The watersheds of the river system mainly sit 500–2000 m above the sea level (Jian et al., 2020a), with the drainage area of 60,992 km^2 and the total length of 577 km (Chen et al., 2014). The Minjiang River basin is characterized by a forested, highland watershed landscape and limited floodplains (Jian et al., 2020b).

The Minjiang River basin is located in the eastern margin of the Asian continent and is under a tectonically stable background (Fig. 2A). Upstream bedrocks therein are mainly composed of Precambrian metamorphic and igneous rocks, Paleozoic sedimentary rocks and Mesozoic igneous rocks, whereas the downstream bedrocks are dominated by Jurassic–Cretaceous volcano-sedimentary rocks with subordinate Late Yanshanian plutons (Li et al., 2014; Jian et al., 2020a). Mountainous regions of the river basin are rich in various mineral resources, such as manganese ore, lead-zinc ore, copper ore, gold-silver ore and Nb—Ta ore (Li, 1986; Huang and Huang, 2002).

The river basin is dominated by subtropical humid monsoon climate, with an annual average temperature of 16–20 °C and an annual rainfall of 1500–2000 mm (Zhu et al., 2018). The lower reaches are often affected by tropical cyclones, which can cause short-term extreme rainfall and flood events (Yin et al., 2010). This results in a high spatial variability of precipitation within one year, i.e., the precipitation in the upstream basin is relatively high from March to June, while that in the downstream basin is relatively high from July to September (Jian et al., 2020a). The Minjiang River has an average flow of 1750 m^3/s and an annual average discharge of suspended sediments of 715.5×10^4 t. Under such climatic conditions, the monthly runoff varies greatly. In flood season (April–July), flow accounts for 68.1% of the annual flux, and SPM transport accounts for 84% of the annual discharge; in dry season (October–February), flow accounts for 14% of the annual flux, and SPM transport accounts for 4.2% of the annual discharge (Liu et al., 2001). Furthermore, the precipitation, runoff and sediment discharge also show remarkable inter-annual variations (Jian et al., 2020a). For example, more rainfall and storms caused by El Nino and La Nina

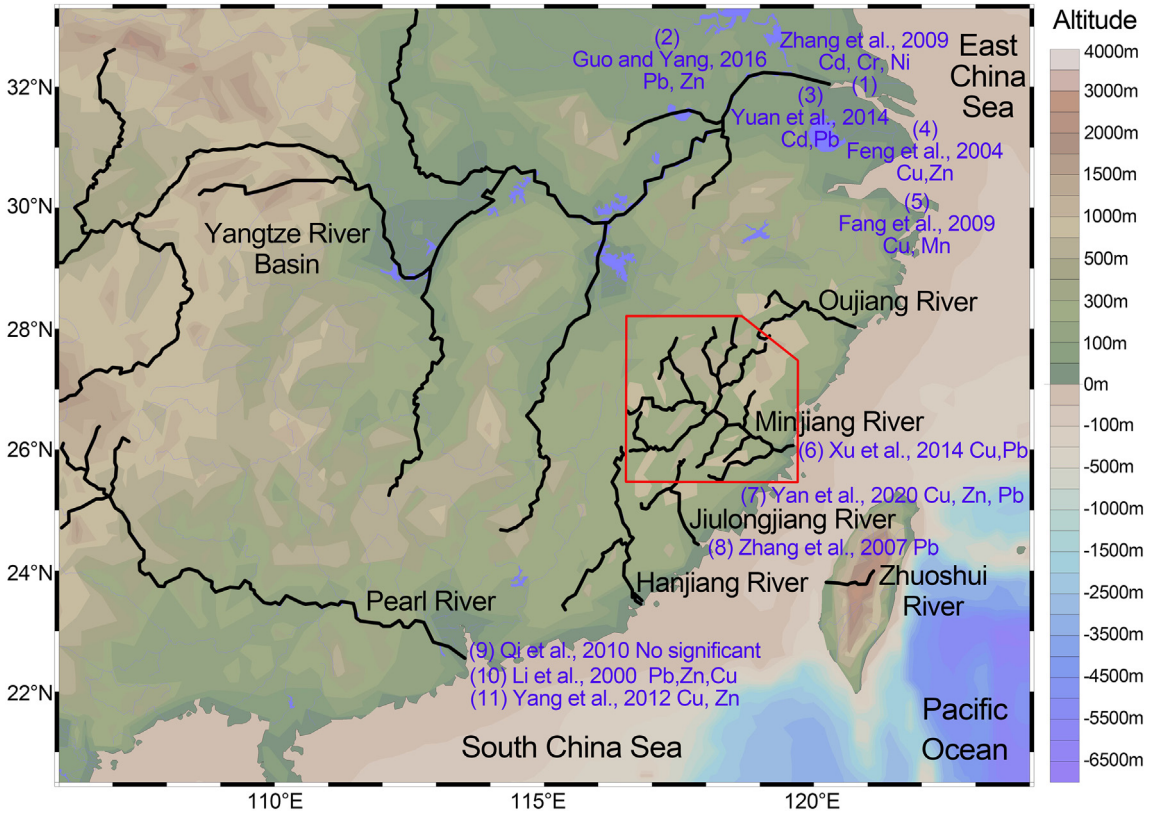

Fig. 1. Location of the Minjiang River and previous heavy metal pollution evaluations on the rivers of southeast China. The Minjiang River, located in the SE China, is close to Oujiang River, Jiulongjiang River and Hanjiang River. (1) Yangtze River intertidal zone, surface sediments (Zhang et al., 2009); (2) Yangtze River catchment, sediment cores (Guo and Yang, 2016); (3) Yangtze River, river surface sediments (Yuan et al., 2014); (4) Yangtze River Estuary, sediment cores (Feng et al., 2004); (5) Hangzhou Bay, surface sediments (Fang et al., 2009); (6) Minjiang River Estuay, sediment cores (Xu et al., 2014b); (7) Quanzhou Bay, sediment cores (Yan et al., 2020); (8) Xiamen Bay, surface sediments (Zhang et al., 2007); (9) Pearl River Estuary, sediment cores (Qi et al., 2010); (10) Pearl River Estuary, sediment cores (Li et al., 2000); (11) Pearl River Estuary, sediment cores (Yang et al., 2012). Note that although the same type samples (i.e., bulk samples from sediment cores) were used, results of the different studies on the same river were quite different. The red frame column area is the study area, which is enlarged in Fig. 2.

phenomenon led to the greater sediment transport of Minjiang River in 2016 than that in 2017 (Jian et al., 2020a). The annual sediment flux shows a decreasing trend in past decades due to increasing human activities (such as dams and reservoirs) (Dai et al., 2009).

About 14 million people live in the area of the Minjiang River basin, mainly distributing in big cities such as Fuzhou, Nanping and Sanming (Fig. 2). In recent years, modern agriculture has developed rapidly in Fujian Province. The use of chemical fertilizer, pesticide and sewage irrigation increased heavy metal pollution in rivers (Cheng et al., 2004). With the continuous industrialization and urbanization in the Minjiang River basin, heavy metal pollution caused by mining, fuel combustion and sewage discharge has enhanced. Furthermore, heavy metal pollution from urban construction and transportation cannot be ignored (Xu et al., 2014b).

3. Materials and methods

3.1. Sample collection

The riverbed sediment and SPM samples were collected from 8 sites and 13 sites (Fig. 2B), respectively. These samples cover major tributaries and different locations of the mainstream. River water was collected at surface of channels and SPM samples were then filtered from river water through 0.45 μm membranes. Riverbed sediment (mud and fine sand) samples were collected from exposed, accessible riverbeds, where the sediment deposit during flood seasons. From October 2016 to October 2017, twenty-five SPM samples were collected in the fixed S04 station (Fig. 2B) which locates at about 50 km away from the river mouth. We confirm that the seasonal sampling process was not affected by tidal currents. For riverbed sediments, fractions of <63 μm were further separated by wet sieving for grain size, mineralogy and geochemical analysis.

3.2. Analytical methods

3.2.1. Grain size and mineralogical analysis

Grain size and mineralogical analysis methods of the riverbed sediment and SPM samples were given by Jian et al. (2020a), using a laser particle size analyzer (Malvern mastersizer 3000) and a Rigaku X-ray diffractometer (Ultima IV) at Xiamen University, respectively. The samples were treated with 30% H_2O_2 and 10% HCl to remove organic matter and carbonate. Then 0.5 mol/L $(NaPO_3)_6$ was added to ensure that the particles were completely dispersed before the grain size test (Yu et al., 2013; Li et al., 2016; Su et al., 2017). For mineralogical analysis, each powdered sample was scanned continuously under the conditions of 40 kV, 30 mA, wave length of 1.54, step width of 0.02° and scanning speeds of 4°/min.

3.2.2. Heavy metal concentration analysis

About 0.5 g of each sample was leached by 1 mol/L acetic acid in a shaker incubator for 24 h at 60 °C to separate the silicate minerals from adsorptive-authigenic components and carbonate minerals. The residue after centrifuging was heated to dryness, weighed, burned in a muffle furnace at 600 °C for 2 h to remove organic matter, and weighed again. A ~50 mg aliquot of each residue sample was then totally digested with a mixture of concentrated HF and HNO_3 in high-pressure Teflon

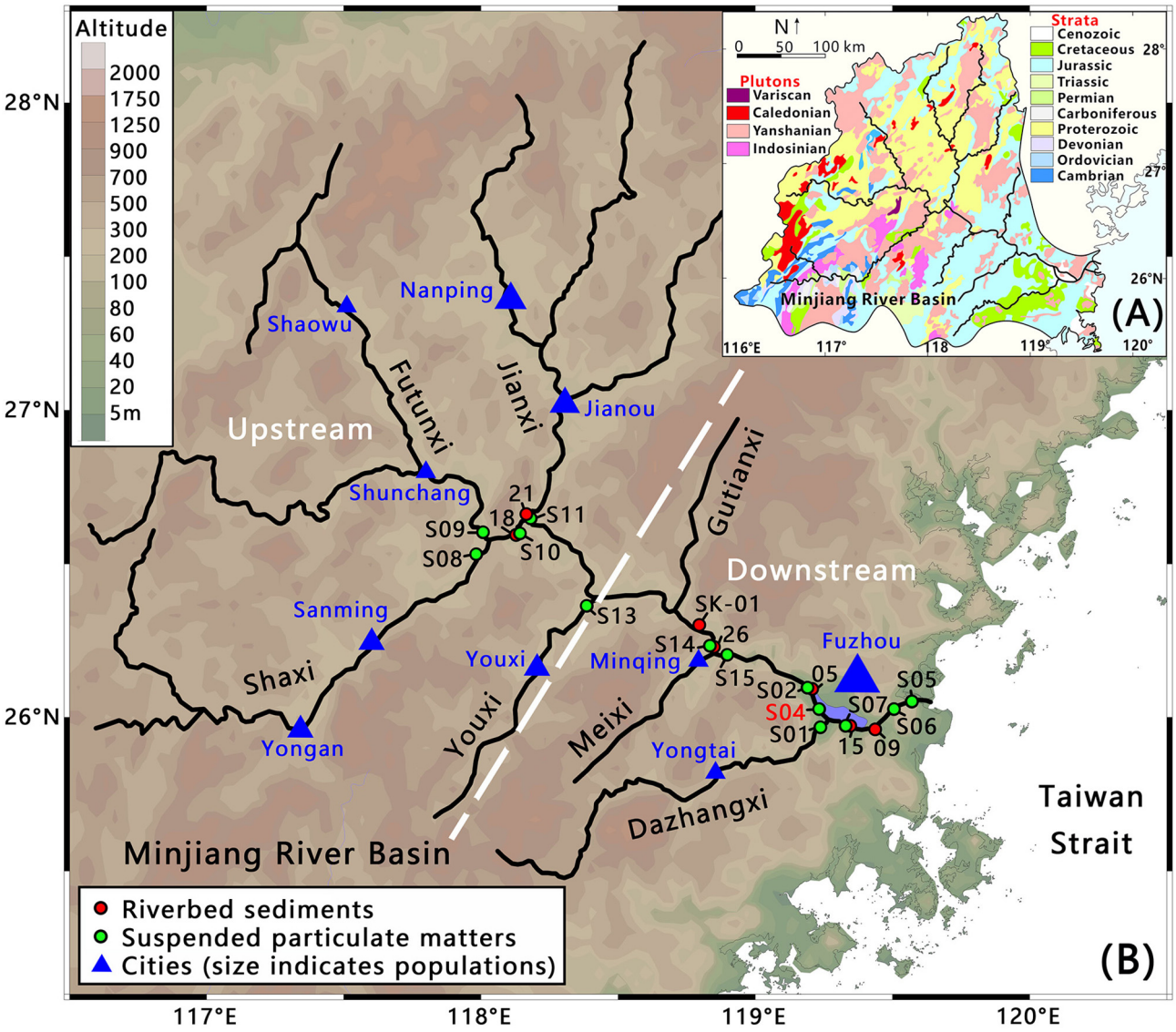

Fig. 2. (A) Geological map of the Minjiang River basin (Jian et al., 2020a). (B) Topography of the Minjiang River basin, major cities and sample locations. The Minjiang River basin is roughly divided into upstream and downstream with Youxi as the boundary. The sample stations were along the Minjiang River. The seasonal SPM samples were collected at the S04 station from October 2016 to October 2017. Note that the S07 and 15 locations (similarly for S02 and 05, S14 and 26, S10 and 18) share the same coordinates. The populations of Jianou, Nanping, Shaowu, Shunchang, Yongan, Sanming, Youxi, Minqing, Yongtai, and Fuzhou are: 0.455, 0.795, 0.277, 0.191, 0.355, 0.388, 0.361, 0.240, 0.254, and 3.897 (million people), respectively (data were collected from the website http://tjj.fujian.gov.cn/tongjinianjian/dz2018/index-cn.htm).

bombs at 190 °C for at least 48 h. The trace elemental concentrations in residual fractions were determined by ICP-MS (Agilent 7900). Blank samples and duplicate samples (separately weighed and digested) were also analyzed in the same batch along with unknown samples to monitor the background level and analytical precision, respectively. The analytical accuracy for each element was monitored by the GSR-5 standard materials (Dou et al., 2016; Guo et al., 2018; Deng et al., 2019).Both the analytical accuracy and precision were estimated to be <10% for all the heavy metals. All pretreatment and measurement steps were carried out in the State Key Laboratory of Marine Geology at Tongji University.

3.3. Environmental assessment methods

3.3.1. Enrichment factor

To evaluate anthropogenic influences of heavy metals in sediments, we use enrichment factor (EF) as an index (Feng et al., 2004; Yan et al., 2020), which is the enrichment degree of heavy metals in analyzed sediments relative to uncontaminated background levels (baseline). It is expressed mathematically by

$$EF = \frac{(Me/\ Re)_{Sample}}{(Me/\ Re)_{Background}}$$

where Me is the heavy metal measured, and Re is a reference element mainly combined in silicate minerals, which is considered to be geochemically conservative and not easy to chemically change in processes of the earth surface system (Jian et al., 2019). Al, Fe, Li, Co, Sc, Ti and Cs are often used as reference elements (He et al., 2019). In this study, Sc was used as a reference element for EF calculation. $(Me/Sc)_{Sample}$ is the metal to Sc ratio for the analyzed sample; $(Me/Sc)_{Background}$ is the natural background value of metal to Sc ratio. We employed the average Sc concentration of the Upper Continental Crust (UCC) of 6.84 mg kg^{-1} as the background value (Taylor and Mclennan, 1985). Average UCC concentrations of heavy metals are seen in Taylor and Mclennan (1985).

The EF index is a useful indicator reflecting the status of environmental contamination. An EF value of 0.5–1.5 (i.e., 0.5 ≤ EF ≤ 1.5)

Table 1
Müller's classification for geoaccumulation index.

I_{geo} value	Class	Quality of sediments
≤0	0	Unpolluted
0–1	1	From unpolluted to moderately polluted
1–2	2	Moderately polluted
2–3	3	From moderately polluted to strongly polluted
3–4	4	Strongly polluted
4–5	5	From strongly polluted to extremely polluted
5–6	6	Extremely polluted

suggests that the heavy metals may be entirely sourced from crustal materials by natural weathering processes. Otherwise, a value of EF greater than 1.5 (i.e., EF > 1.5) suggests that a significant portion of the heavy metal is supplied by non-crustal materials, such as a unique source or anthropogenic contamination (Zhang and Liu, 2002).

3.3.2. Geoaccumulation index

Another criterion to evaluate the heavy metal pollution is the geoaccumulation index (I_{geo}) proposed by Müller (1979), by comparing current concentrations with pre-industrial levels and can be calculated by the following equation:

$$I_{geo} = LOG_2\left(\frac{Ci}{1.5 \times Bi}\right)$$

where C_i is the measured concentration of the heavy metal "i" in sediments and B_i is the geochemical background value of the metal "i". Factor 1.5 is the background matrix correction factor due to lithogenic effects (Zhang et al., 2007; Farkas et al., 2007). We adopted the average UCC values (Taylor and Mclennan, 1985) in geoaccumulation index calculation. Seven classes of geoaccumulation index are distinguished to evaluate heavy metal pollution degrees (Müller, 1981) (Table 1). The highest class (class six) reflects 100-times enrichment above the background values (Zhang et al., 2007).

3.3.3. Potential ecological risk index

We utilize the potential ecological risk index (RI) proposed by Hakanson (1980) was utilized to quantitatively assess the ecological risk degrees of heavy metals in sediments (Guan et al., 2016). In this method, four influencing factors including heavy metal concentration in sediments, heavy metal type, heavy metal toxicity level and sensitivity of water body to heavy metal pollution are considered. Compared with the enrichment factor and geological accumulation index, which only consider the heavy metal concentrations in sediments, the potential ecological risk index can better reflect the environmental quality and ecological risk of river sediments. Thus, it has been widely used in some case studies. RI is defined as follows:

$$E_r^i = T_r^i \times C_f^i = T_r^i \times \left(C_s^i / C_n^i\right)$$

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

where C_s^i is the measured concentration of the heavy metal "i" in sediments, C_n^i is the background value of the metal "i", C_f^i is the single heavy metal pollution factor, E_r^i is the RI of an individual heavy metal, and T_r^i is the biological toxicity factor of the heavy metal, which are defined for Pb, Cd, Cr, Mn, Mo, Zn, V, Co, Ni and Cu as 5, 30, 2, 1, 15, 1, 2, 5, 5 and 5, respectively (Chai et al., 2017; Rehman et al., 2018). The evaluated criteria of RI are classified in Table 2.

3.4. Statistical analysis methods

The Pearson correlation analysis method was employed to determine correlation between heavy metals (Song et al., 2010). A high correlation coefficient indicates a strong correlation (>0.8 is highly relevant). Besides, to explore further and elucidate better factors governing the metal contaminant in the Minjiang River, the elemental data were processed by Principal Component Analysis (PCA) and were decomposed into a correlation matrix (Feng et al., 2004; Qi et al., 2010; Xu et al., 2016a). The Varimax rotation solution method was used to represent the heavy metals data in rotation space, which was beneficial to interpret principal components of the raw datasets. All statistical analyses were performed with the SPSS software (25th version, IBM, USA).

4. Results

4.1. Grain size and mineralogical compositions

Results of grain size and mineralogical analysis from these samples were also given by Jian et al. (2020a). The SPM samples mainly consisted of silt and clay (mean sizes ranging of 2–20 μm, Table A.1 in the Appendix A). The analyzed two riverbed sediments had mean grain sizes of 15 and 17 μm, coarser than the associated SPM samples of 6 and 5 μm, respectively. This is consistent with microscopic observations which showed that the <63 μm fractions of riverbed sediments were coarser than the associated SPM samples (Jian et al., 2020a). The main mineral components of the sediments included quartz, K-feldspar, plagioclase and clay minerals (dominated by kaolinite and illite, Fig. B.1 in the Appendix B). The riverbed sediments therein had relatively lower clay mineral contents, higher feldspar proportions, similar or even lower quartz abundances than the associated SPM samples.

4.2. Heavy metal concentrations

Heavy metal concentration data are shown in Table A.1. Heavy metal concentrations of all the analyzed samples were normalized to the average UCC compositions (Taylor and Mclennan, 1985) and are shown in Table A.2 (in the Appendix A).

4.2.1. Riverbed sediment and SPM samples from different locations in the same season

For riverbed sediment samples from different locations, the ratios of Pb, Mo, Zn, Cr, Mn, V, Co, Ni and Cu to those of UCC had ranges of 3.7–6.7, 1.7–2.8, 1.5–2.8, 0.5–2.2, 0.6–1.2, 0.9–1.9, 0.6–1.4, 0.3–2.3 and 0.6–2.4, respectively (Table A.2). Pb, Mo and Zn in the riverbed sediments were obviously enriched than other heavy metals, compared to UCC. Riverbed sample 16MJ-05 (downstream) had the highest Pb concentration, while Pb in the sample 16MJ-18 (upstream) was the lowest (Fig. 3). Sample 16MJ-26 (middle stream) indicated the lowest

Table 2
The potential ecological risk criteria for heavy metal contamination.

E_r^i value	Class	Level of single metal ecological risk	RI value	Class	Level of comprehensive potential ecological risk
$E_r^i < 40$	1	Low risk	RI < 150	1	Low risk
$40 \le E_r^i < 80$	2	Moderate risk	150 ≤ RI < 300	2	Moderate risk
$80 \le E_r^i < 160$	3	Considerable risk	300 ≤ RI < 600	3	Considerable risk
$160 \le E_r^i < 320$	4	High risk	600 ≤ RI	4	Very high risk
$320 \le E_r^i$	5	Very high risk			

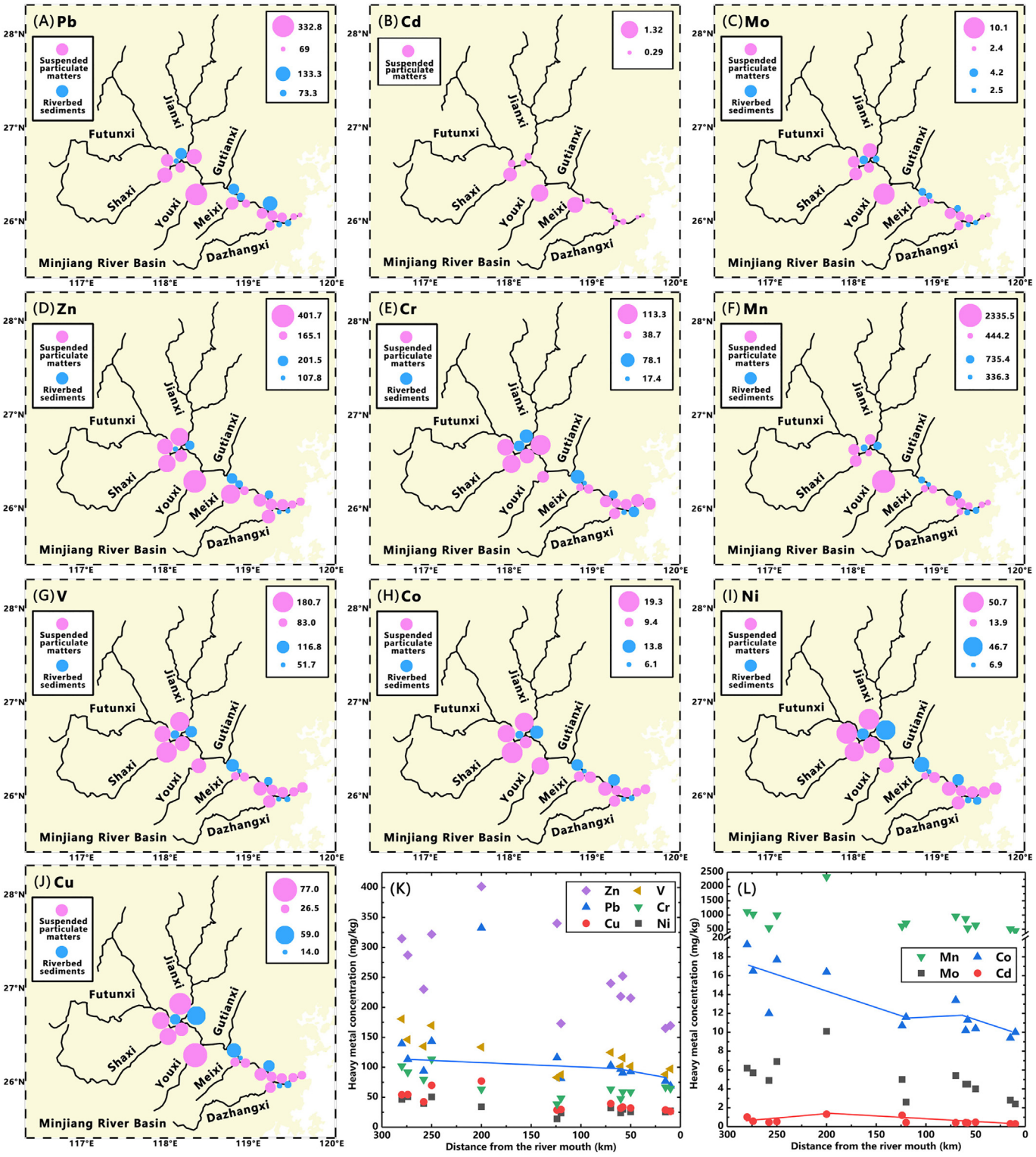

Fig. 3. Spatial variations of the heavy metal concentrations in the analyzed SPM and riverbed sediment samples from the Minjiang River. (A)–(J): Spatial distribution of individual heavy metal in SPM and riverbed sediment samples from the Minjiang River basin. The size of the circle represents the concentration of the heavy metal. (K)–(L): Distribution of heavy metals in SPM samples from upstream to downstream regions. Note that the concentration of Cr, Zn, V, Co, Ni and Cu were significantly higher in upstream samples than those of downstream samples while Pb, Mo and Mn showed no significant difference between upstream and downstream. Besides, heavy metal concentrations increased near the Fuzhou area. Contrasting with other tributaries, Youxi has extremely high values in most heavy metals.

concentrations of Cr, Mn, V, Co, Ni and Cu. The ratios of Pb, Mo, Zn, Cr, Mn, V, Co, Ni, Cu and Cd of SPM samples had ranges of 3.4–16.6, 1.8–6.8, 2.3–5.7, 1.1–3.2, 0.7–3.9, 1.4–3.0, 0.9–1.9, 0.7–2.5, 1.1–3.1 and 0.3–1.3, respectively (Table A.2), and were evidently higher than those of riverbed sediment samples. Pb, Mo and Zn therein were also enriched than the others for the SPM samples. It is surprising that the sample 16MJS-05 (estuary) had the lowest Pb concentrations (Fig. 3). Expect for V, Co, Ni and Cr, the highest concentrations of other heavy metals can be found at sample 16MJS-13 (Youxi, Fig. 3). Detailed spatial distributions of heavy metal concentrations are shown in Fig. 3.

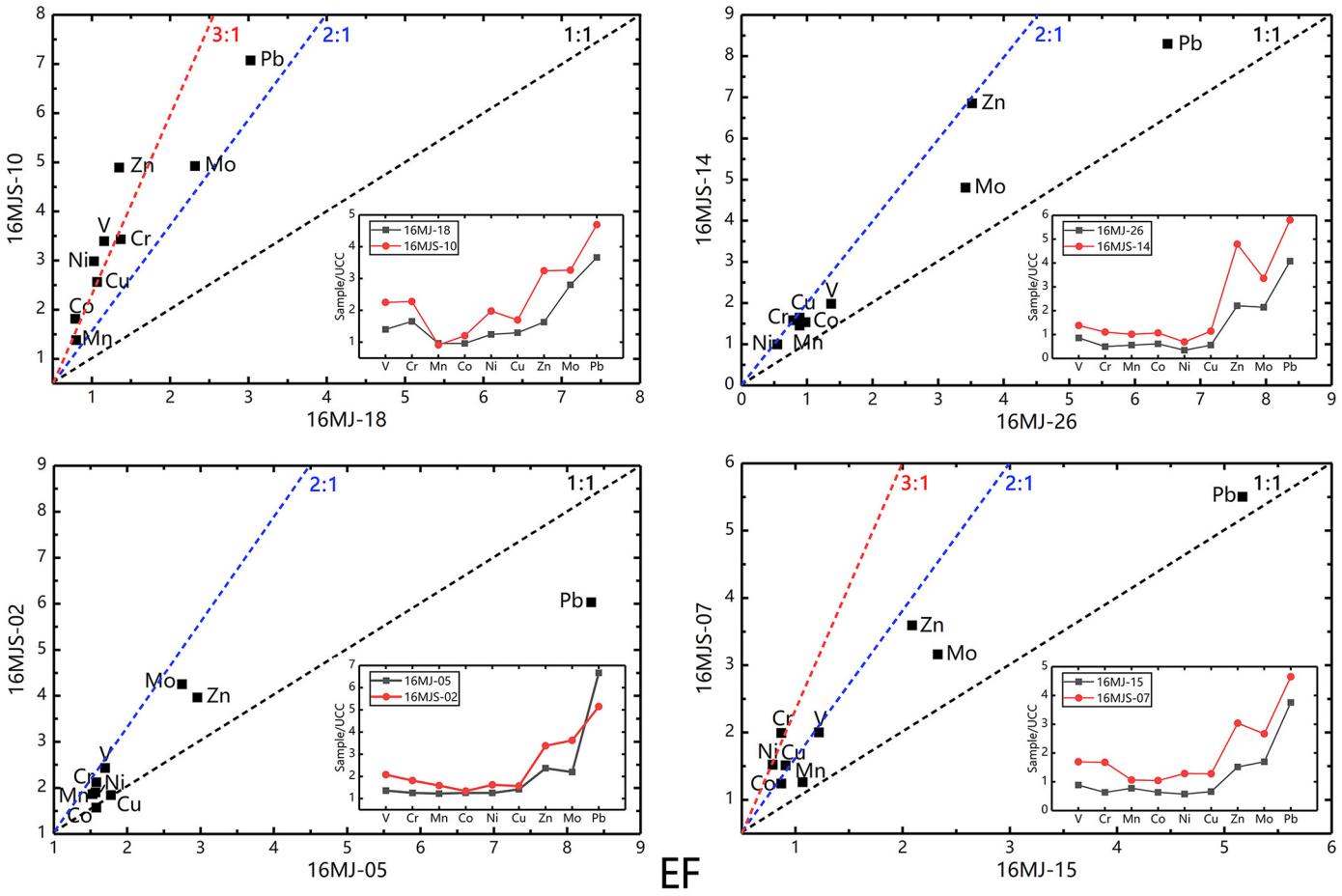

Fig. 4. The EF of heavy metals in the riverbed sediment and SPM samples collected in pair from the same position. Black lines represent the heavy metal pollution of SPM to riverbed sediments is 1, blue lines represent 2 and red lines is 3. The EF of SPM is higher than that of riverbed sediments, particularly for Zn, V, Cr and Ni. The UCC-normalized heavy metal concentration of SPM samples is also higher than that of riverbed sediment samples according to the small figures. Pb in the sample 16MJS-02 is considered as error to ignore.

Figs. 4 & 5 illustrate the comparison of heavy metal concentrations for those SPM and riverbed sediment samples collected in pairs (i.e., at the same location). Note that the SPM samples had higher heavy metal concentrations than the associated riverbed samples. The differences of Pb, Zn and Mo between the SPM and riverbed sediment samples were larger than other heavy metals, while Mn and Co had the minimal differences.

4.2.2. Seasonal SPM samples at the fixed S04 station close to the river mouth

For the SPM samples at the S04 station from different seasons, the ratio ranges of Pb, Mo, Zn, Cr, Mn, V, Co, Ni, Cu and Cd to UCC were 2.0–10.4, 1.5–5.9, 1.3–5.4, 0.7–26.9, 0.8–9.3, 0.8–3.0, 0.5–1.9, 0.5–2.5, 0.6–2.9 and 2.4–8.8, respectively (Table A.2). Most samples were rich in heavy metals compared with the UCC (Fig. 6). Most heavy metals showed similar seasonal variation trends (Fig. 6). The ratios of Cr to UCC had the largest range, varying from 26.9 (a February sample) to 0.7 (a September sample). In particular, Cr and Mn had the highest concentrations in February samples, while other heavy metals (except Mo and Cd) indicated highest concentrations in June samples (Table A.1). September–October samples had relatively low heavy metal concentrations (except Mn).

4.3. Assessment of heavy metal pollution

The enrichment factor and geoaccumulation index-based assessment results of all the analyzed samples are shown in Fig. 7 (also see Table A.3 in the Appendix A). The two indices of riverbed sediments (e.g., I_{geo} values ranging from −2 to 2 indicate not more than moderate pollution) were lower than those of the SPM samples. Samples from different locations showed different types and degrees of heavy metal pollution. The SPM sample 16MJS-13 (Youxi) had the highest EF and I_{geo} values in most heavy metals, such as Pb, Cd, Mo and Zn. EF values of the sample 16MJS-14 (Meixi) were higher only in Cd and Zn, and those of the sample 16MJS-09 (Futunxi) were higher in Cr, V, Co and Ni. The heavy metal pollution levels of the SPM samples showed remarkably seasonal variations and the highest pollution levels for those heavy metals were in different months. Based on the I_{geo} values, most seasonal SPM samples were moderately to strongly polluted for Pb and Cd. The I_{geo} values of V, Co, Ni and Cu for these seasonal samples were mostly lower than 1, revealing unpolluted conditions. While I_{geo} values of Cr and Mn for most seasonal samples were lower than 1.5, several high values (>2.5) appeared in February and March samples.

The results of RI are shown in Table A.3. According to the values of the average of potential ecological risk factor (E_r^i), the ecological risk of the ten heavy metals ranked as the following order: Cd (184.2) > Mo (50.0) > Pb (29.8) > Cu (8.4) > Ni (8.1) > Co (6.5) > V (4.0) > Cr (3.9) > Zn (3.6) > Mn (1.4) in SPM samples of different locations in the Minjiang River. The sample 16MJS-13 had very high potential ecological risk, samples 16MJS-14, 16MJS-8, 16MJS-11 and 16MJS-9 had considerable potential ecological risk, and other samples had moderate potential ecological risk. In seasonal SPM samples, the order of ecological risk of the ten heavy metals was: Cd (148.5) > Mo (49.3) > Pb (24.7) > Cr (7.3) > Cu (7.1) > Ni (6.3) > Co (6.0) > Mn (3.3) > V (3.0) > Zn (2.8). The seasonal SPM samples SS-03

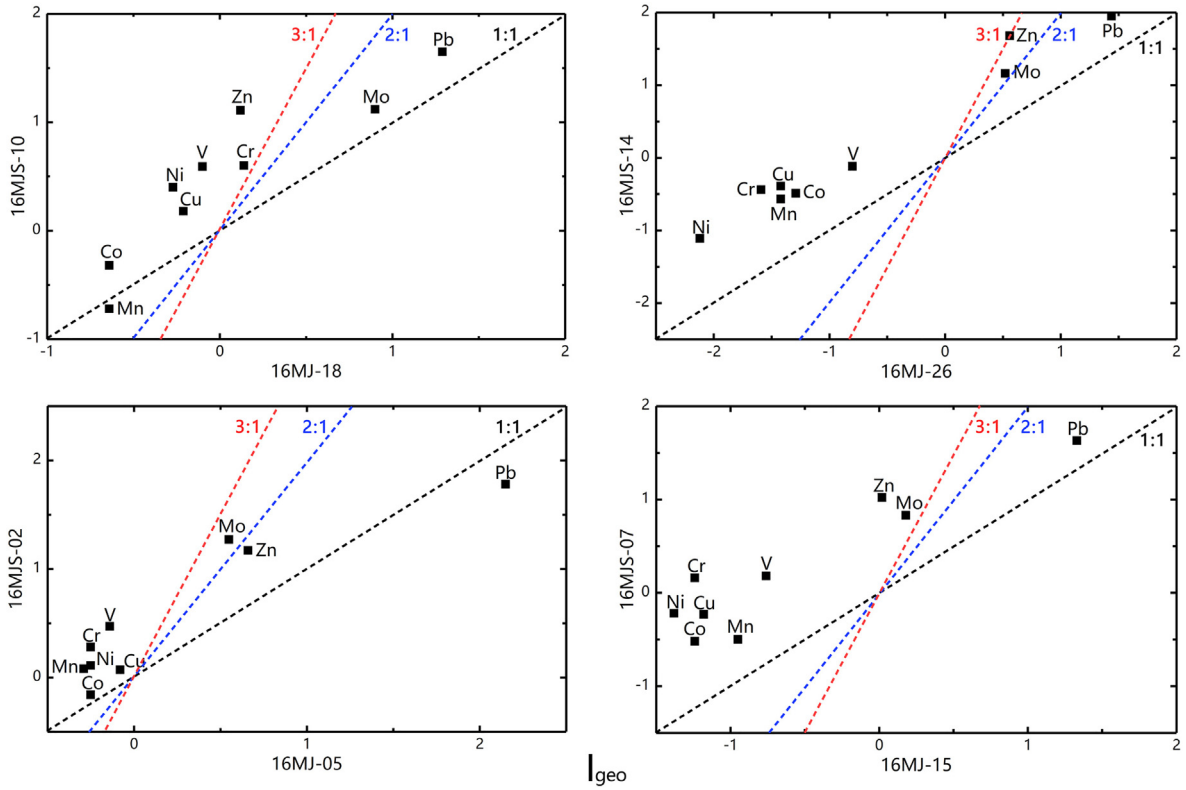

Fig. 5. The I_{geo} of heavy metals in the riverbed sediment and SPM samples collected in pair from the same position. Black lines represent the heavy metal pollution of SPM to riverbed sediments is 1, blue lines represent 2 and red lines is 3. The I_{geo} of SPM is higher than that of riverbed sediments, particularly for Zn, V, Cr and Ni. Pb in sample 16MJS-02 is considered as error to ignore.

(November), SS-05 (December), SS-08 (February), SS-10 (March), SS-14 (May), SS-16 (June), SS-18 (June) and SS-21 (August) had considerable potential ecological risk, only samples SS-23 (September) and SS-25 (October) were in low potential ecological risk levels, and other samples had moderate potential ecological risk. The trend of RI variations for all the samples was similar with EF and I_{geo} values.

5. Discussion

5.1. Previous sedimentary provenance analysis of the Minjiang River SPM and sediments

As mentioned above, the Minjiang River basin is characterized by bedrock lithologic (Fig. 2) and geochemical, as well as climatic (i.e., rainfall) heterogeneity and thus offers an ideal ground for a short-timescale sediment provenance study (Jian et al., 2020a, 2020b). Petrography and detrital zircon U—Pb geochronology of the discharged sand sediments from the Minjiang River indicate strong signatures of the Precambrian crystalline rocks in the upstream regions (Xu et al., 2014a; Xu et al., 2016b; Jian et al., 2020b). This means that the human activities, such as constructions of dams and reservoirs, which are expected to hold back sediments from the upstream regions, probably serve as a minor role in sediment transport of the Minjiang River (Jian et al., 2020b). Furthermore, some trace element ratios (such as La/Sc, Th/Sc, Th/U and $La_{(CN)}/Yb_{(CN)}$) and Sr—Nd isotopes display obviously fluctuant variations in different season SPM samples, indicate seasonal provenance shifts and correspond well with the spatiotemporal variations of precipitation in the catchment. Because of this, the subtropical mountainous rivers in Fujian Province (SE China) are thought to be featured by rapid sediment delivery in response to hydroclimatic changes and have climate-dependent sediment compositional heterogeneity (Jian et al., 2020a, 2020b). However, the new data in this study demonstrate that most particulate heavy metals have different transport behaviors with those natural process-produced sediments.

5.2. Different heavy metal pollution levels between riverbed sediments and suspended particulate matter

The results of SPM and riverbed sediments (<0.063 mm fractions, the same below) collected in pairs (sample locations in Fig. 2) indicate that heavy metal pollution degrees in SPM samples are mostly higher than those in associated riverbed sediment samples, such as heavy metal Zn, V, Cr and Ni (Figs. 4 & 5). Note that the SPM samples have lower average grain sizes than that of the associated riverbed sediments (Table A.1). This means that most particulate heavy metals tend to concentrate in very fine-grained particles of sediments.

Riverine SPM, which is commonly derived from continuous weathering and leaching of bedrock within the watershed and can be derived from recycled flood-plain sediments and resuspended bedload sediments, has been proven to be closely related to waste drainage in some cases (Qiao et al., 2007; Qin et al., 2012; Jian et al., 2020a). Riverine SPM is usually composed of clay- and silt-sized detritus, clay minerals and organic particles including organisms, biological debris, humus, and inorganic particles coated with organic matter (Lu and Allen, 2001; Plach et al., 2011; Xia and Chen, 2012). It is well recognized that heavy metals can largely be adsorbed to surface of particulate matter and thus particles containing large specific surface areas (e.g., clay sized detritus and clay minerals) are expected to have high heavy metal concentrations (Conrad and Chisholm-Brause, 2004; Deng and Zhao, 2017; Li et al., 2018b; Ma et al., 2019b). This is reinforced by the mineralogical analysis results which indicate higher clay mineral (e.g., kaolinite) compositions in SPM samples than the associated riverbed sediments (Fig. B.1). Furthermore, particulate heavy metals also have positive relations with organic matter, which contains many

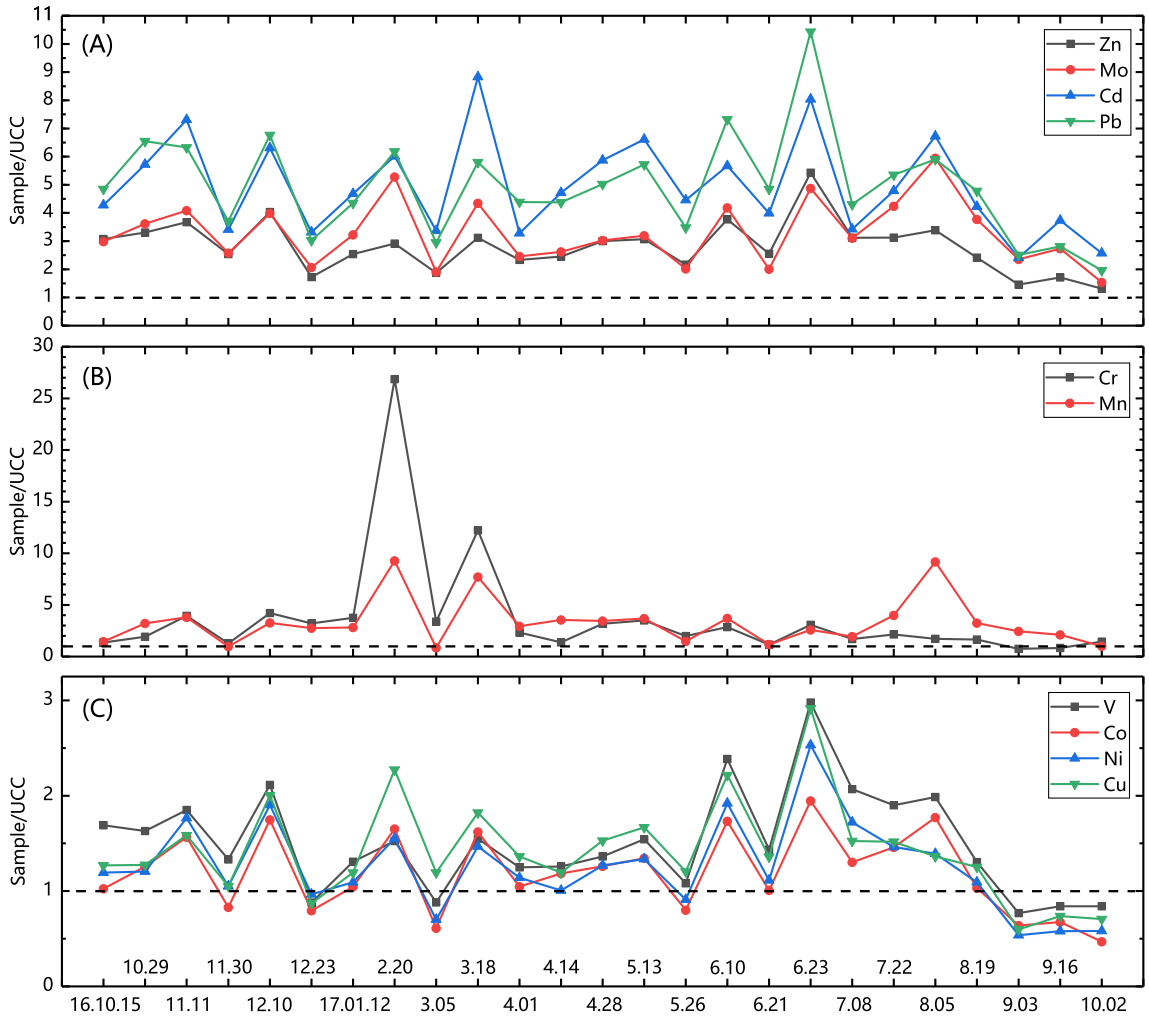

Fig. 6. Heavy metal geochemical results of seasonal SPM samples from the fixed S04 station. (A) The range of values was higher. (B) The range of values varied greatly and differently. (C) The range of values was lower. UCC-normalized heavy metal concentrations change similarly with time for most heavy metals. Note that Cr and Mn have significant different variation trends, and Mn has similar changes with Mo and Cd at some time.

components (e.g., carboxyl, hydroxyl and amino) with good affinity for heavy metals (Zhang et al., 2009; Meers et al., 2009; Wang et al., 2017a). Another possible reason for the low heavy metal concentrations in riverbed sediments is the dilution effect caused by uncontaminated, natural process-produced detritus (Ye et al., 2012), such as silt-sized feldspar and lithic fragments (Fig. B.1, Jian et al., 2020a).

5.3. Spatial variations of particulate heavy metals in the Minjiang River

Both SPM and riverbed sediments results reveal two different spatial variation trends of particulate heavy metal concentrations in the Minjiang River basin (Fig. 3). The first trend is that heavy metals (Cd, Zn, Cr, V, Co, Ni and Cu) show higher concentrations in upstream samples than in downstream samples. High particulate heavy metal concentrations in the upstream regions can be attributed to geological conditions, mining and some industrial activities. Baseline investigations demonstrate that heavy metals in upstream bedrock are more than those in downstream bedrock (Lin, 2003; You, 2007). It is well known that the upstream mountainous regions of the Minjiang River basin are rich in metal mineral resources, such as lead-zinc ore, manganese ore and copper ore (Li, 1986; Huang and Huang, 2002) and mining activities have been conducted for decades. In addition, previous studies reported that many electroplating, chemical and building materials industries around the Yongan and Nanping cities could contribute to high heavy metal concentrations (Chen et al., 2014) and the low heavy metal concentrations in downstream regions are considered to be due to the river dilution and self-purification effects (Song et al., 2010; Chen et al., 2014; Li et al., 2017). Note that a few samples near the Fuzhou city have slightly higher heavy metal concentrations than samples from the midstream and estuary regions (Fig. 3). This might be due to intensive human activities in big cities (e.g., Fuzhou) (Fig. 2B) which produce a great amount of heavy metals. These heavy metals are then discharged into rivers ultimately and exceed the rivers' dilution capacity.

The second trend is that heavy metal concentrations in upstream and downstream samples are similar, such as Pb, Mo and Mn (Fig. 3). Upstream source of Pb, Mo and Mn is probably related to mining, while these heavy metals in the downstream regions are most likely sourced from urban anthropogenic activities (Table 3). The downstream regions of the Minjiang River are densely populated, contributing the second type of heavy metals (Fig. 2). We favor that these regions accept a large number of heavy metals discharged by fuel combustion and domestic sewage, resulting in high heavy metal concentrations in particulate matter (Qiao et al., 2007).

Note that most samples are strongly influenced by anthropogenic contamination because most EF values are higher than 1.5 (especially for heavy metals Pb, Cd and Mo), while heavy metals in some samples (especially in V, Co and Ni) are entirely sourced from natural materials (EF < 1.5) (Fig. 7). Among all the tributaries of the Minjiang River, the Youxi sample (16MJS-13) exhibits the highest heavy metal

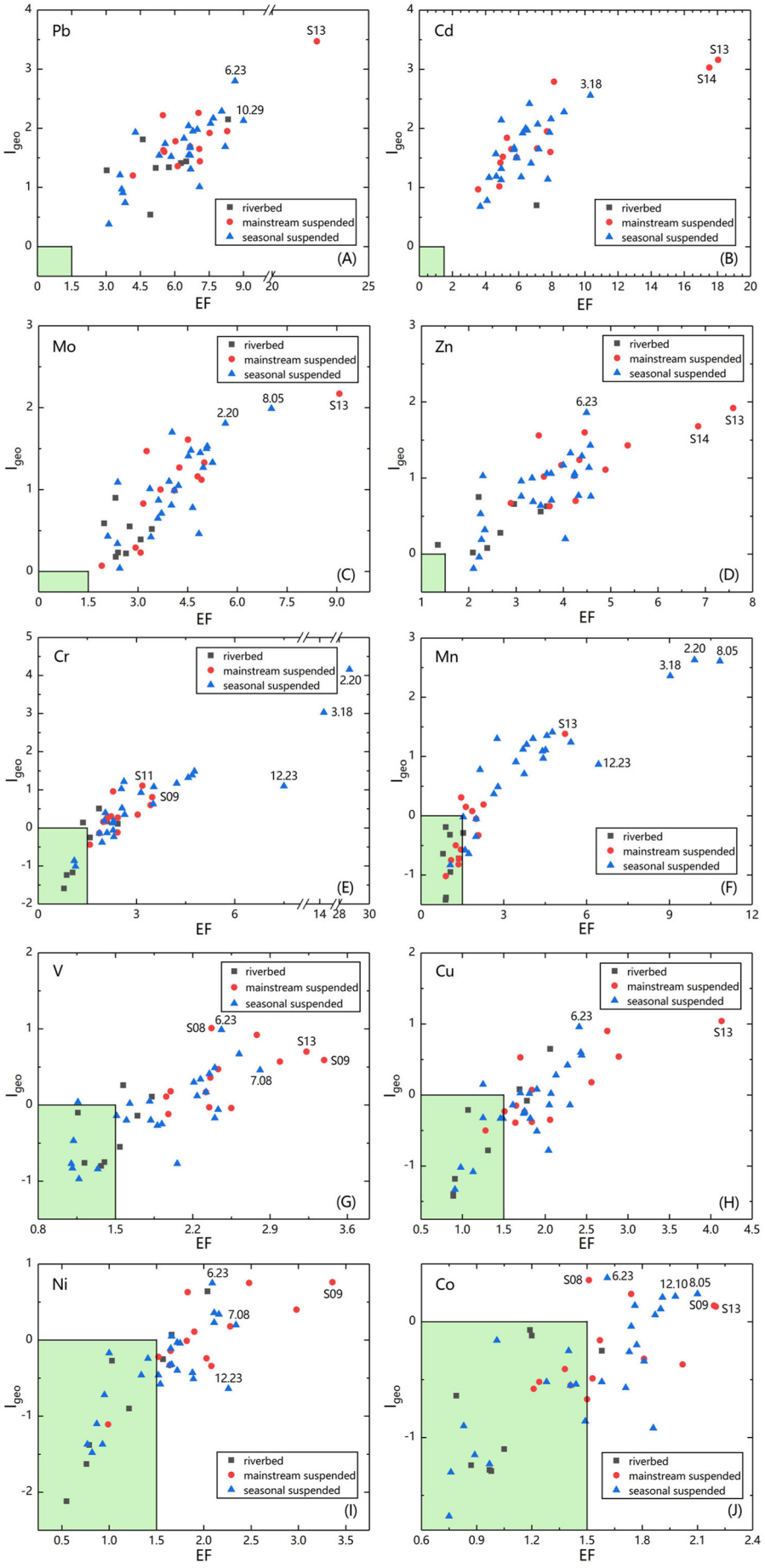
Pb
S13
6.23
10.29
riverbed
mainstream suspended
seasonal suspended
(A)
Cd
S13
S14
3.18
(B)
Mo
8.05
S13
2.20
(C)
Zn
6.23
S13
S14
(D)
Cr
2.20
3.18
S11
S09
12.23
(E)
Mn
2.20
8.05
3.18
S13
12.23
(F)
V
S08
6.23
S13
S09
7.08
(G)
Cu
6.23
S13
(H)
Ni
6.23
S09
7.08
12.23
(I)
Co
S08
6.23
12.10
8.05
S09
S13
(J)
Igeo
EF

Table 3
Possible anthropogenic sources of heavy metal pollution.

Heavy metals	Anthropogenic sources	References
Pb	Fuel combustion, exhaust emissions, mining and smelting of lead-zinc ore	Li et al., 2000; Callender, 2005; Zheng et al., 2008; Xu et al., 2014b; Gong et al., 2018
Zn	Mining and smelting of lead-zinc mine, fuel combustion, exhaust emissions, electroplating, chemical and machinery manufacturing industries	Callender, 2005; Zheng et al., 2008; Che et al., 2017; Gong et al., 2018; Wu et al., 2018; Ma et al., 2019a
V	Fuel combustion, ore mining	Gümgüm et al., 1994; Abdel Ghani et al., 2013
Co	Ore smelting, electroplating, hardware and other industrial activities	Che et al., 2017; Yan, 2018
Ni	Fuel combustion, ore smelting, electroplating, hardware and other industrial activities	Callender, 2005; Che et al., 2017; Yan, 2018
Cu	Mining and smelting of copper, gold and polymetallic ore, fuel combustion, exhaust emissions, electroplating, chemical and machinery manufacturing industries, agriculture and aquaculture	Callender, 2005; Zheng et al., 2008; Yang et al., 2012; Che et al., 2017; Bi et al., 2017; Gong et al., 2018; Ma et al., 2019a
Mo	Fuel combustion, ore mining and smelting	Zhang, 2011; Yu et al., 2011
Cd	Mining and smelting of metal mines, electroplating, chemical industry, atmospheric deposition, agricultural and domestic sewage	Callender, 2005; Zheng, 2007; Zheng et al., 2008; Li et al., 2013; Wu et al., 2018; Elias et al., 2018; Ma et al., 2019a
Cr	Mining of rare earth ore and iron-manganese ore, steel and iron manufacturing, lumbering, metal finishing industries	Callender, 2005; Wei et al., 2015; Che et al., 2017
Mn	Mining of rare earth ore and iron-manganese ore, smelting of copper ore	Callender, 2005; Wei et al., 2015; Che et al., 2017

concentrations and is mostly affected by anthropogenic pollution due to high EF and I_{geo} values (Fig. 7). The potential ecological risk analysis results indicate that the particulate heavy metals in the Minjiang River overall has moderate to considerable potential ecological risks. Cd can cause most severe potential ecological risk in the river (Table A.3), although its concentration was the lowest in all heavy metals. Besides, Mo has moderate potential ecological risk and other heavy metals have comparatively low potential ecological risk. The results of RI indicate that particulate heavy metals in the tributary Youxi (16MJS-13) have very high potential ecological risks and the potential ecological risk was higher in the upstream regions than that in the downstream regions. This implies that long-term mining and industry causes high potential ecological risk (Chai et al., 2017).

5.4. Seasonal variations of particulate heavy metals discharged by the Minjiang River

The heavy metal concentrations of the SPM samples collected at the fixed S04 station (Fig. 2B) in different seasons of 2016–2017 demonstrate strong variations over time (Fig. 6). The fluctuating trends of most heavy metals are similar (i.e., increasing or decreasing isochronously), but some heavy metals (e.g., Cr and Mn) show abnormal concentrations for some samples.

As shown in Fig. B.2 (in the Appendix B), some heavy metals (e.g., Pb, Zn, V and Co) in the SPM samples of October–November 2016, late March–April 2017 and June–August 2017 have relatively higher EF values than those of the SPM samples of other periods. And the average EF values of these heavy metals interestingly display positive correlations with monthly runoff data of the Minjiang River (linear fitting coefficients are positive, Fig. B.3 in the Appendix B). This means a hydrologic control on these heavy metals. Note that most heavy metals of SPM samples in high-runoff periods (i.e., high-water stage or so-called flood season) have higher EF values than those of the low-runoff periods (i.e., low-water stage or so-called dry season) samples (Fig. 8). Therefore, erosion of more intensive heavy metal polluted sediments in the watershed increases in flood seasons than that in dry seasons. Intensive rainfall results in high runoff and high transportation capacity of the river and is expected to wash more particulate heavy metal pollutants into the river (Qiao et al., 2007; Song et al., 2010). These heavy metals have consistent seasonal variation trends of other natural process-produced elements, such as rare earth elements Sc, Th and U, which indicate high La/Yb and Th/U ratios for flood season SPM samples (Jian et al., 2020a). We further find that the flood season SPM samples have EF values closer to the upstream SPM samples, while these EF values in dry season samples are similar with those of the analyzed riverbed sediment samples (Fig. 8). Hence, sediments on riverbeds are probably recycled into SPM during dry seasons, whereas the surface SPM samples in flood seasons are less mixed with riverbed sediments and are likely dominated by SPM directly from the upstream tributaries (rainfalls in March and June of upstream are higher than those of downstream) (Jian et al., 2020a, 2020b). Another reason for the high EF values of flood season SPM samples is that the partition coefficients of SPM–water (ratios of heavy metal concentrations in SPM to those in water) have positive correlations with SPM concentrations for most heavy metals (e.g., Pb) and the SPM concentrations in flood seasons are higher than those in dry seasons (Fig. 8). In this case, heavy metals can be adsorbed to a greater extent on the SPM of flood seasons (Feng et al., 2017; Gogoi et al., 2020).

By contrast, the EF values of Cr and Mn in the seasonal SPM samples don't have positive correlations with runoff of the river (Fig. B.3). High EF values are instead incidentally present in some dry season samples (e.g., Cr, Mn and Mo in February) (Fig. 8). This is due to incidental human pollution (an anthropogenic control) events and are probably related to decreases in temperature, flow velocity and biological activity in dry seasons which reduce the mobility of heavy metals (i.e., desorption from SPM) (Cui, 2000; Wang et al., 2012; Feng et al., 2017). In addition, negative correlations between the partition coefficients of some heavy metals (e.g., Cr) and SPM concentrations (particle induced desorption (PID) mechanism) to some extent can also influence concentrations of those particulate heavy metals during dry season transport (Feng et al., 2017).

According to these results, we propose a transport model of particulate heavy metals by the Minjiang River (Fig. 9). From a spatial perspective, concentrations of most heavy metals (e.g., V and Cr) are comparatively high in the mountainous upstream regions and decrease in downstream regions, but some other heavy metals (e.g., Pb) show no significant spatial variations. In terms of a seasonal timescale, in flood seasons, particulate heavy metal concentrations (e.g., V and Pb) in river SPM are relatively high due to high precipitation and runoff and enhanced erosion of more intensive heavy metal polluted sediments, similar to those natural process-produced elements (Jian et al., 2020a, 2020b), while in dry seasons, heavy metal pollutants are left on the overbanks and exposed riverbeds causing a decrease of the concentrations in SPM. However, some heavy metals (e.g., Cr) are obviously in strongly polluted levels in dry seasons due to incidental anthropogenic contamination emissions. We attribute the diverse seasonal variations

Fig. 7. Scatter plots of EF and I_{geo} of heavy metals. In the green box bounded by EF = 1.5 and I_{geo} = 0, the samples were non-polluting and only had natural sources. The two indices of riverbed sediment samples were mostly lower than these of the other two samples. The seasonal variation ranges of SPM samples were large and the highest EF and I_{geo} values of SPM occurred in different stations and months.

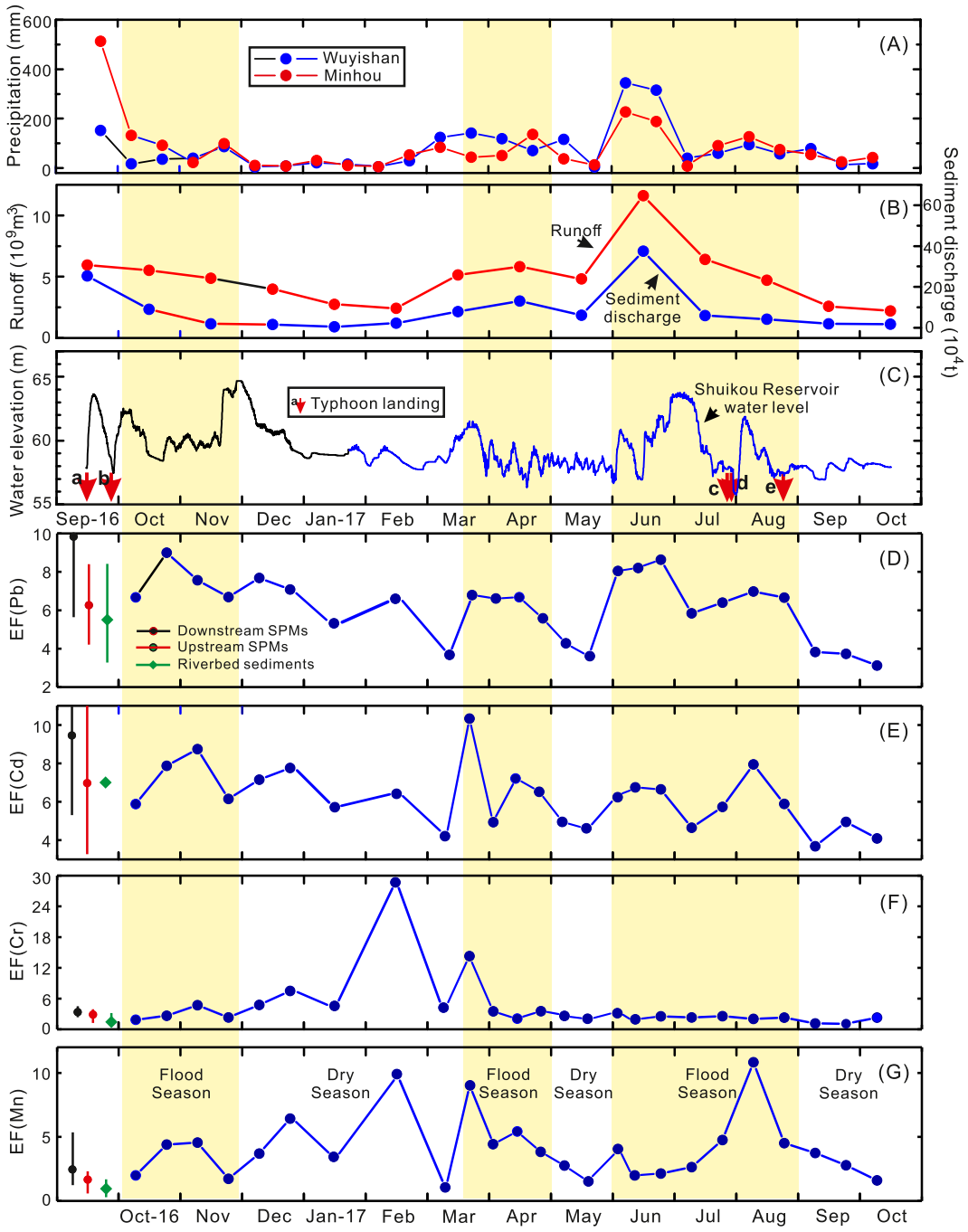

Fig. 8. Seasonal variations of the selected representative heavy metals of the SPM samples from the fixed S04 station. EF index was used. Yellow areas indicate flood season. (A) Semi-monthly precipitation of representative cities in the Minjiang river catchment (Jian et al., 2020a); (B) monthly runoff and sediment discharge of the Minjiang River (Jian et al., 2020a); (C) water level variations of the Shuikou Reservoir (near the SK-01 station), red arrows indicate major typhoon events (a: Meranti, Sept. 2016; b: Megi, Sept. 2016; c: Nesat, Jul. 2017; d: Haitang, Jul. 2017; e: Hato, Aug. 2017) which impacted the catchment (Jian et al., 2020a). (D) EF of Pb; (E) EF of Cd; (F) EF of Cr; (G) EF of Mn. Note that the EF values were generally higher in flood seasons than these in dry seasons, while high increase occurred in February (dry season) for Cd, Cr and Mn, which is closely related to incidental anthropogenic input events.

to a combined action of natural processes and human activities in the Minjiang River basin. This is supported by the PCA analysis results (Table 4, Fig. B.4 in the Appendix B). Two Principal Components account for 86% of the variance, with the first Principal Component (PC1) accounting for 70.9% and the second Principal Component (PC2) for 13.1% (Table 4). The PC1, high loading for Sc and most heavy metals, is considered as a combined action of regular natural-anthropogenic sources. The PC2, which has obviously high loading of Cr and Mn, can be interpreted as the source of incidental anthropogenic input events. Lower loading for Mo and Cd in PC2 suggests that their concentration changes are slightly related to incidental anthropogenic events (Fig. B.4).

5.5. Controls on particulate heavy metal enrichment, transport and discharge by the Minjiang River

Heavy metals in aquatic environments have a variety of natural and anthropogenic sources (Shen et al., 2006; Xu et al., 2018b). Both catchment material erosion and atmospheric deposition can attribute to the riverine heavy metals (Callender, 2005; Wang et al., 2017b). The

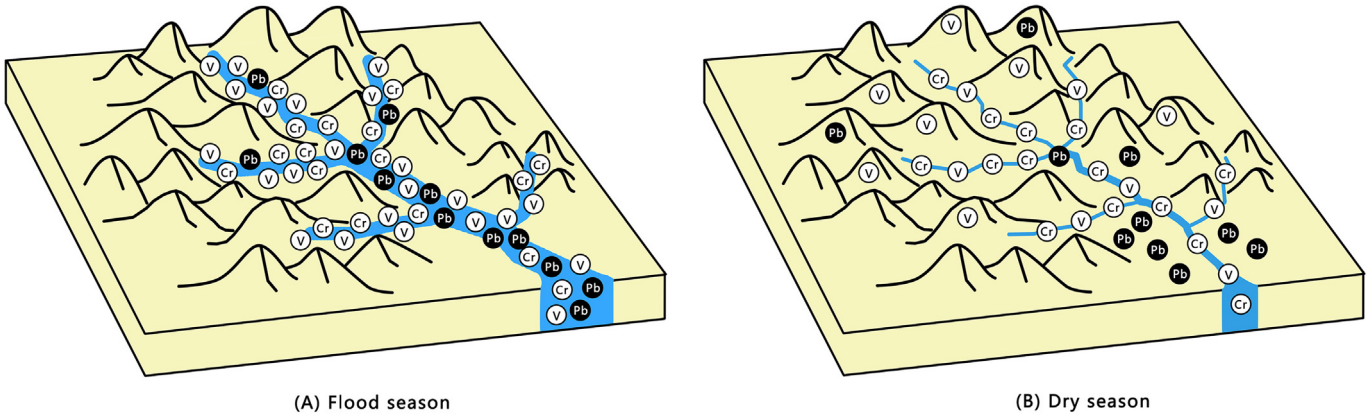

Fig. 9. Particulate heavy metal transport model by the Minjiang River. While and black circles represent different spatial variations while V, Pb and Cr represent different seasonal variations. V and Cr pollution is high in mountainous upstream and low in downstream due to geological background, mining and industry, but Pb pollution has no significant spatial variation because of human activities. (A) In flood season, high precipitation and runoff cause higher heavy metal pollution in river. (B) In dry season, some heavy metal pollutants are left ashore and river pollution decreases (e.g., V and Pb). However, incidental anthropogenic contamination results riverine Cr pollution increase.

major natural source is the crustal materials (including weathering, erosion and volcanic eruption) (Qiao et al., 2007; Qin et al., 2012; Wang et al., 2017a). The principal anthropogenic forcing includes mining (releasing heavy metals as tailings and metal-enriched dust) and smelting (releasing heavy metals as a result of high-temperature refining processes) (Zhang and Liu, 2002; Callender, 2005). Other important anthropogenic sources include fuel combustion, municipal waste incineration, discharge of industrial and domestic sewage, use of commercial fertilizers and pesticides, animal waste and waste-water discharge (Zheng et al., 2008; Che et al., 2017; Gong et al., 2018; Wu et al., 2018; Ma et al., 2019a). We employ the Pearson correlation analysis method to interpret potential sources of the particulate heavy metals in the Minjiang River and the results are shown in Table 5. For the seasonal SPM samples, Pb, Cd, Mo, Zn, V, Co, Ni and Cu show clear high correlations, especially between Pb and Zn ($r = 0.963$, $P < 0.001$), V and Zn ($r = 0.960$, $P < 0.001$), and V and Ni ($r = 0.954$, $P < 0.001$), suggesting that these heavy metals have the same geochemical behaviors or sources (Song et al., 2010). Likewise, Cr shows high correlation with Mn but Mn also shows correlation with Mo ($r = 0.804$, $P < 0.001$), Co ($r = 0.650$, $P < 0.001$) and Cd ($r = 0.626$, $P < 0.001$) (Table 5). According to the possible pollution sources of heavy metals (Table 3) with high Person correlations (Table 5), we contend the following several sources of heavy metal pollution in the Minjiang River: ore mining, industrial activities and urban development. Mining in the watershed and many densely populated cities near channels, which have a great amount of combustion emissions and discharge of agricultural wastewater and domestic sewage, can cause intensive heavy metal pollution of the Minjiang River. Industrial development also accounts for a certain degree of pollution into the Minjiang River.

As mentioned above, particulate heavy metals transported by the Minjiang River display remarkable heterogeneities in species and abundance between riverbed sediment and SPM samples, between upstream and downstream samples and among different season SPM samples (Figs. 3–6). It is well accepted that heavy metals in rivers are carried between particulate and dissolved phases through complicated processes of migration and transformation (e.g., complexation, resuspension and adsorption), and most of them accumulate in the former (Wang et al., 2017b; Borah et al., 2018; Li et al., 2018a). The SPM has loose structure, large specific surface area, colloidal properties and strong adsorption capacity for heavy metals through surface adsorption, chemical precipitation and ion exchange (Plach et al., 2011; Xia and Chen, 2012; Hahn et al., 2016). Thus, SPM is the main carrier and transporter of particulate heavy metals in river systems (Cui, 2000; Yang et al., 2014; Hu et al.,

Table 4
Statistical results of principal component analysis in seasonal SPM samples.

	PC1	PC2
V	0.896	−0.371
Cr	0.450	0.674
Mn	0.577	0.704
Co	0.973	0.042
Ni	0.932	−0.259
Cu	0.925	−0.142
Zn	0.940	−0.290
Mo	0.857	0.227
Cd	0.893	0.284
Pb	0.951	−0.207
Sc	0.684	−0.035
Percentage of variance (%)	70.9	13.1

Table 5
Pearson correlation coefficient of heavy metals in seasonal SPM samples of the Minjiang River.

	V	Cr	Mn	Co	Ni	Cu	Zn	Mo	Cd	Pb
V	1.000									
Cr	0.163	1.000								
Mn	0.275	0.519**	1.000							
Co	0.878**	0.404	0.650**	1.000						
Ni	0.954**	0.313	0.340	0.911**	1.000					
Cu	0.848**	0.396	0.408*	0.868**	0.920**	1.000				
Zn	0.960**	0.268	0.306	0.900**	0.953**	0.880**	1.000			
Mo	0.717**	0.318	0.804**	0.882**	0.712**	0.686**	0.727**	1.000		
Cd	0.661**	0.649**	0.625**	0.851**	0.728**	0.749**	0.786**	0.760**	1.000	
Pb	0.908**	0.287	0.397*	0.906**	0.918**	0.911**	0.963**	0.760**	0.815**	1.000

** At 0.01 level (double tail), the correlation is significant.
* At 0.05 level (double tails), the correlation is significant.

2015). Many factors can affect the heavy metal behaviors during transport, such as the pH and temperature of the river water, the nature and concentration of particulate matter, and the types and contents of heavy metals (Shen et al., 2006; Vaezi et al., 2016; Kumar et al., 2017). Changes in pH can alter the predominant species of heavy metals with different dissolvability and temperature is expected to influence the rate of adsorption and desorption (Cui, 2000; Zhang et al., 2008; Deng and Zhao, 2017). Increasing heavy metal concentrations and SPM concentrations in water also might promote the adsorption of heavy metals (Cui, 2000; Wang et al., 2012; Yang et al., 2014). The adsorption capacity of particulate matter to different heavy metal ions is related to particulate matter's chemical structure, composition and heavy metal adsorption mechanism (e.g., Cu and Pb are mainly bound to organic matter and iron oxides in particulate matter respectively) (Callender, 2005; Liu et al., 2007; Du Laing et al., 2008). All these factors can lead to different heavy metal pollution levels between the riverbed sediments and SPM in the Minjiang River.

The new data in this study obviously demonstrate that the particulate heavy metal pollution level in the Minjiang River was underestimated, compared with the previously published heavy metal data of the river (Hong et al., 2003; Chen, 2010; Cai et al., 2010; Xu et al., 2014b; Chen et al., 2014; Bi et al., 2017). Previous relevant studies, which mostly focused on estuarine surface bulk sediments, suggest overwhelmingly low-level pollution even no pollution for most heavy metals but only prominent Pb pollution. However, based on the SPM results, we argue that the Minjiang River is moderately polluted for most heavy metals but has strong Pb, Cd, Mo, Zn and Cr pollution. We also suggest that incidental anthropogenic emissions cannot be overlooked during short-term sediment transport and discharge of the Minjiang River.

5.6. Implications

The SPM-based heavy metal investigation in this study has at least two implications. First, our findings emphasize that there might be fairly different particulate heavy metal pollution assessment results based on different kinds of particle samples (e.g., bedload, riverbed sediments, SPM from different water depths and SPM over different timescales). As a large number of studies on heavy metal pollution of rivers focus on the estuary and surface bulk samples (Feng et al., 2004; Hung and Hsu, 2004; Marchand et al., 2006; Farkas et al., 2007; Fang et al., 2009; Song et al., 2010; Varol, 2011; Salah et al., 2012; Fu et al., 2012; Mwanamoki et al., 2014; Pheiffer et al., 2014; Yan et al., 2020), we highly recommend high spatiotemporal resolution investigations on SPM samples to better understand particulate heavy metal pollution levels in these rivers. Second, the new heavy metal datasets in this study reveal more complicated enrichment mechanisms and more diversified transport processes of the particulate heavy metals than previously known, in particular for those subtropical mountainous rivers (such the Minjiang River) in high-rainfall regions. The SPM discharged by the rivers, with previously underestimated heavy metal concentrations, can be transported further into marginal seas (due to ocean currents), not only those estuary and delta regions and thus might seriously impact very broad marine ecosystems (Jan et al., 2002; Jan et al., 2006; Wu et al., 2007; Lian et al., 2016; Wang et al., 2017b; Lin et al., 2019).

6. Conclusions

(1) The heavy metal concentrations of SPM samples are higher than those of associated riverbed sediment samples, which can be related to grain size, clay minerals and organic matter of particulate matter and environmental influences.
(2) There are two different spatial variation trends of particulate heavy metal concentrations in the Minjiang River basin. Some heavy metals (Cd, Zn, Cr, V, Co, Ni and Cu) show high concentrations in upstream samples and low concentrations in downstream samples, whereas Pb, Mo and Mn concentrations don't display this variation trend obviously.
(3) The particulate heavy metal concentrations also demonstrate great seasonal variations. Some heavy metal (e.g., Pb, Zn, V and Co) concentrations and related indices suggest a hydrologic control on transport and discharge of the particulate heavy metals by the Minjiang River. By contrast, Cd, Cr, Mn and Mo show abnormally high concentrations in some dry season samples due to incidental anthropogenic input events.
(4) The transport and discharge of particulate heavy metals by the Minjiang River are controlled by both natural and anthropogenic forcings. According to environmental assessment results of EF, I_{geo} and RI, we argue that the Minjiang River is worse polluted by particulate heavy metals than previously known and thus potential ecological risks caused by particulate heavy metals that cannot be ignored.

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CRediT authorship contribution statement

Jiayu Fan: Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Xing Jian:** Conceptualization, Resources, Writing – original draft, Writing – review & editing, Supervision, Funding acquisition. **Fei Shang:** Writing – original draft, Writing – review & editing. **Wei Zhang:** Resources, Writing – original draft, Project administration, Writing – review & editing. **Shuo Zhang:** Writing – review & editing. **Hanjing Fu:** Writing – review & editing.

Declaration of competing interest

We declare that we don't have any conflict of interest.

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