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Multiple grain-size fraction analysis of heavy minerals and the provenance identification of sediments from the abandoned Huanghe River, eastern China*

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Abstract The quantitative analysis of sediment sources in a sink is an important scientific topic and challenge in provenance research. The characteristics of heavy minerals, combined with the geochemical constituents of detrital grains, provide a reliable provenance-tracing approach. We developed a mineral identification method to analyze the multiple grain-size fraction of sediments, from which the elemental geochemistry of hornblende was used to compare the characteristics of sediments from the Huaihe River and Huanghe (Yellow) River in eastern China. Elements that were statistically identified as being able to discriminate sediment provenance were employed to perform a quantitative analysis of the sources of sediments of the abandoned Huanghe River. Results reveal that the Huaihe River is characterized by a high amphibole content of >60% and that the Huanghe and abandoned Huanghe rivers have greater abundances of limonite and carbonate minerals compared with those of the Huaihe River. The contents of trace elements and rare earth elements in hornblende show that the sediments of the abandoned Huanghe River are similar to those of the Huanghe River but different from those of the Huaihe River. Furthermore, chemical mass balance was used to calculate the relative contributions of different provenances of sediment from the abandoned Huanghe River, and nine trace elements of hornblende were identified as discriminators of provenance. Approximately 2% of the hornblende in the abandoned Huanghe River is derived from the Huaihe River and 98% from the Huanghe River. Considering the proportion of hornblende in the total sediment, it is inferred that the contribution of Huaihe River sediment to the abandoned Huanghe River is approximately 0.5%. This study shows that mineral analysis using multiple grain-size fractions (within the wide range of 1Φ to 6Φ) with assessment in elemental geochemistry of hornblende can characterize the provenance of fluvial material in coastal zones.

Keyword: quantitative provenance analysis; heavy mineral; multiple grain-size fraction; hornblende; elemental geochemistry; fluvial sediment; the abandoned Huanghe River

1 INTRODUCTION

Rivers are the main source of detrital sediments in coastal oceans. Identifying sediment sources is crucial for studying terrigenous clastic-transport processes as well as sedimentary fluxes and environmental changes in marine areas (Park et al., 2000; McKee et al., 2004; Maharana et al., 2018). Sediments from large rivers such as the Huanghe (Yellow) River (YR) and Changjiang (Yangtze) River make an important contribution to terrigenous clastic deposits in the eastern marginal seas of China, although the material supply of small- and medium-sized rivers should also be taken into account (Meng et al., 2000; Saito et al., 2001; Yang et al., 2002; Liu et al., 2008b). Previous studies of the sediment source from the marginal seas in eastern China have generally concluded that the abandoned Huanghe River (AY) sediment as material of the YR (Zhao et al., 1991; Meng et al.,

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2000). However, the AY was formed by the southward flow of the YR through the Huaihe River (HR) channel into the Yellow Sea, especially after AD 1494, when the entire YR flowed into the Yellow Sea through the HR, resulting in the merging of the two rivers (Li, 1991; Han, 1999). Thus, the influence of the HR on the formation of the AY delta cannot be disregarded (Gao et al., 2015), and it is necessary to consider the AY as a distinct sediment source from the YR, particularly for considering the source of sediments on the nearshore shelf in northern Jiangsu, China. In addition, the AY delta is a key sediment source for shelf sediments of the southern Yellow Sea (Liu et al., 2010; Wang et al., 2019a). Thus, quantitative characterization of sediments from the AY is important to understanding the sources of sediment in China's eastern marginal seas.

The analysis of detrital heavy minerals has been successfully used for provenance tracing as well as for determining the source area(s) and properties of sediments (Marcinkowski and Mycielska-Dowgiałło, 2013; Huber et al., 2018; Lu et al., 2019; Jagodziński et al., 2020). The crystalline and physical and chemical properties of clastic minerals in rivers are controlled by two factors: the type of source rock and sorting by water transport; and the influence of the sedimentary environment result in different minerals being enriched in different particle-size fractions (Garzanti et al., 2009). Therefore, traditional approaches to the analysis of sediment sources, which have focused on the fine (2Φ–3Φ) and very fine sand (3Φ–4Φ) fractions, are not as comprehensive as multiple grain-size analyses (1Φ–6Φ) of heavy minerals in sediments (Li et al., 2011; Jin et al., 2019b; Wang et al., 2019b).

The study of genetic mineralogy has shown that the crystal structure and morphology, physical properties, and chemical composition of some minerals in sediments have specific typomorphic characteristics that can indicate the sediment source (Xue, 1991). Of such minerals, hornblende is a major rock-forming mineral of magmatic and metamorphic rocks and is the primary heavy-mineral component of siliceous clasts in mid-latitude rivers and shallow seas in China (Jin et al., 2014). Because of the variations in hornblende composition in different basins, this mineral has been widely used to distinguish the provenances of river sediments in the mixed sediments of the sink area (Lee et al., 2003; Jin et al., 2014; Huber et al., 2018). For example, element ratios such as Zr/Nb, Sc/Hf, Rb/Hf, and Ce/Sm in detrital hornblendes from the Huanghe and Changjiang rivers have revealed significant group differences that can be used to distinguish between different possible sources (Jin et al., 2020). In addition, the elemental composition of a single mineral is unaffected by hydrodynamic sorting and sedimentary environment, and thus enables more precise differentiation of sediment sources compared with multiple minerals (Lee et al., 2003; Yang et al., 2004; Safonova et al., 2010; Wang et al., 2016, 2018).

In the present study, heavy minerals were separated and identified based on a wide range of grain sizes (1Φ to 6Φ) rather than narrower range used in the conventional approach (3Φ to 4Φ) (Jin et al., 2019b; Wang et al., 2019b). The abundances of detrital heavy minerals and the geochemical characteristics of hornblende in sediments from the HR, YR, and AY were analyzed and compared. The major, trace, and rare earth elements (REEs) of hornblende were analyzed to determine mineralogical and elemental indicators of provenance for the HR and YR, and the chemical mass balance (CMB) method was used to quantitatively analyze the provenance of the AY. Our study contributes to the methodology used for quantitative sediment provenance analysis and also provides a basis for the characterization of fluvial materials in sediments of the eastern margin of China.

2 MATERIAL AND METHOD

2.1 Sample collection

Fifteen surface-sediment samples were collected from the HR, YR, and AY. Samples from the YR (YR1-YR6) were collected from the estuarine section of the river in May 2015. Those from the HR (HR1-HR4) were taken from the middle section of the river in March 2016. Samples from the AY (AY1-AY5) were obtained from the lower part of the river in March 2016. These samples were collected from riverbanks, floodplains of the rivers. A handheld Qmini A5 GPS with a horizontal accuracy of 1–3 m was used for positioning, and surficial sediments were dug and extracted using a shovel. Each sampling site was 20 cm×20 cm with a depth of 10 cm. Samples were placed in sealed bags in the field and then taken to the laboratory for analysis. The study area and sampling sites are shown in Fig.1.

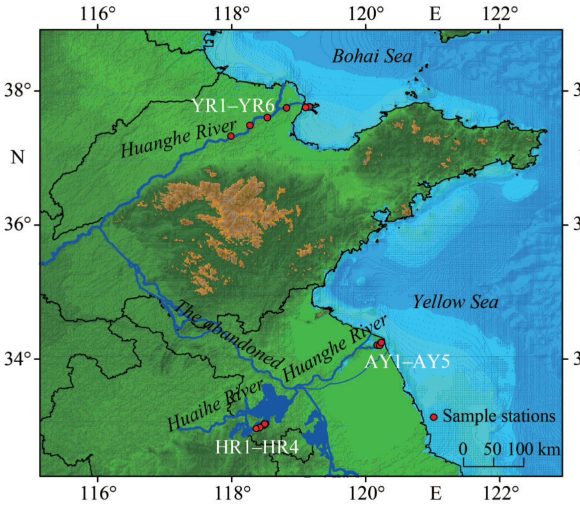

Fig.1 Location of the study area and sampling sites of surficial sediment

2.2 Identification and analysis of minerals in multiple grain-size fractions

Different sieves were used to divide the portion of each sample falling in the size range of 1Φ–5Φ into eight sub-samples (at intervals of 0.5Φ). The 5Φ–6Φ fraction sediment was measured by suspension according to settlement following Stokes's law. Approximately 2 g of each sub-sample was used for the separation of light and heavy minerals. Bromoform ($CHBr_3$, D_4^{20}=2.88–2.89 g/cm^3) was used as the specific-gravity liquid to separate light and heavy minerals at 20±1 °C, with stirring every ~2 min; at 15 min, the mixture was stirred again a total of 3–4 times for the <5Φ fraction. A centrifuge was then used for heavy-mineral separation for the 5Φ–6Φ fraction following the separation of minerals for washing, drying, and weighing (Jin et al., 2019a). Heavy minerals were identified by combining a stereomicroscope and polarizing microscope (Marcinkowski and Mycielska-Dowgiałło, 2013); 300–400 grains were counted for the coarse fraction of ≤4Φ, and 500–800 grains were counted for the fine fraction of >4Φ (Carver and Douglas, 1972). The color, shape, weathering degree, and optical properties of each mineral were described, and the percentage of particles accounted for by each mineral was determined.

Using the percentage of the sediment grain-size fraction and the mass content of heavy minerals as weightings, the sum of the content of each heavy mineral in each grain-size fraction was calculated by weighting as follows (Garzanti et al., 2009; Jin et al., 2019b; Wang et al., 2019b):

$$T_i = \sum\nolimits_{j=1}^{n} Z_j H_j Q_{ij}, \qquad (1)$$

where T_i is the content of heavy mineral (i) in the sample, Z_j is the proportion of particle size (j), Q_{ij} is the percentage of heavy minerals (i) in grain-size fraction (j), and n is the number of grain-size fractions. The aggregate values of the results for each mineral were used as the heavy-mineral statistics for the sample.

2.3 Elemental geochemical analysis of hornblende

An amount of 1 g of detrital heavy minerals of very fine sand size (3Φ–4Φ) was weighed for each sample. Magnetic minerals were separated from non-magnetic minerals with a strong magnet and then placed into diiodomethane (CH_2I_2, D_4^{20}=3.32 g/cm^3) for flotation. The sample suspended in CH_2I_2 was heated with a dilute hydrochloric acid solution (~14%) for 2.5 h to dissolve unwanted minerals such as limonite, magnetite, and hematite. Hornblende was then manually selected under a stereomicroscope to remove rare amphibole, such as tremblite, anthophyllite, and basaltine (Jin et al., 2019a). The amount of hornblende selected from the sediments of each group of samples was a minimum of 50 mg. The purity of hornblende was >95%, and grains containing inclusions or alteration were removed.

Ultrapure water was added dropwise onto the hornblende samples for wetting, following which 1.50 mL of high-purity nitric acid (HNO_3) and 1.50 mL of high-purity hydrofluoric acid (HF) were added. The samples were placed in an oven at 190 °C for 48 h. After cooling, the samples were evaporated to dryness on an electric heating plate; a solution of 1-mL HNO_3 and 1-mL pure water was added, and the samples were placed in an oven at 150 °C to dry for ~8 h (Gao et al., 2010). A 2% HNO_3 solution was added up to 50 g after cooling. Major elements as oxides were determined using inductively coupled plasma-optical emission spectrometry (ICP-OES, ICAP6300) (Gao et al., 2010, 2017). A 10-g sample of the solution to be tested was diluted with deionized water to 20 g to determine trace elements and REEs via ICP-mass spectrometry (ICP-MS, X Series II). The analysis was conducted at the Key Laboratory of Marine Geology and Mineralization, First Institute of Oceanology, Ministry of Natural Resources, according to China's national standard GB/T 20260-2006.

The reagent blank was measured 12 consecutive times, following which the standard deviation was calculated; the concentration value corresponding to 3 times the standard deviation was used as the limit of detection of the method, determined as 0.1–10 ng/g. According to the experimental method, the samples were digested 3 times independently, and the contents of 50 major and trace elements of hornblende were determined; the average value of the three determinations was taken as the final determination value, and the relative error was less than 5%.

The chemical index of alteration (CIA) is an important index for measuring the intensity of chemical weathering and is calculated via the molar proportions of Al_2O_3, CaO^*, Na_2O, and K_2O as follows (Nesbitt and Young, 1982):

$$CIA=[(Al_2O_3/(Al_2O_3+CaO^*+Na_2O+K_2O)]\times 100, \quad (2)$$

where CaO^* represents the amount of CaO in the silicate component. If $CaO<Na_2O$, then CaO is used. If $CaO>Na_2O$, then the mole fraction of Na_2O is considered the mole fraction of CaO^*. $CaO>Na_2O$ in hornblende, so the mole fraction of Na_2O is taken as the value of CaO^*. The higher the CIA, the stronger the degree of chemical weathering.

To compare the differences in the elemental composition of hornblende between the HR and the AY as well as between the YR and the AY, the relative deviation (RD) of each element in hornblende from different rivers was calculated as follows:

$$RD=2\times|(E_{R1}-E_{R2})|/(E_{R1}+E_{R2})\times 100, \quad (3)$$

where E_{R1} and E_{R2} represent the respective amounts of an element in the two rivers. When RD<15%, the difference between the data is small; when RD=15%–20%, the difference between the data is large; and when RD>20%, there is a significant difference between the data (Morton and Hallsworth, 1999).

3 RESULT

3.1 Size distribution and species of heavy minerals in sediments from the Huaihe, Huanghe, and abandoned Huanghe rivers

3.1.1 Distribution of heavy minerals in different grain-size fractions from the three rivers

The main heavy minerals in the sediments from the HR, YR, and AY vary with grain-size fraction (Fig.2). There are few mineral species in medium sand (1Φ–2Φ) and fine sand (2Φ–3Φ), with the main minerals being hydrobiotite, biotite, muscovite, other mica minerals, and heavily weathered limonite. The latter two occur in sheet or plate shapes and have a relatively low specific gravity. In the very fine sand fraction (3Φ–4Φ), the number of mineral species is greater than in the 1Φ–3Φ fractions. Most of these species are amphibole dominated by hornblende and epidote group minerals dominated by epidote, whereas the content of mica is low or nonexistent. The heavy-mineral compositions of coarse silt (4Φ–5Φ) and medium silt (5Φ–6Φ) differ considerably from those of the sand fractions: contents of stable minerals (e. g., garnet, sphene, zircon, apatite, tourmaline, and rutile) and Fe-metallic minerals (mainly ilmenite, magnetite, slightly weathered granular limonite, and hematite) are higher, along with carbonate and pyroxene minerals.

The main minerals differ between grain-size fractions and between rivers. The coarse-grained fractions of the YR and AY have a high proportion of limonite, whereas the coarse-grained minerals of the HR are dominated by mica. In sediments from the YR, hornblende and epidote are found in medium sand sizes and have a high degree of weathering, and carbonate minerals (e.g., magnesite and siderite) account for varied proportions of the fine grain-size fraction (Fig.2b). Carbonate minerals also appear in samples from the AY but are negligible in samples from the HR (Fig.2a). In addition, according to the heavy mineral quality content curves, in sediments from the HR and YR the main grain-size fraction with a high heavy-mineral content is 4.5Φ–5.5Φ, with highest contents of 6.1% and 2.5%, respectively. The grain-size fraction with the highest heavy-mineral content of the AY is 4Φ–5Φ, at 5.4% (Fig.2c). Because of the high content of limonite in the medium sand fraction (1Φ–2Φ), the weight percentages of heavy minerals in samples from the YR and AY are relatively high in this grain-size fraction. The grain-size frequency curves show that the samples from the HR and YR have the highest contents in the interval of 3.5Φ–4.5Φ, whereas the AY samples are coarser, with peaks at 3Φ–4Φ.

3.1.2 Comparative analysis of heavy-mineral contents in sediments from the Huaihe, Huanghe, and abandoned Huanghe rivers

Here, we compare the contents of the main heavy

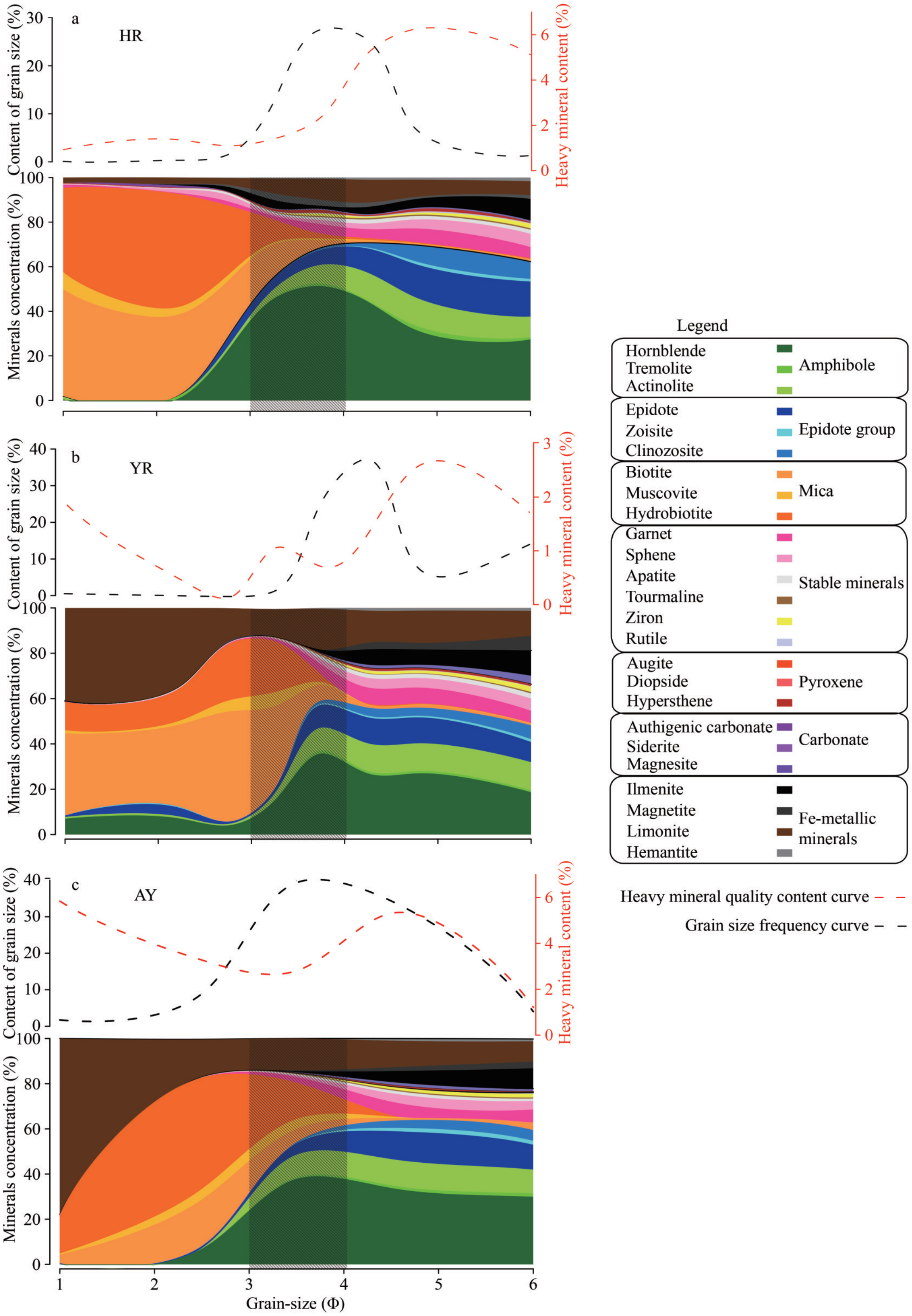

Fig.2 Distribution of the main heavy minerals in different grain-size fractions and average contents in samples from the Huaihe (HR) (a), Huanghe (YR) (b), and abandoned Huanghe (AY) (c) rivers

The shadowed region represents the results of conventional analysis using a narrow range of particle size (3Φ–4Φ).

minerals in sediments from the three studied rivers (Table 1). The HR has the highest heavy-mineral weight content (HMC) (3.30%), followed by the AY (2.21%) and the YR (1.44%), as the average percentage

Table 1 Comparison of the main heavy-mineral contents (as a percentage of all heavy minerals) and indicators of sediment characteristics for samples from the Huaihe (HR), Huanghe (YR), and abandoned Huanghe (AY) rivers

Mineral classification (main minerals)	Mean content (content of main minerals)		
	HR (*n*=4)	YR (*n*=6)	AY (*n*=5)
Amphibole (Hbl) (%)	65.95 (55.60)	37.11 (28.40)	38.02 (27.86)
Epidote group (Ep) (%)	11.90 (9.61)	15.72 (10.58)	7.71 (6.07)
Mica (Bt/Hb) (%)	3.36 (Bt 1.61)	3.82 (Bt 2.47)	23.16 (Hb 15.70)
Pyroxene (Di) (%)	0.31 (0.26)	0.91 (0.87)	0.30 (0.17)
Carbonate (Mgs) (%)	0.11 (0.11)	1.79 (1.76)	0.50 (0.48)
Stable minerals (Grt) (%)	7.21 (3.27)	13.78 (6.43)	5.90 (2.80)
Fe-metallic minerals (Lm) (%)	9.54 (6.77)	24.68 (14.25)	23.04 (19.99)
UM (%)	68.30	39.65	57.89
SM (%)	7.21	13.78	5.90
UM/SM	12.95	2.88	9.81
ZRT	0.66	1.56	0.79
HMC (%)	3.30	1.44	2.21

Hbl: hornblende; Ep: epidote; Bt: biotite; Hb: hydrobiotite; Di: diopside; Mgs: magnesite; Grt: garnet; Lm: limonite; UM: unstable minerals (including hornblende, actinolite, biotite, hydrobiotite, augite, and hypersthene); SM: stable minerals (including garnet, sphene, apatite, tourmaline, zircon, and rutile); ZRT = zircon + rutile + tourmaline; HMC: heavy-mineral weight content.

of the sample weights for each river. The heavy-mineral assemblage of the HR is hornblende+epidote+limonite, that of the YR is hornblende+limonite+epidote, and that of the AY is hornblende+limonite+hydrobiotite. The characteristic mineral of the three rivers is garnet. The amphibole particle content (including hornblende, actinolite, and tremolite) in samples from the HR (65.95%) is much higher than those for the YR (37.11%) and AY (38.02%). Mica contents of samples from the HR and the YR are 3.36% and 3.82%, respectively, and for the AY are as high as 23.16%. Mineral identification and grain-size statistics show that mica is most enriched in fractions coarser than very fine sand and is less common in sediments with silt-sized grains as the main component. The contents of carbonate minerals in sediments from the YR and AY (1.79% and 0.50%, respectively) are higher than those of the HR (0.11%). Limonite contents in samples from the YR and AY (14.25% and 19.99%, respectively) are second only to hornblende. The limonite content in the HR is only 6.77%. These results show that sediment from the YR and AY has inherited some of the characteristics of loess from the Loess Plateau (the most important source area of the YR sediments). In addition, the stable minerals from the YR samples accounted for 13.78% of the total content of heavy minerals, higher than those from the AY (5.90%) and HR (7.21%).

The ratio of unstable minerals to stable minerals (UM/SM) reflects the degree of weathering of clastic sediments and is an indicator of provenance (Wang et al., 2006). Values of UM/SM for heavy minerals from the HR are higher than those from the AY and YR, indicating that the weathering of sediments in the HR is weaker and that unstable minerals are mostly preserved. The zircon-rutile-tourmaline (ZRT) index is the sum of the contents of these three extremely stable minerals. The larger the value, the farther the sediment has been transported and the greater the maturity of heavy minerals (Yang and Li, 1999). The ZRT index of heavy minerals from the YR is 1.56, compared with 0.66 for the HR and 0.79 for the AY, suggesting that minerals from the YR have greater maturity and have been transported farther.

3.2 Elemental geochemical analysis of clastic hornblende from the Huaihe, Huanghe, and abandoned Huanghe rivers

3.2.1 Comparative analysis of major elements

The main major elements (as oxides) of hornblende in the analyzed samples are Al_2O_3, CaO, TFe_2O_3 (total Fe), and MgO, of which the TFe_2O_3 content is the highest, between 15.9% and 16.9% in samples from the three rivers, followed by CaO and MgO (Fig.3). The secondary major elements (as oxides) are K_2O, MnO, Na_2O, TiO_2, and P_2O_5. The contents

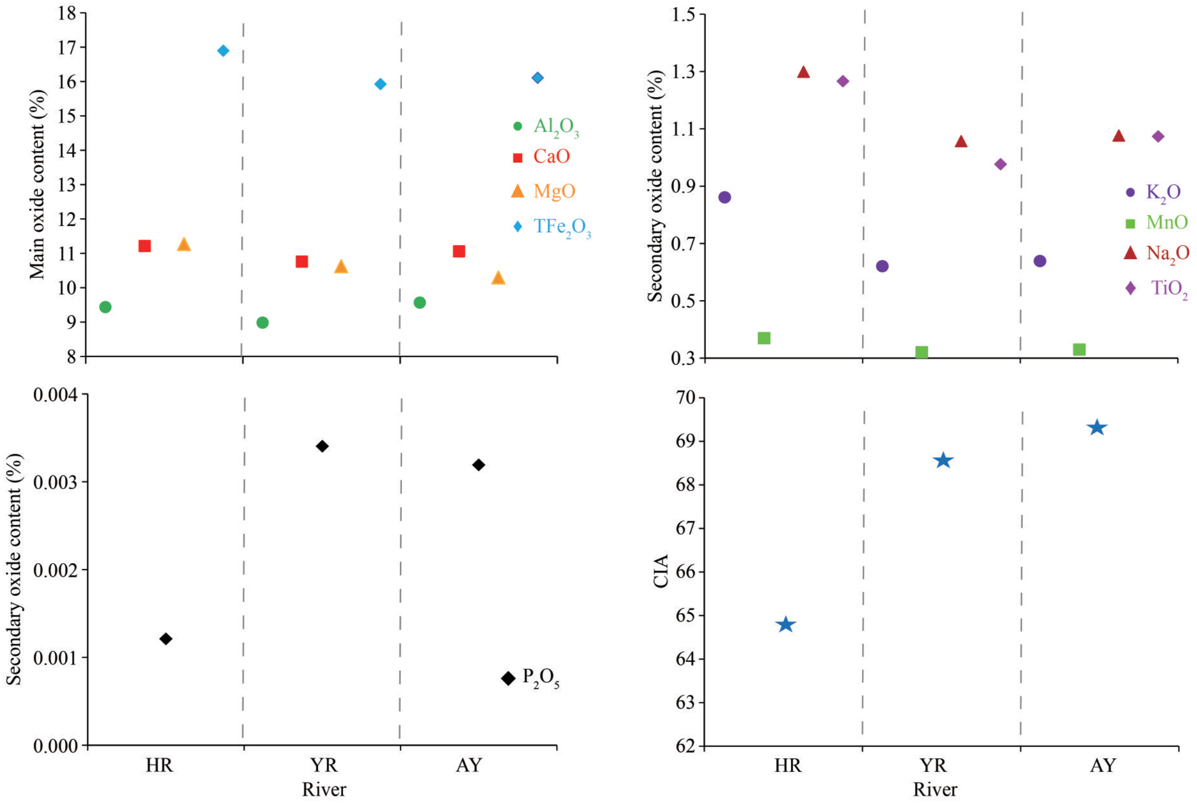

Fig.3 Comparison of major-element contents and chemical alteration index (CIA) of hornblende from sediment samples of the Huaihe (HR), Huanghe (YR), and abandoned Huanghe (AY) rivers

of secondary major elements in hornblende from YR and AR samples are similar to each other and differ from those of HR samples. The contents of K_2O, MnO, Na_2O, and TiO_2 in HR samples are higher than those of samples from the other two rivers, although the P_2O_5 content in HR samples is lower than those in samples from the YR and AR. The average CIA of hornblende is 65 in HR samples and 69 in both YR and AY samples, indicating that hornblende in sediments from the AY and YR is more strongly weathered compared with the HR.

3.2.2 Comparative analysis of trace elements

Trace-element contents in hornblende of sediment samples from the three studied rivers are reported in Table 2. The contents of iron group elements such as V and Cr are high; these elements are mostly substituted into the crystal lattice with Fe and Mg elements in chain-like silicate minerals, so are enriched in hornblende, reaching 10^{-4} in the studied samples (Huber et al., 2018). Zn and Sr are the main trace elements of hornblende, followed by Ba, Zr, Sc, Co, Ni, Cu, Ga, Nb, and Pb. The contents of elements at the 10^{-6} level are Li, Be, Ge, Pb, Hf, and Th; the contents of other elements Cd, CS, Ta, W, Tl, and U are 10^{-7} or below.

The RD values of six elements (Ba, Sr, Be, Cu, Nb, and U) between the HR and the AY are greater than 20% (Table 2), the relative deviations of five elements (Zn, Cd, Cs, Ta, and Tl) lie between 15% and 20%, indicating a large difference between the HR and the AY. The relative deviation values of 22 trace elements for hornblende from the YR and the AY are all less than 15%, indicating hornblende grains from sediment samples of these two rivers have similar chemical characteristics.

3.2.3 Comparative analysis of rare earth elements

Detrital hornblende from sediments from the three rivers, analyzed using ICP-MS, contains light rare earth elements (LREEs) such as La, Ce, Pr, Nd, Sm, Eu, and heavy rare earth elements (HREEs) such as Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y. The highest total REE (ΣREE) content is 318.36×10^{-6} for samples from the HR, compared with 213.84×10^{-6} for samples from the YR and 247.22×10^{-6} for samples from the AY. The mean ΣLREE/ΣHREE value for the HR is 4.38, which is higher than that

Table 2 Mean trace-element contents (10^{-6}) and relative deviation (RD) values for hornblende from sediment samples from the Huaihe (HR), Huanghe (YR), and abandoned Huanghe (AY) rivers

Element	HR_{mean}	AY_{mean}	YR_{mean}	$RD_{HR\&AY}$	$RD_{YR\&AY}$
Ba	82.38	59.13	60.97	32.86	3.08
Sr	100.80	123.55	82.58	20.28	39.75
Zr	61.05	69.08	61.80	12.34	11.13
Li	5.71	6.34	7.28	10.46	13.81
Be	2.03	1.57	1.58	25.71	0.58
Sc	76.77	76.48	75.00	0.38	1.95
V	331.15	351.50	340.44	5.96	3.20
Cr	280.12	275.56	292.11	1.64	5.83
Co	48.68	45.81	46.22	6.09	0.90
Ni	91.28	81.72	88.69	11.04	8.17
Cu	10.33	7.93	6.87	26.37	14.31
Zn	205.78	169.71	182.75	19.22	7.40
Ga	22.82	20.84	20.26	9.06	2.83
Ge	2.87	2.81	2.73	2.32	2.81
Rb	7.97	7.36	6.81	8.02	7.74
Nb	21.04	15.59	15.07	29.79	3.39
Cd	0.31	0.27	0.28	16.24	4.15
Cs	0.19	0.22	0.22	18.31	0.86
Hf	2.74	2.50	2.58	9.25	3.21
Ta	0.84	0.72	0.78	15.69	8.98
W	0.51	0.49	0.43	2.97	13.20
Tl	0.04	0.04	0.04	15.06	4.25
Pb	11.16	10.12	8.85	9.76	13.41
Th	1.04	1.16	0.89	10.77	26.06
U	0.35	0.47	0.39	29.58	18.48

for both the AY (3.99) and YR (3.59), indicating that the HR is more enriched in LREEs than the YR and AY (Table 3).

Chondrite is considered to be comparable with the original material composition of Earth in that its rare earth elements are not fractionated, so a comparison with chondrite data provides a direct indication of the degree of differentiation of REEs in hornblende. Chondrite data for comparison in this study were obtained from Masuda et al. (1973). Chondrite-normalized REE patterns for the analyzed hornblendes from sediment samples of the three rivers are presented in Fig.4 and are characterized by enrichment in LREEs (La-Nd), negative Eu anomalies, and an essentially flat HREE (Ho-Lu) pattern. The REE patterns for the YR and the AY are similar to each other, in agreement with the results of major and trace elements, and the pattern for the AY lies between those for the HR and YR. Moreover, results for the AY hornblende may have been affected by a contribution of hornblende from the HR. Samples

Table 3 Contents of rare earth element in hornblende from sediment samples of the Huaihe (HR), Huanghe (YR), and abandoned Huanghe (AY) rivers (in $\times 10^{-6}$ except for ΣLREE/ΣHREE)

REEs	HR	YR	AY
La	28.48	16.68	22.88
Ce	86.82	52.91	64.73
Pr	13.46	8.35	9.79
Nd	61.03	39.49	44.44
Sm	14.57	10.06	10.97
Eu	3.14	2.03	2.30
Gd	14.23	10.25	11.08
Tb	2.34	1.76	1.89
Dy	12.22	9.52	10.14
Ho	2.65	2.09	2.23
Er	6.97	5.55	5.93
Tm	1.05	0.83	0.90
Yb	6.78	5.32	5.76
Lu	1.02	0.83	0.87
Y	63.61	48.16	53.30
ΣREE	318.36	213.84	247.22
ΣLREE	207.49	129.52	155.11
ΣHREE	47.26	36.15	38.80
ΣLREE/ΣHREE	4.38	3.59	3.99

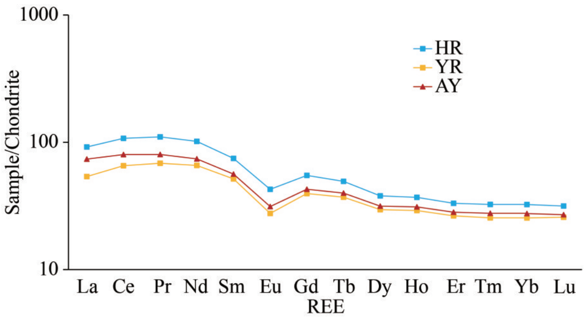

Fig.4 Chondrite-normalized REE patterns for hornblende (sample means) from sediments from the Huaihe (HR), Huanghe (YR), and abandoned Huanghe (AY) rivers

from all three rivers show a negative Eu anomaly, indicating the loss of Eu from hornblende, with the size of the anomaly being in the order of YR>AY>HR.

4 DISCUSSION

4.1 Advantages of the multiple grain-size fraction method

The multiple grain-size fraction method, in which sediments are sifted into multiple grain-size fractions, enables the identification of mineral species and produces content statistics for sub-samples corresponding to the different grain-size fractions. Table 4 provides a comparison of the clastic sediment content and heavy-mineral species for the wide (1Φ–6Φ) and conventional narrow (3Φ–4Φ) ranges of grain size. The mean amount of sediment in the 1Φ–6Φ fraction as a percentage of the total sediment sample varies from 82.42% to 94.88%, compared with 21.92%–37.42% for the 3Φ–4Φ fraction. This very fine sand size fraction (3Φ–4Φ), which encompasses only a small proportion of the overall grain-size distribution, does not represent the characteristics of the entire sediment sample. In comparison, the much wider grain-size fraction (1Φ–6Φ) contains the vast majority of the sediment sample and is therefore representative of the sample overall.

Furthermore, more heavy-mineral species are identified by the multiple-fraction method compared with the single-fraction method. The traditional method of heavy-mineral analysis with a single fraction (3Φ–4Φ) gives results that are suitable for sediment samples dominated by very fine sand. However, for coarser or finer sediments with a small proportion of very fine sand (3Φ–4Φ), or sediments with a full range of grain sizes, this method does not reflect the mineralogical characteristics of the sediment overall.

Table 4 Comparison between the 1Φ–6Φ and 3Φ–4Φ fractions in terms of the sediment content (as a percentage of the total sediment sample) and the number of heavy-mineral species

Rivers	Sediment content (%)		Heavy-mineral species (number)	
	1Φ–6Φ	3Φ–4Φ	1Φ–6Φ	3Φ–4Φ
HR	82.42	21.92	22	17
YR	85.18	26.33	25	20
AY	94.88	37.42	23	16

4.2 Qualitative provenance analysis for sediments of the abandoned Huanghe River

Sediment in the YR is sourced mainly from its middle reaches. The middle reaches of the YR basin account for two-thirds of the total basin area and are dominated by the Loess Plateau, which is covered with 100–200-m thick loess and is the most important source of YR sediments. In addition to loess, the middle reaches also contribute a small amount of sediment from Triassic chalk and clasolite (Fig.5) (Ren and Shi, 1986). The grain size and mineral composition of the YR estuary sediments show a strong loess signal, with high contents of carbonate minerals and limonite. The HR originates from the Tongbai and Funiu mountains, which are a part of the western Dabie Orogen (Ma, 2002). Magmatic rocks of various type are widely distributed in the HR basin, accounting for two-thirds of the exposed area of rocks, and some metamorphic rocks, limestone, and clastic rocks also occur (Compilation Committee of Huaihe Chronicles of Huaihe River, Huaihe Water Resources Commission, Ministry of Water Resources, 2000). The southern tributaries of the HR flow through the high-ultrahigh-pressure metamorphic zone of the eastern Dabie Orogen. Metamorphosed granites are widely exposed along the southern tributaries along with eclogite xenoliths (Compilation Committee of Huaihe Chronicles of Huaihe River, Huaihe Water Resources Commission, Ministry of Water Resources, 2000). The main minerals in eclogite are Mg-Al garnet and omphacite, observed as pink garnets in the sediments of the HR. Holocene loess sediments and alluvium are widely distributed in the northern tributaries of the upper reaches of the HR (Ma, 2002). Sediments of the AY are influenced by the YR, with a small contribution from the HR. Owing to the different basin environments and source rock types of the HR and YR basins, the typical heavy minerals and elemental geochemistry of hornblende from each river show distinct characteristics.

To analyze the similarities and differences between hornblende from the three rivers, we used principal component analysis (PCA). PCA utilizes mathematical dimensionality reduction or feature extraction methods to linearly transform the original numerous and somewhat correlated original variables, extract a smaller number of important but

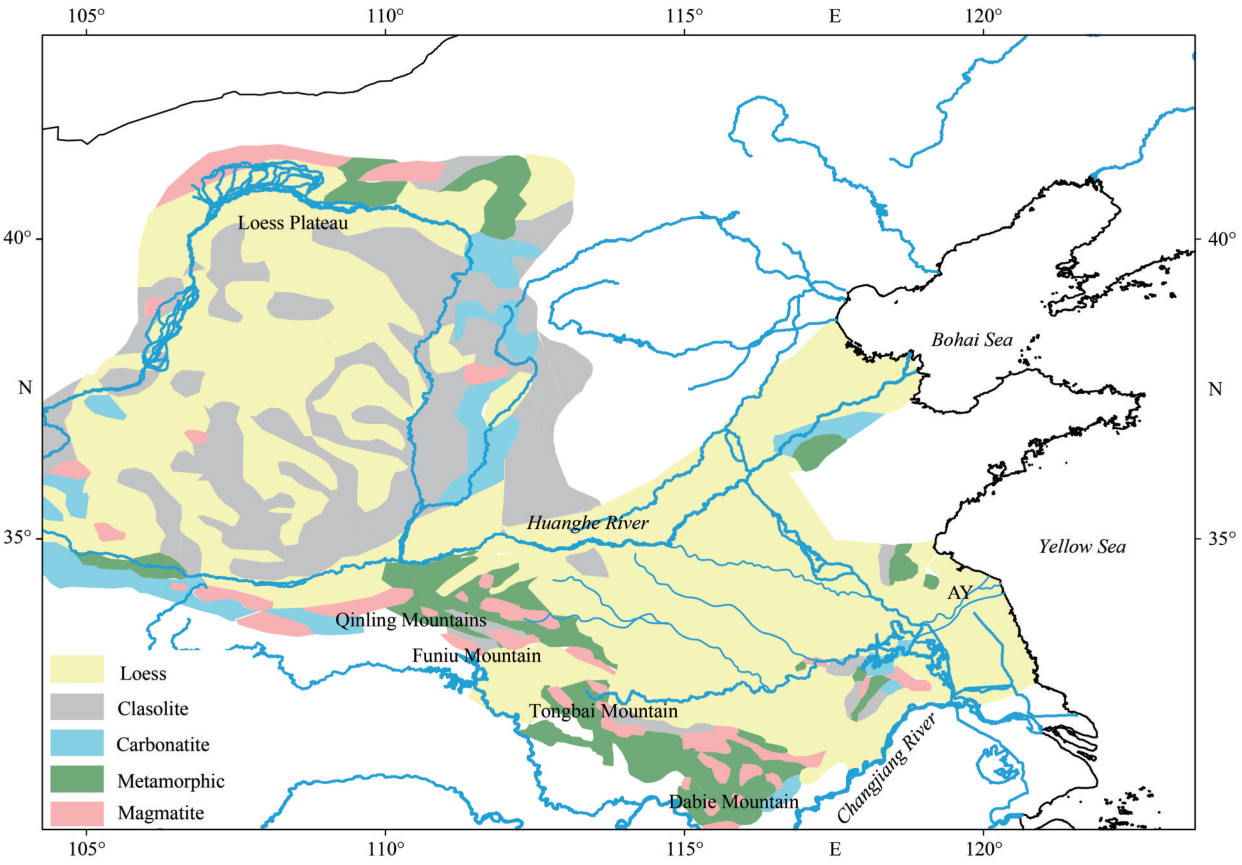

Fig.5 Geological map of the Huaihe, Huanghe, and abandoned Huanghe (AY) rivers

Modified after Ma, 2002.

uncorrelated variables, use a smaller number of representative factors to explain the main information of numerous variables, and provide information on the source of the object (Aitchison, 1986; Montero-Serrano et al., 2010; Von Eynatten et al., 2012; Wang et al., 2020).

The results of PCA showed that the first two principal components (PC1+PC2) of trace elements in hornblende from sediments from the three rivers explain 73% of the total variance in element contents (Fig.6). PC1 accounts for 51.1% of the total variance and has a strong positive loading on Ga, Cd, Pb, and Ge, elements that are characteristic of hornblende from the HR, and on Ba, Zn, Ta, Li, and Cs, which are characteristic of hornblende from the YR. PC2 accounts for 21.9% of the total variance, and its negative loading is correlated with Sc, Cr, and Zr. A plot of the two components shows that the HR samples all fall in the first quadrant and differ significantly from the YR samples, whereas the difference between samples from the YR and AY is small (Fig.6).

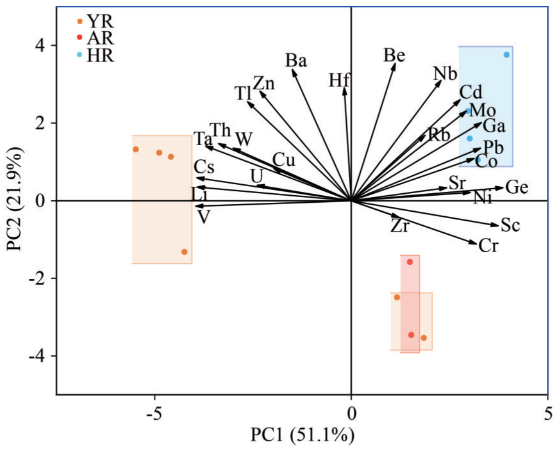

Fig.6 The biplot of trace-element content in hornblende

4.3 Quantitative provenance analysis for sediments from the abandoned Huanghe River

4.3.1 Potential provenance areas and establishment of reliable provenance-tracing indicators

In quantitative provenance analysis, the potential number of provenance areas should be first determined. The HR, which is located between the

Huanghe and the Changjiang River basins, has played an important role in generating the northern Jiangsu Plain. However, the sediment deposited by the HR is difficult to identify because the YR has repeatedly breached and changed its course; the latter has also captured the former, and has discharged into the southern Yellow Sea. The last time of capture of the HR by the YR lasted for more than 700 years (AD 1128–1855) (Compilation Committee of Huaihe Chronicles of Huaihe River, Huaihe Water Resources Commission, Ministry of Water Resources, 2000). After the YR discharged into the Bohai Sea to the north in 1855, the original channel of the YR was termed the AY, and the delta that formed in northern Jiangsu was named the AY Delta (Compilation Committee of Huaihe Chronicles of Huaihe River, Huaihe Water Resources Commission, Ministry of Water Resources, 2000). Thus, the HR and the YR currently belong to different basins despite once having had close contact because of the AY. The sedimentary materials brought to the AY were sourced mainly from the YR, with some contribution by the HR (Han, 1999). In addition, since the delta and mouth of the YR returned to the Bohai Sea to the north, the AY delta has been eroded as a result of a large proportion of its sediment supply having been cut off. This sediment has now become an important source of material deposited in the coastal area of Jiangsu and the continental shelf of the southern Yellow Sea (Zhang et al., 2014). Therefore, when the AY was formed, there were two primary sources of material supply: the YR and HR, as well as small local rivers.

As a second aspect of quantitative provenance analysis, it is necessary to establish reliable and effective indicators or discriminators of provenance. In the multiple grain-size fraction method, the heavy-mineral content in each grain-size fraction can be obtained. In addition, the elemental content of mineral species (e.g., hornblende) can be determined. Both the heavy-mineral content and the elemental content of hornblende can give useful information for discriminating sediment provenance.

Our calculations show that hornblende accounts for the largest proportion of heavy minerals in mid-latitude fluvial clastic sediments from eastern China, with weighted mean values of 55.60% in the HR, and 28.40% and 27.86% in the YR and AY, respectively (Table 1). Isomorphism of hornblende can occur as a result of the distinct geology underlying different river basins, which leads to differences in the trace-element contents of clastic hornblende, with trace elements being stable within this mineral once it is formed (Huber et al., 2018).

Screening of potential provenance discrimination indexes of 25 trace elements in hornblende from the HR and the YR was divided into three steps. The first step was to conduct a multi-independent non-parametric Kruskal-Wallis (K-S) test for elements in hornblende in the YR and HR sources, retaining elements with differences at a level of confidence of $P<0.05$. The second step was to calculate the coefficient of variation (CV) to test the degree of internal dispersion of element contents in hornblende; more specifically, the selected indicators have little difference in the same provenance. Elements with a CV of >15% were eliminated. The CVs of trace elements in hornblende from the YR and HR are all less than 10%, indicating low internal dispersion of trace element contents. The third step was to perform relative deviation (RD) analysis, removing those element contents with RD of <15% and retaining those elements with a large content difference (Zhang and Liu, 2016; Gholami et al., 2019; Yang et al., 2020).

Nine trace elements of hornblende in sediment samples from the HR and YR simultaneously met the three conditions of the non-parameter test K-S, CV, and RD (Table 5): Ba, Be, Cu, Zn, Ga, Rb, Nb, Cd, and W. These elements can be used reliably for quantitative analysis of provenance and thus as effective provenance discriminators for sediments of the HR and YR.

Table 5 Provenance-discriminating trace elements of hornblende from the Huanghe (HR) and Huaihe (YR) rivers

Element	K-S	YR-cv	HR-cv	RD
Ba	0.03	6.93	6.34	31.61
Be	0.03	11.82	4.91	35.24
Cu	0.03	5.97	13.46	44.13
Zn	0.03	12.10	2.98	18.91
Ga	0.03	6.53	1.66	15.11
Rb	0.03	8.40	5.91	24.34
Nb	0.03	7.76	6.08	39.23
Cd	0.03	9.76	7.83	21.88
W	0.04	5.73	6.06	18.71

4.3.2 Quantitative analysis of the contributions of the Huanghe and Huaihe rivers to sediment of the abandoned Huanghe River

The content differences of the nine discriminating elements between the provenance and sink sediments were compared to determine the sources of AY sediments. The CMB method was used to quantitatively analyze the sources (Kelley and Nater, 2000; Massoudieh and Kayhanian, 2013). The difference between each discriminating element and application of the law of conservation of mass allows determination of the contribution of different sources by detecting the contents of components (in this case, trace elements of hornblende) in the sink area (Chen et al., 2013; Al-Naiema et al., 2018). Specifically, the content of a component in the sink area is the linear sum of the product of the content of that component in each source and the contribution proportion:

$$C_i=\sum_{j=1}^{J}F_{ij}S_j, \tag{4}$$

where C_i is the content of the discriminating element i in the sediment of the sink area, S_j is the contribution percentage of provenance j to sediment in the sink area, F_{ij} is the average content of the discriminating element i in provenance j, and J is the number of provenances.

Using the discriminating elements presented in Table 5, the least squares method was employed to quantitatively analyze the sources of the AY, and the relative contribution of each source was obtained by converting the sum of the different contributions to 100%. According to our calculations, the contribution of hornblende from the YR to the AY is ~98%, and the contribution of hornblende from the HR to the AY is ~2%.

Hornblende from the HR accounted for ~2% of the AY, so the sediment supply from the HR to the AY was calculated as follows:

$$C_{\mathrm{h}}=\frac{A_{\mathrm{h}}\times S_{\mathrm{h}}}{A_{\mathrm{h}}\times S_{\mathrm{h}}+A_{\mathrm{H}}\times S_{\mathrm{H}}}, \tag{5}$$

where C_{h} is the contribution percentage of hornblende from the HR, A_{h} is the hornblende content in HR sediment, S_{h} is the sediment supply from the HR, A_{H} is the hornblende content in YR sediment, and S_{H} is the sediment supply from the YR.

The relative content of hornblende in each grain-size fraction was weighted according to the particle-size probability of sediments, and the mass contents of heavy minerals were used as weights to calculate the sum of the contents of hornblende in each grain-size fraction and obtain the total content of hornblende within the range of grain sizes of the sediment samples. According to Eq.1, the hornblende content is 0.4% in the YR and 1.6% in the HR. According to Eq.5, the sediment contributions of the HR and YR to the AY are 0.5% and 99.5%, respectively.

According to previous research, the average annual sediment discharge of the YR was 10.5×10^8 t during 1128–1855 AD (Ren, 2006). Although there is a lack of data on the sediment discharge of the HR in the historical period, the climate of the basin and the source of material supply have not changed greatly over this time; therefore, modern observation data can be used as a proxy for historical data, with the modern average annual sediment discharge being 721.8×10^4 t (Hay, 1998; Liu et al., 2008a). The percentage of sediment discharge between the HR and YR is 0.7% for the historical data, which is not significantly different from the percentage of 0.5% determined in this study. Therefore, hornblende content as a percentage of total sediment and hornblende trace-element composition can be used as effective discriminators for provenance analysis.

5 CONCLUSION

Our multiple grain-size fraction analysis of heavy minerals in sediments from three rivers in eastern China reveals that the Huaihe River (HR) is characterized by high contents of amphibole minerals, the Huanghe River (YR) has a high content of Fe-metallic minerals and stable minerals, and the abandoned Huanghe River (AY) has high contents of mica and Fe-metal minerals. The chemical index of alteration of heavy minerals and the ratio of unstable to stable minerals revealed that the weathering degree and maturity of heavy minerals from the YR are higher than those from the HR, with those from the AY being intermediate between the other two rivers.

Elemental geochemical analysis shows that hornblende from the YR and AY is more highly weathered than that from the HR. Trace-element contents of hornblende from the HR and YR differ substantially. The content of REEs in hornblende from the HR is the highest of the three rivers. Although REE contents in hornblende from both the

YR and AY are similar, those from the abandoned Huanghe River are also influenced by material supply from the HR.

Nine trace elements in hornblende were identified statistically as elements able to discriminate sediment provenance. These trace elements in hornblende from the AY sediment reveal a hornblende contribution of 98% from the YR and 2% from the HR. Furthermore, the supply of sediment from the HR to the AY is calculated as 0.5%, and that from the YR as 99.5%. The results show that the geochemical composition of hornblende can be used as a reliable discriminator for quantitative provenance analysis.

6 DATA AVAILABILITY STATEMENT

Data are available on request to the authors.

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