



Seasonal geochemical heterogeneity of sediments from a subtropical mountainous river in SE China

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ABSTRACT

The Zhe-Min mountainous rivers in SE China are characterized by stable tectonics, small–mesoscale drainage basins, relatively homogenous bedrocks, dense vegetation cover and high rainfall conditions. These rivers deliver loads of sediments and nutrients to the oceans and always indicate considerably high variability of sediment flux on short-term timescales. However, short-term sediment compositional variations and transport mechanism by these mountainous rivers remain poorly understood. Here we target the Minjiang River, use mineralogical and trace element geochemical data of suspended particulate matters (SPMs) within a one-year hydrological cycle (from October 2016 to October 2017), riverbed sediments (< 0.063 mm fractions) and SPMs from the mainstream and major tributaries, to investigate their provenance and transport process. The results indicate that the SPMs in different seasons have strong seasonal geochemical heterogeneity, with highly positive correlations between several trace element ratios (e.g. $La_{(CN)}/Yb_{(CN)}$ and Th/Sc) and runoff of the river. Riverbed sediments and SPMs collected in pairs have distinct geochemical compositions. The riverbed sediments have relatively low $La_{(CN)}/Yb_{(CN)}$ (ca. 10–15) and Th/Sc (ca. 2–5) ratios, whereas the SPMs have $La_{(CN)}/Yb_{(CN)}$ and Th/Sc ratio ranges of ca. 14–17 and ca. 4–7, respectively. These findings suggest a predominant hydrologic control on the Minjiang River sediment transport and geochemical compositions on a seasonal timescale. We conclude that exposed riverbed sediments are probably selectively recycled in dry-season sediment transport, whereas the flood-season SPMs are less mixed with riverbed sediments and are likely dominated by particulates directly from those tributaries and the upper mainstream. We favor that the Zhe-Min mountainous rivers are featured by rapid sediment delivery in response to hydroclimatic changes and have similar sediment transport behaviors with those small mountainous rivers in tectonically active regions of the subtropical East Asia (e.g. Taiwan). This case study also highlights the importance of multi-spatio-temporal scale, heterogeneous river input of chemical elements to the oceans.

1. Introduction

Rivers serve as the major link between land and sea, historically discharging annually ca. 36,000 km³ of freshwater and about 20 billion tons of sediments and solutes (including ca. 0.38 Gt total organic carbon) to the global ocean (Ludwig et al., 1996; Milliman and Farnsworth, 2011). The transport of materials by rivers, which provides essential information both on surface processes (physical, chemical, biological and anthropogenic) of the continents and on the amount and nature of terrigenous inputs to the oceans (Martin and Meybeck, 1979; Gaillardet et al., 1999a; Bridge and Demicco, 2008), is particularly crucial to better understanding of land-ocean interaction, geochemical

cycle and global change.

Large rivers dominate most of the global population and economy in the world. These rivers were originally considered to account for the great mass of water and sediment discharges to the global ocean and thus have been widespread concerned for decades (Meybeck, 1982; Gaillardet et al., 1999a, 1999b; Nilsson et al., 2005; Bianchi and Allison, 2009; Viers et al., 2009). Many large rivers tend to experience relatively little seasonal or inter-annual variability in sediment discharge, by regulating short-term meteorological events through their watersheds (Meybeck et al., 2003; Walling and Fang, 2003). By contrast, most mountainous rivers behave quite differently from large rivers. Rivers running across mountains are characterized by steep

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gradients, relatively small drainage basins and the general lack of downstream sediment storage, resulting in greater erosion rate and less space for sediment accommodation than those large rivers. Therefore, mountainous rivers have been recently recognized as much more important contributors of clastic sediments and particulate organic matters to the ocean than previously thought (e.g. Milliman and Syvitski, 1992; Kao and Liu, 1996; Wheatcroft et al., 1997; Carey et al., 2005; Coynel et al., 2005; Kao and Milliman, 2008; Korup, 2012). Because of these watershed characteristics, mountainous rivers always indicate considerably high variability in sediment fluxes on seasonal- or event-timescales (Farnsworth and Milliman, 2003; Milliman and Kao, 2005; Gislason et al., 2006; Hilton et al., 2008; Sadeghi et al., 2008; Hatten et al., 2012; Conaway et al., 2013; Goñi et al., 2013). However, transport mechanism and short-term compositional variability of the delivered sediments by mountainous rivers remain poorly understood.

Erosion, transport and yield of river sediments are a function of precipitation, runoff, tectonics, bedrock lithology, basin morphology and vegetation cover, and even human activity (Milliman and Syvitski, 1992; Walling and Fang, 2003; Leithold et al., 2006; Kao and Milliman, 2008). While seasonal variations of sediment export by mountainous rivers are usually related to hydroclimatic factors (e.g. seasonal rainfall and runoff) or differential denudation of sub-watersheds (Mano et al., 2009; López-Tarazón et al., 2010; Hatten et al., 2012; Navratil et al., 2012; Goñi et al., 2013), triggers for an extreme sediment transport event generally include volcanic eruptions, earthquake- and storm-driven landslide episodes, catastrophic dam breaks and typhoon-generated mega-floods (Milliman and Kao, 2005; Hilton et al., 2008; Townsend-Small et al., 2008; Korup, 2012 and references therein; Milliman et al., 2017; Su et al., 2018). Most previous studies focused on small mountainous rivers in tectonically active regions, such as the western coast of North America, New Zealand, Japan and Taiwan (e.g. Carey et al., 2005; Sadeghi et al., 2008; Conaway et al., 2013; Milliman et al., 2017), and emphasized the significance of event-triggered extreme sediment export, whereas short-term monitoring of sediment transport by mountainous rivers in relatively stable regions is rarely reported. On a global scale, rivers running through tropical and subtropical mountains (most in South and East Asia) deliver the greatest loads of sediments, organic carbon and nutrients to the ocean (Milliman and Farnsworth, 2011). This raises awareness of the importance of mountainous rivers in low latitudes, especially the high rainfall regions.

It is well known that the low latitude East Asia develops two distinct types of river systems (Fig. 1A). These include tectonically stable continental large rivers (e.g. Pearl and Mekong rivers) and tectonically active mountainous rivers (e.g. rivers on the Taiwan island) (Yang et al., 2014; Bi et al., 2015). The Zhe-Min Rivers, such as Oujiang, Minjiang and Jiulongjiang Rivers (Fig. 1B), as important exorheic, mountainous rivers along the SE coast of China, are developed on the stable East Asia continent but have obviously different watershed topography (Fig. 1B) and climate from the large rivers. These river basins are instead under similar subtropical climatic conditions with the Taiwan rivers (but different geological settings), which are currently dominated by the East Asian monsoon system and high-frequency tropical cyclone activities (Yin et al., 2010; Chang et al., 2012). How the Zhe-Min mountainous rivers deliver sediments to the marginal seas under such backgrounds remains poorly known.

In this study, we target the Minjiang River (Fig. 1) and present grain size, mineralogical, petrographic and trace element geochemical results and corresponding interpretations for suspended particulate matters (SPMs) within a one-year hydrological cycle (from October 2016 to October 2017) at a fixed location in the lower reach and for riverbed sediments and SPMs from the mainstream and major tributaries (Fig. 2A). The aims are to: (1) evaluate the spatial and seasonal variations in geochemical compositions of the Minjiang River-delivered sediments and (2) decipher sediment transport process and mechanism of this tectonically stable, subtropical mountainous river on seasonal timescales.

2. Regional setting

The Minjiang River, as the largest river in Fujian Province, SE China, originates from the Wuyi Mountains and flows east into the East China Sea (Fig. 1B). The watersheds of the river system mainly sit 500–2000 m above the sea level and spread over the region of 25–29° north latitude and 116–120° east longitude (Fig. 2), with the drainage area of ca. 61,000 km². The river basin is dominated by subtropical humid monsoon climate and has average annual air temperature of 16–20 °C and annual precipitation of 1500–2000 mm (Zhu et al., 2018). The catchment is always influenced by tropical cyclones (Yin et al., 2010; Chang et al., 2012), which can trigger short-term extreme rainfall and flood events, especially in the downstream regions. This kind of climatic background leads to highly spatial variability of rainfall in different seasons, i.e. relatively high precipitation in the upstream watershed during the March–June and intensive rainfall in the downstream watershed during July–September (Yu, 2002).

The Minjiang River basin is on the Cathaysia block under a tectonically stable background. The upstream (such as the tributaries Jianxi, Futunxi and Shaxi) bedrocks mainly comprise Precambrian metamorphic and igneous rocks, Paleozoic sedimentary rocks and Mesozoic igneous rocks (Fig. 1C–D), whereas the downstream (such as the tributaries Meixi and Dazhangxi) bedrocks are dominated by Jurassic–Cretaceous volcano-sedimentary rocks with subordinate Late Yanshanian (Cretaceous) plutons (Chen and Jahn, 1998; Li et al., 2014).

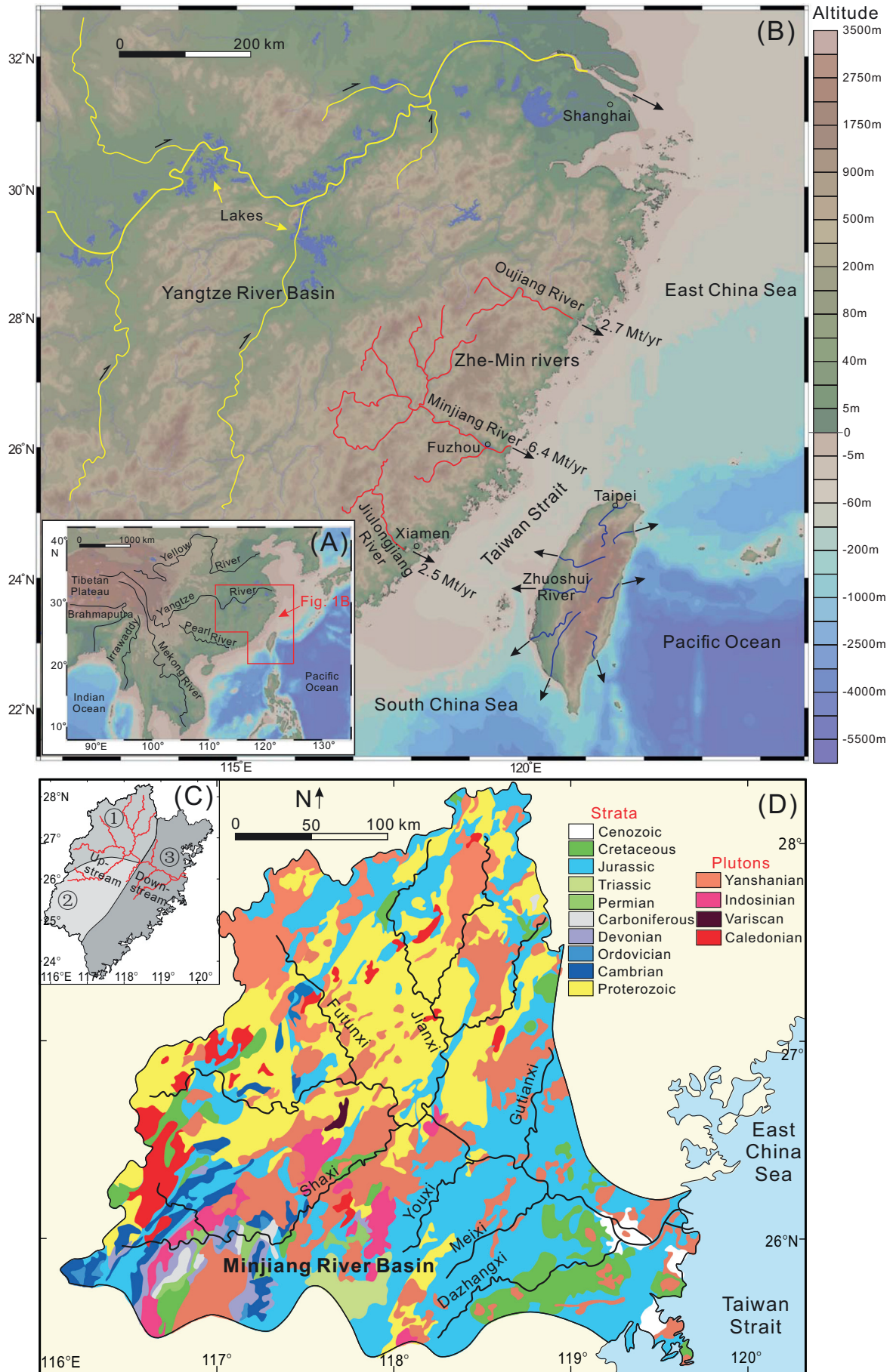
The Minjiang River historically export ca. 54 billion m³ of fresh water and ca. 6 million tons of sediment annually (MWRPRC, 2016). However, sediment discharge of the river shows a decreasing trend due to increasing human activities (such as construction of dams and reservoirs (Fig. 2A)) along the river in past decades (Dai et al., 2009). Almost 70–80% of rainfall and runoff occur in the period of April to September each year (Zhu et al., 2018) and the sediment discharge flux displays remarkably seasonal and inter-annual variability (Fig. 2B). Due to the influence of El-Nino and La-Nina climate phenomena, much more precipitations and rainstorm events happened in 2016 than those in previous and following years (ICMWRPRC, 2016, 2017). This resulted in much higher sediment discharge by the Minjiang River in 2016 than that in 2017 (Fig. 2B).

3. Samples and analytical methods

To obtain SPM samples, river water was collected at surface of river channels using a clean water sampler. SPMs were then filtered from the river water through 0.45 μm membranes. Twenty-five SPM samples were collected nearly semimonthly from October 2016 to October 2017 at the fixed S04 station in the downstream river channel, about 50 km away from the river mouth (Fig. 2A). Note that the S04 station was not influenced by tide flow from the ocean during each sampling. We also collected 13 SPM samples from the major tributaries and different locations along the mainstream (Fig. 2A) in October 2016. Furthermore, mud and fine sand sediment samples (Fig. 2A) were collected from exposed, accessible riverbeds where sediment was deposited during high water levels. Fractions of < 63 μm were further separated by wet sieving for grain size, mineralogical, petrographic and geochemical analysis.

SPM and riverbed sediment (< 63 μm fractions, the same below) smears were made for petrography analysis. Each targeted sample was evenly placed at a glass, wet by dripping about 0.1 ml water and then dried at 110 °C. Canada balsam was employed to adhere the samples to the glass. The smears were observed under a polarizing microscope after consolidation.

Grain size distributions of the SPMs and riverbed sediments were analyzed by a Malvern laser particle analyzer (Mastersizer 3000) at Xiamen University. The samples were treated with 30% H₂O₂ and 10% HCl to remove organic matter and carbonate components, respectively. Addition of 0.5 M (NaPO₃)₆ and ultrasonic dispersion were employed to



(caption on next page)

Fig. 1. Location and background of the East Asia river-sea system and the Minjiang River. (A) East Asia continental margins and major rivers on the East Asia continent (modified from Jian et al., 2020). These rivers were mainly derived from the Tibetan Plateau. (B) Representative subtropical mountainous rivers (red lines: major Zhe-Min rivers; blue lines: major Taiwan rivers) into the sea in SE China. Black arrows indicate the directions of the sediment discharge. (C) Stratigraphic divisions of the Fujian Province, SE China. 1) Proterozoic; 2) Paleozoic; 3) Jurassic–Cretaceous. (D) Geological map of the Minjiang river basin (Jian et al., 2020). Note that the upstream river basin is dominated by Proterozoic and Paleozoic rocks, while the downstream regions mainly consist of Jurassic–Cretaceous rocks. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

ensure complete disaggregation of particles before the grain size analysis.

Mineralogical analysis of the SPMs and riverbed sediments was carried out by using a Rigaku X-ray diffractometer (Ultima IV) at Xiamen University and followed the analytical procedures given by Jian et al. (2014, 2019). Each powdered sample was continuously scanned under the conditions of 40 kV, 30 mA, wave length of 1.54, step width of 0.02° and scanning speeds of 4°/min.

A total of 45 samples were selected for trace element geochemistry analysis (Table A1 in the Appendix A). All the samples were powdered by using an agate mortar and were treated by 1 N acetic acid for 24 h to remove carbonate components. The residues were dried at 50 °C and then combusted in a muffle furnace at 600 °C for 2 h. The pretreated samples were then accurately weighed and completely digested by HNO₃-HF mixture acid solutions. An Agilent 7900 ICP-MS at State Key Laboratory of Marine Geology (Tongji University) was employed for trace element analysis. The GSR-5 standard materials were used to monitor analytical quality. Both the analytical accuracy and precision were estimated to be < 10% for most trace elements. The detailed treatment and analytical procedures were given in Guo et al. (2018) and Deng et al. (2019).

4. Results

Representative photomicrographs of the riverbed sediments and SPMs are shown in Fig. 3A–B. The results indicate that the < 63 μm fractions of riverbed sediments have relatively larger detritus than the associated SPMs, and are rich in blocky quartz, feldspar and lithic fragments, whereas the SPMs contain a certain amount of flaky micas (Fig. 3A–B). Laser particle analyzer-based grain size analysis results show that the SPMs have diverse grain size distributions (Fig. A1 in the Appendix B) and are mainly composed of silt and clay (mean sizes ranging of 2–20 μm, Fig. 3C). Furthermore, the analyzed two riverbed sediments have mean grain sizes of 14–17 μm (Fig. 3C).

The XRD analysis results of the selected SPMs and riverbed sediments indicate that these sediments are mainly composed of quartz, K-feldspar, plagioclase and clay minerals (dominated by kaolinite and illite) (Fig. A2 in the Appendix B). The SPMs therein have comparatively higher clay mineral and lower feldspar compositions than the associated riverbed sediment samples. Besides, the SPM samples are surprising to have similar or even higher quartz abundances (46%–64%, averaging in 56%) than the associated riverbed sediment samples (46%–59%, averaging in 53%).

Trace- and rare earth element data are shown in Table A1. Element concentrations of all the analyzed samples were normalized to the Upper Continental Crust (UCC) compositions (Taylor and McLennan, 1985) and are shown in Fig. 4. Fig. 5 illustrates the comparison of trace element concentrations and representative element ratios for those SPM and riverbed sediment samples collected in pairs (i.e. at the same location). Overall, the Minjiang River sediments have roughly similar UCC-normalized trace element patterns with the Post-Archean Australian Shale (PAAS) and the SPMs discharged by the Yangtze, Pearl and Mekong rivers (Fig. 4). The analyzed samples display variable large ion lithophile element (LILE) compositions (e.g. remarkable depletion in Sr and enrichment in Rb and Cs relative to UCC). Most samples are rich in rare earth elements (REEs) compared with the UCC (Fig. 4).

The SPM samples have comparatively higher LILE and REE and lower high field strength element (HFSE, e.g. Zr and Hf) concentrations

than the associated riverbed samples (Fig. 5). Riverbed sample 16MJ-18 has higher heavy REEs (e.g. Tm, Yb and Lu) and much higher HFSE concentrations than sample 16MJS-10 (Fig. 5), likely due to detrital zircon enrichment in the riverbed sediment phases. Note that the SPM samples from upper and lower tributaries and different locations of the river mainstream in the same season have indistinguishable elemental ratio values, such as Th/Sc (3.6–7.0), La_(CN)/Yb_(CN) (13.3–16.6), Th/U (4.5–5.7) and La/Sc (8.0–13.7) ratios, which are obviously higher than those of riverbed sediment samples (Fig. 6; Table A1). However, the SPM samples from different seasons collected at the fixed station S04 (Fig. 2A) have variable Th/Sc, La_(CN)/Yb_(CN), Th/U and La/Sc ratios, with the ranges of 2.2–5.1, 7.5–17.6, 3.8–5.1 and 4.8–11.6, respectively. Furthermore, several riverbed sediment samples (16MJ-15, 16MJ-18 and 16MJ-26) show relatively high Hf abundances (11–24) and Zr/Sc values (51–62) (Table A1).

5. Discussion

5.1. Sediment geochemical variability of the Minjiang River catchment on multi-spatial scales

Since Sc is generally rich in mafic igneous rocks (Taylor and McLennan, 1985; McLennan et al., 1993), relatively low Sc abundances (varying from 4.7–14.7 ppm, averaging 8.9 ppm) of all the samples reveal a predominant felsic source for the river sediments (Fig. 4; Table A1). The strong contribution of felsic source rocks is also reinforced by low La/Th ratios (varying from 1.8 to 2.9, averaging 2.2) and Chondrite-normalized REE patterns (i.e. classical light REE enrichment, negative Eu anomaly and flat heavy REE distribution) of the analyzed samples, showing the characteristics of PAAS-like sediments (Figs. 4–6; Fig. A3 in the Appendix B). Although the upstream and downstream regions of the river catchment are characterized by different bedrocks (Fig. 1), our data demonstrate that the SPMs from upper tributaries (e.g. Jianxi, Shaxi and Youxi) have similar REE, Th and Sc features (e.g. the La/Th, Th/Sc, La_(CN)/Yb_(CN), Th/U and La/Sc value ranges are indistinguishable) to those from lower tributaries (Fig. 6).

By contrast, the upper tributary SPMs have different LILE features from the lower tributary SPMs. Specifically, the upper tributary samples have obviously higher Rb/Sr, α_{Sr} ($\alpha_{Sr} = [Nd/Sr]_{sed}/[Nd/Sr]_{UCC}$) and α_{Ba} ($\alpha_{Ba} = [Th/Ba]_{sed}/[Th/Ba]_{UCC}$) values than the lower tributary samples (Fig. A4 in the Appendix B). It is well known that Sr and Ba are very active during water-rock interactions and thus tend to concentrate in weathering solution as weathering progresses (Canfield, 1997; Gaillardet et al., 1997), while Rb, Nd and Th (the magmatic compatibility of Nd and Th is close to that of Sr and Ba, respectively) are relatively immobile and are preferentially retained in weathering residual solid products. Hence, these trace element-based proxies have been proposed as useful indicators for evaluating chemical weathering intensity; and to some extent higher Rb/Sr, α_{Sr} and α_{Ba} values indicate stronger chemical weathering (Chen et al., 1999; Gaillardet et al., 1999b; Yang et al., 2004; Jin et al., 2006; Guo et al., 2018). Although the hydrodynamic sorting during sediment transport, which can lead to different sediment grain sizes, can impact geochemistry-based weathering proxies (Xiong et al., 2010; Bouchez et al., 2011a, 2011b; Garzanti et al., 2011; von Eynatten et al., 2012; Jian et al., 2013; Guo et al., 2018), the analyzed upstream and downstream SPMs have similar and quite small ranges of mean grain sizes (Fig. 3C) and no obvious relationship is observed between the grain sizes and the Rb/Sr ratios

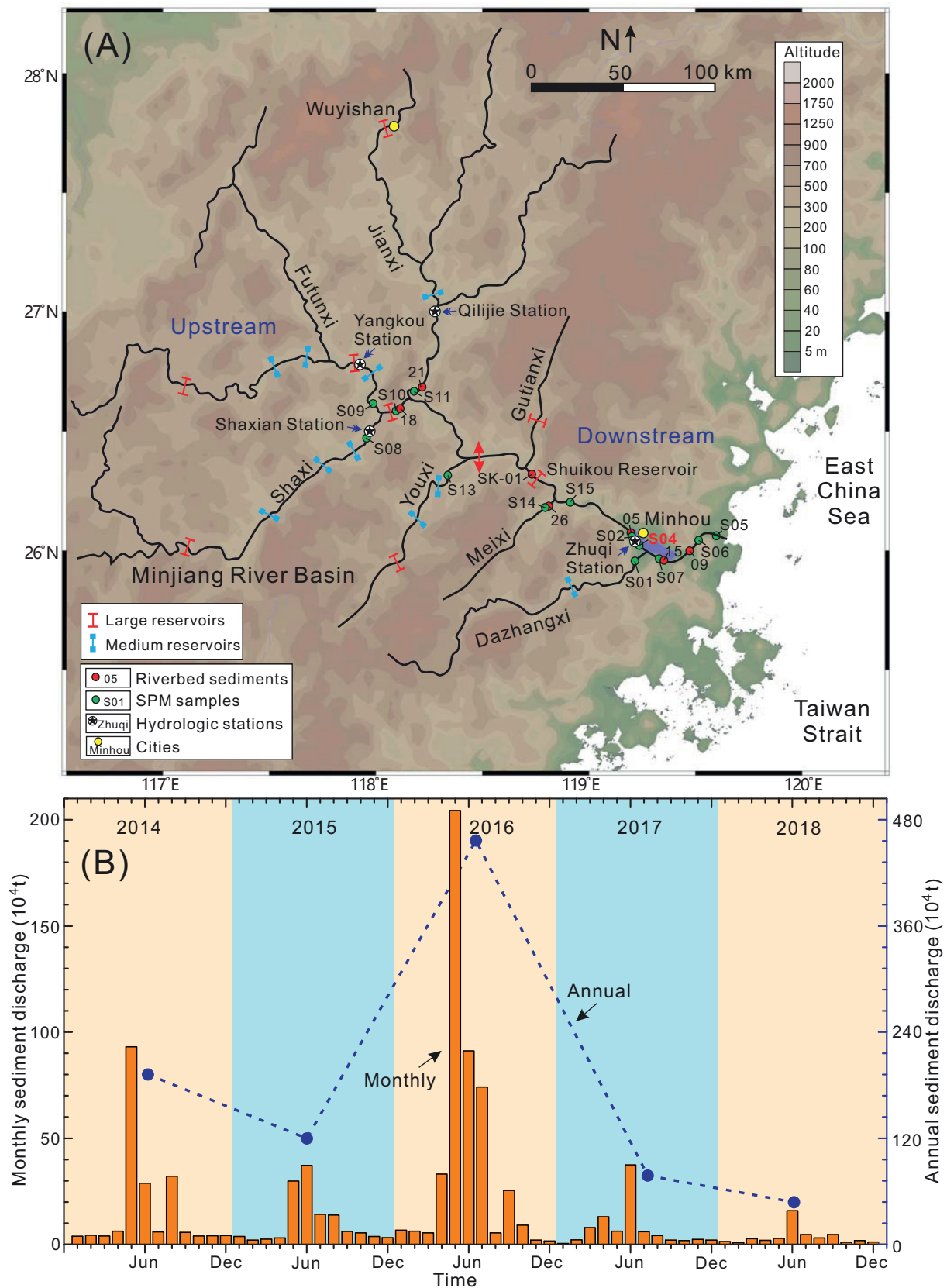


Fig. 2. (A) Topography of the Minjiang river basin, major water reservoirs, representative hydrologic station locations and sample locations. We roughly divide the river basin into upstream and downstream regions based on the geological settings (the boundary is indicated by the red line with double sided arrows). Note that different season SPMs were collected at the S04 station, where the tidal currents consistently could not reach during the sampling seasons. The locations of reservoirs are from the Fujian water resources information network (<http://slt.fujian.gov.cn>). (B) Annual and monthly sediment discharge of the Minjiang River during 2014–2018 (data from the Ministry of Water Resources, the People's Republic of China). The discharged sediment flux shows obviously seasonal and inter-annual variability. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

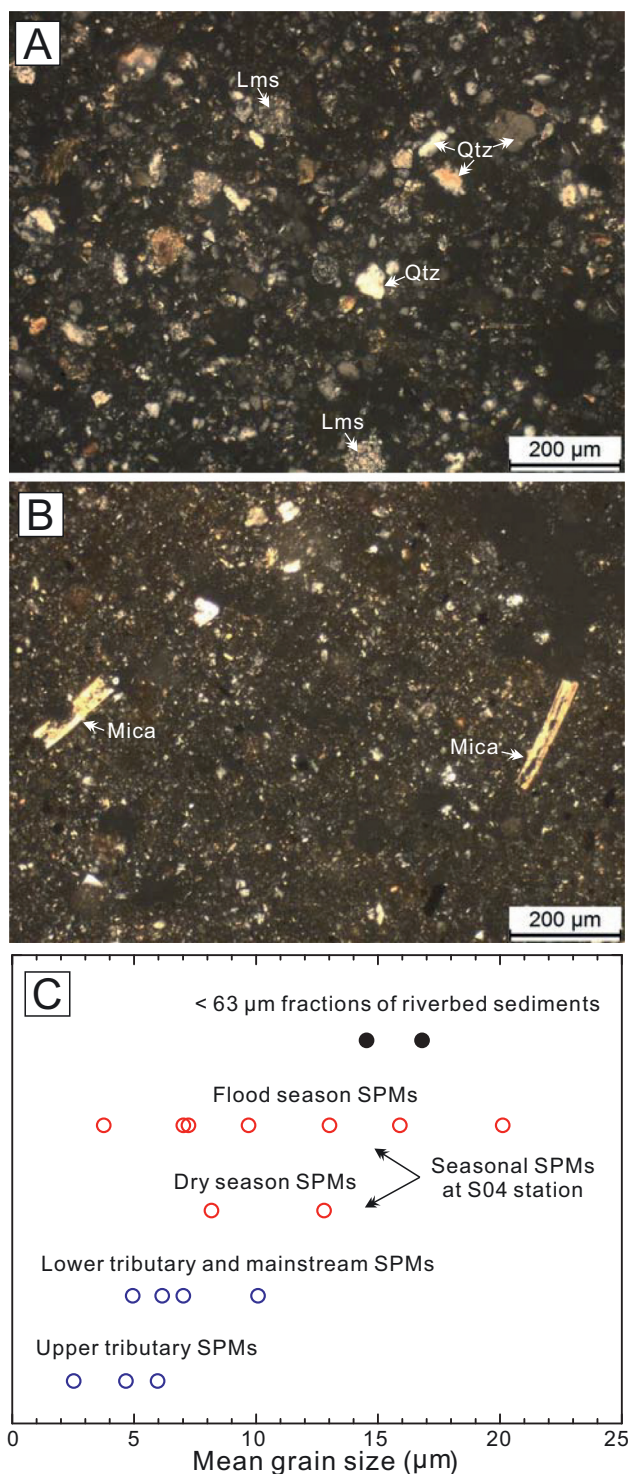


Fig. 3. Representative photomicrographs of the analyzed riverbed sediments and SPMs. (A) Riverbed sediment sample 16MJ-26 (< 63 μm fractions). (B) SPM sample 16MJS-14. (C) Mean grain size values of all the analyzed samples. Qtz: quartz; Lms: metasedimentary lithic fragment.

(Fig. A4), indicating a minor control of hydrodynamic sorting on the SPM chemical weathering intensity proxies. In this case, the geochemical data suggest that the upstream regions (with Rb/Sr, α_{Sr} and α_{Ba} value ranges of 3.2–4.9, 13.3–22.0 and 3.7–7.2 (Table A1; Fig. A4), respectively) of the Minjiang River basin experience stronger chemical weathering than the downstream regions (with Rb/Sr, α_{Sr} and α_{Ba} value ranges of 2.6–3.4, 11.6–12.4 and 2.5–3.9, respectively). This is

consistent with clay mineralogy results which reveal that the upstream river sediments have higher kaolinite/(illite + chlorite) ratios than the downstream sediments (Xu et al., 2013; Feng et al., 2014; our unpublished data). Potential explanations for the spatial difference of chemical weathering intensity of the river basin include, 1) the upstream river basin has higher annual precipitation than the downstream regions due to the monsoon-dominated climate and topography variations (Yu, 2018); 2) the downstream (and coast) regions always suffer from typhoon-induced rainstorms which probably increase landslide, soil erosion and physical weathering (Wang et al., 2012; Huang and Cheng, 2013) and thus result in relatively low chemical weathering intensity in the downstream regions.

Our data also show that the SPMs have quite different trace element concentrations and ratios from the associated riverbed sediments (Figs. 4–6). This is most likely attributed to the hydrodynamic sorting process during sediment transport and deposition in which quartz, feldspar and some heavy minerals tend to place in riverbed sediments, while SPMs are generally dominated by phyllosilicates (e.g. clay minerals and micas) (Bouchez et al., 2011a, 2011b; Garzanti et al., 2011). The inference is reinforced by the differential petrographic compositions of the SPM and riverbed sediment samples, which show that the SPMs are rich in clay minerals and flaky micas (Fig. 3A–B). A great number of studies have pointed out that modern sediments or ancient sedimentary rocks with the same sources but different grain sizes (or different size fractions of one sediment sample) are expected to have distinct major, trace and rare earth element concentrations and ratios (e.g. Cullers, 1994, 1995; Nesbitt et al., 1996; Vital and Statterger, 2000; Singh, 2009; Garzanti et al., 2010; Lupker et al., 2011; Jian et al., 2013; Guo et al., 2018; Deng et al., 2019). Physical sorting can dominate transport and deposition of sediments based on grain size, shape and density and thus results in mineralogical and geochemical differentiation (Garzanti et al., 2010). For instance, all the riverbed samples in this study have much higher Zr/Sc values than the associated SPM samples. This can be attributed to the enrichment of zircon (Belousova et al., 2002) in the riverbed phases. The overwhelmingly higher concentrations of REEs and LILEs in most SPMs than associated riverbed sediment samples (Fig. 5), are due to the comparative enrichment of clay minerals and flaky micas in the suspended phases (Fig. 3A–B). This is supported by the XRD-based mineral composition analysis results which indicate relatively high kaolinite (and similar quartz contents) compositions in the SPMs (Fig. A2; Fig. 5). Although most previous studies emphasized quartz (or calcite) dilution resulting in low concentrations for most trace elements in bedload or sandy sediments (e.g. McLennan et al., 1990; Cullers, 1995; Singh and Rajamani, 2001; Roddaz et al., 2006; Bouchez et al., 2011a, 2011b), we favor the major control of phyllosilicates on sediment trace element concentrations in this study. Some trace elements, such as REEs, can easily enter the clay mineral lattice or be adsorbed onto the clay mineral surface, which can result in one to two orders of magnitude higher trace element concentrations of clays than those of quartz and feldspar (Condie, 1991; Vital et al., 1999; Garzanti et al., 2011).

5.2. Geochemical and provenance variability of the Minjiang River-discharged sediments on seasonal timescales

The geochemical results of SPMs collected at the S04 station (Fig. 2A) in different seasons of 2016–2017 demonstrate strong variations in some trace elements (Fig. 7). Specifically, the SPMs of October–November 2016, late March–April 2017 and June–August 2017 have relatively higher La/Sc (most in the range of 7.7–11.6), Th/Sc (3.6–5.1), Th/U (4.5–5.1) and La_(CN)/Yb_(CN) (14.0–17.6) ratio values than the SPMs of other periods (the La/Sc, Th/Sc, Th/U and La_(CN)/Yb_(CN) ratios ranging 4.8–8.4, 2.2–4.7, 3.8–4.7 and 7.5–14.6, respectively) (Table A1).

Long-term monitoring results demonstrate a decrease trend in sediment discharge of the Minjiang River during the past decades and

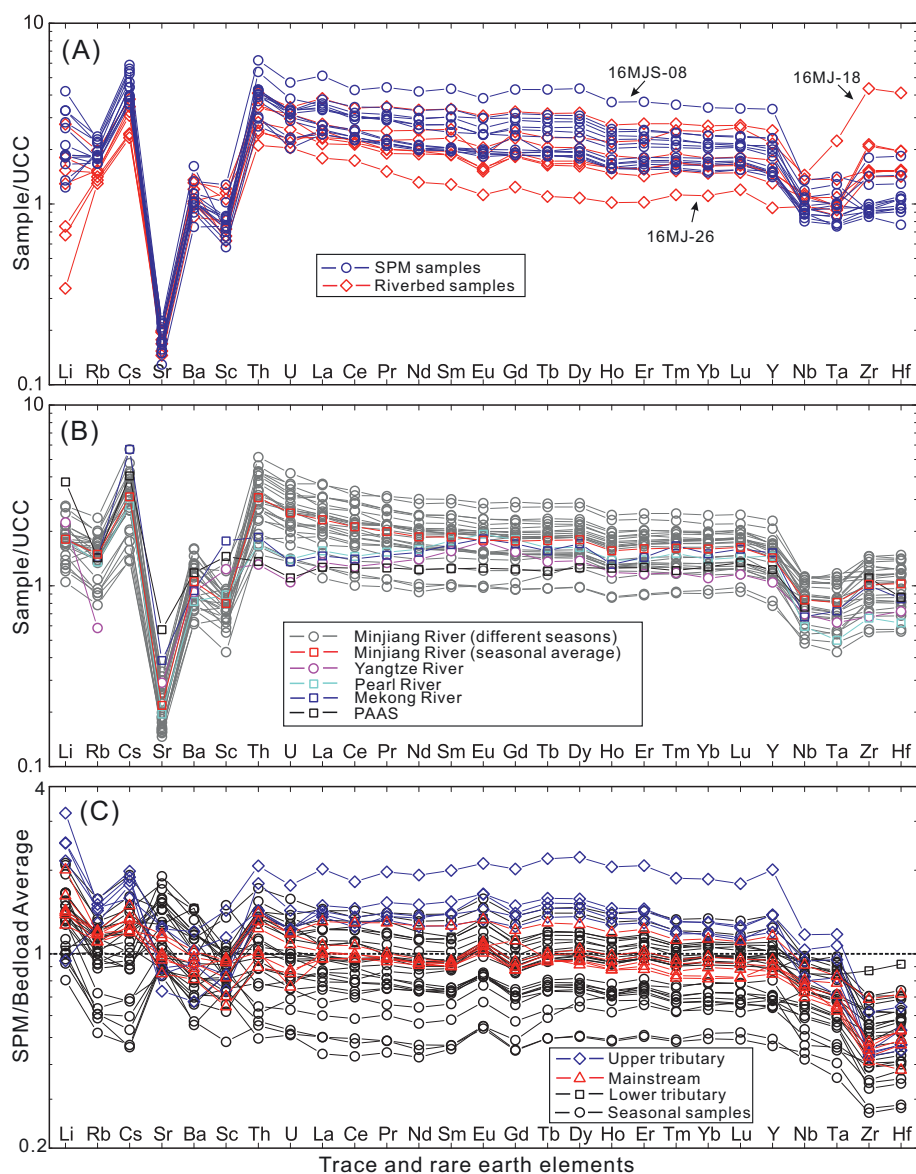


Fig. 4. Trace element geochemical results of all the Minjiang River sediments and SPMs. (A) All the tributary and mainstream samples; (B) all the SPM samples from different seasons collected at the S04 station (Fig. 2A); (C) trace element concentrations of all the SPM samples normalized by average values of the riverbed samples. All the samples show significant Sr depletion relative to the Upper Continental Crust (UCC) compositions, revealing relatively intensive chemical weathering of the catchment. The sediments have similar UCC-normalized trace element patterns with the Post-Archean Australian Shale (PAAS). Sample 16MJ-18 indicate obvious enrichments of Zr and Hf, probably due to high zircon abundance in the analyzed sample. The UCC and PAAS data are from Taylor and McLennan (1985). The data of Yangtze, Pearl and Mekong river SPMs are from Gaillardet et al. (1999b); Zhang and Wang (2001) and Yang et al. (2003).

human activities (such as construction of dams and reservoirs) are regarded as a principal cause for the decline of sediment flux (Dai et al., 2009; MWRPRC, 2016). The Shuikou Reservoir, as the most important reservoir in the middle mainstream (Fig. 2A), optionally increased flood-discharge (marked by sharp declines of the water level) after several strong rainfall events during the sampling seasons (Fig. 7). However, the geochemical signals of the seasonal SPMs did not respond to the water release events (Fig. 7). This means that the dams and reservoirs play a minor role in geochemical compositions of the discharged SPMs on the seasonal timescales. Other human activities, such as land use and river sand mining, might seasonally vary, but are hard to be precisely quantified for this mesoscale river basin. Furthermore, there is no obvious correlation between these trace element ratios and mean grain sizes (Fig. A5 in the Appendix B), implying that hydrodynamic sorting-induced mineralogical differences might not be the major control on these chemical elements. These element ratios (e.g. monthly average values of $La_{(CN)}/Yb_{(CN)}$ and Th/Sc) display highly positive correlations with monthly runoff data of the river instead (Fig. 8). Therefore, we suggest a hydrologic control on the trace element geochemical compositions (i.e. seasonal heterogeneity) of the Minjiang River-discharged SPMs. As expected, the Minjiang River displayed comparatively high water-discharges during those three periods

with high sediment trace element ratios (Fig. 7). The late March–April and June–August 2017 therein were wet seasons when the river basin experienced high precipitations (Fig. 7). Although October–November is generally regarded as a comparatively dry season and the river basin is always rainless in this period, the October–November 2016 Minjiang River catchment had much more precipitations than the same period of previous and following years (ICMWRPRC, 2016, 2017). Furthermore, two typhoon-triggered rainstorm events (Meranti and Megi typhoons) happened in late September 2016 (Fig. 7). These climatic backgrounds and events resulted in fairly high runoff conditions of the Minjiang River in October–November 2016 (Fig. 7).

We further find that the SPMs of high-runoff period (i.e. high-water stage or so-called flood season) have similar La/Sc , Th/Sc , Th/U and $La_{(CN)}/Yb_{(CN)}$ ratio values with the upper and lower tributary SPMs, whereas these element ratios of the low-runoff period (i.e. low-water stage or so-called dry season) samples are close to or even lower than those of the analyzed riverbed sediments (Fig. 7). This reveals distinct provenance and transport mechanisms of the Minjiang River-delivered sediments in flood and dry seasons. We favor that solid materials on exposed riverbeds and overbanks are selectively recycled during the transport process in dry seasons. For example, detrital zircons, as common ultra-dense grains ($> 4.5 \text{ g/cm}^3$) in bedload sediments

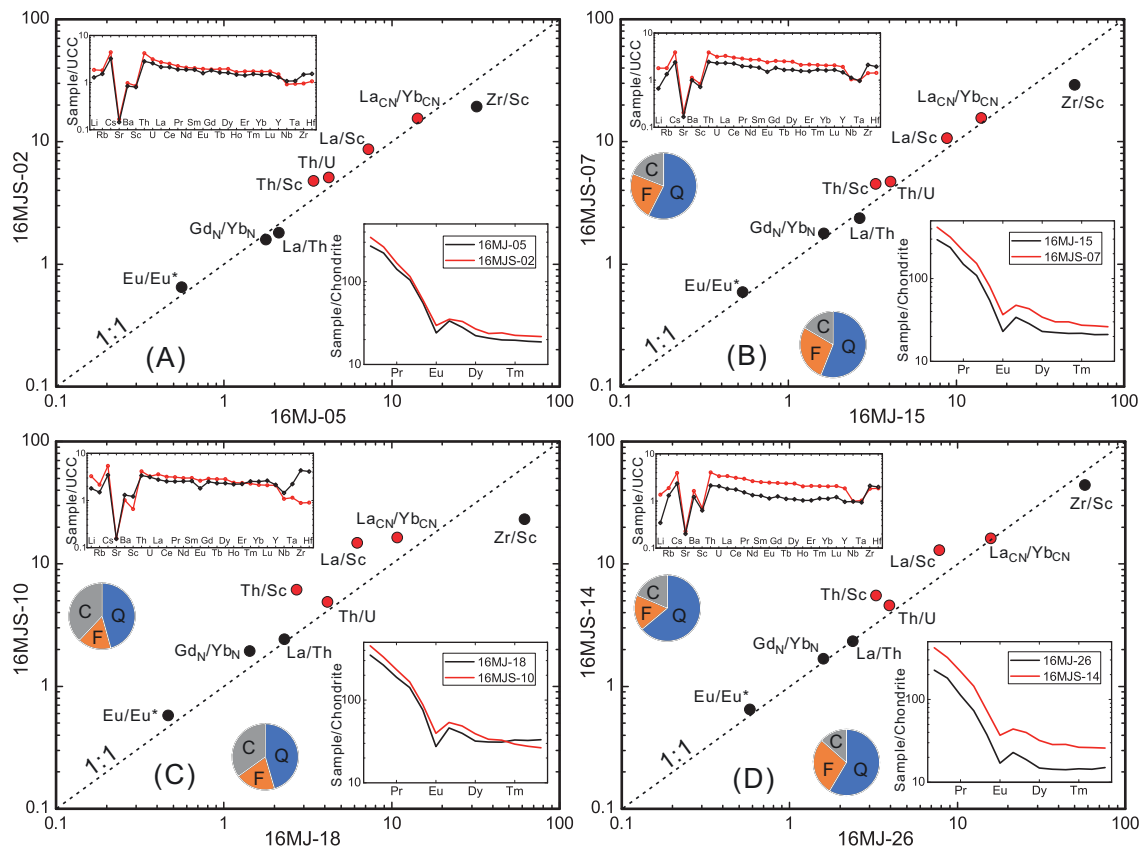


Fig. 5. Comparison of UCC-normalized trace element distribution patterns, chondrite-normalized REE patterns and representative trace element ratios between riverbed and SPM samples collected in pairs (i.e. at the same location). Samples 16MJ-05, 16MJ-15, 16MJ-18 and 16MJ-26 are riverbed sediment samples. Note that the SPMs have relatively higher Th/Sc, La/Sc, Th/U and $La_{(CN)}/Yb_{(CN)}$ (ratio of Chondrite-normalized values) ratios than the associated riverbed sediments. Higher Zr/Sc ratios for the riverbed sediments indicate more sedimentary recycling during the transport process. The chondrite (CI carbonaceous chondrite) data are from McDonough and Sun (1995). Pie charts indicate relative abundances of quartz (Q), feldspar (F) and clay minerals (C) (mainly including kaolinite and illite) based on XRD data interpretations. For the raw XRD patterns, refer to Fig. A2 in the Appendix B.

(Garzanti et al., 2010), are probably not largely resuspended in shallow water of the river, since the dry season SPM samples have similar Zr and Hf concentrations and Zr/Sc ratios with the wet season samples (Table A1). Therefore, we contend that the surface water of the Minjiang River carries mixed riverbed-suspended sediment load into marginal seas under such conditions (Fig. 9A–B). By contrast, in flood seasons, surface water of the Minjiang River carries sediments that are most directly derived from the SPMs of those tributaries and upper mainstream (Fig. 9C–D). Although we only investigated the SPMs from the surface water, SPMs within the river water column are probably less mixed and have highly geochemical heterogeneity in such conditions (e.g. Bouchez et al., 2011a, 2011b; Garzanti et al., 2011; Luo et al., 2012).

Furthermore, LILEs-based proxies (e.g. Rb/Sr and α_{Ba}) and our unpublished Sr–Nd isotope data (Jian et al., 2020) indicate that the upstream bedrocks likely contributed more materials for the Minjiang River-discharged SPMs than the downstream bedrocks during high precipitation seasons in the upstream regions (e.g. March–early April and May–June 2017) (Fig. A6 in the Appendix B), conversely the discharged SPMs were mainly derived from downstream regions during typhoon-dominated seasons (e.g. July–August 2017) (Fig. A6). This means that the geochemical compositions of the seasonal SPMs correspond well with the spatiotemporal variations of precipitation and therefore climate variability also likely serves as a major control to regulate the transport process and compositions of the Minjiang River sediment into the East China Sea (Jian et al., 2020).

5.3. Implications for sediment erosion and transport by subtropical mountainous rivers in East Asia

Physical erosion of mountainous regions, especially those tectonically active mountains under humid climatic conditions, contributes to supply fresh mineral surfaces to weathering environments and thus play a significant role in regulating global carbon cycles as well as climate on both modern and geologic timescales (e.g. France-Lanord and Derry, 1997; Dadson et al., 2003; Riebe et al., 2004; Galy et al., 2007; Willenbring and von Blanckenburg, 2010). Note that the modern global erosion rate is usually inferred from suspended load flux of rivers (e.g. Galy and France-Lanord, 2001). The occurrence of exposed riverbed sediment recycling into suspended load under low runoff conditions (i.e. dry seasons) of the Minjiang River (Fig. 9) suggests that the contributions of these kinds of mountainous rivers to the global erosion rate would sometimes be overestimated. This case study highlights that the short-term transport mechanism variability should be taken into account when evaluating erosion rate of continental mountains.

Given that most large rivers (e.g. Yangtze, Ganga and Amazon rivers) have large flood plains, deltas and widespread lakes, short-term sediment transport by those rivers can always be modulated by their large drainage basins and therefore the flux and compositions of their discharged sediments tend to display relatively little seasonal or inter-annual variability (Meybeck et al., 2003; Walling and Fang, 2003). In this case, sediments probably experience a complex sedimentary recycling and trapping history and much of the sediment load can be stored for long time periods (e.g. Dosseto et al., 2006; Li et al., 2016). By contrast, mountainous rivers (e.g. rivers on the Taiwan island) have

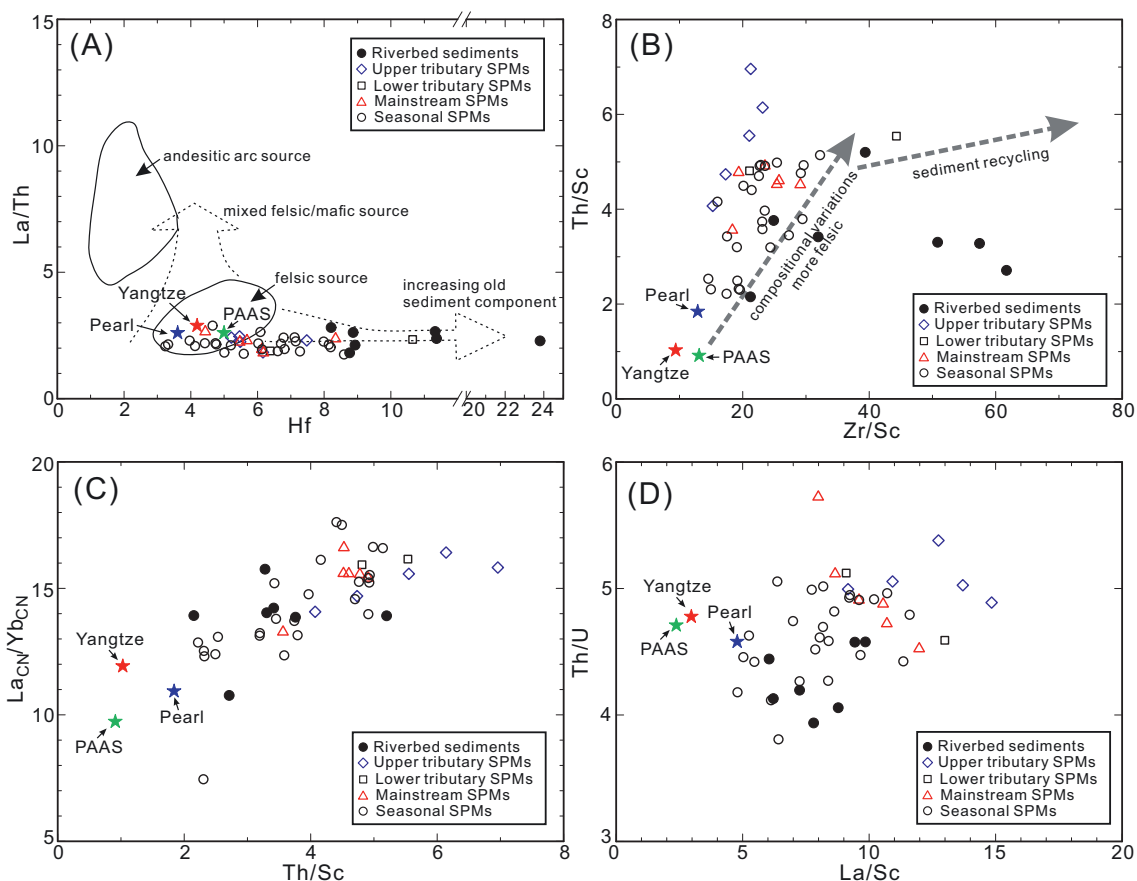


Fig. 6. Representative element ratio binary diagrams for the analyzed sediment samples. (A) La/Th vs. Hf (after Floyd and Leveridge, 1987); (B) Th/Sc and Zr/Sc (after McLennan et al., 1993); (C) La_{CN}/Yb_{CN} vs. Th/Sc; (D) Th/U vs. La/Sc. Note that the flood-season SPMs from upper and lower tributaries have similar Th/Sc, La/Yb, La/Sc and Th/U ratios, but show much higher ratio values than the riverbed sediment samples.

relatively small drainage basins, steep gradients and limited downstream sediment storage. They are more likely and easily to respond to short-term (e.g. seasonal- or event- timescales) environmental changes (Milliman and Syvitski, 1992). Under these backgrounds, sediments generally undergo less-recycling and fast-transport and the discharged sediment compositions always display great variations over different spatial-temporal scales (Mano et al., 2009; Hatten et al., 2012; Navratil et al., 2012; Goñi et al., 2013; Li et al., 2016; Milliman et al., 2017; Deng et al., 2019).

The Minjiang River provides a typical example of mountainous rivers in which hydrologic variability, especially climate-induced hydrologic changes, play an important role in short-term sediment transport and compositions of discharged sediments (Jian et al., 2020). As the sediment transport models shown in Fig. 9, changing river flow conditions throughout the hydrological cycle influence the sorting process of SPMs, which can explain the seasonal variability of geochemistry observed in the river surface SPMs. Previous studies on other Zhe-Min Rivers (e.g. Jiulongjiang and Mulanxi Rivers) and Taiwan rivers also underlined hydrologic controls on material transport over short-term timescales (e.g. Milliman and Kao, 2005; Hilton et al., 2008; Wei et al., 2016; Milliman et al., 2017; Su et al., 2018; Deng et al., 2019). Although the Zhe-Min rivers (Fig. 1) are developed on the tectonically stable East Asia continent (similar to the Yangtze and Pearl rivers), they likely display similar sediment transport behavior with those small mountainous rivers in tectonically active regions of the tropical and subtropical East Asia where the modern climate is mainly controlled by the East Asian monsoon system and incidental tropical cyclones (Yin et al., 2010; Chang et al., 2012). In addition to the seasonal geochemical heterogeneity of the sediments into the seas, the

Zhe-Min mountainous rivers are also supposed to have differential sediment transport regimes and compositional variability on inter-annual (e.g. the difference between October 2016 and October 2017, shown in Fig. 7) and event (e.g. typhoon-triggered rainstorms and meta-floods) scales.

5.4. Implications for geochemical cycles at the land-ocean interface

As stated above, the Minjiang River-discharged sediments demonstrate strong spatial and seasonal geochemical heterogeneity (Figs. 4–7). We also found the highly heterogeneous geochemical compositions between riverbed and SPM samples collected in pairs (Fig. 5). A great number of investigations on the small mountainous rivers in East Asia indicate huge geochemical variability of sediments into the oceans over event-timescales and multi-spatial scales (e.g. Hilton et al., 2008; Milliman et al., 2017; Deng et al., 2019). Although there hasn't been a relatively long-term (e.g. several decades) geochemical monitoring data on SPMs from these mountainous rivers, these rivers show strong inter-annual variability of sediment fluxes (Fig. 2B; Kao and Milliman, 2008; Dai et al., 2009) and are expected to have sediment geochemical heterogeneity on inter-annual timescales. Collectively, these findings and assumptions suggest the importance of multi-spatio-temporal scale, heterogeneous mountainous river input of chemical elements (as well as isotopes) to the ocean.

There have been several attempts to establish a comprehensive database on chemical compositions of SPMs in world rivers (Martin and Meybeck, 1979; Martin and Whitfield, 1983; Gaillardet et al., 1999b; Viers et al., 2009). We contend that the seasonal geochemical heterogeneity, as well as the highly variable sediment discharge over time by

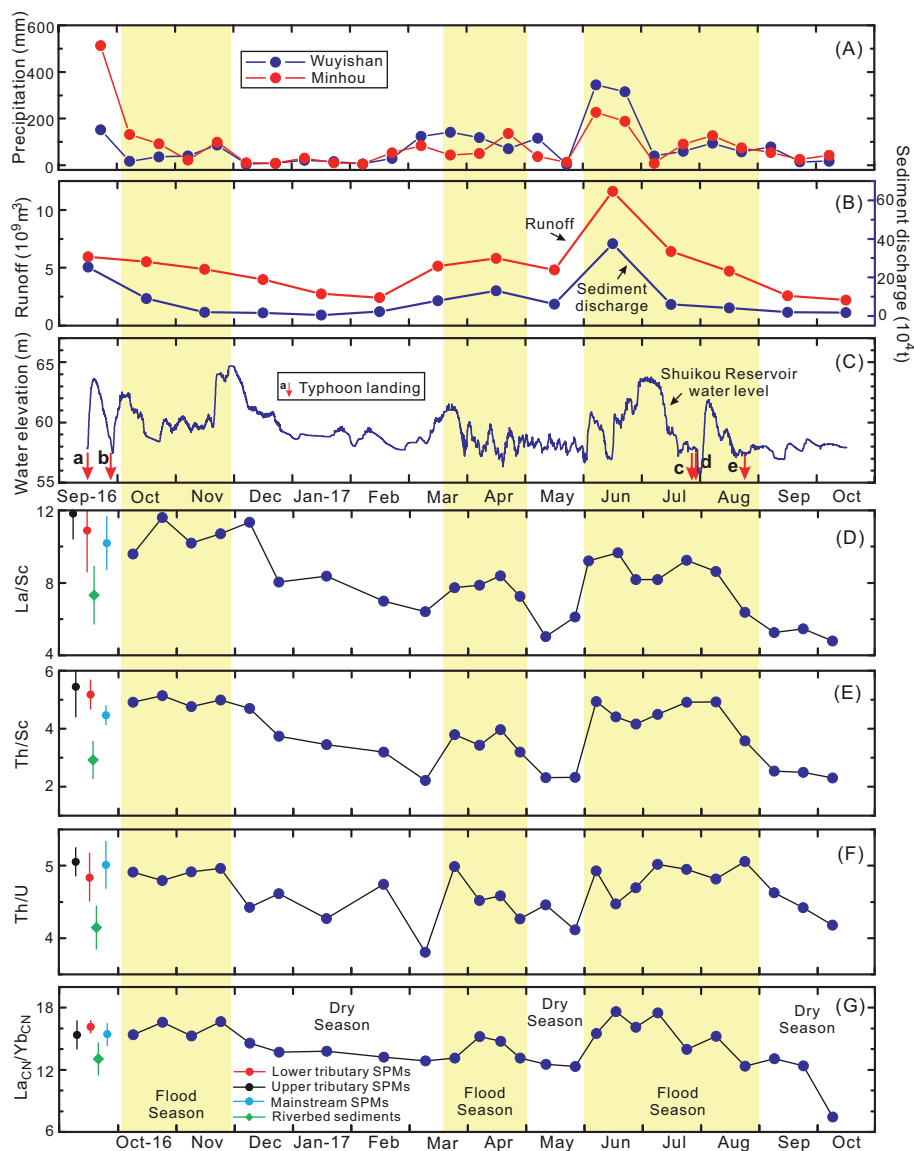


Fig. 7. Seasonal variations of representative trace element ratios of the analyzed SPMs from the fixed S04 station. (A) Semi-monthly precipitation of representative cities in the Minjiang river catchment (data from the National Meteorological Information Center website (<http://data.cma.cn>)); (B) monthly runoff and sediment discharge of the Minjiang River (indicated by data of the Zhuqi station, from MWRPRC (2016)); (C) water level variations of the Shuikou Reservoir (Fig. 2A), 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. (D) La/Sc; (E) Th/Sc; (F) Th/U; (G) $La_{(CN)}/Yb_{(CN)}$. Note that the Minjiang River-discharged sediments during flood seasons (yellow bands) have relatively high La/Sc, Th/Sc, Th/U and $La_{(CN)}/Yb_{(CN)}$ values, which are close to those of upper and lower tributary SPMs. By contrast, the dry season discharged sediments display similar geochemical compositions with the riverbed sediments. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

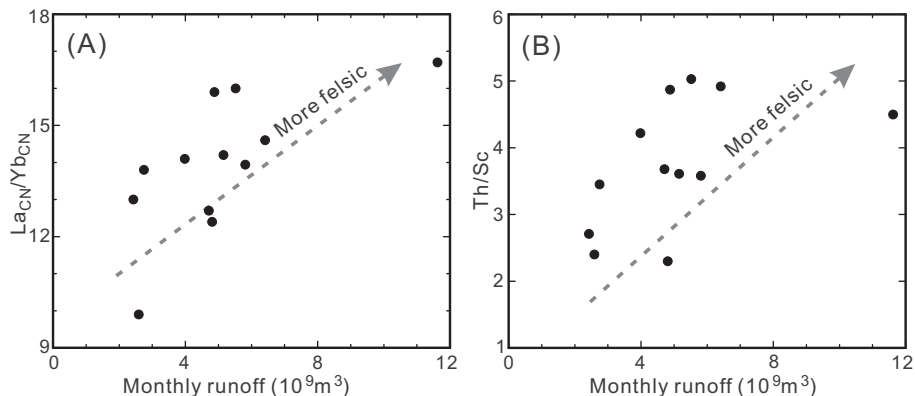


Fig. 8. Relations between seasonal SPMs element ratios and river runoff. (A) $La_{(CN)}/Yb_{(CN)}$ vs. monthly runoff; (B) Th/Sc vs. monthly runoff. The mean values of element ratios of SPMs each month were calculated for the comparison.

the mountainous rivers, can intensively influence elemental fluxes and budget at the land-ocean interface. Furthermore, our findings underline that the multi-spatio-temporal scale elemental and isotopic variations of the high-sediment-output mountainous rivers should be considered in reconstructions of geological and climatic events utilizing elemental

and isotopic geochemistry data of ancient clastic sedimentary records.

6. Conclusions

This study combines grain size analysis, mineralogy, petrography

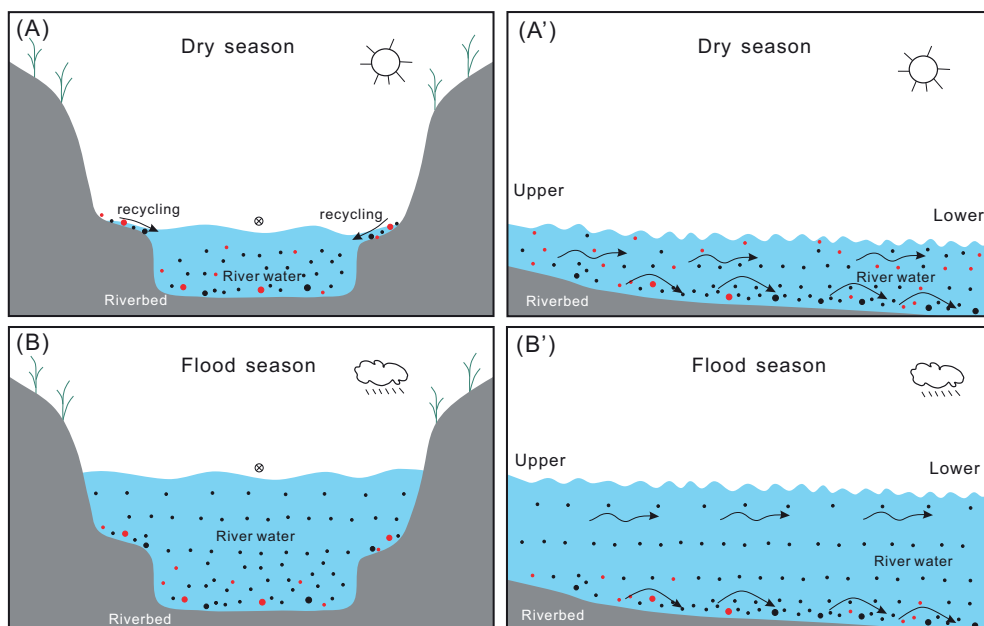


Fig. 9. Sediment transport model for the Minjiang River. (A) and (B) indicate cross sections of the river during dry and flood seasons, respectively, and (A') and (B') illustrate corresponding sediment transport status (one-way arrows indicate sediment transport directions). Note that sediments (indicated by red dots) on exposed riverbeds and overbanks are probably selectively recycled in the transport process during dry seasons, whereas the discharged surface SPMs in flood seasons are less mixed with bedload sediments and are likely dominated by particulates directly from those tributaries and the upper mainstream. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and trace element geochemistry of riverbed sediments and SPMs from the Minjiang River, SE China and yields the following conclusions:

- 1) The upper and lower tributary SPMs of the Minjiang River have different LILE features but similar REE, Th and Sc features. These SPMs have significant differences of most trace element concentrations and ratios from the associated (i.e. collected in pairs) riverbed sediments, due to the influence of hydrodynamic sorting process which results in the relative enrichment of clay minerals in the suspended-load phases.
- 2) The discharged SPMs in a one-year hydrological cycle demonstrate strong geochemical variations, with highly positive correlations between $La_{(CN)}/Yb_{(CN)}$ and Th/Sc ratios and the river runoff data. SPMs during high-runoff periods therein have similar La/Sc, Th/Sc, Th/U and $La_{(CN)}/Yb_{(CN)}$ ratio values with the upper and lower tributary SPMs, whereas these element ratios of samples in low-runoff periods are close to or even lower than those of the analyzed riverbed sediments.
- 3) The results of the discharged SPMs in different seasons indicate a hydrologic control on the sediment transport and geochemical compositions of the Minjiang River. This river has similar sediment transport behavior to those small mountainous rivers in tectonically active regions of the tropical and subtropical East Asia. Our findings also highlight the importance of multi-spatio-temporal scale, heterogeneous river input of chemical elements to the oceans.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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