2016

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Publication Details
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Disciplines
Medicine and Health Sciences | Social and Behavioral Sciences

Publication Details

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This journal article is available at Research Online: http://ro.uow.edu.au/smhpapers/4266
The time scale of river sediment source-to-sink processes in East Asia

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Abstract

Knowledge of river sediment recycling provides important constraints on continent weathering and earth surface processes. In this study, we estimate the “comminution age” of sediments from the Changjiang (Yangtze River) and two small mountainous rivers in Taiwan based on their lithogenic \((^{234}\text{U}/^{238}\text{U})\) ratio. The \((^{234}\text{U}/^{238}\text{U})\) distributions in the Changjiang catchment are overall related to sediment grain size and chemical weathering regime, while \((^{234}\text{U}/^{238}\text{U})\) ratios in Taiwan rivers mainly depend on erosion/denudation processes. The comminution age constrains the time scale of sediment source-to-sink processes in catchments from sediment weathering/denudation to transportation, and finally deposition. Our results indicate that the comminution ages vary from 250 to 600 krys for the Changjiang sediments and ~110 krys for the Taiwan sediments. Different comminution ages are associated with contrasting erosion and weathering regimes and diverse topography between the large Changjiang catchment and small mountainous Taiwan basins. The longer comminution age of the Changjiang sediment is an interacting effect of a longer erosion/weathering history and sediment trapping effect (and thus slow transfer rate) created by broad floodplains and lakes in the middle and lower reaches. The shorter comminution age of the Taiwan sediment results from fast sediment denudation and transport associated with strong tectonic uplift, typhoon climate and steep topography. As these two major river systems dominate the sedimentology in East Asia continent margin, the distinct geological and topographical settings between the Changjiang and Taiwan river systems result in different sediment “source to sink” transport processes. This work presents a systematic and quantitative constraint on the time-scale of river sediment transfer process in East Asia, and also provides new insight into weathering
Regimes and sediment transport in monsoon climate-dominated continent and Island.

**Key words:**

$^{234}$U/$^{238}$U activity ratio, comminution age, Changjiang (Yangtze River), Taiwan rivers, sediment source-to-sink transport, erosion and weathering
1. Introduction

The East Asia continental margin links the Eurasian continent and the northwest Pacific Ocean, and is characterized by broad continental shelves and huge terrigenous sediment input from adjacent rivers. The river-dominated marginal sea, e.g. East China Sea (ECS), witnesses the complex sediment source-to-sink transport and sedimentary environmental changes during the late Quaternary (Li et al., 2014). The two distinct river systems that determine the sediment source-to-sink processes in the ECS include the Changjiang River (Yangtze River), the largest river in Asia, and several small mountainous rivers, especially those on Taiwan Island (Yang et al., 2015). A better understanding of the sediment source-to-sink transport processes from these river systems is critical to decipher the sedimentary evolution in the East Asia continental margin (Bi et al., 2015; Li et al., 2015). Although many previous studies focused on the sediment provenance and late Quaternary paleoenvironmental changes in the ESC and adjacent region (Yang et al., 2003; Liu et al., 2010; Li et al., 2014; Yang et al., 2015), the sediment transfer mechanism from land to sea and its difference between river systems have rarely been reported. In particular, the knowledge of sediment transfer time scale in the Changjiang and Taiwan river systems is very limited (Li et al., 2016).

Although a number of isotopic geochronological approaches have been successfully employed in measuring the ages of rock formations, they have rarely been used to constrain the time scale of sediment transport process (Rasmussen, 2005). This is primarily due to complex sedimentary processes from weathering and erosion, sediment transport and recycling to post-depositional diagenetic alteration, which makes it difficult to reliably date clastic sediments (DePaolo et al., 2012). Probing into the physical or
chemical changes that occur at or near the time of sediment formation is therefore of
great importance for the assessment of the time scale of sediment recycling.

Previous studies of dating clastic sedimentary rocks involve the uranium and thorium
radioactive decay series (Vigier et al., 2001; Chabaux et al., 2003; Dosseto et al., 2006;
Granet et al., 2010) and cosmogenic nuclides (Robert Bierman and Nichols, 2004;
DiBiase et al., 2010), which provide the first estimation of sediment transfer processes in
terms of time scale. Uranium isotope disequilibria provides a possibility of measuring the
time that has elapsed subsequent to the bedrock disintegrated by physical weathering to
small grains, which is coined as “comminution age” by DePaolo et al. (2006). The
comminution age method has been applied to North Atlantic deep sea sediments
(DePaolo et al., 2006; Maher et al., 2006), paleo-river channel sediments (Dosseto et al.,
2010; Lee et al., 2010; Handley et al., 2013a; Handley et al., 2013b) and Antarctica ice
cores (Aciego et al., 2011), which has yielded different timescales of sediment transfer
across various geological settings. The reported sediment transport time is determined by
many factors, but mainly by the change of sediment provenance in response to sea level
fluctuation (DePaolo et al., 2006; Lee et al., 2010), catchment erosion (Dosseto et al.,
2010) and climate (Handley et al., 2013b), and the uncertainties of the calculation itself
(DePaolo et al., 2012; Handley et al., 2013a).

In this study, sediment comminution ages of the Changjiang River and two rivers in
Taiwan Island are calculated by the sediment \( \frac{^{234}U}{^{238}U} \) activity ratios, and furthermore,
major constraints of the sediment comminution ages of the Changjiang and Taiwan rivers
are investigated. This study applies the comminution age method to these two contrasting
river systems in East Asia, and quantitatively constrains the time scale of sediment
source-to-sink processes within these river basins. The major recognition of this study will contribute to the understanding of sedimentary evolution in the west Pacific margin.

2. Comminution age theory

U-series disequilibria refers to the fractionation between different nuclides within a decay chain resulting in a non-steady state condition (steady state is known as secular equilibrium) (Ivanovich and Harmon, 1992; Bourdon et al., 2003; Dosseto, 2015). When $^{238}$U decays to $^{234}$Th in a mineral grain, the alpha decay can result in the physical displacement of the $^{234}$Th nuclide from the original position, which is also known as the $\alpha$-recoil effect. This process leads to the subsequent depletion of $^{234}$U relative to $^{238}$U (Kigoshi, 1971). If the fraction of $^{234}$Th ejected due to recoil is reliably estimated, the time since the grain reached its current size can be estimated, provided uranium in the mineral is uniformly distributed. The disequilibria between $^{238}$U and $^{234}$U can be used to calculate the comminution age using the following equation (DePaolo et al., 2006):

$$t_{com} = -\frac{1}{\lambda_{234}} \ln \left[ \frac{A_{meas} - (1 - f_a)}{A_0 - (1 - f_a)} \right]$$  \hspace{1cm} (1)

where $\lambda_{234}$ is the $^{234}$U decay constant (in yr$^{-1}$) and $f_a$ is the recoil loss factor. $A_{meas}$ and $A_0$ are the measured ($^{234}$U/$^{238}$U) activity ratio (U-series isotopic ratio in parenthesis denotes activity ratio) of the sediment while $A_0$ is the initial ($^{234}$U/$^{238}$U) of the source rock. The $t_{com}$ is the comminution age, which defines the time elapsed since physical erosion of the bedrock into fine-grained (typically <50 μm diameter) particles (DePaolo et al., 2006). As ($^{234}$U/$^{238}$U) will reach a grain-size-dependent steady state value after about four half lives of $^{234}$U. This method of determining a comminution age is limited to less than ∼1 Ma (Lee et al., 2010). For riverine sediment, this comminution age integrates the storage time
of sediment in regolith, transport in rivers, and possible temporary storage in an alluvial
plain (i.e. floodplain) (Dosseto et al., 2010), which overall represents the time of
sediment source-to-sink processes in the catchment.

For coarse-grained sediment, $^{234}$U–$^{238}$U disequilibria is only measurable at the grain
edge because the grain radius is much larger than the recoil length of ~30 nm (theoretical
$\alpha$-recoil length of feldspar). Preferential loss of $^{234}$U relative to $^{238}$U may also occur via
leaching during weathering of the source rock and/or sediment transport process.
Although DePaolo et al. (2006) and Maher et al. (2006) have argued that $^{234}$U depletion
can be determined by $\alpha$-recoil alone, a number of studies suggest (or have assumed) that
preferential leaching from damaged tracks of the mineral lattice is significant and worth
considering (Eyal and Olander, 1990; Chabaux et al., 2008; Bourdon et al., 2009;
Andersen et al., 2013).

3. River settings

Among the numerous rivers draining into the west Pacific, the Changjiang is the
largest river on the East Asian continent and also one of the largest rivers in the world. It
originates from the Tibet Plateau and has a drainage area up to 1.8×10$^6$ km$^2$ (Fig. 1a).
Geologically, the Changjiang catchment comprises complex rock types including
Archean metamorphic rocks, Jurassic sandstone, Paleozoic carbonate and sedimentary
rocks, Mesozoic–Cenozoic igneous and clastic rocks, and Quaternary detrital sediments
(Yang et al., 2004). The long-term average of annual sediment discharge of the
Changjiang River is about 470 Mt·yr$^{-1}$ (Table 1). The major part of the Changjiang-
derived sediment is trapped in the estuary to build a large delta and/or deposited on the
adjacent ECS shelf, while the remainder may be transported eastward to the west Pacific.
The small rivers in East Asia also deliver huge terrigenous sediments into the ECS, especially the mountainous rivers of Taiwan Island (Kao and Milliman, 2008). Taiwan is characterized by strong tectonic uplift at a rate of 5−10 mm/yr (Shin and Teng, 2001), and high rates of physical erosion up to 3–6 mm/yr (Dadson et al., 2003). Together with frequent typhoon and earthquake events, the rivers in Taiwan discharge >180 Mt·yr\(^{-1}\) of sediment to the surrounding marginal seas and make the Taiwan Island one of the highest sediment yields in the world (Kao and Milliman, 2008). The Taiwan river basins are mainly composed of sedimentary rocks and epimetamorphic rocks including sandstone, shale, slate and phyllite, with rare outcrops of volcanic rocks (Teng, 1990). The Zhuoshui (also named Chuoshui) River in the west is the largest in Taiwan, and has a total length of 190 km (Fig. 1; Table 1). It originates from the Central Mountain Range at an elevation of ~3,000 m and enters the Taiwan Strait. The Lanyang River in the east originates from the northeast Central Mountain Range and forms a large fluvial plain named the Yilan plain near its estuary (Fig. 1; Table 1).

4. Samples and methods

A total of 20 sediment samples were collected from three different rivers in East Asia, including 12 from the Changjiang River (Fig. 1a) on mainland China and 8 from two Taiwan Rivers, Zhuoshui and Lanyang (Fig. 1b). All the sediment samples were collected from the river floodplain except the two suspended samples from Jingzhou (JZ) and Jiujiang (JJ) in the mid-Changjiang mainstream that were filtered in situ by 0.45 μm filter. Approximately 2–5 g of sediment was wet sieved and the fraction <50 μm in diameter was separated for this study. Following Dosseto et al. (2010), the <50 μm fraction was then ignited at 550°C overnight (~8 hours) in order to eliminate organic
matter, and then leached with 1.5 N HCl for 6 hours in order to remove the uranium previously bound to organic matter, exchangeable fraction and any carbonates. The residues were finally leached using 0.04 M NH$_2$OH-HCl at pH=2 for 5 hours to dissolve the Fe-Mn oxides before elemental and ($^{234}$U/$^{238}$U) measurement. The net effect of the HCl-leaching procedure has been checked and it does not generate additional $^{234}$U-$^{238}$U disequilibria to the sample (DePaolo et al., 2006)

A portion (~0.15 g) of sediment <50 μm fraction was used for grain size measurement. The samples were leached by 1N HCl and 10% H$_2$O$_2$ to remove the carbonate and organic components, respectively. Then the residues were dispersed by (NaPO$_3$)$_6$ before the grain size measurement. Sediments size distributions were measured using a Beckman Coulter LS230 particle size analyzer at the State Key Laboratory of Marine Geology, Tongji University.

For geochemical analysis, about 0.05 g subsample was separated from the sequential leaching residues for element analysis. The residual samples were completely digested in a mixture of concentrated (HF+HNO$_3$) mix acid. The concentrations of major and trace elements were respectively determined by ICP-AES (IRIS Advantage, Thermo) and ICP-MS (VG-X7, Thermo) at the State Key Laboratory of Marine Geology, Tongji University. The precision and accuracy were monitored by national geostandards GSR-5, GSR-6 and GSR-9 from National Research Center. The analytical precision is generally better than 1–2% for major elements and better than 1–3% for trace elements. The U content for total procedure is < 0.01% of sample level.

($^{234}$U/$^{238}$U) activity ratios were determined on a Nu Plasma multi-collector ICP-MS at the Radiogenic Isotope Facility, the University of Queensland. Approximately 0.1 g of the
pretreated sediment material for each sample was digested using ultra-pure distilled HF+HNO₃+HCl acid mix in high-pressure Teflon bombs at 180°C. U was separated by using conventional anion-exchange columns made of TRU resin following standard chromatographic techniques described in (Edwards et al., 1987). Accuracy was assessed by analyzing the international U metal standard CRM-112A. Repeated measurements of the CRM-112A solution during this study give a mean \( \left( ^{234}\text{U}/^{238}\text{U} \right) \) value of 0.9639±0.0015 (N=9; 2σ), which is in agreement with the certified value of 0.9627, and consistent with values reported in the previous study (Hellstrom, 2003). The U procedural blank for the digestion and column separation procedure is 3.3±2.2 pg, which is negligible for samples containing ppm-level U.

5. Results

5.1 U concentrations and \( \left( ^{234}\text{U}/^{238}\text{U} \right) \) activity ratios

The U concentrations for the Changjiang samples range from 2.41 ppm to 6.17 ppm, with an average value of 3.28 ppm (Table 2). The highest U concentration is from a Panzhihua (PZH) sample collected from the upstream of the Changjiang River, with weak variations among the other samples. The U concentrations for the Taiwan samples vary from 3.25 ppm to 5.00 ppm, with an average of 3.75 ppm, which is a little higher than the average of the Changjiang River samples.

The \( \left( ^{234}\text{U}/^{238}\text{U} \right) \) activity ratios for the Changjiang sediments range from 0.8544 to 0.9364 (mean 0.8976) and decrease gradually from the upstream section of the river (Jinshajiang reaches) to the Three Gorges region in the middle reaches, with the exception of the sample from Zigui (ZG) which shows much higher \( \left( ^{234}\text{U}/^{238}\text{U} \right) \) than the
adjacent samples (Fig. 2a). Downstream the Three Gorges, the \(^{234}\text{U}/^{238}\text{U}\) increases from Jingzhou (JZ) to the estuary. The \(^{234}\text{U}/^{238}\text{U}\) ratios for the Taiwan samples vary from 0.9299 to 1.0017, and the mean value is 0.9569, which is higher than that of the Changjiang sediments. For the Taiwan river sediments, the \(^{234}\text{U}/^{238}\text{U}\) generally decreases from the source region to the estuary in the Zhuoshui River, but shows less variation in the Lanyang River (Fig. 2b).

It is notable that the \(^{234}\text{U}/^{238}\text{U}\) for the sample ZS1 collected from the upmost Zhuoshui River is >1. This suggests that the pre-leaching may not effectively remove all the non-detrital matter for this sample with current treatment (Dosseto et al., 2006; Handley et al., 2013a; Handley et al., 2013b). As no further assessment is performed in this study to verify the effectiveness of the pre-leaching, it is not warranted that the \(^{234}\text{U}/^{238}\text{U}\) for the other samples may also be higher than their true \(^{234}\text{U}/^{238}\text{U}\) values. Recently, Handley et al. (2013a), Suresh et al. (2014) and Martin et al. (2015) make great efforts to improve the leaching procedure. A mild HF/HCl leaching after the traditional sequential extraction is recommend for future study (Martin et al., 2015).

### 5.2 Grain size distribution

The mean grain size distributions for all the samples are listed in Table 2 and Appendix Table 1. The average of mean grain size for the Changjiang sediments is ~15.1 μm and the size variations are in agreement with the variations of \(^{234}\text{U}/^{238}\text{U}\) (Fig. 2a). The average of mean grain size for the Taiwan river sediments is slightly finer (~12.4 μm) but still in the same range as the Changjiang River samples. Unlike the Changjiang River samples, the mean grain size for the Taiwan river sediments has a poor correlation with their \(^{234}\text{U}/^{238}\text{U}\) (Fig. 2b).
5.3 Calculation of $f_\alpha$ and comminution age

The $\alpha$-recoil loss factor, $f_\alpha$, has been identified as a critical factor that may cause a large uncertainty in the comminution age methodology (DePaolo et al., 2006; DePaolo et al., 2012; Handley et al., 2013a; Handley et al., 2013b). In this study, $f_\alpha$ has been determined using the weighted geometric method based on grain size distribution (DePaolo et al., 2006), following:

$$f_\alpha = \int_{L}^{r_{max}} X(r)\beta(r)\lambda_s(r) \frac{3}{4} \left( \frac{L}{r} - \frac{L^3}{12r^3} \right) dr \ (2)$$

where $L$ is the $^{234}$Th $\alpha$-recoil length, $r$ is the grain radius (in μm). $X(r)$ is the volume fraction of grains with radius $r$. $\beta(r)$ is the aspect ratio of the grain and $\lambda_s$ is the surface roughness factor. Assumptions are required for the aspect ratio ($\beta$), surface roughness ($\lambda_s$) and $\alpha$-recoil length $L$. The aspect ratio ($\beta$) is previously assumed to range between 1 for the largest grain and 10 for the smallest grain (DePaolo et al., 2006; Dosseto et al., 2010), while surface roughness factor ($\lambda_s$) is expected to be 10 for the largest grains and 1 for the smallest grains (Handley et al., 2013a). The calculated $f_\alpha$ ranges from 0.108 to 0.285, which is generally within the published $f_\alpha$ ranges (0.135–0.19 in DePaolo et al. (2006), 0.06–0.143 in Dosseto et al. (2010), 0.143–0.225 in Aciego et al. (2011), 0.003–0.063 in Handley et al. (2013a) and 0.009–0.682 in Handley et al. (2013b)). Providing $A_0=1$, the comminution ages are thus calculated using Eq. (1).

The result of comminution ages are shown in Table 2. The comminution age for the Changjiang River sediments varies from 253±59 kyrs to 578±234 kyrs with an average of 360±100 kyrs. In contrast, comminution ages for the Taiwan river sediments are much
younger, which is 113±20 on average. In particular, the comminution age of Zhuoshui river sediment from the source region (ZS1) yields a negative value due to \(\frac{^{234}U}{^{238}U} > 1\).

As discussed in section 5.1, it is not warranted that the \(\frac{^{234}U}{^{238}U}\) may be larger than their true values as the leaching has not been checked for its effectiveness. If that was the case, then the comminution age calculated may be even longer as some non-detrital \(\frac{^{234}U}{^{238}U}\) may bias our comminution age calculation.

6. Discussion

6.1 \(^{234}\text{Th}\) recoil length and activity ratio of the source rocks \((L\text{ and } A_0)\)

The \(^{234}\text{Th}\) \(\alpha\)-recoil length \((L)\) is a critical parameter for the comminution age model, but the value is poorly constrained because \(L\) largely depends on grain mineralogy. Although \(L\) has been estimated using a number of approaches in previous studies (Maher et al., 2006; Dosseto and Schaller, 2016), a simply presumed \(L\) is widely used to calculate the comminution age. As the sediments in previous studies are complicated in mineralogy, the theoretical value of \(L=30\) nm for feldspar is usually used to yield a general estimation of comminution age. In this study, the mineralogy of the Changjiang and Taiwan river sediments have not been determined, thus the intermediate and theoretical \(L\) following previous studies is referred in the \(f_\alpha\) calculation.

The initial \(\frac{^{234}U}{^{238}U}\) value of the parent rocks \((A_0)\) is crucial for the calculation of comminution age in addition to \(f_\alpha\). The assumption that \(A_0 = 1\) has been widely adopted in earlier studies (DePaolo et al., 2006; Maher et al., 2006; Dosseto et al., 2010; Lee et al., 2010; DePaolo et al., 2012). Vigier and Bourdon (2011) suggested that the assumption of initial secular equilibrium for bulk rocks is overall valid. Afterwards, DePaolo et al.
(2012) measured the \(^{234}\text{U}/^{238}\text{U}\) in rocks taken from glacial outwash, which was supposed to represent freshly ground rock. The result showed that \(^{234}\text{U}/^{238}\text{U}\) in glacial outwash yielded an average \(^{234}\text{U}/^{238}\text{U}\) value close to 1.00±0.01 independent of its lithology. However, Handley et al. (2013b) argued that sedimentary rocks may not always be in secular equilibrium when analyzing some bedrocks from Proterozoic Flinders Ranges. Meanwhile, Handley et al. (2013a) calculated that the 2% uncertainties for \(A_0\) can result in ±178 kyrs uncertainties in comminution age.

The Changjiang River drains the typical topography of the China continent, with three-grade relief terraces spanning 3500–5000 m, 500–2000 m and <500 m in elevation (Fig. 1a). The large drainage basin, complex topography and diverse lithology of the Changjiang basin make it difficult, if not impossible, to find a proper bedrock to represent the \(A_0\) for the whole basin. Before \(A_0\) can be precisely determined, \(A_0=1\) has to be assumed in our comminution age calculation though it may bear a large uncertainty. In contrast, Taiwan has a steep topography and is subject to rapid tectonic uplift, resulting in high erosion rates of up to 3–6 mm/yr (Dadson et al., 2003). The small mountainous rivers in Taiwan have the highest sediment yields in the world (Kao and Milliman, 2008). The \(^{234}\text{U}/^{238}\text{U}\) of the sediment (ZS1) from the source region of Zhuoshui River is 1.0017 ± 0.0017, verifying our assumption of \(A_0=1\) for the Taiwan sediments to some extent.

### 6.2 \(^{234}\text{U}/^{238}\text{U}\) distributions in Changjiang and Taiwan river sediments

According to the sediment comminution model (DePaolo et al., 2006), when a small mineral grain is produced by erosion, the bulk \(^{234}\text{U}/^{238}\text{U}\) starts to decrease. If the grain size does not change substantially over time, the \(^{234}\text{U}/^{238}\text{U}\) ratio will eventually reach a steady state value which is a function of the grain size (or surface-to-volume ratio). In
In this study, ($^{234}$U/$^{238}$U) ratios in both the Changjiang and Taiwan river sediments show distinct variations within each river basin.

The ($^{234}$U/$^{238}$U) for the Changjiang sediments decrease gradually from the upstream section (including Jinshajiang reaches) of the river to the Three Gorges region in the middle reaches regardless of the sample from Zigui (ZG), and then increases from Jingzhou (JZ) to the estuary (Fig. 2a). It is noteworthy that the ($^{234}$U/$^{238}$U) value for ZG sample is abnormally higher than those of the samples from adjacent regions. The decrease of ($^{234}$U/$^{238}$U) in the upstream Changjiang can be interpreted by sediment comminution process during its transport from upstream to downstream, while the ($^{234}$U/$^{238}$U) of sediments from the mid-lower Changjiang reaches are distributed in a different way. A correlation analysis between ($^{234}$U/$^{238}$U) and mean grain size indicates that ($^{234}$U/$^{238}$U) has a similar trend to grain size with a correlation coefficient of $R^2=0.57$ (Fig. 3a) in the Changjiang sediments. On this account, the abnormally high ($^{234}$U/$^{238}$U) for ZG is primarily due to its much larger mean grain size than the neighboring samples.

For the Taiwan river sediments, the ($^{234}$U/$^{238}$U) generally decreases from the source region to the estuary in the Zhuoshui River, but displays less variation in the Lanyang River (Fig. 2b). The ($^{234}$U/$^{238}$U) distributions in Taiwan rivers are overall consistent with the comminution model. However, the correlation between ($^{234}$U/$^{238}$U) and mean grain size for the Taiwan river sediments is not significant (Fig. 3a).

Moreover, ($^{234}$U/$^{238}$U) ratios in the Changjiang and Taiwan rivers are also related to the elevations of these river basins, in particular for the mountainous or highland regions, e.g. the Zhuoshui River basin. With the decline of the basin elevation, ($^{234}$U/$^{238}$U) ratios in river sediments display clear decreasing trends in Taiwan and upstream Changjiang.
region (Fig. 2). Note that the $\left( ^{234}\text{U}/^{238}\text{U} \right)$ of ZG sample is extremely larger than the adjacent samples, which is probably caused by the large differences in mean grain size between these samples. Besides, the ZG sample is from the mountainous Three Gorges region that is likely to produce fresh rock clast grains with high $\left( ^{234}\text{U}/^{238}\text{U} \right)$ values.

A further analysis between sediment $\left( ^{234}\text{U}/^{238}\text{U} \right)$ and the elevations at sampling sites reveals that $\left( ^{234}\text{U}/^{238}\text{U} \right)$ has different distributions in the highland region (>100 m in this study) and lowland region (<100 m) (Fig. 3b), in particular for the Changjiang sediments. This result implies that the basin topography may exert an important impact on sediment $\left( ^{234}\text{U}/^{238}\text{U} \right)$ ratio and its comminution process. The mountainous/highland region is commonly steep in terrain and characterized by faster transport rate than erosion rate. This kind of region is termed as “weathering-limited” erosion regime (Gilbert, 1877). In a weathering limited region, once a small grain was produced by erosion, it was easily mobilized and transported in the catchment. Therefore, the sediment $\left( ^{234}\text{U}/^{238}\text{U} \right)$ values start to decrease gradually with the decline of elevation in Taiwan and upstream Changjiang region.

By contrast, the $\left( ^{234}\text{U}/^{238}\text{U} \right)$ values obtained from the mid-lower Changjiang sections increased from the middle reach to the estuary. The terrain in the mid-lower Changjiang basin is mainly lowland and/or plain regions after the Three Gorges, with the well development of lakes and flood plains (Fig. 1a). Moreover, this region is primarily located in the subtropical and temperate climate zone, and is prevailed by subtropical monsoon climate. Compared to the upstream region, the weathering regime in the mid-lower Changjiang valley is more “transport-limited” (Bi et al., 2015). The weathering products are less mobilized in the mid-lower Changjiang reaches as a result of basin
trapping, and thus undergo a longer residence time and are subject to stronger chemical weathering under a more favorable monsoon climate (Li and Yang, 2010; Shao et al., 2012). On this basis, the distribution of ($^{234}$U/$^{238}$U) and sediment comminution process in the mid-lower Changjiang reaches may be influenced by the chemical weathering.

To verify this hypothesis, the Chemical Index of Alteration (CIA) (Nesbitt and Young, 1982) that is widely used to assess the degree of chemical weathering (Li and Yang, 2010), was calculated and compared with ($^{234}$U/$^{238}$U). The higher CIA value reflects a stronger chemical weathering degree. A negative correlation between ($^{234}$U/$^{238}$U) and CIA ($R^2=0.76$) exists for the Changjiang River sediments (Fig. 4), which supports our hypothesis that the increasing ($^{234}$U/$^{238}$U) from the middle Changjiang reaches (transport-limited region) to the estuary is primarily affected by chemical weathering. It is worth noting that two out of five samples from the upper Changjiang River fall outside the 99% confidence interval of the ($^{234}$U/$^{238}$U) versus CIA plot (Fig. 4), which suggests that the ($^{234}$U/$^{238}$U) in the upper reaches of the river are poorly correlated to CIA than in the mid-lower reaches.

The CIA of Taiwan river sediments is as high as that of the Changjiang sediments from the mid-lower reaches, but the ($^{234}$U/$^{238}$U) is much higher for Taiwan river sediments (Fig. 4). The comparison and indications of CIA between the Changjiang and Taiwan river sediments are previously discussed by a number of studies (Li and Yang, 2010; Shao et al., 2012; Bi et al., 2015). In this research, no apparent relationship is found between ($^{234}$U/$^{238}$U) and CIA for Taiwan sediments, especially in the Zhuoshui River. However, excluding the samples from the mid-lower Changjiang reaches, the upper Changjiang sediments together with the sediments from Taiwan rivers display an
interesting positive correlation between \((^{234}\text{U}/^{238}\text{U})\) and CIA although the correlation coefficient is not high \((R^2=0.36;\ \text{Fig.}\ 4)\). Considering that the upper Changjiang basin and Taiwan catchments can be attributed to the weathering-limited region (Bi et al., 2015), our observations suggest that the \((^{234}\text{U}/^{238}\text{U})\) in river sediments can be greatly influenced by chemical weathering and erosion processes, and yield distinct distributions in weathering-limited and transport-limited regions.

For the transport-limited region (e.g. the mid-lower Changjiang reaches), a longer sediment residence time (indicated by low \((^{234}\text{U}/^{238}\text{U})\) value) tends to induce a strong chemical weathering due to the low sediment mobility. In comparison, for the weathering-limited region (e.g. Taiwan catchments), the sediment may suffer from extreme erosion due to the monsoon climate and high denudation rate (Table 1) but fast sediment transfer. The fast crush (mainly physical erosion) of fresh rocks may accelerate the sediment weathering. As a result, the CIA for Taiwan river sediment is high even in a short sediment residence time (high \((^{234}\text{U}/^{238}\text{U})\) value). However, a recent study by Dou et al. (2016) indicates the high CIA for Taiwan river sediments might reflect the reworking of older altered sediments from terraces and floodplains (i.e. sediment recycling). Taking this into account, the relation between \((^{234}\text{U}/^{238}\text{U})\) and weathering index (like CIA) is complicated as the proxy for weathering may integrate multiple sediment cycles while the sediment residence time recorded by \((^{234}\text{U}/^{238}\text{U})\) is limited to <1 Myr. Therefore, the comparison between CIA and \((^{234}\text{U}/^{238}\text{U})\) reveals that the \((^{234}\text{U}/^{238}\text{U})\) may evolve in different ways under different weathering regimes. However, more thorough studies are needed to elucidate the underlying mechanisms.
6.3 Contrasting sediment source-to-sink processes between the Changjiang and Taiwan river catchments

It is noticed that the calculated comminution ages for the Changjiang and Taiwan river sediments are much different, but the large uncertainties associated with the theoretic assumptions and the ($^{234}$U/$^{238}$U) calculation have to be taken into account. For a preliminary comparison of the sediment source-to-sink processes within a catchment and between different basins, some important information can be obtained according to the distinct comminution ages. The comminution ages for the Changjiang River sediments range from 250 to 600 kyrs, and vary between the upper (~300 kyrs) and mid-lower reaches (~410 kyrs) (Fig. 5a). This difference in comminution age largely depends on different erosion/weathering regime, as well as basin topography. The upper Changjiang basin, in particular the Jinshajiang catchment, drains the eastern portion of the Tibetan Plateau, which is the highest plateau on Earth with fast physical erosion (Chappell et al., 2006; He et al., 2014). Thus, the sediments from the upper Changjiang River have relatively short comminution ages. In contrast, the mid-lower Changjiang River is primarily situated on the Yangtze Craton, which has been relatively stable since the end of the middle Triassic (Jia, 2013). The eastern lowland is characterized by well-developed alluvial plains in the mid-lower valley, which may slow down the transport of sediments (Bi et al., 2015). Furthermore, two large freshwater lakes (Dongting and Poyang Lakes) are located in the middle Changjiang reaches, which may also increase sediment retention in the basin (Dai et al., 2005). As a whole, the sediments in the mid-lower Changjiang basin may have undergone longer and more complex sediment recycling relative to the sediments in the upper catchment, which consequently result in longer comminution ages.
The comminution ages for the Zhuoshui and Lanyang River sediments in Taiwan range from 70 to 200 kyrs, but do not show much topography-dependent due to the large errors in comminution age calculation (Fig. 5b). The comminution ages for the Zhuoshui River sediments generally decrease from the upstream to estuary, with the exception of the sample ZS3 that has the longest comminution age in the Zhuoshui River. The longer comminution age for ZS3 is primarily because of the sediment contribution from its tributary, Qingshui River, which supplies a large volume of sediment sourced from the lowland region. In contrast, the comminution ages for the Lanyang River sediments are more constant with small variations and are somewhat higher than those of the Zhuoshui River. The Lanyang River is located in the orogenic wedge, and it is thus characterized by relatively low erosion rates compared to the Zhuoshui River from the central part of the mountain belt (Siame et al., 2011).

The comminution ages for the Changjiang River sediments (250−600 kyrs) are significantly longer than those for the Zhuoshui and Lanyang River sediments (<200 kyrs) because of different erosion and weathering regimes in variable climatic and tectonic settings. Compared to the relatively slow sediment transfer rate in the large Changjiang River catchment on mainland China, the sedimentary environment in Taiwan is highly dynamic. Here, fluvial transport is fast as active tectonic activity causes high uplift rates and extreme climate events facilitating strong erosion and mass movements (Dadson et al., 2003; Siame et al., 2011; Egholm et al., 2013). Under these conditions, sediments from the Taiwan rivers are easily comminuted from the provenance rocks because frequent earthquakes and landslides mobilize large amounts of sediments from less weathered and deeper saprolite (Dosseto et al., 2014). In addition, the mobilized
sediments are quickly transported to the sea during high rainfall events, especially while frequent typhoons impact the steep topography in Taiwan. These processes will ultimately generate the sediments from Taiwan with younger “comminution ages”.

The different sediment transfer processes for these two river systems can also be seen from the hydrological data. For example, the total flux of suspended solid of the Changjiang River is about 470 Mt·yr⁻¹ based on long-term observations, which is more than 10 times that of the Zhuoshui River (Table 1). However, when river basin area is taken into account, the “sediment yields” (defined as the total sediment flux divided by drainage area) for the Zhuoshui and Lanyang Rivers are estimated to be 400 and 20 times higher than the Changjiang River, respectively.

It is necessary to note that all our comparisons based on the calculated comminution ages bear unexpected large uncertainties due the assumptions inherent in the method. It is difficult, if not impossible, to determine the true comminution age before all the key parameters can be quantitatively derived rather than using the assumptions and theoretical values. Although this study may not substantially contribute to improve the accuracy of comminution age calculation, it offers a systematic attempt of quantifying the time scale of sediment source-to-sink processes in East Asia where the various tectonic and climate settings exist in different catchments. The comparison between the Changjiang and Taiwan river sediment demonstrates that the comminution age cannot be considered as a precise dating method without the robust constraints on key parameters in the calculation. The uncertainties of this method await more studies.

8. Conclusions
The East Asian continental margin is characterized by both large (e.g. Changjiang) and small mountainous river systems. This study investigated the sediment comminution ages of these two river systems using \(^{234}\text{U}/^{238}\text{U}\). The \(^{234}\text{U}/^{238}\text{U}\) ratios range from 0.8544 to 0.9364 for the Changjiang River sediments, and from 0.9299 to 1.0017 for the Zhuoshui and Lanyang River sediments of Taiwan. The \(^{234}\text{U}/^{238}\text{U}\) distributions in the Changjiang basin are overall related to sediment grain size and chemical weathering, while \(^{234}\text{U}/^{238}\text{U}\) values in Taiwan rivers primarily depend on erosion/denudation processes in the catchments. The \(^{234}\text{U}/^{238}\text{U}\) in river sediments may evolve in different ways under variable weathering regimes within a catchment or between different basins.

The calculated comminution ages vary from 250 to 600 kyrs for the Changjiang River sediments and ~ 110 kyrs for the sediments from Taiwan. These differences are tightly related to contrasting erosion/weathering processes and diverse topography between the large Changjiang basin on the mainland and Taiwan Island. The longer comminution ages for the Changjiang basin are the result of a longer sediment recycling history and complex sediment trapping in the well-developed alluvial plains and lakes in the mid-lower reaches. In contrast, the younger comminution ages for the Taiwan sediments are caused by fast sediment denudation and transport as a result of strong tectonic uplift, typhoon-dominated climate and steep topography.

In summary, our study offers a quantitatively constraint on the time scales of sediment source-to-sink transport processes in East Asia, and provides an important understanding of the mechanism of sediment recycling in two distinct river systems. However, better constraints on key parameters for comminution age calculation are required to make precisely calculation.
Acknowledgments

We thank Heather Handley and an anonymous reviewer for their valuable comments and Francois Chabaux for the editorial handling of the manuscript. We thank James T. Liu and Yuan-Pin Chang for their support in the field to Taiwan Island. We thank Yuexing Feng for the help on isotopic data measurement. This work was supported by the Natural Science Foundation of China (41306040; 41225020, 41376049, U1606401), National Programme on Global Change and Air-Sea Interaction (GASI-GEOGE-03), and China Post-doctor Science Foundation (2015M570384).
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Table 1 Generals of the Changjiang, Zhuoshui and Lanyang Rivers.

<table>
<thead>
<tr>
<th>Rivers</th>
<th>Area $10^3$ km$^2$</th>
<th>Length km</th>
<th>$Q^a$ km$^3$.yr$^{-1}$</th>
<th>TSS$^a$ Mt.yr$^{-1}$</th>
<th>Sediment yield$^b$ t.km$^{-2}$.yr$^{-1}$</th>
<th>Erosion rate mm.yr$^{-1}c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Changjiang</td>
<td>1,800</td>
<td>6,300</td>
<td>900</td>
<td>470</td>
<td>300</td>
<td>0.7$^c$ (western)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.03–0.07$^c$ (eastern)</td>
</tr>
<tr>
<td>Zhuoshui</td>
<td>3.1</td>
<td>190</td>
<td>6.1</td>
<td>38</td>
<td>123,000</td>
<td>3–6$^d$</td>
</tr>
<tr>
<td>Lanyang</td>
<td>0.98</td>
<td>73</td>
<td>2.8</td>
<td>6.5</td>
<td>6,600</td>
<td>1–3$^e$</td>
</tr>
</tbody>
</table>

Q: river water discharge; TSS: total suspended solids

b. Sediment yield is simply defined as TSS divided by drainage area.
c. Erosion rates of the mid-lower reaches (eastern) and upper reaches (western) of the Changjiang are from Chappell et al. (2006).
d. Data of erosion rate referred from Dadson et al. (2003).
e. Date of erosion rate referred from Siame et al. (2011).
Table 2 Sample locations, U concentrations, \(^{234}\text{U}/^{238}\text{U}\) activity ratios, mean grain size, \(f_a\) and \(t_{\text{com}}\)

<table>
<thead>
<tr>
<th>Sample (Abbr.)</th>
<th>Lat. (°)</th>
<th>Lon. (°)</th>
<th>U (mg/kg)</th>
<th>(^{234}\text{U}/^{238}\text{U})</th>
<th>2SD</th>
<th>Mean grain size (μm)</th>
<th>(f_a)</th>
<th>(t_{\text{com}}) (kyr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shigu (SG)</td>
<td>26.87</td>
<td>99.97</td>
<td>2.64</td>
<td>0.9300</td>
<td>0.0009</td>
<td>15.77</td>
<td>0.137</td>
<td>253±59</td>
</tr>
<tr>
<td>Panzhihua (PZH)</td>
<td>26.61</td>
<td>101.80</td>
<td>6.17</td>
<td>0.9221</td>
<td>0.0013</td>
<td>16.75</td>
<td>0.135</td>
<td>306±78</td>
</tr>
<tr>
<td>Luzhou (LZ)</td>
<td>28.90</td>
<td>105.45</td>
<td>3.41</td>
<td>0.9079</td>
<td>0.0010</td>
<td>15.52</td>
<td>0.166</td>
<td>286±70</td>
</tr>
<tr>
<td>Wanzhou (WZ)</td>
<td>30.81</td>
<td>108.38</td>
<td>3.10</td>
<td>0.8974</td>
<td>0.0014</td>
<td>12.47</td>
<td>0.167</td>
<td>335±90</td>
</tr>
<tr>
<td>Zigui (ZG)</td>
<td>30.85</td>
<td>110.98</td>
<td>2.85</td>
<td>0.9364</td>
<td>0.0011</td>
<td>21.55</td>
<td>0.120</td>
<td>267±64</td>
</tr>
<tr>
<td>Jingzhou (JZ)</td>
<td>30.32</td>
<td>112.23</td>
<td>2.41</td>
<td>0.8544</td>
<td>0.0011</td>
<td>3.79</td>
<td>0.285</td>
<td>253±59</td>
</tr>
<tr>
<td>Wuhan (WH)</td>
<td>30.55</td>
<td>114.29</td>
<td>3.19</td>
<td>0.8715</td>
<td>0.0009</td>
<td>12.81</td>
<td>0.172</td>
<td>487±168</td>
</tr>
<tr>
<td>Jiujiang (JJ)</td>
<td>29.73</td>
<td>115.98</td>
<td>3.23</td>
<td>0.8699</td>
<td>0.0011</td>
<td>14.41</td>
<td>0.210</td>
<td>341±92</td>
</tr>
<tr>
<td>Datong (DT)</td>
<td>30.77</td>
<td>117.64</td>
<td>2.97</td>
<td>0.8919</td>
<td>0.0011</td>
<td>13.14</td>
<td>0.168</td>
<td>364±102</td>
</tr>
<tr>
<td>Nanjing (NJ)</td>
<td>31.98</td>
<td>118.64</td>
<td>2.81</td>
<td>0.8844</td>
<td>0.0014</td>
<td>15.86</td>
<td>0.164</td>
<td>429±134</td>
</tr>
<tr>
<td>Yangzhong (YZ)</td>
<td>32.31</td>
<td>119.75</td>
<td>3.23</td>
<td>0.8808</td>
<td>0.0014</td>
<td>17.19</td>
<td>0.148</td>
<td>578±234</td>
</tr>
<tr>
<td>Chongming (CM)</td>
<td>31.36</td>
<td>121.79</td>
<td>3.31</td>
<td>0.9250</td>
<td>0.0013</td>
<td>22.13</td>
<td>0.108</td>
<td>422±130</td>
</tr>
<tr>
<td>Lanyang-1 (LY1)</td>
<td>24.71</td>
<td>121.72</td>
<td>3.70</td>
<td>0.9299</td>
<td>0.0012</td>
<td>7.74</td>
<td>0.217</td>
<td>138±27</td>
</tr>
<tr>
<td>Lanyang-2 (LY2)</td>
<td>24.64</td>
<td>121.57</td>
<td>3.40</td>
<td>0.9496</td>
<td>0.0016</td>
<td>15.18</td>
<td>0.148</td>
<td>147±29</td>
</tr>
<tr>
<td>Lanyang-3 (LY3)</td>
<td>24.43</td>
<td>121.38</td>
<td>5.00</td>
<td>0.9399</td>
<td>0.0014</td>
<td>9.77</td>
<td>0.192</td>
<td>133±26</td>
</tr>
<tr>
<td>Zhuoshui-1 (ZS1)</td>
<td>23.97</td>
<td>121.11</td>
<td>4.09</td>
<td>1.0017</td>
<td>0.0017</td>
<td>11.11</td>
<td>0.159</td>
<td>-4±1</td>
</tr>
<tr>
<td>Zhuoshui-2 (ZS2)</td>
<td>23.79</td>
<td>120.91</td>
<td>3.45</td>
<td>0.9697</td>
<td>0.0011</td>
<td>9.41</td>
<td>0.176</td>
<td>67±12</td>
</tr>
<tr>
<td>Zhuoshui-3 (ZS3)</td>
<td>23.79</td>
<td>120.65</td>
<td>3.25</td>
<td>0.9507</td>
<td>0.0014</td>
<td>19.67</td>
<td>0.120</td>
<td>188±40</td>
</tr>
<tr>
<td>Zhuoshui-4 (ZS4)</td>
<td>23.81</td>
<td>120.47</td>
<td>3.41</td>
<td>0.9531</td>
<td>0.0016</td>
<td>10.99</td>
<td>0.163</td>
<td>120±23</td>
</tr>
<tr>
<td>Zhuoshui-5 (ZS5)</td>
<td>23.82</td>
<td>120.21</td>
<td>3.72</td>
<td>0.9603</td>
<td>0.0009</td>
<td>15.38</td>
<td>0.140</td>
<td>118±22</td>
</tr>
</tbody>
</table>

Abbreviation for sample ID is shown in bracket; Lat: latitude; Log: longitude; SD: standard deviation.

The comminution age error at the 2σ level was calculated by propagating the error on the measured \(^{234}\text{U}/^{238}\text{U}\) ratio and a 16% uncertainty for \(f_a\) follow Dosseto et al. (2010).
Figure captions

Figure 1
Map of East Asia and marginal seas showing the Changjiang River in China mainland (a), and Zhuoshui and Lanyang Rivers in Taiwan (b). Abbreviation for sample names are listed in Table 2.

Figure 2
Distributions of \(^{234}\text{U}/^{238}\text{U}\) and mean grain size in the river sediments and basin elevations for the Changjiang River (a) and Zhuoshui and Lanyang rivers in Taiwan (b).

Figure 3
Plots show the correlations of \(^{234}\text{U}/^{238}\text{U}\) activity ratio with mean grain size and (a) and elevations at sample locations (b).

Figure 4
A plot of CIA versus \(^{234}\text{U}/^{238}\text{U}\) in the river sediments. Red solid line indicates the linear regression for the Changjiang sediments, with 99% confidence interval shown in pink shade area. Blue solid line indicates the linear regression for the Taiwan sediments and upper-Changjiang sediments, with 99% confidence interval shown in grey dash lines.

Figure 5
Downstream distributions of comminution ages of sediments along the Changjiang (a) and Taiwan rivers (b). Topographic elevation for the Changjiang basin and Taiwan Island are also shown in grey solid lines. Note that the axis of comminution age is shown in a reverse order for a better comparison.
The time scale of river sediment source-to-sink processes in East Asia

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Abstract

Knowledge of river sediment recycling provides important constraints on continent weathering and earth surface processes. In this study, we estimate the “comminution age” of sediments from the Changjiang (Yangtze River) and two small mountainous rivers in Taiwan based on their lithogenic \(^{234}\text{U}/^{238}\text{U}\) ratio. The \(^{234}\text{U}/^{238}\text{U}\) distributions in the Changjiang catchment are overall related to sediment grain size and chemical weathering regime, while \(^{234}\text{U}/^{238}\text{U}\) ratios in Taiwan rivers mainly depend on erosion/denudation processes. The comminution age constrains the time scale of sediment source-to-sink processes in catchments from sediment weathering/denudation to transportation, and finally deposition. Our results indicate that the comminution ages vary from 250 to 600 kyrs for the Changjiang sediments and ~110 kyrs for the Taiwan sediments. Different comminution ages are associated with contrasting erosion and weathering regimes and diverse topography between the large Changjiang catchment and small mountainous Taiwan basins. The longer comminution age of the Changjiang sediment is an interacting effect of a longer erosion/weathering history and sediment trapping effect (and thus slow transfer rate) created by broad floodplains and lakes in the middle and lower reaches. The shorter comminution age of the Taiwan sediment results from fast sediment denudation and transport associated with strong tectonic uplift, typhoon climate and steep topography. As these two major river systems dominate the sedimentology in East Asia continent margin, the distinct geological and topographical settings between the Changjiang and Taiwan river systems result in different sediment “source to sink” transport processes. This work presents a systematic and quantitative constraint on the time-scale of river sediment transfer process in East Asia, and also provides new insight into weathering
regimes and sediment transport in monsoon climate-dominated continent and Island.

**Key words:**

$^{234}$U/$^{238}$U activity ratio, comminution age, Changjiang (Yangtze River), Taiwan rivers, sediment source-to-sink transport, erosion and weathering
1. Introduction

The East Asia continental margin links the Eurasian continent and the northwest Pacific Ocean, and is characterized by broad continental shelves and huge terrigenous sediment input from adjacent rivers. The river-dominated marginal sea, e.g. East China Sea (ECS), witnesses the complex sediment source-to-sink transport and sedimentary environmental changes during the late Quaternary (Li et al., 2014). The two distinct river systems that determine the sediment source-to-sink processes in the ECS include the Changjiang River (Yangtze River), the largest river in Asia, and several small mountainous rivers, especially those on Taiwan Island (Yang et al., 2015). A better understanding of the sediment source-to-sink transport processes from these river systems is critical to decipher the sedimentary evolution in the East Asia continental margin (Bi et al., 2015; Li et al., 2015). Although many previous studies focused on the sediment provenance and late Quaternary paleoenvironmental changes in the ESC and adjacent region (Yang et al., 2003; Liu et al., 2010; Li et al., 2014; Yang et al., 2015), the sediment transfer mechanism from land to sea and its difference between river systems have rarely been reported. In particular, the knowledge of sediment transfer time scale in the Changjiang and Taiwan river systems is very limited (Li et al., 2016).

Although a number of isotopic geochronological approaches have been successfully employed in measuring the ages of rock formations, they have rarely been used to constrain the time scale of sediment transport process (Rasmussen, 2005). This is primarily due to complex sedimentary processes from weathering and erosion, sediment transport and recycling to post-depositional diagenetic alteration, which makes it difficult to reliably date clastic sediments (DePaolo et al., 2012). Probing into the physical or
chemical changes that occur at or near the time of sediment formation is therefore of great importance for the assessment of the time scale of sediment recycling.

Previous studies of dating clastic sedimentary rocks involve the uranium and thorium radioactive decay series (Vigier et al., 2001; Chabaux et al., 2003; Dosseto et al., 2006; Granet et al., 2010) and cosmogenic nuclides (Robert Bierman and Nichols, 2004; DiBiase et al., 2010), which provide the first estimation of sediment transfer processes in terms of time scale. Uranium isotope disequilibria provides a possibility of measuring the time that has elapsed subsequent to the bedrock disintegrated by physical weathering to small grains, which is coined as “comminution age” by DePaolo et al. (2006). The comminution age method has been applied to North Atlantic deep sea sediments (DePaolo et al., 2006; Maher et al., 2006), paleo-river channel sediments (Dosseto et al., 2010; Lee et al., 2010; Handley et al., 2013a; Handley et al., 2013b) and Antarctica ice cores (Aciego et al., 2011), which has yielded different timescales of sediment transfer across various geological settings. The reported sediment transport time is determined by many factors, but mainly by the change of sediment provenance in response to sea level fluctuation (DePaolo et al., 2006; Lee et al., 2010), catchment erosion (Dosseto et al., 2010) and climate (Handley et al., 2013b), and the uncertainties of the calculation itself (DePaolo et al., 2012; Handley et al., 2013a).

In this study, sediment comminution ages of the Changjiang River and two rivers in Taiwan Island are calculated by the sediment \((^{234}\text{U}/^{238}\text{U})\) activity ratios, and furthermore, major constraints of the sediment comminution ages of the Changjiang and Taiwan rivers are investigated. This study applies the comminution age method to these two contrasting river systems in East Asia, and quantitatively constrains the time scale of sediment
source-to-sink processes within these river basins. The major recognition of this study will contribute to the understanding of sedimentary evolution in the west Pacific margin.

2. Comminution age theory

U-series disequilibria refers to the fractionation between different nuclides within a decay chain resulting in a non-steady state condition (steady state is known as secular equilibrium) (Ivanovich and Harmon, 1992; Bourdon et al., 2003; Dosseto, 2015). When $^{238}\text{U}$ decays to $^{234}\text{Th}$ in a mineral grain, the alpha decay can result in the physical displacement of the $^{234}\text{Th}$ nuclide from the original position, which is also known as the $\alpha$-recoil effect. This process leads to the subsequent depletion of $^{234}\text{U}$ relative to $^{238}\text{U}$ (Kigoshi, 1971). If the fraction of $^{234}\text{Th}$ ejected due to recoil is reliably estimated, the time since the grain reached its current size can be estimated, provided uranium in the mineral is uniformly distributed. The disequilibria between $^{238}\text{U}$ and $^{234}\text{U}$ can be used to calculate the comminution age using the following equation (DePaolo et al., 2006):

$$t_{com} = -\frac{1}{\lambda_{234}} \ln \left[ \frac{A_{meas} - (1 - f_a)}{A_0 - (1 - f_a)} \right]$$  \hspace{1cm} (1)

where $\lambda_{234}$ is the $^{234}\text{U}$ decay constant (in yr$^{-1}$) and $f_a$ is the recoil loss factor. $A_{meas}$ and $A_0$ are the measured ($^{234}\text{U}/^{238}\text{U}$) activity ratio (U-series isotopic ratio in parenthesis denotes activity ratio) of the sediment while $A_0$ is the initial ($^{234}\text{U}/^{238}\text{U}$) of the source rock. The $t_{com}$ is the comminution age, which defines the time elapsed since physical erosion of the bedrock into fine-grained (typically $<50$ μm diameter) particles (DePaolo et al., 2006). As ($^{234}\text{U}/^{238}\text{U}$) will reach a grain-size-dependent steady state value after about four half lives of $^{234}\text{U}$. This method of determining a comminution age is limited to less than $\sim 1$ Ma (Lee et al., 2010). For riverine sediment, this comminution age integrates the storage time
of sediment in regolith, transport in rivers, and possible temporary storage in an alluvial
plain (i.e. floodplain) (Dosseto et al., 2010), which overall represents the time of
sediment source-to-sink processes in the catchment.

For coarse-grained sediment, $^{234}$U–$^{238}$U disequilibria is only measurable at the grain
edge because the grain radius is much larger than the recoil length of ~30 nm (theoretical
$\alpha$-recoil length of feldspar). Preferential loss of $^{234}$U relative to $^{238}$U may also occur via
leaching during weathering of the source rock and/or sediment transport process.

Although DePaolo et al. (2006) and Maher et al. (2006) have argued that $^{234}$U depletion
can be determined by $\alpha$-recoil alone, a number of studies suggest (or have assumed) that
preferential leaching from damaged tracks of the mineral lattice is significant and worth
considering (Eyal and Olander, 1990; Chabaux et al., 2008; Bourdon et al., 2009;
Andersen et al., 2013).

3. River settings

Among the numerous rivers draining into the west Pacific, the Changjiang is the
largest river on the East Asian continent and also one of the largest rivers in the world. It
originates from the Tibet Plateau and has a drainage area up to $1.8 \times 10^6$ km$^2$ (Fig. 1a).

Geologically, the Changjiang catchment comprises complex rock types including
Archean metamorphic rocks, Jurassic sandstone, Paleozoic carbonate and sedimentary
rocks, Mesozoic–Cenozoic igneous and clastic rocks, and Quaternary detrital sediments
(Yang et al., 2004). The long-term average of annual sediment discharge of the
Changjiang River is about 470 Mt·yr$^{-1}$ (Table 1). The major part of the Changjiang-
derived sediment is trapped in the estuary to build a large delta and/or deposited on the
adjacent ECS shelf, while the remainder may be transported eastward to the west Pacific.
The small rivers in East Asia also deliver huge terrigenous sediments into the ECS, especially the mountainous rivers of Taiwan Island (Kao and Milliman, 2008). Taiwan is characterized by strong tectonic uplift at a rate of 5−10 mm/yr (Shin and Teng, 2001), and high rates of physical erosion up to 3−6 mm/yr (Dadson et al., 2003). Together with frequent typhoon and earthquake events, the rivers in Taiwan discharge >180 Mt·yr⁻¹ of sediment to the surrounding marginal seas and make the Taiwan Island one of the highest sediment yields in the world (Kao and Milliman, 2008). The Taiwan river basins are mainly composed of sedimentary rocks and epimetamorphic rocks including sandstone, shale, slate and phyllite, with rare outcrops of volcanic rocks (Teng, 1990). The Zhuoshui (also named Chuoshui) River in the west is the largest in Taiwan, and has a total length of 190 km (Fig. 1; Table 1). It originates from the Central Mountain Range at an elevation of ~3,000 m and enters the Taiwan Strait. The Lanyang River in the east originates from the northeast Central Mountain Range and forms a large fluvial plain named the Yilan plain near its estuary (Fig. 1; Table 1).

4. Samples and methods

A total of 20 sediment samples were collected from three different rivers in East Asia, including 12 from the Changjiang River (Fig. 1a) on mainland China and 8 from two Taiwan Rivers, Zhuoshui and Lanyang (Fig. 1b). All the sediment samples were collected from the river floodplain except the two suspended samples from Jingzhou (JZ) and Jiujiang (JJ) in the mid-Changjiang mainstream that were filtered in situ by 0.45 μm filter. Approximately 2−5 g of sediment was wet sieved and the fraction <50 μm in diameter was separated for this study. Following Dosseto et al. (2010), the <50 μm fraction was then ignited at 550°C overnight (~8 hours) in order to eliminate organic
matter, and then leached with 1.5 N HCl for 6 hours in order to remove the uranium previously bound to organic matter, exchangeable fraction and any carbonates. The residues were finally leached using 0.04M NH$_2$OH-HCl at pH=2 for 5 hours to dissolve the Fe-Mn oxides before elemental and ($^{234}\text{U}/^{238}\text{U}$) measurement. The net effect of the HCl-leaching procedure has been checked and it does not generate additional $^{234}\text{U}-^{238}\text{U}$ disequilibria to the sample (DePaolo et al., 2006).

A portion (~0.15 g) of sediment <50 μm fraction was used for grain size measurement. The samples were leached by 1N HCl and 10% H$_2$O$_2$ to remove the carbonate and organic components, respectively. Then the residues were dispersed by (NaPO$_3$)$_6$ before the grain size measurement. Sediments size distributions were measured using a Beckman Coulter LS230 particle size analyzer at the State Key Laboratory of Marine Geology, Tongji University.

For geochemical analysis, about 0.05 g subsample was separated from the sequential leaching residues for element analysis. The residual samples were completely digested in a mixture of concentrated (HF+HNO$_3$) mix acid. The concentrations of major and trace elements were respectively determined by ICP-AES (IRIS Advantage, Thermo) and ICP-MS (VG-X7, Thermo) at the State Key Laboratory of Marine Geology, Tongji University. The precision and accuracy were monitored by national geostandards GSR-5, GSR-6 and GSR-9 from National Research Center. The analytical precision is generally better than 1–2% for major elements and better than 1–3% for trace elements. The U content for total procedure is < 0.01% of sample level.

($^{234}\text{U}/^{238}\text{U}$) activity ratios were determined on a Nu Plasma multi-collector ICP-MS at the Radiogenic Isotope Facility, the University of Queensland. Approximately 0.1 g of the
pretreated sediment material for each sample was digested using ultra-pure distilled
HF+HNO$_3$+HCl acid mix in high-pressure Teflon bombs at 180°C. U was separated by
using conventional anion-exchange columns made of TRU resin following standard
chromatographic techniques described in (Edwards et al., 1987). Accuracy was assessed
by analyzing the international U metal standard CRM-112A. Repeated measurements of
the CRM-112A solution during this study give a mean ($^{234}$U/$^{238}$U) value of
0.9639±0.0015 (N=9; 2σ), which is in agreement with the certified value of 0.9627, and
consistent with values reported in the previous study (Hellstrom, 2003). The U procedural
blank for the digestion and column separation procedure is 3.3±2.2 pg, which is
negligible for samples containing ppm-level U.

5. Results

5.1 U concentrations and ($^{234}$U/$^{238}$U) activity ratios

The U concentrations for the Changjiang samples range from 2.41 ppm to 6.17 ppm,
with an average value of 3.28 ppm (Table 2). The highest U concentration is from a
Panzhihua (PZH) sample collected from the upstream of the Changjiang River, with weak
variations among the other samples. The U concentrations for the Taiwan samples vary
from 3.25 ppm to 5.00 ppm, with an average of 3.75 ppm, which is a little higher than the
average of the Changjiang River samples.

The ($^{234}$U/$^{238}$U) activity ratios for the Changjiang sediments range from 0.8544 to
0.9364 (mean 0.8976) and decrease gradually from the upstream section of the river
(Jinshajiang reaches) to the Three Gorges region in the middle reaches, with the
exception of the sample from Zigui (ZG) which shows much higher ($^{234}$U/$^{238}$U) than the
adjacent samples (Fig. 2a). Downstream the Three Gorges, the \((^{234}\text{U}/^{238}\text{U})\) increases from Jingzhou (JZ) to the estuary. The \((^{234}\text{U}/^{238}\text{U})\) ratios for the Taiwan samples vary from 0.9299 to 1.0017, and the mean value is 0.9569, which is higher than that of the Changjiang sediments. For the Taiwan river sediments, the \((^{234}\text{U}/^{238}\text{U})\) generally decreases from the source region to the estuary in the Zhuoshui River, but shows less variation in the Lanyang River (Fig. 2b).

It is notable that the \((^{234}\text{U}/^{238}\text{U})\) for the sample ZS1 collected from the upmost Zhuoshui River is \(>1\). This suggests that the pre-leaching may not effectively remove all the non-detrital matter for this sample with current treatment (Dosseto et al., 2006; Handley et al., 2013a; Handley et al., 2013b). As no further assessment is performed in this study to verify the effectiveness of the pre-leaching, it is not warranted that the \((^{234}\text{U}/^{238}\text{U})\) for the other samples may also be higher than their true \((^{234}\text{U}/^{238}\text{U})\) values. Recently, Handley et al. (2013a), Suresh et al. (2014) and Martin et al. (2015) make great efforts to improve the leaching procedure. A mild HF/HCl leaching after the traditional sequential extraction is recommend for future study (Martin et al., 2015).

### 5.2 Grain size distribution

The mean grain size distributions for all the samples are listed in Table 2 and Appendix Table 1. The average of mean grain size for the Changjiang sediments is \(~15.1\) μm and the size variations are in agreement with the variations of \((^{234}\text{U}/^{238}\text{U})\) (Fig. 2a). The average of mean grain size for the Taiwan river sediments is slightly finer \(~12.4\) μm but still in the same range as the Changjiang River samples. Unlike the Changjiang River samples, the mean grain size for the Taiwan river sediments has a poor correlation with their \((^{234}\text{U}/^{238}\text{U})\) (Fig. 2b).
5.3 Calculation of $f_\alpha$ and comminution age

The $\alpha$-recoil loss factor, $f_\alpha$, has been identified as a critical factor that may cause a large uncertainty in the comminution age methodology (DePaolo et al., 2006; DePaolo et al., 2012; Handley et al., 2013a; Handley et al., 2013b). In this study, $f_\alpha$ has been determined using the weighted geometric method based on grain size distribution (DePaolo et al., 2006), following:

$$f_\alpha = \int_{L}^{r_{\text{max}}} X(r)\beta(r)\lambda_s(r) \frac{3}{4} \left( \frac{L}{r} - \frac{L^3}{12r^3} \right) dr \tag{2}$$

where $L$ is the $^{234}$Th $\alpha$-recoil length, $r$ is the grain radius (in μm). $X(r)$ is the volume fraction of grains with radius $r$. $\beta(r)$ is the aspect ratio of the grain and $\lambda_s$ is the surface roughness factor. Assumptions are required for the aspect ratio ($\beta$), surface roughness ($\lambda_s$) and $\alpha$-recoil length $L$. The aspect ratio ($\beta$) is previously assumed to range between 1 for the largest grain and 10 for the smallest grain (DePaolo et al., 2006; Dosseto et al., 2010), while surface roughness factor ($\lambda_s$) is expected to be 10 for the largest grains and 1 for the smallest grains (Handley et al., 2013a). The calculated $f_\alpha$ ranges from 0.108 to 0.285, which is generally within the published $f_\alpha$ ranges (0.135–0.19 in DePaolo et al. (2006), 0.06–0.143 in Dosseto et al. (2010), 0.143–0.225 in Aciego et al. (2011), 0.003–0.063 in Handley et al. (2013a) and 0.009–0.682 in Handley et al. (2013b)). Providing $A_0=1$, the comminution ages are thus calculated using Eq. (1).

The result of comminution ages are shown in Table 2. The comminution age for the Changjiang River sediments varies from 253±59 kyrs to 578±234 kyrs with an average of 360±100 kyrs. In contrast, comminution ages for the Taiwan river sediments are much
younger, which is 113±20 on average. In particular, the comminution age of Zhuoshui river sediment from the source region (ZS1) yields a negative value due to \((^{234}\text{U}/^{238}\text{U}) > 1\). As discussed in section 5.1, it is not warranted that the \((^{234}\text{U}/^{238}\text{U})\) may be larger than their true values as the leaching has not been checked for its effectiveness. If that was the case, then the comminution age calculated may be even longer as some non-detrital \((^{234}\text{U}/^{238}\text{U})\) may bias our comminution age calculation.

6. Discussion

6.1 \(2^{34}\text{Th}\) recoil length and activity ratio of the source rocks \((L\text{ and }A_0)\)

The \(2^{34}\text{Th}\) \(\alpha\)-recoil length \(L\) is a critical parameter for the comminution age model, but the value is poorly constrained because \(L\) largely depends on grain mineralogy. Although \(L\) has been estimated using a number of approaches in previous studies (Maher et al., 2006; Dosseto and Schaller, 2016), a simply presumed \(L\) is widely used to calculate the comminution age. As the sediments in previous studies are complicated in mineralogy, the theoretical value of \(L=30\) nm for feldspar is usually used to yield a general estimation of comminution age. In this study, the mineralogy of the Changjiang and Taiwan river sediments have not been determined, thus the intermediate and theoretical \(L\) following previous studies is referred in the \(f_\alpha\) calculation.

The initial \((^{234}\text{U}/^{238}\text{U})\) value of the parent rocks \((A_0)\) is crucial for the calculation of comminution age in addition to \(f_\alpha\). The assumption that \(A_0 = 1\) has been widely adopted in earlier studies (DePaolo et al., 2006; Maher et al., 2006; Dosseto et al., 2010; Lee et al., 2010; DePaolo et al., 2012). Vigier and Bourdon (2011) suggested that the assumption of initial secular equilibrium for bulk rocks is overall valid. Afterwards, DePaolo et al.
(2012) measured the \(^{234}\text{U}/^{238}\text{U}\) in rocks taken from glacial outwash, which was supposed to represent freshly ground rock. The result showed that \(^{234}\text{U}/^{238}\text{U}\) in glacial outwash yielded an average \(^{234}\text{U}/^{238}\text{U}\) value close to 1.00±0.01 independent of its lithology. However, Handley et al. (2013b) argued that sedimentary rocks may not always be in secular equilibrium when analyzing some bedrocks from Proterozoic Flinders Ranges. Meanwhile, Handley et al. (2013a) calculated that the 2% uncertainties for \(A_0\) can result in ±178 kyrs uncertainties in comminution age.

The Changjiang River drains the typical topography of the China continent, with three-grade relief terraces spanning 3500–5000 m, 500–2000 m and <500 m in elevation (Fig. 1a). The large drainage basin, complex topography and diverse lithology of the Changjiang basin make it difficult, if not impossible, to find a proper bedrock to represent the \(A_0\) for the whole basin. Before \(A_0\) can be precisely determined, \(A_0=1\) has to be assumed in our comminution age calculation though it may bear a large uncertainty. In contrast, Taiwan has a steep topography and is subject to rapid tectonic uplift, resulting in high erosion rates of up to 3–6 mm/yr (Dadson et al., 2003). The small mountainous rivers in Taiwan have the highest sediment yields in the world (Kao and Milliman, 2008). The \(^{234}\text{U}/^{238}\text{U}\) of the sediment (ZS1) from the source region of Zhuoshui River is 1.0017 ± 0.0017, verifying our assumption of \(A_0=1\) for the Taiwan sediments to some extent.

6.2 \(^{234}\text{U}/^{238}\text{U}\) distributions in Changjiang and Taiwan river sediments

According to the sediment comminution model (DePaolo et al., 2006), when a small mineral grain is produced by erosion, the bulk \(^{234}\text{U}/^{238}\text{U}\) starts to decrease. If the grain size does not change substantially over time, the \(^{234}\text{U}/^{238}\text{U}\) ratio will eventually reach a steady state value which is a function of the grain size (or surface-to-volume ratio). In
this study, ($^{234}$U/$^{238}$U) ratios in both the Changjiang and Taiwan river sediments show distinct variations within each river basin.

The ($^{234}$U/$^{238}$U) for the Changjiang sediments decrease gradually from the upstream section (including Jinshajiang reaches) of the river to the Three Gorges region in the middle reaches regardless of the sample from Zigui (ZG), and then increases from Jingzhou (JZ) to the estuary (Fig. 2a). It is noteworthy that the ($^{234}$U/$^{238}$U) value for ZG sample is abnormally higher than those of the samples from adjacent regions. The decrease of ($^{234}$U/$^{238}$U) in the upstream Changjiang can be interpreted by sediment comminution process during its transport from upstream to downstream, while the ($^{234}$U/$^{238}$U) of sediments from the mid-lower Changjiang reaches are distributed in a different way. A correlation analysis between ($^{234}$U/$^{238}$U) and mean grain size indicates that ($^{234}$U/$^{238}$U) has a similar trend to grain size with a correlation coefficient of $R^2=0.57$ (Fig. 3a) in the Changjiang sediments. On this account, the abnormally high ($^{234}$U/$^{238}$U) for ZG is primarily due to its much larger mean grain size than the neighboring samples.

For the Taiwan river sediments, the ($^{234}$U/$^{238}$U) generally decreases from the source region to the estuary in the Zhuoshui River, but displays less variation in the Lanyang River (Fig. 2b). The ($^{234}$U/$^{238}$U) distributions in Taiwan rivers are overall consistent with the comminution model. However, the correlation between ($^{234}$U/$^{238}$U) and mean grain size for the Taiwan river sediments is not significant (Fig. 3a).

Moreover, ($^{234}$U/$^{238}$U) ratios in the Changjiang and Taiwan rivers are also related to the elevations of these river basins, in particular for the mountainous or highland regions, e.g. the Zhuoshui River basin. With the decline of the basin elevation, ($^{234}$U/$^{238}$U) ratios in river sediments display clear decreasing trends in Taiwan and upstream Changjiang
region (Fig. 2). Note that the \(^{234}\text{U}/^{238}\text{U}\) of ZG sample is extremely larger than the adjacent samples, which is probably caused by the large differences in mean grain size between these samples. Besides, the ZG sample is from the mountainous Three Gorges region that is likely to produce fresh rock clast grains with high \(^{234}\text{U}/^{238}\text{U}\) values.

A further analysis between sediment \(^{234}\text{U}/^{238}\text{U}\) and the elevations at sampling sites reveals that \(^{234}\text{U}/^{238}\text{U}\) has different distributions in the highland region (>100 m in this study) and lowland region (<100 m) (Fig. 3b), in particular for the Changjiang sediments. This result implies that the basin topography may exert an important impact on sediment \(^{234}\text{U}/^{238}\text{U}\) ratio and its comminution process. The mountainous/highland region is commonly steep in terrain and characterized by faster transport rate than erosion rate. This kind of region is termed as “weathering-limited” erosion regime (Gilbert, 1877). In a weathering limited region, once a small grain was produced by erosion, it was easily mobilized and transported in the catchment. Therefore, the sediment \(^{234}\text{U}/^{238}\text{U}\) values start to decrease gradually with the decline of elevation in Taiwan and upstream Changjiang region.

By contrast, the \(^{234}\text{U}/^{238}\text{U}\) values obtained from the mid-lower Changjiang sections increased from the middle reach to the estuary. The terrain in the mid-lower Changjiang basin is mainly lowland and/or plain regions after the Three Gorges, with the well development of lakes and flood plains (Fig. 1a). Moreover, this region is primarily located in the subtropical and temperate climate zone, and is prevailed by subtropical monsoon climate. Compared to the upstream region, the weathering regime in the mid-lower Changjiang valley is more “transport-limited” (Bi et al., 2015). The weathering products are less mobilized in the mid-lower Changjiang reaches as a result of basin
trapping, and thus undergo a longer residence time and are subject to stronger chemical weathering under a more favorable monsoon climate (Li and Yang, 2010; Shao et al., 2012). On this basis, the distribution of $^{234}\text{U}/^{238}\text{U}$ and sediment comminution process in the mid-lower Changjiang reaches may be influenced by the chemical weathering.

To verify this hypothesis, the Chemical Index of Alteration (CIA) (Nesbitt and Young, 1982) that is widely used to assess the degree of chemical weathering (Li and Yang, 2010), was calculated and compared with $^{234}\text{U}/^{238}\text{U}$. The higher CIA value reflects a stronger chemical weathering degree. A negative correlation between $^{234}\text{U}/^{238}\text{U}$ and CIA ($R^2=0.76$) exists for the Changjiang River sediments (Fig. 4), which supports our hypothesis that the increasing $^{234}\text{U}/^{238}\text{U}$ from the middle Changjiang reaches (transport-limited region) to the estuary is primarily affected by chemical weathering. It is worth noting that two out of five samples from the upper Changjiang River fall outside the 99% confidence interval of the ($^{234}\text{U}/^{238}\text{U}$) versus CIA plot (Fig. 4), which suggests that the $^{234}\text{U}/^{238}\text{U}$ in the upper reaches of the river are poorly correlated to CIA than in the mid-lower reaches.

The CIA of Taiwan river sediments is as high as that of the Changjiang sediments from the mid-lower reaches, but the $^{234}\text{U}/^{238}\text{U}$ is much higher for Taiwan river sediments (Fig. 4). The comparison and indications of CIA between the Changjiang and Taiwan river sediments are previously discussed by a number of studies (Li and Yang, 2010; Shao et al., 2012; Bi et al., 2015). In this research, no apparent relationship is found between $^{234}\text{U}/^{238}\text{U}$ and CIA for Taiwan sediments, especially in the Zhuoshui River. However, excluding the samples from the mid-lower Changjiang reaches, the upper Changjiang sediments together with the sediments from Taiwan rivers display an
interesting positive correlation between ($^{234}\text{U}/^{238}\text{U}$) and CIA although the correlation coefficient is not high ($R^2=0.36$; Fig. 4). Considering that the upper Changjiang basin and Taiwan catchments can be attributed to the weathering-limited region (Bi et al., 2015), our observations suggest that the ($^{234}\text{U}/^{238}\text{U}$) in river sediments can be greatly influenced by chemical weathering and erosion processes, and yield distinct distributions in weathering-limited and transport-limited regions.

For the transport-limited region (e.g. the mid-lower Changjiang reaches), a longer sediment residence time (indicated by low ($^{234}\text{U}/^{238}\text{U}$) value) tends to induce a strong chemical weathering due to the low sediment mobility. In comparison, for the weathering-limited region (e.g. Taiwan catchments), the sediment may suffer from extreme erosion due to the monsoon climate and high denudation rate (Table 1) but fast sediment transfer. The fast crush (mainly physical erosion) of fresh rocks may accelerate the sediment weathering. As a result, the CIA for Taiwan river sediment is high even in a short sediment residence time (high ($^{234}\text{U}/^{238}\text{U}$) value). However, a recent study by Dou et al. (2016) indicates the high CIA for Taiwan river sediments might reflect the reworking of older altered sediments from terraces and floodplains (i.e. sediment recycling). Taking this into account, the relation between ($^{234}\text{U}/^{238}\text{U}$) and weathering index (like CIA) is complicated as the proxy for weathering may integrate multiple sediment cycles while the sediment residence time recorded by ($^{234}\text{U}/^{238}\text{U}$) is limited to $<1$ Myr. Therefore, the comparison between CIA and ($^{234}\text{U}/^{238}\text{U}$) reveals that the ($^{234}\text{U}/^{238}\text{U}$) may evolve in different ways under different weathering regimes. However, more thorough studies are needed to elucidate the underlying mechanisms.
6.3 Contrasting sediment source-to-sink processes between the Changjiang and Taiwan river catchments

It is noticed that the calculated comminution ages for the Changjiang and Taiwan river sediments are much different, but the large uncertainties associated with the theoretic assumptions and the \( ^{234}\text{U}/^{238}\text{U} \) calculation have to be taken into account. For a preliminary comparison of the sediment source-to-sink processes within a catchment and between different basins, some important information can be obtained according to the distinct comminution ages. The comminution ages for the Changjiang River sediments range from 250 to 600 kyrs, and vary between the upper (~300 kyrs) and mid-lower reaches (~410 kyrs) (Fig. 5a). This difference in comminution age largely depends on different erosion/weathering regime, as well as basin topography. The upper Changjiang basin, in particular the Jinshajiang catchment, drains the eastern portion of the Tibetan Plateau, which is the highest plateau on Earth with fast physical erosion (Chappell et al., 2006; He et al., 2014). Thus, the sediments from the upper Changjiang River have relatively short comminution ages. In contrast, the mid-lower Changjiang River is primarily situated on the Yangtze Craton, which has been relatively stable since the end of the middle Triassic (Jia, 2013). The eastern lowland is characterized by well-developed alluvial plains in the mid-lower valley, which may slow down the transport of sediments (Bi et al., 2015). Furthermore, two large freshwater lakes (Dongting and Poyang Lakes) are located in the middle Changjiang reaches, which may also increase sediment retention in the basin (Dai et al., 2005). As a whole, the sediments in the mid-lower Changjiang basin may have undergone longer and more complex sediment recycling relative to the sediments in the upper catchment, which consequently result in longer comminution ages.
The comminution ages for the Zhuoshui and Lanyang River sediments in Taiwan range from 70 to 200 kyrs, but do not show much topography-dependent due to the large errors in comminution age calculation (Fig. 5b). The comminution ages for the Zhuoshui River sediments generally decrease from the upstream to estuary, with the exception of the sample ZS3 that has the longest comminution age in the Zhuoshui River. The longer comminution age for ZS3 is primarily because of the sediment contribution from its tributary, Qingshui River, which supplies a large volume of sediment sourced from the lowland region. In contrast, the comminution ages for the Lanyang River sediments are more constant with small variations and are somewhat higher than those of the Zhuoshui River. The Lanyang River is located in the orogenic wedge, and it is thus characterized by relatively low erosion rates compared to the Zhuoshui River from the central part of the mountain belt (Siame et al., 2011).

The comminution ages for the Changjiang River sediments (250–600 kyrs) are significantly longer than those for the Zhuoshui and Lanyang River sediments (<200 kyrs) because of different erosion and weathering regimes in variable climatic and tectonic settings. Compared to the relatively slow sediment transfer rate in the large Changjiang River catchment on mainland China, the sedimentary environment in Taiwan is highly dynamic. Here, fluvial transport is fast as active tectonic activity causes high uplift rates and extreme climate events facilitating strong erosion and mass movements (Dadson et al., 2003; Siame et al., 2011; Egholm et al., 2013). Under these conditions, sediments from the Taiwan rivers are easily comminuted from the provenance rocks because frequent earthquakes and landslides mobilize large amounts of sediments from less weathered and deeper saprolite (Dosseto et al., 2014). In addition, the mobilized
sediments are quickly transported to the sea during high rainfall events, especially while frequent typhoons impact the steep topography in Taiwan. These processes will ultimately generate the sediments from Taiwan with younger “comminution ages”.

The different sediment transfer processes for these two river systems can also be seen from the hydrological data. For example, the total flux of suspended solid of the Changjiang River is about 470 Mt·yr⁻¹ based on long-term observations, which is more than 10 times that of the Zhuoshui River (Table 1). However, when river basin area is taken into account, the “sediment yields” (defined as the total sediment flux divided by drainage area) for the Zhuoshui and Lanyang Rivers are estimated to be 400 and 20 times higher than the Changjiang River, respectively.

It is necessary to note that all our comparisons based on the calculated comminution ages bear unexpected large uncertainties due the assumptions inherent in the method. It is difficult, if not impossible, to determine the true comminution age before all the key parameters can be quantitatively derived rather than using the assumptions and theoretical values. Although this study may not substantially contribute to improve the accuracy of comminution age calculation, it offers a systematic attempt of quantifying the time scale of sediment source-to-sink processes in East Asia where the various tectonic and climate settings exist in different catchments. The comparison between the Changjiang and Taiwan river sediment demonstrates that the comminution age cannot be considered as a precise dating method without the robust constraints on key parameters in the calculation. The uncertainties of this method await more studies.

8. Conclusions
The East Asian continental margin is characterized by both large (e.g. Changjiang) and small mountainous river systems. This study investigated the sediment comminution ages of these two river systems using \((^{234}\text{U}/^{238}\text{U})\). The \((^{234}\text{U}/^{238}\text{U})\) ratios range from 0.8544 to 0.9364 for the Changjiang River sediments, and from 0.9299 to 1.0017 for the Zhuoshui and Lanyang River sediments of Taiwan. The \((^{234}\text{U}/^{238}\text{U})\) distributions in the Changjiang basin are overall related to sediment grain size and chemical weathering, while \((^{234}\text{U}/^{238}\text{U})\) values in Taiwan rivers primarily depend on erosion/denudation processes in the catchments. The \((^{234}\text{U}/^{238}\text{U})\) in river sediments may evolve in different ways under variable weathering regimes within a catchment or between different basins.

The calculated comminution ages vary from 250 to 600 kyrs for the Changjiang River sediments and ~110 kyrs for the sediments from Taiwan. These differences are tightly related to contrasting erosion/weathering processes and diverse topography between the large Changjiang basin on the mainland and Taiwan Island. The longer comminution ages for the Changjiang basin are the result of a longer sediment recycling history and complex sediment trapping in the well-developed alluvial plains and lakes in the mid-lower reaches. In contrast, the younger comminution ages for the Taiwan sediments are caused by fast sediment denudation and transport as a result of strong tectonic uplift, typhoon-dominated climate and steep topography.

In summary, our study offers a quantitatively constraint on the time scales of sediment source-to-sink transport processes in East Asia, and provides an important understanding of the mechanism of sediment recycling in two distinct river systems. However, better constraints on key parameters for comminution age calculation are required to make precisely calculation.
Acknowledgments

We thank Heather Handley and an anonymous reviewer for their valuable comments and Francois Chabaux for the editorial handling of the manuscript. We thank James T. Liu and Yuan-Pin Chang for their support in the field to Taiwan Island. We thank Yuexing Feng for the help on isotopic data measurement. This work was supported by the Natural Science Foundation of China (41306040; 41225020, 41376049, U1606401), National Programme on Global Change and Air-Sea Interaction (GASI-GEOGE-03), and China Post-doctor Science Foundation (2015M570384).
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Vigier, N., Bourdon, B. (Eds.), 2011. Constraining rates of chemical and physical erosion using U-series radionuclides. Handbook of Environmental Isotope Geochemistry,


Table 1 Generals of the Changjiang, Zhuoshui and Lanyang Rivers.

<table>
<thead>
<tr>
<th>Rivers</th>
<th>Area</th>
<th>Length</th>
<th>Q$^a$</th>
<th>TSS$^a$</th>
<th>Sediment yield$^b$</th>
<th>Erosion rate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10$^3$ km$^2$</td>
<td>km</td>
<td>km$^3$·yr$^{-1}$</td>
<td>Mt·yr$^{-1}$</td>
<td>t·km$^{-2}$·yr$^{-1}$</td>
<td>mm·yr$^{-1}$c</td>
</tr>
<tr>
<td>Changjiang</td>
<td>1,800</td>
<td>6,300</td>
<td>900</td>
<td>470</td>
<td>300</td>
<td>0.7$^c$ (western)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.03–0.07$^c$ (eastern)</td>
</tr>
<tr>
<td>Zhuoshui</td>
<td>3.1</td>
<td>190</td>
<td>6.1</td>
<td>38</td>
<td>123,000</td>
<td>3–6$^d$</td>
</tr>
<tr>
<td>Lanyang</td>
<td>0.98</td>
<td>73</td>
<td>2.8</td>
<td>6.5</td>
<td>6,600</td>
<td>1–3$^e$</td>
</tr>
</tbody>
</table>

Q: river water discharge; TSS: total suspended solids

b. Sediment yield is simply defined as TSS divided by drainage area.
c. Erosion rates of the mid-lower reaches (eastern) and upper reaches (western) of the Changjiang are from Chappell et al. (2006).
d. Data of erosion rate referred from Dadson et al. (2003).
e. Date of erosion rate referred from Siame et al. (2011).
Table 2 Sample locations, U concentrations, ($^{234}$U/$^{238}$U) activity ratios, mean grain size, $f_a$ and $t_{com}$

<table>
<thead>
<tr>
<th>Sample (Abbr.)</th>
<th>Lat. (°)</th>
<th>Lon. (°)</th>
<th>U (mg/kg)</th>
<th>($^{234}$U/$^{238}$U)</th>
<th>2SD ±2σ</th>
<th>Mean grain size (μm)</th>
<th>$f_a$ (kyr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shigu (SG)</td>
<td>26.87</td>
<td>99.97</td>
<td>2.64</td>
<td>0.9300</td>
<td>0.009</td>
<td>15.77</td>
<td>0.137</td>
</tr>
<tr>
<td>Panzhihua (PZH)</td>
<td>26.61</td>
<td>101.80</td>
<td>6.17</td>
<td>0.9221</td>
<td>0.0013</td>
<td>16.75</td>
<td>0.135</td>
</tr>
<tr>
<td>Luzhou (LZ)</td>
<td>28.90</td>
<td>105.45</td>
<td>3.41</td>
<td>0.9079</td>
<td>0.0010</td>
<td>15.52</td>
<td>0.166</td>
</tr>
<tr>
<td>Wanzhou (WZ)</td>
<td>30.81</td>
<td>108.38</td>
<td>3.10</td>
<td>0.8974</td>
<td>0.0014</td>
<td>12.47</td>
<td>0.167</td>
</tr>
<tr>
<td>Zigui (ZG)</td>
<td>30.85</td>
<td>110.98</td>
<td>2.85</td>
<td>0.9364</td>
<td>0.0011</td>
<td>21.55</td>
<td>0.120</td>
</tr>
<tr>
<td>Jingzhou (JZ)</td>
<td>30.32</td>
<td>112.23</td>
<td>2.41</td>
<td>0.8544</td>
<td>0.0011</td>
<td>3.79</td>
<td>0.285</td>
</tr>
<tr>
<td>Wuhan (WH)</td>
<td>30.55</td>
<td>114.29</td>
<td>3.19</td>
<td>0.8715</td>
<td>0.0009</td>
<td>12.81</td>
<td>0.172</td>
</tr>
<tr>
<td>Jiujiang (JJ)</td>
<td>29.73</td>
<td>115.98</td>
<td>3.23</td>
<td>0.8699</td>
<td>0.0011</td>
<td>14.41</td>
<td>0.210</td>
</tr>
<tr>
<td>Datong (DT)</td>
<td>30.77</td>
<td>117.64</td>
<td>2.97</td>
<td>0.8919</td>
<td>0.0011</td>
<td>13.14</td>
<td>0.168</td>
</tr>
<tr>
<td>Nanjing (NJ)</td>
<td>31.98</td>
<td>118.64</td>
<td>2.81</td>
<td>0.8844</td>
<td>0.0014</td>
<td>15.86</td>
<td>0.164</td>
</tr>
<tr>
<td>Yangzhong (YZ)</td>
<td>32.31</td>
<td>119.75</td>
<td>3.23</td>
<td>0.8808</td>
<td>0.0014</td>
<td>17.19</td>
<td>0.148</td>
</tr>
<tr>
<td>Chongming (CM)</td>
<td>31.36</td>
<td>121.79</td>
<td>3.31</td>
<td>0.9250</td>
<td>0.0013</td>
<td>22.13</td>
<td>0.108</td>
</tr>
<tr>
<td>Lanyang-1 (LY1)</td>
<td>24.71</td>
<td>121.72</td>
<td>3.70</td>
<td>0.9299</td>
<td>0.0012</td>
<td>7.74</td>
<td>0.217</td>
</tr>
<tr>
<td>Lanyang-2 (LY2)</td>
<td>24.64</td>
<td>121.57</td>
<td>3.40</td>
<td>0.9496</td>
<td>0.0016</td>
<td>15.18</td>
<td>0.148</td>
</tr>
<tr>
<td>Lanyang-3 (LY3)</td>
<td>24.43</td>
<td>121.38</td>
<td>5.00</td>
<td>0.9399</td>
<td>0.0014</td>
<td>9.77</td>
<td>0.192</td>
</tr>
<tr>
<td>Zhuoshui-1 (ZS1)</td>
<td>23.97</td>
<td>121.11</td>
<td>4.09</td>
<td>1.0017</td>
<td>0.0017</td>
<td>11.11</td>
<td>0.159</td>
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<tr>
<td>Zhuoshui-2 (ZS2)</td>
<td>23.79</td>
<td>120.91</td>
<td>3.45</td>
<td>0.9697</td>
<td>0.0011</td>
<td>9.41</td>
<td>0.176</td>
</tr>
<tr>
<td>Zhuoshui-3 (ZS3)</td>
<td>23.79</td>
<td>120.65</td>
<td>3.25</td>
<td>0.9507</td>
<td>0.0014</td>
<td>19.67</td>
<td>0.120</td>
</tr>
<tr>
<td>Zhuoshui-4 (ZS4)</td>
<td>23.81</td>
<td>120.47</td>
<td>3.41</td>
<td>0.9531</td>
<td>0.0016</td>
<td>10.99</td>
<td>0.163</td>
</tr>
<tr>
<td>Zhuoshui-5 (ZS5)</td>
<td>23.82</td>
<td>120.21</td>
<td>3.72</td>
<td>0.9603</td>
<td>0.0009</td>
<td>15.38</td>
<td>0.140</td>
</tr>
</tbody>
</table>

Abbreviation for sample ID is shown in bracket; Lat: latitude; Log: longitude; SD: standard deviation

The comminution age error at the 2σ level was calculated by propagating the error on the measured ($^{234}$U/$^{238}$U) ratio and a 16% uncertainty for $f_a$ follow Dosseto et al. (2010).
Figure captions

Figure 1
Map of East Asia and marginal seas showing the Changjiang River in China mainland (a), and Zhuoshui and Lanyang Rivers in Taiwan (b). Abbreviation for sample names are listed in Table 2.

Figure 2
Distributions of \( ^{234}U/^{238}U \) and mean grain size in the river sediments and basin elevations for the Changjiang River (a) and Zhuoshui and Lanyang rivers in Taiwan (b).

Figure 3
Plots show the correlations of \( ^{234}U/^{238}U \) activity ratio with mean grain size and (a) and elevations at sample locations (b).

Figure 4
A plot of CIA versus \( ^{234}U/^{238}U \) in the river sediments. Red solid line indicates the linear regression for the Changjiang sediments, with 99% confidence interval shown in pink shade area. Blue solid line indicates the linear regression for the Taiwan sediments and upper-Changjiang sediments, with 99% confidence interval shown in grey dash lines.

Figure 5
Downstream distributions of comminution ages of sediments along the Changjiang (a) and Taiwan rivers (b). Topographic elevation for the Changjiang basin and Taiwan Island are also shown in grey solid lines. Note that the axis of comminution age is shown in a reverse order for a better comparison.
Appendix table

Click here to download Background dataset for online publication only: Appendix Table_Grain size distribution.xlsx
Fig. 1

Map of East Asia showing topographical features and rivers. The map highlights regions such as the Tibetan Plateau, the Changjiang River, and the East China Sea. Key geographical locations include Tibet, China Mainland, and Japan. The map also indicates the distribution of elevations and the directions of rivers and seas.
Fig. 2

(a) Elevation (km) vs. Longitude (°) showing the Three Gorges area with different reaches labeled: Jinshajiang reaches, Upper reaches, and Middle and Lower reaches. The mean grain size and (\(^{234}\)U/\(^{238}\)U) ratio are indicated by different markers.

(b) Elevation (km) vs. Longitude (°) for the Central Range and Zhuoshui, showing the Lanyang area and mean grain size and (\(^{234}\)U/\(^{238}\)U) ratio.
Fig. 3

(a) Plot showing the relationship between mean grain size (µm) and the ratio of $^{234}$U/$^{238}$U, with a linear regression line $y=0.0043x+0.83$ (R²=0.57).

(b) Graph comparing the relationship between elevation (m) and the ratio of $^{234}$U/$^{238}$U, with different categories labeled as "Transport-limited" (<100m lowland) and "Weathering-limited" (>100m highland).
Fig. 4

\[ y = -4.28x + 1.18 \ (R^2 = 0.76) \]

\[ y = 2.74x + 0.75 \ (R^2 = 0.36) \]
Fig. 5

(a) Changjiang

(b) Central Range