Geochemistry of river-borne clays entering the East China Sea indicates two contrasting types of weathering and sediment transport processes

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Abstract The East China Sea is characterized by wide continental shelf receiving a huge input of terrigenous matter from both large rivers and mountainous rivers, which makes it an ideal natural laboratory for studying sediment source-to-sink transport processes. This paper presents mineralogical and geochemical data of the clays and bulk sediments from the rivers entering the East China Sea, aiming to investigate the general driving mechanism of silicate weathering and sediment transport processes in East Asian continental margin. Two types of river systems, tectonically stable continental rivers and tectonically active mountainous rivers, coexist in East Asia. As the direct weathering products, clays can better reflect the silicate weathering regimes within the two river systems. Provenance rock types are not the dominant factor causing silicate weathering intensity difference existed in the East Asian rivers. The silicate weathering intensity of tectonically stable river basins is primarily driven by monsoon climate, and the sediment transfer is relatively slow because of natural trapping process and increasing damming effect. The geochemistry of these river-borne sediments can thus indicate paleo-weathering intensities in East Asian continent. In contrast, silicate weathering intensity in tectonically active mountainous rivers is greatly limited by strong physical erosion despite the high temperature and highest monsoon rainfall. The factors controlling silicate weathering in tectonically active catchments are complex and thus, it should be prudent to use river sediment records to decipher paleoclimate change. These two different silicate weathering regimes and sediment transport processes are manifestations of the landscape evolution and overall dominate the sedimentation in Asian continental margin.

1. Introduction

Chemical weathering is an important earth surface process linking the interactions between the Earth’s spheres since it controls the evolution of climate through the consumption of atmospheric carbon dioxide [Walker et al., 1981; Berner, 1992; Gaillardet et al., 1999a,b], shapes the Earth’s landscape through physical denudation and soil production [Heimsath et al., 1997], modifies the composition of the upper continental crust [Rudnick and Gao, 2003; Lee et al., 2008; Liu and Rudnick, 2011], and alters the chemical composition of riverwater and seawater [Hodell et al., 1990].

Extensive studies on river geochemistry have been carried out to examine the weathering mechanisms and sinks of atmospheric CO₂ in various drainage basins. Most of the world's large rivers, including the Amazon, Changjiang (Yangtze River), Huanghe (Yellow River), Congo, Mississippi, and Ganges-Brahmaputra rivers, have been studied separately or comparatively [Berner, 1992; Dupré et al., 1996; Canfield, 1997; Gaillardet et al., 1999a, 1999b; Galy and Fance-Lanord, 1999; Vital and Stattegger, 2000; Yang et al., 2004; Bouchez et al., 2011, 2012]. Generally, dissolved components in rivers, rather than river-borne particles, are considered to estimate chemical denudation rates because they come directly from source rock (soil) weathering although the riverwater chemistry may be contaminated by anthropogenic activities. Over the past decade, quantitative model analysis of rock weathering that integrates the effects of runoff, erosion, temperature, biological processes and predicts weathering-induced changes in physical and chemical properties of soils based on solute-derived weathering data from catchments, have made plentiful and substantial achievements.
geochemistry, Geophysics, Geosystems

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250,000 km² in area. Rocks include Jurassic sandstone, Mesozoic igneous rocks, Paleozoic marine and Quaternary fluviolacustrine sedimentary rocks. Logically, the Changjiang catchment comprises complex rock types including Paleozoic carbonate rocks, Jurassic sandstone, Jurassic basalt, and Mesozoic igneous rocks. The samples were collected from ten major rivers entering the ECS, namely, from north to south, the Changjiang River, rivers in Zhejiang-Fujian Province (Lingjiang, Oujiang, Feiyunjiang, Minjiang, and Mulantxi) and in Taiwan (Lanyangxi, Wuxi, Dajiaxi, and Zhuoshuixi) (Figure 1 and Table 1). These river basins span temperate and subtropical zones typical of monsoon climate regimes, and encompass various tectonic units and geologic settings. Basically, these rivers can be classified into two types: continental rivers in mainland China including the Changjiang and those in southeast Chinese coastal areas with tectonically stable catchments, and the mountainous rivers in the tectonically active island of Taiwan [Limp et al., 2011].

In this study, we report the mineralogical and elemental compositions of the clays and bulk sediments from representative rivers entering the ECS, and examine the major controlling factors of river sediment chemistry. The major purpose of this study is to investigate the intensity of silicate weathering registered in these fluvial sediments (chemical alteration relative to source rocks), and further to reveal the general driving mechanism of silicate weathering in East Asian continental margin where complex tectonics and active sediment source-to-sink processes coexist.

2. Study Area

The samples were collected from ten major rivers entering the ECS, namely, from north to south, the Changjiang River, rivers in Zhejiang-Fujian Provinces (Lingjiang, Oujiang, Feiyunjiang, Minjiang, and Mulanxi) and in Taiwan (Lanyangxi, Wuxi, Dajiaxi, and Zhuoshuixi) (Figure 1 and Table 1). These river basins span temperate and subtropical zones typical of monsoon climate regimes, and encompass various tectonic units and geologic settings. Basically, these rivers can be classified into two types: continental rivers in mainland China including the Changjiang and those in southeast Chinese coastal areas with tectonically stable catchments, and the mountainous rivers in the tectonically active island of Taiwan [Milliman and Farnsworth, 2011].

2.1. The Changjiang and Small Rivers Draining Southeast China

The Changjiang originates from the Tibetan Plateau and its catchment is primarily situated on the Yangtze Craton. The catchment area is about 1.8 × 10⁶ km², more than 1/5 of the land area of China continent. Geologically, the Changjiang catchment comprises complex rock types including Paleozoic carbonate rocks, Jurassic sandstone, Mesozoic igneous rocks, Paleozoic marine and Quaternary fluviolacustrine sedimentary rocks [Yang et al., 2004]. The Permian Emeishan basalt in the upper Changjiang valley occupies about 250,000 km² in area [Zhang et al., 2006], which is the largest igneous province in China and also the unique
provenance rock type in the Changjiang catchment. Besides, the Paleozoic marine carbonates also widely occur in the upper drainage basin, with an area of about 400,000 km². Various metamorphic and igneous rocks, especially Mesozoic granites, are widely distributed in the midlower basins \[Wang et al., 2013\].

Figure 1. A schematic map showing the East China Sea and surrounding rivers with the sampling locations. CDW: Changjiang Diluted Water; CC: East China Sea Coastal Current; TWC: Taiwan Warm Current; (a) Lingjiang; (b) Oujiang; (c) Feiyunjiang; (d) Minjiang; (e) Mulanxi; (f) Lanyangxi; (g) Wuxi; (h) Dajiaxi; (i) Zhuoshuixi; The gray shadowed areas indicate muddy deposition in the East China Seas. (1): The mud area on the inner shelf of the East China Sea; (2): The mud area off the Changjiang Estuary; (3): The mud area to the southwest of Cheju Island (modified from Qin [1996]).

Table 1. Basic Parameters of the Typical Rivers Entering the East China Sea

<table>
<thead>
<tr>
<th>Rivers</th>
<th>Length (km)</th>
<th>Drainage Area (10³ km²)</th>
<th>Gradient (%)</th>
<th>Mean Temperature (°C)</th>
<th>Mean Rainfall (mm/yr)</th>
<th>Water Discharge (10⁸ m³)</th>
<th>Mean Runoff (mm/yr)</th>
<th>Sediment Load (10⁶ t/yr)</th>
<th>Sediment Yield (t/km²/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Changjiang*</td>
<td>6380</td>
<td>1800</td>
<td>0.10</td>
<td>15.0</td>
<td>1100</td>
<td>8964b</td>
<td>500</td>
<td>39000</td>
<td>217</td>
</tr>
<tr>
<td>Lingiang¹</td>
<td>198</td>
<td>6.6</td>
<td>0.30</td>
<td>17.5</td>
<td>1500</td>
<td>51.7</td>
<td>783</td>
<td>112</td>
<td>170</td>
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<tr>
<td>Oujiang¹</td>
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<td>0.28</td>
<td>19.0</td>
<td>1746</td>
<td>202b</td>
<td>1056</td>
<td>273</td>
<td>152</td>
</tr>
<tr>
<td>Feiyunjiang¹</td>
<td>176</td>
<td>3.7</td>
<td>0.60</td>
<td>17.9</td>
<td>1850</td>
<td>40</td>
<td>1183</td>
<td>42</td>
<td>114</td>
</tr>
<tr>
<td>Minjiang²</td>
<td>577</td>
<td>61</td>
<td>0.04</td>
<td>19.0</td>
<td>1717</td>
<td>584b</td>
<td>951</td>
<td>637</td>
<td>104</td>
</tr>
<tr>
<td>Mulanxi²</td>
<td>168</td>
<td>1.7</td>
<td>0.47</td>
<td>19.5</td>
<td>1637</td>
<td>15.2</td>
<td>921</td>
<td>47</td>
<td>276</td>
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<tr>
<td>Zhuoouxi³</td>
<td>186</td>
<td>3.2</td>
<td>1.84</td>
<td>22.2</td>
<td>2232</td>
<td>61</td>
<td>1200</td>
<td>6387</td>
<td>19959</td>
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<tr>
<td>Wuxi³</td>
<td>117</td>
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<td>2.22</td>
<td>23.0</td>
<td>1800</td>
<td>37.3b</td>
<td>1800</td>
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<td>140</td>
<td>1.2</td>
<td>2.77</td>
<td>23.0</td>
<td>2200</td>
<td>26b</td>
<td>2000</td>
<td>403</td>
<td>3358</td>
</tr>
<tr>
<td>Lanyangxi³</td>
<td>73</td>
<td>1</td>
<td>4.84</td>
<td>22.0</td>
<td>3046</td>
<td>27.7b</td>
<td>2860</td>
<td>798</td>
<td>7980</td>
</tr>
</tbody>
</table>

¹Data from Changjiang Water Resources Commission (http://www.cjw.com.cn/).  
²Data from references Kao and Milliman [2008] and Milliman and Farnsworth [2011] and Water Resources Agency, Ministry of Economic Affairs, Taiwan (http://www.wra.gov.tw/).  
³Data from Zhejiang Province Water Resources Bulletin [Zhejiang Provincial Water Management Institute, 2012].  
⁴Data from Fujian Province Water Resources Bulletin [Fujian Provincial Water Management Institute, 2012].
The Asian monsoon climate characterizes the catchment, resulting in variable rainfall over the catchment at seasonal scale, with mean annual precipitation of 1100 mm and temperature of 15°C. The midlower basins are strongly influenced by the East Asian monsoon climate, which yields the mean annual temperature of 16–18°C, while the headwater is influenced by the Indian monsoon in the highland and cold climate results in the mean annual temperature <4°C. Based upon long-term (1950–2010) hydrologic observation at Datong gauge station in the lower mainstream, the water and sediment discharges of the Changjiang average at 8964×10^6 m^3/yr and 390×10^6 t/yr, respectively, which makes it the largest source of terrigenous material to the ECS (Table 1). However, the sediment flux of the Changjiang has been rapidly decreasing since the impoundment of the Three Gorges Reservoir in 2003, averaging at only about 155×10^6 t/yr over the last ten years (2003–2013) (Changjiang Water Resources Commission, http://www.cjw.com.cn/).

The small rivers entering the coastal area of southeast China drain the hilly regions in Zhejiang and Fujian Provinces, with altitudes around 1000 m at the headwaters of the rivers. Influenced by the East Asian monsoon and typhoon events, these catchments have mean annual precipitation between 1500–1800 mm and annual temperature of about 17.5–19.5°C (Table 1). These catchments have relatively homogeneous source rocks, which primarily consist of Yanshanian granite and Quaternary detrital sediments, while metamorphic rocks and limestone occur sporadically. The water discharges of these coastal rivers vary between 15.2×10^6 and 51.7×10^6 m^3/yr and sediment fluxes range from 0.42×10^6 to 6.37×10^6 t/yr, much smaller than those of the Changjiang. The estuaries of these rivers are strongly influenced by semidiurnal tide, with mean tidal ranges of about 4–6 m.

2.2. Small Mountainous Rivers on the High-Standing Island of Taiwan

The island of Taiwan is well known for its strong tectonic uplift at a rate of 5–10 mm/yr [Shin and Teng, 2001], and high physical erosion rates up to 3–7 mm/yr [Dadson et al., 2003; Fuller et al., 2003]. In this sense, some studies suggest that Taiwan Island has reached a steady state in terms of tectonic uplift and physical erosion [Hilley and Strecker, 2004; Konstantinovskaia and Malavieille, 2005]. In addition, Taiwan has frequent typhoons and earthquakes [Wu and Kuo, 1999]. Consequently, it has by far the highest sediment yield in the world, resulting in a tremendous sediment load (about 180–380 Mt/yr) discharged by the small mountainous rivers into the surrounding marginal seas including the ECS [Liu et al., 2009, 2013; Milliman and Farnsworth, 2011]. Most of these fluvial sediments are rapidly transferred to the sea in just a few days during typhoon events, and hyperpycnal flow is an important way for sediment transport to deep sea [Liu et al., 2009, 2012, 2013]. In general, the concentration of suspended sediment in the rivers is below 5–10 g/L during the calm days before typhoon but it can reach 200 g/L during typhoon [Kao and Milliman, 2008; Milliman and Kao, 2005]. Source rocks of Taiwan river basins are mainly composed of sedimentary rocks and epimetamorphic rocks including sandstone, shale, slate and phyllite, with rare occurrence of acidic rocks.

3. Materials and Methods

3.1. Sampling Plans

From March to November 2011, a total of 21 river bank sediment samples were taken from the lower reaches and estuarine areas of the main rivers. For comparison, a total of 15 bank sediment samples were collected from four major rivers in Taiwan in 2011 and 2013 (Wuxi, Dajiaxi and Zhuoshuixi in western Taiwan and Lanyangxi in eastern Taiwan (Figure 1 and supporting information Table S1). All of these sediments were collected from the river bank where can be flooded during high water level stages, and the sampling localities are less influenced by strong fluvial currents and vegetation coverage. All the samples were collected by scooping the top 5 cm of the river bank sediments using a clean polyethylene spade, and then transferred to the laboratory and kept in a cold repository. In addition, 51 suspended sediment samples were collected weekly from April 2008 to 2009 at the lower Changjiang mainstream near Nantong City (Figure 1), approximately 90 km away from the river mouth (supporting information Table S1). All of the suspended samples were taken from the main river channel. At each sampling site, about 25 L of river water was collected from 1 m below the river surface and filtered through a 0.45 μm membrane of cellulose acetate to collect particulate matter. All of these samples were dried at low temperature (40°C) in the laboratory.

The studied rivers have much variable hydrodynamic conditions, which result in significant difference in sediment grain size within a specific river system and between these different rivers. The resulting sorting effect would have a considerable impact on geochemical compositions of the fluvial sediments.
[Garzanti et al., 2008; Bouchez et al., 2011]. In this study, the clay fraction was chosen for mineralogical and geochemical analyses for reducing the grain-size and hydrodynamic sorting effects on sediment composition. The element geochemical data of clayey sediments from most of these East Asian rivers will be reported for the first time. In general, clay may better integrate the weathering products in the source area [Chamley, 1989]. A quantity of 10% H2O2 was added to the sediment samples gradually with stirring, accomplished by successive rinsing with deionized water to deflocculation. Particles smaller than 2 μm were separated from the bulk sediment by the pipette method following the Stoke’s Law at room temperature. The separated suspension was concentrated by centrifuging at 7500 rotations per minute for 10 minutes and then smeared on glass slides to air dry. For comparison, the bulk samples from the Minjiang, Lanyangxi and Zhuoshuixi as well as the suspended samples from the Changjiang at Nantong were analyzed for elemental concentrations.

3.2. Mineralogical and Geochemical Analyses

3.2.1. X-Ray Diffraction (XRD)

The mineral compositions in the clay fraction were estimated by X-ray diffraction (XRD) measurements in the State Key Laboratory of Mineral Deposits Research at Nanjing University. The XRD was conducted by a Rigaku D/Max-III X-ray diffractometer equipped with a Cu-target tube and a curved graphite monochromator, operating at 37.5 kV and 20 mA. The analyses were run from 3° to about 40° (2θ) with a step size of 0.02° (2θ). A side-packing method proposed by the National Bureau of Standards was used to prepare the powder (nonoriented) mounts. Mineral identification was made by Jade software version 9.0 and mineral contents were semiquantitatively estimated by the whole-pattern fitting method using Siroquant software. Replicate analyses of a few selected samples yield a precision of about 2% (1σ).

3.2.2. Elemental Analysis

For better revealing the provenance character of terrigenous sediments discharged into the sea, 1M HCl (hydrochloric acid) was used to leach the sediment samples following the method of [Dou et al., 2010]. Using the method, the authigenic, biogenic and adsorptive components including weakly-bound or nondegradable components can be largely removed from the samples and concentrated in the leachate [Loring and Rantala, 1992; Song and Choi, 2009]. About 0.2 g powdered dry samples were leached with 20 ml 1 M HCl in a shaker incubator for 6 h at 60°C. The residues of the leached samples were rinsed using deionized water through four runs of centrifugation in centrifuge tube until the pH value of supernatant near neutral. Then, the supernatant in each run was collected and then the residues were heated to dryness at 50°C. All of the residues were combusted in a muffle furnace at 600°C for 2 h in order to remove the organic matter before acid digestion. Afterward, 30 mg samples were digested with 4 ml HNO3 and 1 ml HClO4 for 24 h in a tightly closed Teflon vessel on a hot plate at less than 160°C, heated to dryness, and then digested with a mixture of 4 ml HF and 1 ml HClO4. Afterward, the solution was evaporated to dryness, and eluted with 10 ml 1% HNO3. The concentrations of the major elements in the leachate and residual fractions were determined by ICP-AES (IRIS Advantage, Thermo Elemental) and trace elements by ICP-MS (VG-X7, Thermo Elemental) in the State Key Laboratory of Marine Geology at Tongji University. The analytical precision and accuracy were monitored by the national geostandard GSD-9. Accuracy was calculated as 100×(Xmeas−Xstd)/Xstd with Xmeas as the measured concentration and Xstd as the certified value. The analytical precision (one standard deviation) was determined by the repeated analysis (n=4) of geostandard GSD-9 (supporting information Table S2). The accuracy of element analysis is better than 3% with the exception of Sc (9.8%) and the precision is better than 4% (supporting information Table S2).

3.2.3. Chemical Index of Alteration (CIA)

The rivers involved in this study cover a wide spectrum of geographical and climatic zones from a continuum between warm temperate and subtropical (Changjiang) to subtropical zones (Minjiang and Taiwan rivers) [Chen and Wang, 1996]. Previous studies on the bulk sediment chemistry of these rivers have suggested variable intensities of silicate weathering in different catchments in East China [Li and Yang, 2010; Shao et al., 2012]. In this study, more geochemical proxies are used to evaluate the silicate weathering intensity registered in the sediments from these rivers.

The Chemical Index of Alteration (CIA) proposed by Nesbitt and Young [1982] is defined as $\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O}) \times 100$ (molecular proportions, with CaO* being CaO content in silicate fraction of the sample). The CIA actually reflects the degree of plagioclase weathering to clays and it can indicate the integrated weathering history of silicates in a catchment. The $\text{Al}_2\text{O}_3 - (\text{CaO}^* + \text{Na}_2\text{O}) - \text{K}_2\text{O}$ (A-CN-K)
diagram has also been widely used to indicate silicate weathering in river basins [von Eynatten et al., 2003; Yang et al., 2004; Liu et al., 2007; Li and Yang, 2010; Shao et al., 2012; Chetelat et al., 2013; Clift et al., 2014].

3.2.4. The Weathering (Mobility) Indices of \( \alpha \text{Na}, \alpha \text{K}, \) and \( \alpha \text{Ba} \)

Gaillardet et al. [1999a] introduced a separate index \( \alpha \text{Me} \) for each mobile element (Me) by comparing its concentration to that of an immobile element which has magmatic compatibility close to that of the mobile element, such as Na and Sm, K and Th, Mg and Al, Ba and Th, etc. In this study, this index was used to discuss the effect of source rock differentiation on the intensity of silicate weathering in the fluvial sediments. The main advantage of this index is that the ratio of elements with similar magmatic compatibilities can minimize the effects of igneous differentiation that exists on the assemblage of parent rocks drained by the river. For instance, if large scale chemical variations exist in the Upper Continental Crust (UCC), then ratios such as Sm/Na are expected to be less variable than ratios such as Th/Na because Th and Na have very different compatibilities [Gaillardet et al., 1999a]. As with CIA, this weathering index is defined for each soluble element and the weathering intensities calculated for each specific element can be compared. This study focuses on the weathering indices of Na, K and Ba which are denoted by the symbol \( \alpha \) followed by the elements of interest (Me).

\[
\begin{align*}
\alpha \text{Na} &= \frac{(\text{Sm/Na})_{\text{sed}}}{(\text{Sm/Na})_{\text{UCC}}} \\
\alpha \text{K} &= \frac{(\text{Th/K})_{\text{sed}}}{(\text{Th/K})_{\text{UCC}}} \\
\alpha \text{Ba} &= \frac{(\text{Th/Ba})_{\text{sed}}}{(\text{Th/Ba})_{\text{UCC}}}
\end{align*}
\]

To better reflect the degree of silicate weathering in this study, here we use the subscripts “sed” and “UCC” to indicate the concentrations of investigated elements in the residual fractions of the fluvial sediments studied and in the upper continental crust [Rudnick and Gao, 2003], respectively. \( \alpha \text{Me} \) values greater than 1 indicate a depletion while the values lower than 1 indicate an enrichment of the specific mobile element (Me) with respect to UCC.

3.2.5. High Field Strength Element Ratios

High field strength elements, such as Th, Sc and Cr, have extremely low concentrations in river water and seawater and are predominantly concentrated in clastic sediments [Taylor and McLennan, 1995]. The immobile element is relatively stable under supergene environment and thus, immobile element ratios can more precisely reflect the geochemical compositions of source rocks and decipher the sediment provenances. Because of their difference in compatibility during magmatic processes, Cr and Sc contents normalized to Th are good tracers of magmatic differentiation. As Cr and Sc are more compatible than Th, the residual melt during differentiation is enriched in Th relative to Co and Cr. Thus, The Cr/Th and Sc/Th ratios in sediments may be used as indicators of the mafic versus felsic natures of the parent rocks. In view of this, the ratios of these immobile trace element such as Cr/Th and Sc/Th will be used in this study to investigate the source rock constraints on sediment chemistry and weathering intensity.

4. Results

4.1. Mineral Contents in the <2 \( \mu \)m Fraction of River Sediments

The <2 \( \mu \)m clay fraction consist mostly of clay minerals, i.e., 30–60% of illite, 7–54% of kaolinite, and 6–23% of chlorite. The total contents of quartz and feldspar reach 6–22% on average. The contents of calcite and hematite are generally lower than 5%. Without the ethylene-glycol solvation experiment on the <2 \( \mu \)m fraction, the contents of smectite cannot be accurately estimated in this study. Nevertheless, previous studies suggest that smectite is minor (0–6%) in the clays from the Changjiang and coastal rivers in Zhejiang and Fujian Provinces, and Taiwan’s riverine sediments do not contain smectite [Yang et al., 2003; Xu et al., 2009; Li et al., 2012].

Overall, the clay mineral contents in the Changjiang are dominated by illite (with an average of 56%), similar to those in the Lingjiang, Oujiang and Feiyunjiang rivers in Zhejiang Province. The average contents of other minerals in these rivers are about 15% for kaolinite, 13% for chlorite and a combined total of 14% for quartz and feldspar. In comparison, the mineral contents in the coastal rivers in Fujian Province are remarkably different, yielding the highest contents of kaolinite, i.e., 54% and 29% on average in the Minjiang and Mulanxi rivers, respectively. The average contents of chlorite in these two rivers are 10% and 11%,
respectively, close to that of the Changjiang, while the total contents of illite, quartz and feldspar are much lower in these two southern rivers than in the Changjiang and other rivers in Zhejiang Province.

As for the <2 μm fractions of Taiwan’s riverine sediments, they contain higher quartz and feldspar, with a combined average content of 21%, while kaolinite content is only about 10%, much lower than those from the rivers in mainland China.

4.2. Element Chemistry in the Riverine Sediments

The concentrations of selected elements in the clays and bulk sediments from these rivers are given in supporting information Tables S3 and S4. The acid leaching result indicates that both clay and bulk sediments show relatively consistent element distributions. Elements including K, Na, Al, Sc and Th are predominantly concentrated in the residual factions, while Ca is more concentrated in the leachate fractions and accounts for over 90% of the total concentrations (Figure 2a). The residual/bulk ratio for an element means the value obtained by dividing the concentration of the element in the residual fraction to that in the bulk sample. Overall, the elements in clays have lower residual/bulk ratios than those in the bulk samples.

To better reveal the difference in elemental compositions of the residues between the small rivers and the Changjiang, the elemental concentrations in the clays from these rivers were normalized to the averages of the Changjiang clay (Figure 2b). For the bulk samples, their elemental concentrations were normalized to the averages of the suspended sediment samples of the Changjiang collected at Nantong. Apparently, the clays of the Lingjiang, Oujiang and Feiyunjiang have elemental concentrations closer to the Changjiang, while the Minjiang clay is relatively enriched in Al, Sm, Eu and Th, and depleted in K, Na, Ca, Cr, and Ba. For the bulk samples, K, Na, Ca, Al, Sc, Cr, Ba, Sm and Eu are more or less depleted in the Minjiang sediments while Th is relatively enriched. Overall, the rivers in Zhejiang Province have similar sediment chemistry with the Changjiang, different from those southern rivers in Fujian Province.

The samples collected from Taiwan rivers have much variable element chemistry. The clays have the highest residual/bulk ratios for Cr, Ba, Sm and Eu, and the residues of Taiwan river clays are notably rich in K, Na, Ba, Sm and Eu, but depleted in Ca (Figure 2a). Meanwhile, the Taiwan bulk samples have higher Na and Sm concentrations, and lower K, Ca, Al, Sc, Cr and Th than the Changjiang samples (Figure 2b).

4.3. Proxy Indicators of Silicate Weathering Intensity

4.3.1. The Chemical Index of Alteration (CIA)

The CIA values of all the bulk samples range from 60.0 (Zhuoshuixi) to 83.0 (Minjiang) and average at 72.6. On the A-CN-K diagram, the bulk sediments fall on a line parallel to the A-CN line, with the Minjiang bulk samples being closer to the A apex (Figure 3). The distribution trend of the bulk samples suggests that integrated silicate weathering in the Changjiang catchment is overall more intense than in Taiwan’s river basins, both showing the removal of Ca and Na from silicate minerals while K-bearing minerals remain almost
intact. The mean CIA value of the Changjiang suspended samples is comparable with the world river average [Li and Yang, 2010].

The CIA values of the fluvial clays vary between 77.4 (Lanyangxi) and 90.6 (Minjiang) with an average of 81.5. The CIA values of the Changjiang clay, similar to those of the Lingjiang, Oujiang, Feiyunjiang and Mulanxi rivers, range from 85.8 to 89.2 and average at 87.2. The highest (90.6) and lowest (78.0) mean CIA values are found in the clays of Minjian and Taiwan rivers, respectively (supporting information Table S6). The A-CN-K diagram clearly shows different intensities of silicate weathering between the bulk sediments and their clay fractions. The weathering trend of the clays is closer and somewhat parallel to the A-K line, reflecting strong removal of K-bearing minerals relative to the bulk samples. The Minjiang clay is closer to the A apex (kaolinite apex), suggesting the most intense silicate weathering and high kaolinite content, which is confirmed by the XRD results (Figure 4). In contrast, Taiwan’s river clays have the lowest CIA values, suggesting less intense silicate weathering, although the latitude of the island is lower than other river basins in mainland China (Figure 1).

4.3.2. The Weathering (Mobility) Indexes of Na, Ak, and Ba
For the bulk samples, Changjiang and Taiwan rivers have more or less similar Na, Ak and Ba values, while the Minjiang still has the highest values (supporting information Table S6). Compared to the bulk samples, the clays are more sensitive to the difference of chemical weathering registered in the fluvial sediments [Qiu et al., 2014]. All the river clays, except for the Minjiang (Na = 20), have similar Na values, ranging from 3.6 to 6.5 (Figure 5). The river clays have obviously different K and Ba values, with the highest values in the Minjiang and the lowest in Taiwan rivers. Good correlations between K, Ba and CIA in the studied clays (Figure 5) demonstrate that these geochemical indices can provide valuable and consistent information on the degree of silicate weathering.

5. Discussions
5.1. Source Rock Constraints on Sediment Chemistry and Silicate Weathering Intensity
Despite the different opinions on the dominant mechanism of chemical weathering, the key factors include source rock type, climate regime, tectonic and topographic settings, vegetation, soil development, and

Figure 3. The A-CN-K diagram showing the weathering trends (dotted arrows) of the clay and bulk samples from the rivers studied. The data of world river average are from Li and Yang [2010]; the data of upper continental crust (UCC) are from Rudnick and Gao [2003].
human activities as well [Gibbs, 1970; Meybeck, 1987; Stollard, 1995; White and Blum, 1995; Gaillardet et al., 1999a, 1999b; Oliva et al., 2003]. In this study, all of the samples were pretreated by 1M HCl before the complete digestion, and thus, most of the authigenic, biogenic and adsorptive components including weakly bound or nondetrital fractions are removed from the bulk samples and then concentrated in the leachate [Loring and Rantala, 1992]. The clays have much lower residual/bulk ratios than those in the bulk samples, demonstrating that the clays can adsorb more mobile elements relative to the bulk sediments during weathering and sedimentary processes (Figure 2). Meanwhile, the clay fraction was selected for mineralogical and geochemical analyses, which can significantly reduce the grain-size and hydrodynamic sorting effects on sediment composition. In consequence, the residues in this study can basically represent the detrital components in sediments and thus, reflect source rock compositions and silicate weathering intensity in the drainage basins.

The rock type is the first factor, if not a dominant one, in chemical denudation [Meybeck, 1987]. Numerous studies further highlight the control of source rock types on river water chemistry [Gaillardet et al., 1999b; Chetelat et al., 2008]. However, the causal links between chemical weathering and source rock types is difficult to define by sediment chemistry, as suggested by Li and Yang [2010], because the heterogeneous parent rocks cause the variable compositions of source rocks subject to weathering in different catchments, as is the case in China. In view of this, we investigate the relationships between different indices of weathering intensity and discuss the role of lithology on the apparent loss of soluble elements during weathering for the representative rivers entering the ECS.

5.1.1. Sediment Provenance

The Changjiang basin is characterized by complex and variable source rock types, while the smaller catchments of the Lingjiang, Ouijiang and Feiyunjiang in Zhejiang Province have relatively homogeneous source rocks dominated by granites. Despite the different geologic settings, this study reveals that mineralogical and geochemical compositions of the clays in these rivers are quite similar.

The modern Changjiang discharges most of its sediment between June and September to the river mouth to develop a large delta, while about 30–50% of the river-derived fine sediment escapes the estuarine trapping and is transported primarily southeastward by the coastal current to inner shelf off Zhejiang and Fujian Provinces [Huh and Su, 1999]. Compared with the sediment load of the Changjiang, the rivers in Zhejiang Province have much smaller sediment fluxes, mostly between 0.42 and 2.73 × 10^6 t/yr based on long-term hydrological observations. Furthermore, the sediment loads into the ECS of these rivers have been rapidly decreasing over the last decade mainly due to extensive dam construction in their catchments [Milliman and Farnsworth, 2011]. In addition, these rivers in Zhejiang Province are strongly influenced by tide with the tidal ranges of about 4–8 m, which can pump a large volume of the Changjiang-derived fine sediments into

![Figure 4. Relationship between the Chemical Index of Alteration (CIA) of clayey sediments and mineral composition (a) Kaolinite; (b) Quartz + Feldspar.](image-url)
their estuaries and even lower reaches [Li et al., 1993; Guan et al., 1998; Guan et al., 2005]. Therefore, the river clays in Zhejiang Province measured in this study are probably contaminated by the Changjiang-derived clay, which consequently results in the similarity of mineralogical and geochemical compositions between them.

Based on the Cr/Th versus Sc/Th plot (Figure 6), both the bulk and clay sediments of the studied rivers have provenance rocks falling along a line defined by the felsic-mafic end-member trend. At one hand, the Changjiang drainage basin has source rocks close to the average compositions of East China Upper Continental Crust (ECUC) and is in line with Upper Continental Crust (UCC) [Rudnick and Gao, 2003; Yan and Chi, 2005]. On the other hand, the catchments of the Mulanxi and Minjiang rivers in Fujian Province have source rocks closer to the felsic end-member (granitic rocks from the Cathysian block), which is consistent with provenance geology in their watersheds.

It is interesting to note that the sediments from Taiwan rivers, especially their clay fractions, are similar to those of the rivers in mainland China, all having source rocks close to the average composition of ECUC (Figure 6). We thus infer that sediments of Taiwan contain recycled continental crustal material from Southeast China and the basement rocks of Taiwan [Lan et al., 2002].
5.1.2. Influence of Lithology on Silicate Weathering Indices

In order to explore the effects of source rock types on silicate weathering, we focus on the index $a_{\text{Ba}}$ given that it correlates well with other weathering indices (Figure 5). By definition, $a_{\text{Ba}} = \frac{(\text{Th}/\text{Ba})_{\text{sed}}}{(\text{Th}/\text{Ba})_{\text{UCC}}}$. The ratio can indicate the depletion or enrichment of Th/Ba in the investigated sediment with respect to UCC, the representative of the unweathered source rocks within a large river such as the Changjiang. For those rivers draining small catchments, like the Minjiang and Taiwan rivers, they have relatively homogeneous source rocks that cannot be represented by the UCC model composition. Nevertheless, the calculation of $a_{\text{Ba}}$ can be corrected by using initial Ba/Th ratio of source rock rather than $(\text{Ba/Th})_{\text{UCC}}$, which can further reduce the effects of source rock difference [Chetelat et al., 2013]. For the correction of $a_{\text{Ba}}$, the Ba/Th ratio of both clayey and bulk samples has been plotted versus the Eu anomalies $(\text{Eu/Eu}^*)$ [Gao et al., 1998]. Both Eu and Ba are mainly hosted in the felsic components during the process of magmatic differentiation but Ba is preferentially released into solution during surficial processes whereas Eu and Th are basically retained in weathering residues. Ba is relatively depleted in both clayey and bulk samples compared to the trend line, which can represent the unweathered source rocks in the catchments, that passes through most widely distributed igneous rocks in South China. The values of initial Ba/Th ratio of the source rocks for each river can be estimated by vertical projection of each sample point on the trend line (Figure 7).

The corrected Ba/Th ratios of the source rocks range from 47.4 to 68.8 for the clays, which compares favorably with the value of 59.8 for the UCC [Rudnick and Gao, 2003]. According to the notation of $a_{\text{Ba}}$, the corrected weathering index $a_{\text{Ba}}$ of the Changjiang, Minjiang and Taiwan river clays are 2.8, 5.6 and 1.5, respectively (Figure 8), which are obtained by normalizing the $(\text{Ba/Th})_{\text{sed}}$ ratio against the recalculated Ba/Th ratios of the source rocks. Comparing the difference of $a_{\text{Ba}}$ values before and after the correction, the Minjiang bulk samples show a change of $a_{\text{Ba}}$ index from 6.8 to 5.6, while for the other river samples, the corrected $a_{\text{Ba}}$ values are close to the uncorrected ones (Figure 8 and supporting information Table S6). Based on the absolute values of corrected $a_{\text{Ba}}$, the silicate weathering in the Minjiang catchment is still the strongest, while Taiwan river basins have weaker weathering than the Changjiang catchment. Similar $a_{\text{Ba}}$ values in a given river before and after the correction suggests that the silicate weathering intensity registered in these river sediments has a poor correlation with diverse source rock types in these river basins. In other words, the provenance rock types apparently do not exert a dominant role on silicate weathering intensity difference in these catchments with different geological and climate settings.

5.2. Major Constraints of Silicate Weathering in the River Basins

The rivers entering East China’s marginal seas are mostly latitudinal oriented and span across different climate zones from north to south. Previous studies suggest that climate (average temperature, precipitation and runoff) is the dominant factor controlling silicate weathering intensity in these catchments while...
tectonic activity and physical erosion exert minor roles (Yang et al., 2004; Li and Yang, 2010; Shao et al., 2012; Qiu et al., 2014). This is likely true when considering that the river basins in East China mainland are all located in tectonically stable terrains. The majority of the Changjiang catchment lies in the Yangtze Craton and the small rivers entering the coastal area of southeast China run through South China fold system, both of which belong to stable blocks in mainland China relative to Taiwan Island. Based on the observed sediment fluxes, these rivers in mainland China generally have lower total denudation rates in their catchments, averaging at 104-276 t/km²/yr (Table 1). Under lower denudation rates, the relationship between chemical weathering flux and physical erosion is approximately linear (West et al., 2005, 2012; Gabet and Mudd, 2009), and the chemical weathering rate is greatly limited by the supply of weatherable material and is sensitive to the change of physical erosion rate (Riebe et al., 2004; West et al., 2005; West, 2012). In the current study, these mainland rivers have weaker denudation with a very narrow range, which may limit the silicate chemical weathering rates and may not cause huge difference in silicate chemical weathering intensity in these catchments.

The investigated mainland rivers are all latitudinally distributed and subject to variable monsoon climate, which causes the values of mean annual temperature, precipitation and runoff in these river basins to increase with decreasing latitudes (Figure 9). For instance, the mean temperature, precipitation and runoff are respectively 15°C, 1100 mm/yr and 500 mm/yr in the Changjiang catchment, but reach 19°C, 1717 mm/yr and 951 mm/yr in the Minjiang basin. In this study, all the weathering indices of both clay and bulk samples of the rivers in mainland China exhibit good inter-basinal correlations with the average temperature, precipitation and runoff, confirming the previous viewpoint that monsoon climate predominantly determines the integrated silicate weathering intensity in mainland China reflected by sediment chemistry (Figure 9) (Li and Yang, 2010; Shao et al., 2012). Under such a weathering environment with a low denudation rate (i.e., supply limited), the residence time of sediment in the catchment tends to be longer. Thus, the higher temperature and abundant rainfall will promote more dissolution of silicate minerals before their

Figure 7. Relationships linking europium (Eu) anomaly measured in the studied rivers with Ba/Th ratio. The coefficients of the regression line are calculated based on the data compiled from the associated gabbroic intrusions from the ELIP (Xu et al., 2001) and the granitic rocks from the Cathysian block (Li et al., 2003; Wang et al., 2007; Shellnutt et al., 2009; Zhang et al., 2012).

Figure 8. The histogram showing the values of weathering index $\alpha_{Ba}$ before and after the correction of source rocks. The world average of $\alpha_{Ba}$ (0.77, weighted based on river sediment yields) is from reference (Gaillardet et al., 1999a).
Figure 9. Comparisons of the weathering indices including Na, K, Ba and CIA with mean annual temperature (Temp.), precipitation (Prec.), runoff, sediment yield and gradient between the studied rivers. Correlations between these weathering indices and climate parameters are better for the mainland rivers than for Taiwan rivers (shaded area), suggesting different driving forces for silicate weathering in these two types of catchments.
removal from the weathering profiles. Consequently, higher rainfall, precipitation and average temperature in the Minjiang catchment in southern China lead to more intense silicate weathering (Figure 9), and thus the highest mean values of CIA and kaolinite content (Figure 4). It is generally recognized that kaolinite is mainly weathered from feldspar, mica and pyroxene after strong chemical eluviation by acidic medium under hot and humid climate [Singer, 1984; Thiry, 2000; Velde and Meunier, 2008], which is exactly the climate condition in the Minjiang watershed. Therefore, we suggest that the silicate weathering in the mainland rivers with tectonically stable catchments is primarily driven by monsoon climate (transport-limited). Therefore, it will be possible to use geochemistry of river-borne sediments weathered from the long-term stable terrains to evaluate paleo-weathering intensities. Such an attempt has been recently made by Wan et al. [2015] and Lupker et al. [2013] using the core data from the continental slope of the South China Sea and the Bengal Fan respectively.

It is noteworthy that, although Taiwan Island has the highest rainfall, average temperature and precipitation in the studied settings, silicate weathering intensity in their river basins revealed by sediment chemistry is the weakest (Figure 9). In particular, Taiwan rivers are situated at lower latitudes than the Minjiang but the intensity of integrated silicate weathering registered in Taiwan river sediments is much weaker than that of the Minjiang. Even the Changjiang basin farther to the north has higher silicate weathering intensity than Taiwan catchments. Therefore, other factors other than monsoon climate must be invoked for driving the silicate weathering in Taiwan Island.

It is noteworthy that the silt-laden mountainous rivers in Taiwan have very high chemical weathering fluxes. For instance, the silicate cation weathering rate of the Liwu River flowing through the east flank of the Central Range can reach 18 t km$^{-2}$ yr$^{-1}$, which is one of highest observed value in the world’s rivers draining felsic rocks [Calmels et al., 2011]. It is well recognized that strong tectonic uplift, frequent typhoon events and storm or earthquake-triggered landslides in Taiwan Island cause the strongest physical erosion in the world [Dadson et al., 2003; Milliman and Kao, 2005; Selvaraj and Chen, 2006; Kao and Milliman, 2008]. The physical erosion rate in Taiwan Island averages at 3–7 mm/yr [Dadson et al., 2003; Fuller et al., 2003], while the maximum physical erosion rates in the Changjiang Basin is about 0.066–0.175 mm/yr in the middle valley [Huang et al., 2013]. Strong land erosion in Taiwan river basins has been also inferred from the sedimentary records in the ECS, Taiwan Strait and South China Sea [Kao and Liu, 1996; Leithold et al., 2006; Huh et al., 2011]. The average sediment yield in the drainage basins of the studied Taiwan rivers reaches 8700 t km$^{-2}$/yr (Figure 9). The residence time of Taiwan river sediments in their watersheds is thus short because the eroded sediments are rapidly transferred to the sea and less stored in the basin [Huh et al., 2009, 2011].

The recent theoretical model analysis suggests that the chemical weathering fluxes can continue to increase even at the highest denudation rates [West, 2012]. High physical weathering rates in Taiwan will continuously create fresh rock surfaces, which may potentially enhance the chemical weathering defined by major ion fluxes in river waters [Gaillardet et al., 1999b]. In this sense, extremely high chemical fluxes in Taiwan river waters do indicate strong chemical weathering in response to intensive physically erosion and fast exhumation in the small mountainous catchments [Bey et al., 2001]. Rapid dissolution of some soluble salts and instable rocks under high temperature and abundant precipitation may cause the high chemical fluxes of river waters in Taiwan. However, in fact the weathering zone is not always contained entirely within the regolith, especially at the highest denudation rates [West, 2012]. In tectonically active Taiwan Island with rapid uplift of fractured rocks, the groundwater system have actually made great contribution to total chemical weathering flux [Calmels et al., 2011]. Yet the chemical weathering degree of regolith is actually not so high as reflected by the river sediment chemistry in this study.

As the CIA values of the fluvial clays decrease, the total contents of quartz and feldspar increase (Figure 4). If the parent rocks experienced intense chemical weathering and long sedimentary recycling, the feldspars would be destructed and altered totally to clay minerals [Fedo et al., 1995; Nesbitt et al., 1996], causing the increase of CIA. The high contents of feldspar and quartz in Taiwan river clays also indicate that enhanced physical erosion in Taiwan renders the rock-forming minerals less weathered due to the shorter sediment residence time in the small and steep catchments.

In such a mountainous setting with strong physical denudation and rapid sediment transfer from basin to sea, the silicate weathering in Taiwan river basins are largely restricted (weathering-limited). The short contact time of exposed fresh rocks with fluids due to strong physical erosion cannot result in significant...
alteration of feldspars to clay minerals during silicate weathering [Selvaraj and Chen, 2006]. Therefore, the fluvial sediments in Taiwan may experience shorter sedimentary recycling and residence time since their weathering from source rocks than those in the mainland China rivers. The latter are located in tectonically stable terrains and most of the sediments subject to weathering have already experienced multiple weathering and erosion cycles [Gaillardet et al., 1999a; Li and Yang, 2010; Shao et al., 2012]. Some studies suggest that under the strong physical denudation, temperature and runoff-related kinetics may dominate the silicate weathering fluxes [Riebe et al., 2004; West et al., 2005; West, 2012]. Nevertheless, the intensity of silicate weathering of present-day river particles from the active mountain belt, such as Taiwan Island, is largely restricted by incomplete water-rock interaction because of the limited chemical reaction time. Thus, the factors governing silicate weathering in active mountainous catchments are complex, and using sedimentary records to decipher paleoclimate change must be careful.

5.3. Two Types of Silicate Weathering and Sediment Transport Processes in East Asian Continental Margin

The East China Sea, as a typical river-dominated marginal sea in East Asia, is important in receiving and delivering a significant amount of terrigenous matter from the Eurasian continent to the Okinawa Trough and even to the West Pacific. This study reveals two types of river systems in East Asia in terms of silicate weathering regimes and sediment transport processes in the watersheds: the tectonically stable continental rivers versus tectonically active mountainous rivers (Figure 10a).

The tectonically stable continental rivers are represented by those rivers in mainland China. The intensity of silicate weathering in these river basins is primarily driven by monsoon climate and varies with latitudes. Among which, the Changjiang as a representative, is characterized by the mode of “large river/delta – wide shelf – huge terrigenous input – slower sediment transfer – strong anthropogenic impact” (Figure 10b). The Changjiang has a large river basin primarily situated on the stable Yangtze Craton, and developed a large...
delta by accumulating the enormous river sediment in its estuary and on the subaqueous delta [Dai et al., 2013]. The sediment source-to-sink transport in the Changjiang catchment is relatively slow, and the sedimentary recycling is strong because a large proportion of the sediments eroded from the upper valley are trapped in the midlower basins, such as in the floodplains and widely distributed lakes. The floodplain weathering may exert an important influence on river water chemistry and alter the composition of river sediments into the sea [Bouchez et al., 2012]. The floodplain weathering in the mainland river basins of East China is beyond the research scope of this paper, but it deserves research attention in the following study.

In addition, more than 50,000 reservoirs/dams have been constructed in the Changjiang catchment over the last five decades including the world largest one, the Three Gorges Dam. Intense damming activities also characterize the small catchments along southeast coastal China. The effects of damming on retaining river sediments have been widely documented in recent years [Yang et al., 2006; Yang et al., 2007; Dai et al., 2013; Yang et al., 2014]. Both the natural process and anthropogenic activities result in the long sediment recycling in the continental river basins located in the tectonically stable terrains. In consequence, the silicate weathering processes in tectonically stable continental river basins are mainly controlled by Asian monsoon climate, are typical transport-limited weathering regime. Overall, the mainland river basins covering different latitudes constitute a consistent weathering spectrum from the intermediate to strong weathering intensities [Shao et al., 2012; Qiu et al., 2014].

In contrast, tectonically active mountainous rivers are represented by those in Taiwan, characterized by the mode of “mountainous river – narrow shelf – huge terrigenous input – rapid sediment transfer by extreme weather events” (Figure 10b). Taiwan rivers have small catchments and short lengths but large sediment fluxes because of intense physical erosion resulting from high topographic relief and frequent typhoon-earthquake impacts on the landforms. Most of the sediments from these mountainous rivers experience shorter residence time and sedimentary recycling in their small watersheds, which implies that the silicate weathering intensity reflected by sediment chemistry in Taiwan is greatly limited by strong physical weathering on a short-term scale (typical weathering-limited regime). As a result of intense physical erosion but weak-intermediate silicate weathering in such small mountainous river systems, massive sediment discharges to the sea, mostly induced by hyperpycnal flow, lead to good preservation and quick burial of terrestrial organic carbon on continental shelves [Hilton et al., 2010, 2011; Liu et al., 2013]. This may represent another kind of river-sea interaction in terms of the transport pathway and depositional pattern of terrestrial organic matter in the marginal sea.

In summary, both tectonically stable continental rivers and tectonically active mountainous rivers coexists in East Asia, which constitutes two general silicate weathering regimes with different driving mechanisms and sediment source-to-sink transport processes. Both are important in governing the sedimentation and material cycle in East Asian continental margin and probably on a global scale. The East China Sea can therefore serve as an ideal natural laboratory for studying river-sea interaction and weathering-transport mechanisms at variable temporal and spatial scales.

6. Conclusions

In this study, we report elemental and mineral compositions in the clay and bulk sediments separated from the rivers entering the East China Sea, with the major purposes of investigating the mechanisms and processes of silicate weathering and sediment transport processes in their river basins.

Based upon the geochemical compositions of the fluvial sediments, the studied rivers can be classified into three categories: the Changjiang and small rivers in Zhejiang Province, the Minjiang in Fujian Province, and Taiwan rivers. The Changjiang and small rivers in Zhejiang have similar sediment compositions mainly because strong tide pumps the Changjiang-derived fine sediments into the estuaries and lower reaches of these small rivers. The meager sediment fluxes of these small coastal rivers make it difficult to reveal the real mineralogical and chemical compositions of the river sediments discharged into the sea. The Minjiang in south China is less affected by the Changjiang-derived sediment and has experienced the strongest silicate weathering in its catchment. Taiwan rivers have similar latitudes with the Minjiang but much weaker weathering than the later and even weaker than the Changjiang in the north.

The clays in the rivers are more sensitive indicators of silicate weathering in the catchments than the bulk samples are. Provenance rock types are not the dominant factor causing different intensities of silicate
weathering in the East Asian rivers. As a whole, two types of river systems coexist in East Asian continental margin: tectonically continental rivers (such as those in mainland China) and tectonically active mountainous rivers (e.g., Taiwan rivers). The silicate weathering process in the tectonically stable catchments is predominantly driven by Asian monsoon climate, and the sediment transfer from land to sea is relatively slow because of the long sediment residence time and sedimentary recycling in the basins (transport-limited weathering regime). The sediment geochemistry of these rivers may be used as the indication of paleoweathering and climate change. In comparison, the silicate weathering intensity in the tectonically active mountainous watersheds is greatly limited by strong physical denudation and fast sediment transfer (weathering-limited regime), although the fast and frequent exposure of fresh rock surface may account for high chemical fluxes of river water. The chemical weathering in tectonically active catchments is governed by complex factors and thus, it should be cautious to use sedimentary records for the reconstruction of paleoclimate change. These two river systems may represent two general driving mechanisms of silicate weathering and sediment source-to-sink transport processes on the Earth’s surface, and both governs the sedimentation and material cycle in East Asian continental margin.

Acknowledgments

Data supporting all figures and the analytical quality are available in supporting information Tables S1–S5. The data for Table 1 are available at the web of Changjiang Water Resources Commission (http://www.cjw.com.cn/), Water Resources Agency, Ministry of Economic Affairs, Taiwan (http://www.wra.gov.tw/), Zhejiang Provincial Water Management Institute (2012), and Fujian Provincial Water Management Institute (2012). The SRTM 90 m Digital Elevation Data for Figure 10b are available from the CGDIAR-CSI (http://srtm.csi.cgiar.org/). This work was supported by National Natural Science Foundation of China (grants 41376049 and 41225020) and Continental Shelf Drilling Program (grant GZH201100202). We thank Cinty Lee, Xiao-Ming Liu and the anonymous reviewers for their valuable comments which greatly improve the manuscript.

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