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# Riverine Sediment Geochemistry as Provenance Fingerprints along the Eastern Coast of China: Constraint, Approach, and Application

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**Abstract:** Sediment geochemistry is affected by sediment granularity and chemical partition, which may greatly influence the accuracy of sediment source identification. In this study, we analyzed Ca, Co, Zr, V, Cr, Ti, Sc, Th, and Al in the sediments of major rivers and a Holocene core along the eastern coast of China to reveal the constraints on sediment geochemistry related to granularity and chemical partition and to try and identify the sources of the sediments present in the core. The results reveal that the element concentrations have a significant positive correlation with Al concentration in all these riverine sediments. There are significant differences in the element contents of the bulk sample and the residual sediment leached with acid, including in their ratios. The ratios of Cr/Th–Sc/Th, which is often used for provenance discrimination, reveal that uncertainty of provenance discrimination will increase if the impact of sediment granularity and chemical phase on the index system is not considered. We applied this geochemical approach for provenance on a Holocene core of the East China Sea using Cr/Th–Al, Sc/Th–Al, Ti/Zr–Al, and Cr/Th–Sc/Th with the same granularity as the residue sediments. Based on this approach, we identified the core sediments to have had a large component derived from the western Taiwanese rivers since the mid-Holocene. This study sheds light on the sediment geochemistry used to identify the provenance of marginal seas with multiple rivers entering them.

**Keywords:** provenance discrimination; grain size; acid-leached fraction; Yangtze River; western Taiwanese rivers; inner-shelf mud wedge of the East China Sea

## 1. Introduction

Sediment provenance is the core scientific issue of marine geology and paleoenvironmental research in the marginal seas of China as it is closely related to physical–chemical processes, the sedimentary system, and the geomorphological evolution of the continental shelf [1–3]. Sediment geochemistry is often used to trace the source of marine sediment due to its inheritance of distinct geochemical signatures, mostly from the source rocks of the river catchments [4–7]. However, the geochemical composition of riverine and marine sediments is controlled by many complex factors besides the mineralogical compositions of the source rocks in the catchment, including weathering history, hydrodynamic sorting, postdepositional diagenesis, anthropogenic inputs, and biogenic processes during transportation and deposition [8–10]. The changes in geochemical composition caused by these processes are ultimately manifested by the sediment grain size and chemical phase. Therefore, using

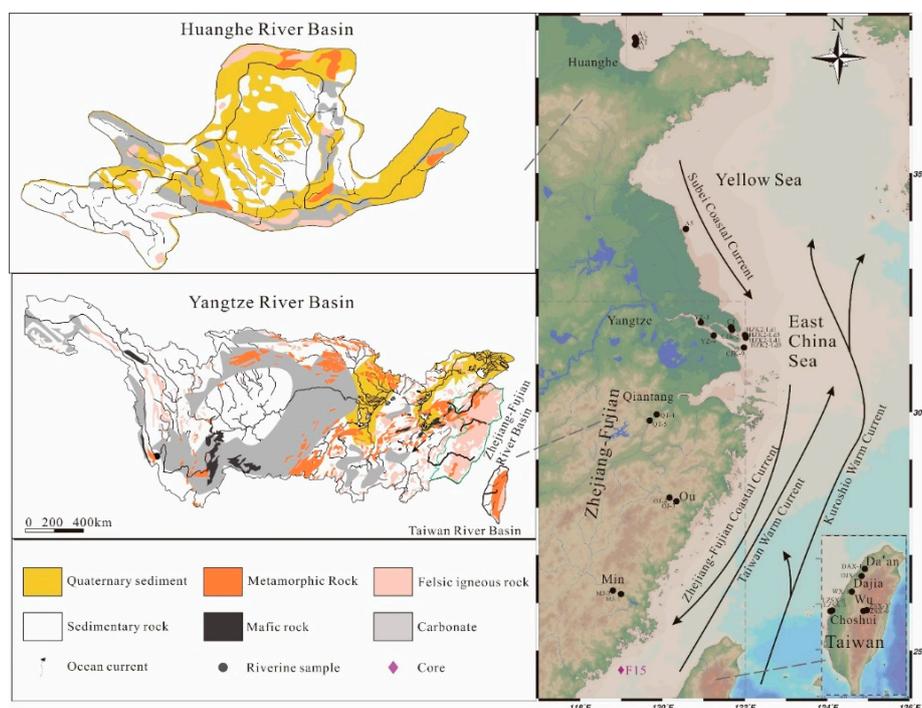
geochemical indicators to trace provenance often leads to uncertainties or an inaccurate identification of the source if the abovementioned factors are not fully considered [11–13].

Many studies have used the geochemical analogy of modern riverine sediment to identify sediment sources of the cores along the eastern coast of China [7,14–22]. The key to provenance discrimination is to set up an analogy of the geochemical fingerprints using potential riverine sediments. However, few studies cover sediment geochemistry of all the rivers involved due to the number of rivers entering the East China marginal seas. Many articles have compared the geochemical characteristics between some of these rivers, including large rivers such as the Huanghe and Yangtze as well as small mountainous rivers from the Zhejiang–Fujian coast and also Taiwan, where most rivers are large rivers [5,23–27]. Of note, the granularity and chemical phase of sediments used for geochemistry are not uniform in these studies. Many studies have used the geochemical analogy of modern riverine sediment to identify sediment sources of the cores along the eastern coast of China [7,14–22]. However, recent studies have revealed the heterogeneity of geochemical composition caused by grain size related to hydrodynamic sorting and chemical phases in the riverine sediments [12,23,27–31]. Therefore, it is necessary to review the impact of sediment grain size and chemical phase on the geochemistry of the rivers along the eastern coast of China, which will help to better understand their sediment sources.

The present study aims to systematically tease out the geochemical characteristics of the major rivers along the eastern coast of China, which are constrained by grain size and chemical partition, and improve the geochemical approach for provenance discrimination. The approach is further applied to identify the source of the sediment in a Holocene core from the inner shelf of the East China Sea.

## 2. Study Area

There are many rivers flowing into the sea from the eastern coast of China, which provide a great deal of sediment to the marginal seas. Basically, these rivers can be classified into two types: large river systems, such as the Yangtze and Huanghe, and short, mountainous river systems from the Zhejiang–Fujian provenance and Taiwan (Figure 1). Their sediment fluxes vary greatly, and their dispersal in the marginal seas is controlled by the ocean current system (Table 1).



**Figure 1.** Geological background and sampling locations. The base map is derived from Ocean Data view. The current system is modified from [32]. The geology maps of drainage basins are sourced from [33].

**Table 1.** Hydrological characteristics of major rivers along the eastern coast of China.

River	Length (km)	Area (10 <sup>3</sup> km <sup>2</sup> )	Water Discharge (10 <sup>8</sup> m <sup>3</sup> /Year)	Sediment Load (10 <sup>4</sup> t/Year)
Huanghe [34]	5464	682	336	97,800
Yangtze1 [34]	6380	1800	8931	36,800
Qiantang [34]	688	24	291	670
Oujiang [34]	388	18	221	289
Minjiang [34]	577	56	573	599
Choshui [35]	186	3.2	61	6387
Dajiayi [35]	140	1.2	26	403
Daanxi [35]	98	0.8	12	497
Wuxi [35]	117	2.0	37.3	679

The Chinese marginal seas are mostly fed with sediment from the Huanghe and Yangtze, although the Yangtze input has declined to 150 Mt/year following the closure of the Three Gorges Dam in 2003 (Table 1; [36,37]). Sediments from these two large rivers as well as those from Zhejiang–Fujian are transported southward by the Chinese Coastal Current (including Subei and Zhejiang–Fujian Coastal Currents) [3,32,38] (Figure 1). Correspondingly, several mud depocenters are formed along the eastern coast of China, such as Huanghe distal mud in the Yellow Sea and a 1000 km long inner-shelf mud wedge of the East China Sea [38,39] (Figure 1). Sediments derived from western Taiwanese rivers disperse northward along the coast, driven by the Taiwan Warm Current [20,40–42] (Figure 1). These rivers, represented by the Choshui River—the largest river in Taiwan—can actually provide 50–200 Mt/year to the Taiwan Strait, although they have much smaller catchments than the Yangtze River [43,44] (Table 1).

### 3. Materials and Methods

A total of 27 surface samples of riverine sediments were taken from the lower reaches and estuaries of 9 rivers: Huanghe, Yangtze, Qiantang, Ou, and Min from the East China mainland and Choshui, Dajia, Daan, and Wuxi from western Taiwan (Figure 1; Supplementary Table S1). In addition, published geochemical data of the lower reaches and estuaries of these rivers (154 samples) were also collected for analysis. All detailed information can be found in Supplementary Table S1.

Core F15 with a length of 2.5 m was obtained in the southernmost part of the inner-shelf mud wedge of the East China Sea at a water depth of 47.1 m (Figure 1). A total of 60 samples were collected at an interval of 4 cm. For details of the chronostratigraphic framework and lithology of this core, see [45].

Geochemical data from previous studies are focused on the samples of the whole or <63 µm fraction of the sediments of these rivers. In this study, we selected sediment fractions of <45 µm and <4 µm for geochemical analysis as a supplement. The two fractions (<45 µm and <4 µm) were extracted by screening with a 45 µm aperture sieve and pipetting following Stokes' law after drying, respectively. Then, enough 1N HCl and 30% H<sub>2</sub>O<sub>2</sub> were used to remove the labile components and organic matter, leaving the residual sediment. After dried at 40 °C, the residue was transferred into digestion tanks for wet digestion, heated to 160 °C with 5 mL of nitric acid, perchloric acid, and hydrofluoric acid. The volume was fixed to 50 mL with 1% nitric acid after the acid was washed out for geochemical testing using an inductively coupled plasma mass spectrometer (ICP-MS) from Thermo Fisher VG-X7 (Thermo Fisher Scientific, Waltham, MA, USA). The relative standard deviation of the test results was below 2% as monitored by international standards of 1ppb Rh and Re and international standard samples of GSD-5, GSD-6, and GSD-9. The pretreatment of samples was completed in the State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai, China, and geochemical testing was carried out in the State Key Laboratory of Marine Geology, Tongji University, Shanghai, China.

The geochemical composition of all the core sediment samples (<45 µm) was analyzed following the above treatment. The grain size distribution of the samples was measured using a laser particle analyzer

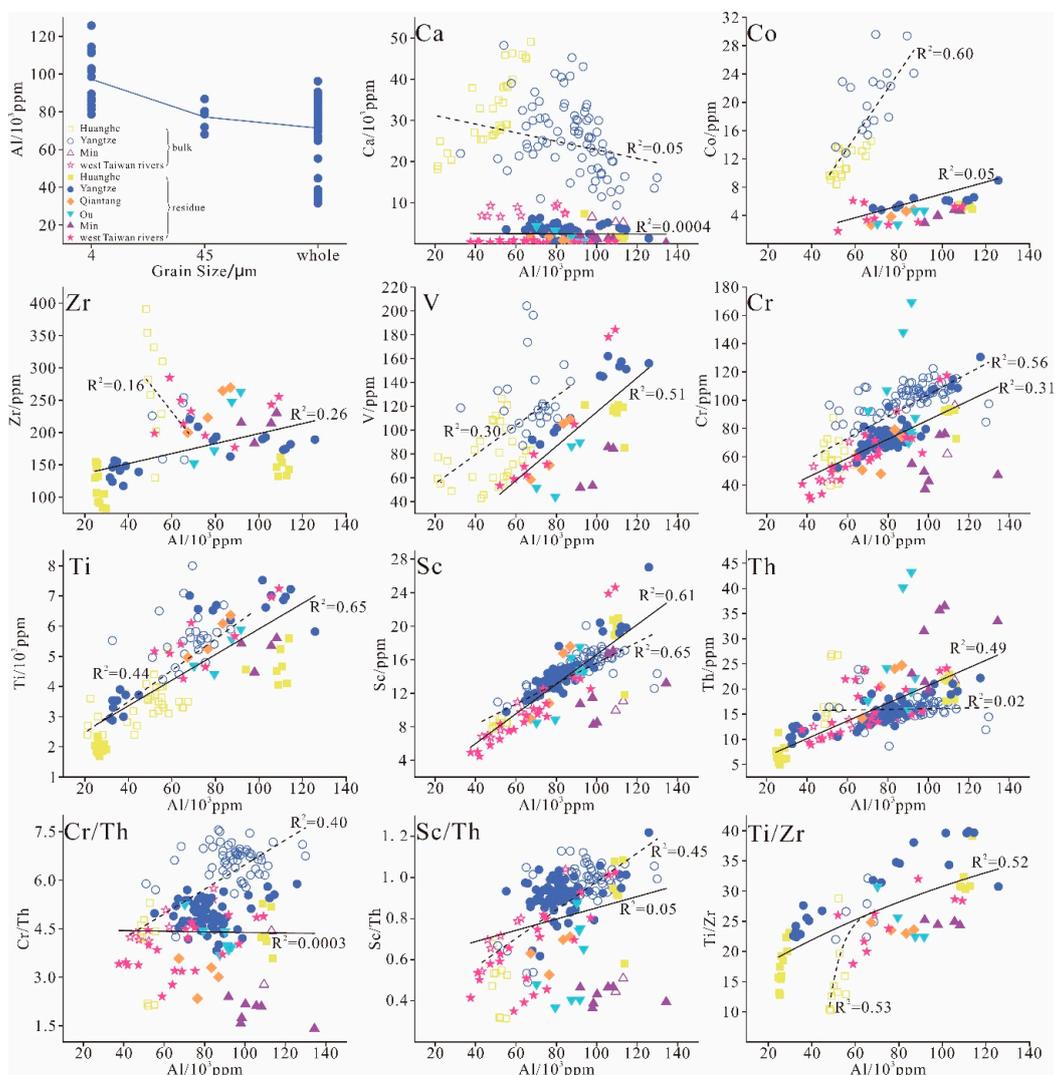
(LS13320) (Beckman Coulter Inc, Brea, CA, USA) with a measurement range of 0.02–2000  $\mu\text{m}$ . Before testing, samples were pretreated with 10%  $\text{H}_2\text{O}_2$  and 1 mol/L HCl to remove organic matter and carbonates. Sodium hexametaphosphate ( $\text{NaPO}_3$ )<sub>6</sub> was used to disaggregate samples via ultrasonic dispersion.

The present study mainly focused on the geochemical elements and elemental ratios that are often used as fingerprints to trace sediment sources, including Al, Ca, Ti, Zr, V, Cr, Co, Sc, Th, and Cr/Th, Sc/Th, and Ti/Zr [23,30,46,47].

## 4. Results

### 4.1. Sediment Geochemistry of the Major Rivers along the Eastern Coast of China

Al is an inert element during the process of weathering and increases with the thinning of sediment particles. It is thus generally used as an indicator of sediment grain size [48,49]. The results show that, in the Yangtze sediments, Al generally decreases with the coarsening of sediment grain size (Figure 2). Therefore, Al is also used as an approximate alternative to grain size to evaluate the sediment geochemistry of major rivers along the eastern coast of China in this study.



**Figure 2.** Geochemical elements and elemental ratios of riverine sediments from the eastern coast of China. The dotted line represents the element trend with Al of the bulk phase, and the solid line represents that of the residual phase.

#### 4.1.1. Geochemical Characteristics Related to Chemical Partition

Ca and Co of riverine sediments from the East China mainland and western Taiwan were found to show a significant constraint in chemical partition in that their contents in the residual phase are much lower than those in the bulk (Figure 2). The difference in Ca between the residue and the bulk is the most significant in the large rivers of Yangtze and Huanghe but is lower in mountainous rivers, such as the western Taiwanese rivers (Figure 2). The differences of Zr, V, and Cr are relatively small, but their contents in the bulk phase are still higher than those in the residual phase (Figure 2). There is little difference in Ti, Sc, and Th between the bulk and the residue, both in large rivers and in mountainous rivers (Figure 2). There are also differences in the elemental ratios of Sc/Th, Cr/Th, and Ti/Zr between the bulk and the residue, especially in Cr/Th (Figure 2).

#### 4.1.2. Geochemical Characteristics Related to Sediment Grain Size

These results show that disparities between the bulk and the residue occur in most of the elements and ratios (Figure 2). This section, therefore, analyzes the grain-size effect of elements based only on the geochemical data of the residual phase.

The relationships between the elements and Al contents were analyzed using the Pearson's correlation method with a bilateral *t*-test (Table 2). Results show that the elements Co, Zr, V, Cr, Ti, Sc, and Th all have a significant positive correlation with Al at a 0.05 level; the only exception is Ca (Table 2), which is also reflected in Figure 2. The correlation coefficient between Co and Al in the Zhejiang–Fujian rivers can reach over 0.6, while the value for Zr and Al is close to 0.7 in the Yangtze River. The other elements—V, Cr, Ti, Sc, and Th—all show a correlation coefficient of above 0.5 with Al in each river. The elemental ratios of Sc/Th, Cr/Th, and Ti/Zr are also positively correlated to Al at the 0.05 level (Table 2). Among them, the correlation coefficients of Sc/Th and Ti/Zr with Al are close to 0.4 and 0.9, respectively, in the Yangtze sediments, while those of Sc/Th, Cr/Th, and Ti/Zr can be over 0.6 in the western Taiwanese rivers (Table 2).

**Table 2.** Correlation between the elements and Al (grain size) in the residual phase of the riverine sediments.

Element \ River	Huanghe	Yangtze	Zhejiang–Fujian	Western Taiwan	All
Ca	-	−0.224	−0.538 *	−0.195	−0.020
Co	-	0.237	0.656 *	0.292	0.229
Zr	0.509 *	0.696 **	0.298	0.055	0.511 **
V	-	0.950 **	0.321	0.963 **	0.712 **
Cr	-	0.848 **	−0.209	0.893 **	0.556 **
Ti	0.965 **	0.719 **	0.264	0.787 *	0.806 **
Sc	-	0.900 **	0.284	0.914 **	0.781 **
Th	0.960 **	0.823 **	0.492 *	0.649 **	0.697 **
Cr/Th	-	0.219	−0.655 **	0.554 **	−0.016
Sc/Th	-	0.358 **	−0.287	0.758 **	0.227 *
Ti/Zr	0.924 **	0.896 **	−0.344	0.669 *	0.722 **

\*\* correlation significant at the 0.01 level (bilateral); \* correlation significant at the 0.05 level (bilateral); - indicates not tested due to insufficient samples.

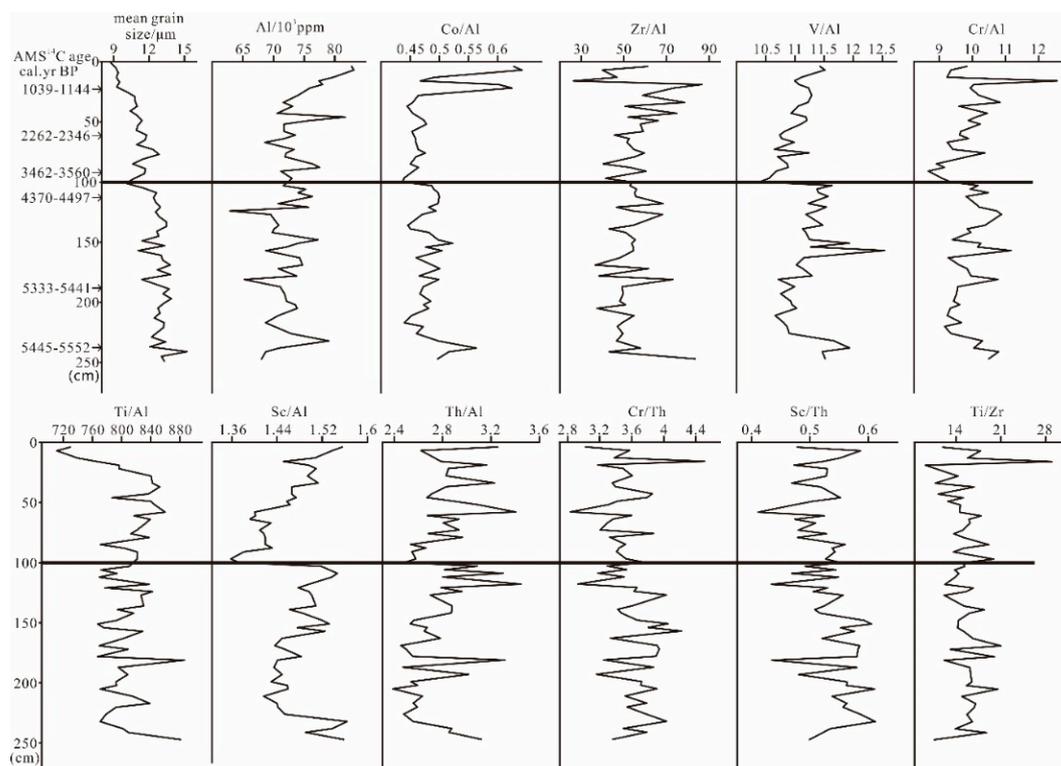
#### 4.1.3. Geochemical Differences of Riverine Sediments along the Eastern Coast of China

Geochemical data of the residual phase reveal that the Yangtze sediment is rich in Ti, V, and Cr, while Zr and Th are abundant in the Zhejiang–Fujian and western Taiwanese rivers (Figure 2). There is relatively little Ti, V, and Cr in the Zhejiang–Fujian rivers. The content of most elements in the Huanghe sediments are generally low, particularly in the very fine sediments (Figure 2). The elemental ratios of Sc/Th, Cr/Th, and Ti/Zr are usually highest in the Yangtze sediments, while they are lowest in the Zhejiang–Fujian and western Taiwanese rivers (Figure 2). It is worth noting that fine-grained (clay)

elements and ratios of the western Taiwanese rivers are very similar to those of the Yangtze sediments, such as Ti, V, Cr, Sc, Sc/Th, and Cr/Th, rather than those of the whole sediments on their own (Figure 2).

#### 4.2. Geochemical Characteristics of Core F15 Sediments in the Southernmost Inner-Shelf Mud Wedge

The geochemical results of the residual phase in core F15 showed that, overall, it is relatively stable upward. However, a small change occurs at a core depth of 100 cm (Figure 3). The contents of V, Cr, Co, and Sc of the upper core sediments are lower than those of the lower core, as are the ratios of Sc/Th and Cr/Th (Figure 3). Zr presents an increasing trend in the upper core, while Ti/Zr changes little over the core (Figure 3).



**Figure 3.** Mean grain size and geochemical characteristics of the residual sediments (<45 µm) in core F15. AMS  $^{14}\text{C}$  age and granularity information are cited from [45].

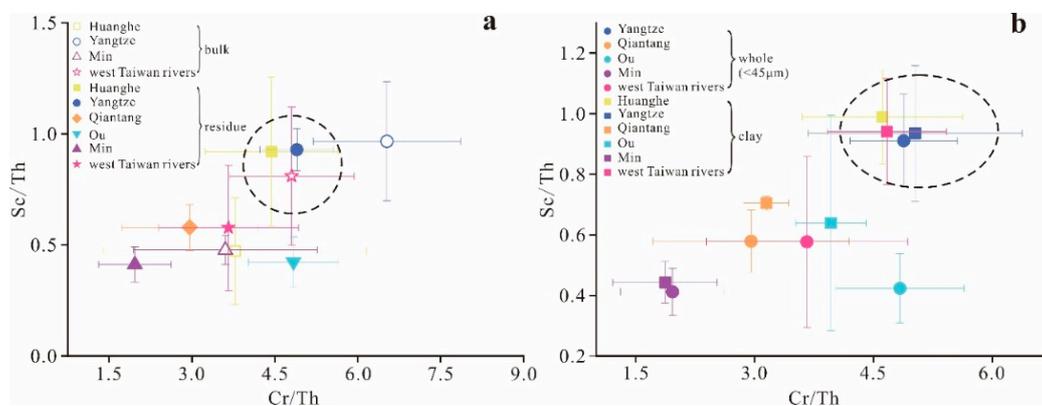
## 5. Discussion

### 5.1. Geochemical Provenance Fingerprint Constrained by Chemical Partition and Sediment Grain Size

The geochemical composition of riverine sediments along the eastern coast of China is largely influenced by the sediment grain size and chemical partition (Figure 2; Table 2). This means that it is necessary to be careful when using geochemical fingerprints to trace sediment sources in the marginal sea.

Except for the elements existing in the lattice of stable minerals, those elements in the adsorbed and oxide-bounded forms in sediments easily migrate during weathering, transportation, and deposition [22,27,50]. The two forms of element occurrence in the bulk sediments can be separated into leached and residual phases using the acid leaching method. The leached fraction contains unstable elements that migrate during natural weathering processes, and the remnant elements from the lattice of stable minerals are concentrated in the residual phase. Significant differences of Ca and Co between the bulk and the residual phase in Figure 2 reveal that these two elements mostly occur in the leached fraction of riverine sediments along the China coast. Ca is usually used as a provenance fingerprint of Huanghe sediments [5,51–53]. Actually, it is highly susceptible to migration

during the hypergene process [12,22,49]. Ca is often enriched in carbonate minerals, such as calcite and dolomite, which are present in large amounts in the Huanghe and Yangtze sediments, causing a large amount of Ca to be stored in the leached fraction [5,54,55]. Co has an ion-exchange index of 0.14 and is easily adsorbed on the surface of clay minerals [56,57]. These are the reasons Co is mostly concentrated in the leached fraction and increases with the fining of sediment grain size in the bulk (Figure 2). When riverine sediments enter the sea, a series of marine environment processes, such as redox, adsorption and desorption, dissolution, and biology, will cause a significant change of the geochemical components in the leached fraction, such as the abovementioned Ca and Co. It is easy for the provenance discrimination of marine sediments to be biased if these elements from the leached fraction or bulk are used, whereas elements such as Ti, Sc, and Th are relatively stable with little disparity between the bulk and the residual phase, whether in large rivers or in short mountainous rivers (Figure 2). These elements are not easily affected by epigenesis and benefit from their high ionic potential, high field strength, and small radius [22,58]. Therefore, the ratios of Sc/Th, Cr/Th, and Ti/Zr are often used to discriminate sediment provenances [4,23,46]. However, the error of provenance discrimination is still present if chemical phases of the elemental ratios are mixed to set up riverine source endmembers, as revealed by Figure 4. Taking the western Taiwanese rivers and Yangtze River as examples, the values of Ti/Zr are almost the same between the Yangtze bulk sediment and the residual phase of western Taiwanese rivers, and the Sc/Th–Cr/Th diagram also reveals the overlapped ranges of the bulk sediment from the western Taiwanese rivers and the Yangtze residual sediment (Figures 2 and 4a). Overall, chemical partition has a great effect on sediment provenance discrimination and cannot be ignored when identifying the sediment sources of China’s marginal seas and adjacent regions using geochemical indexes. It is suggested that the geochemistry of the residual sediments is a better choice for tracing sediment sources because it is inherited mostly from parent rocks in the source region and the simultaneously eliminated maximum disturbance of the marine environmental and biological processes in the sink region.



**Figure 4.** Effects of chemical partition (a) and grain size (b) on sediment provenance discrimination (error bar for  $2\sigma$ ).

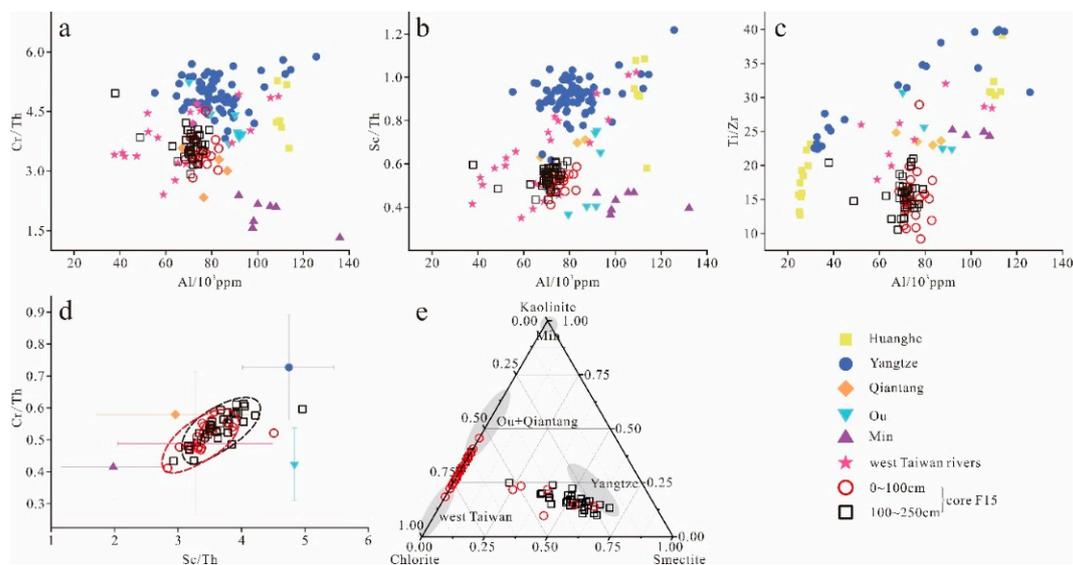
Sediment granularity is closely related to the parent rocks of the source region and hydrodynamics during fluvial/marine transportation and deposition [59]. The differences in sediment geochemical compositions are possibly caused by sediment granularity instead of sediment source in the coastal regions, such as in the Yangtze estuarine sand bar and subaqueous mud delta [49,60–63]. Therefore, strong grain-size effect of elements can increase the uncertainty of sediment source discrimination [48,49]. The present study shows that most elements in the riverine residual sediments from the eastern coast of China have strongly positive correlations with granularity as well as the elemental ratios (Figure 2; Table 2). The Cr/Th–Sc/Th ratios reveal that the impact of elemental grain-size effect on provenance discrimination is not significant in the Yangtze River because the ratios of the whole and clay samples fall in the same range (Figure 4b). In the western Taiwanese rivers, the ratios show a significant

disparity between the whole sediment and clay, and the clay even falls into the range of the Yangtze River (Figure 4b). It can be seen that the grain-size effect of elements should not be ignored for geochemical source discrimination, especially for the marginal seas with multiple river inputs and developed current systems.

### 5.2. Geochemical Disparity of Riverine Sediments along the Eastern Coast of China and the Approach Proposed for Provenance Determination

As mentioned above, the geochemistry of the residual phase of riverine sediments is mainly derived from the parent rocks. Abundant Ti, V, and Cr with mafic origin in the Yangtze residues were possibly inherited from the large basalt province (E' Meishan basalt) of the upper reach [46,54,57] (Figures 1 and 2). Comparatively, Zr and Th are more enriched in the residues of Zhejiang and Fujian rivers as well as in the western Taiwanese rivers (Figure 2). The two elements are often concentrated in the mineral of zircon, which is widely distributed in felsic igneous rocks and sedimentary rocks due to its strong weathering resistance [57]. The East China mainland and Taiwan are dominated by felsic igneous rocks of the Mesozoic and low-grade sedimentary metamorphic rocks of the Himalayan, respectively, causing their riverine sediments to be rich in Zr and Th [64] (Figures 1 and 2). It is worth noting that there is a significant geochemical disparity between the whole sediments and clay of the western Taiwanese rivers (Figures 2 and 4). The geochemistry of the clay fraction is very similar to that of the Yangtze sediments from Sc/Th–Cr/Th and Zhejiang–Fujian riverine sediments from Ti/Zr, suggesting Taiwan's sedimentary metamorphic rocks originated partially from the regenerative continental crust of East China [23,64,65] (Figures 2 and 4).

On the basis of the above discussion, it is suggested that the sediment geochemistry in the residual phase could be used as riverine source tracers, considering the grain-size effect of elements, in which the ratios of Ti/Zr, Sc/Th, and Cr/Th related to the content of Al are recommended as provenance indicators due to their greater stability and discrimination among these rivers. The Cr/Th–Sc/Th diagram is also suggested for provenance identification, provided that the sediment fraction is uniform (Figures 4 and 5).



**Figure 5.** Sediment provenance discrimination of core F15 based on the residual phase using the approaches of Cr/Th–Al (a), Sc/Th–Al (b), Ti/Zr–Al (c), and Cr/Th–Sc/Th (d) with sediment <45  $\mu\text{m}$ . (e) Provenance discrimination from clay minerals of core F15 [45].

### 5.3. Provenance Discrimination of Core F15: An Application of the Geochemical Approach

Core F15 is located in the southernmost point of the inner-shelf mud wedge, which possibly receives sediments transported from both the East China mainland and Taiwan by the Zhejiang–Fujian Coastal Current (ZFCC) and Taiwan Warm Current (TWC) [18,20,41,45]. Previous studies have shown that fine sediments of <45–63  $\mu\text{m}$  could be transported southward by the winter ZFCC [45,66,67]. Therefore, in this study, we have tried to trace the provenance of <45  $\mu\text{m}$  residual sediments using the abovementioned geochemical solution.

All the discrimination diagrams of Ti/Zr–Al, Cr/Th–Al, Sc/Th–Al, and Sc/Th–Cr/Th suggest that the major source of core F15 is from the western Taiwanese rivers rather than the Yangtze River, but not excluding Zhejiang–Fujian rivers, since the mid-Holocene (Figure 5a–d). This is also supported by studies of the modern sediment sources of this region [20]. In addition, the geochemical change at a core depth of 100 cm is not obviously reflected in the discrimination diagrams, suggesting there has been no significant shift of sediment sources (Figures 3 and 5).

Based on the ratios of Sc/Th–Cr/Th, it seems that the sediment source of the lower core (>100 cm core depth) is biased slightly toward the Yangtze endmember, implying that Yangtze-derived sediments contributed more strongly in the lower core section before 4.0 cal kyr BP (Figure 5d). However, the clay minerals of this core in our previous study showed a much clearer signal of Yangtze-derived sediments in the lower core section [45] (Figure 5e). This suggests that clay and silt come from different sources in the core location. Compared with the sediment fraction of <45  $\mu\text{m}$ , clay is more easily transported southward by ZFCC over long distances, which are more sensitive to the ZFCC variation during the Holocene. The source of the clay does not represent the sediment source of the core because it only accounts for less than 10% of the sediment. In comparison, the source of sediments within 45  $\mu\text{m}$  is more representative as it is >60%. Overall, this region received less sediment from the Yangtze River during the mid-Holocene.

## 6. Conclusions

The sediment geochemistry of the Huanghe, Yangtze, Qiantang, Ou, Min, and western Taiwanese rivers along the eastern coast of China is significantly affected by chemical partition and granularity. Most elements and elemental ratios show a notable difference between the bulk and the residual phase, especially Ca, Co, Zr, V, Cr, and Cr/Th. Co, Zr, V, Cr, Ti, Sc, and Th, as well as the elemental ratios, all present a positive correlation with Al, suggesting that the grain-size effect of elements is significant in these riverine sediments. In addition, the ratios of Cr/Th–Sc/Th, which is often used for provenance discrimination, reveal that uncertainty of provenance discrimination will increase if sediment granularity and chemical phase are not uniform in these rivers. Therefore, we suggest that the ratios of Ti/Zr, Cr/Th, and Sc/Th in the residual sediments should be used as provenance discrimination indexes, considering the grain-size effect of elements (content of Al).

Based on the provenance discrimination diagrams of Ti/Zr–Al, Cr/Th–Al, Sc/Th–Al, and Sc/Th–Cr/Th in the residual sediments, the sediment source since the mid-Holocene of core F15 in the southernmost point of the inner-shelf mud wedge is identified as mainly the western Taiwanese rivers rather than the Yangtze River.

**Supplementary Materials:** The following are available online at <http://www.mdpi.com/2075-163X/10/1/29/s1>, Table S1: Geochemical element contents of the main rivers along the East China coast.

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