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Key Points:
- Trace elements and Sr-Nd isotopes are used to ascertain sediment provenance for modern Huanghe River in China
- Four major source terranes are recognized and discriminated based on their unique Nd isotopic compositions
- The heterogeneity of Nd isotopes in the river sediments reveals the sediment routing regime under anthropogenic activity

Supporting Information:
Supporting Information may be found in the online version of this article.

Correspondence to:
S. Yang,
syyang@tongji.edu.cn

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Sediment Routing and Anthropogenic Impact in the Huanghe River Catchment, China: An Investigation Using Nd Isotopes of River Sediments

Yulong Guo[1], Chao Li[1], Chenyu Wang[1,2], Juan Xu[1], Chenyang Jin[1], and Shouye Yang[1]

[1]State Key Laboratory of Marine Geology, Tongji University, Shanghai, China, [2]Earth Surface Geochemistry, Helmholtz Centre Potsdam GFZ German Research Centre for Geosciences, Potsdam, Germany

Abstract The Huanghe once had a sediment flux of >1,000 Mt/yr, but this has decreased by ~90% as its river sediment routing systems have undergone dramatic changes influenced by human activities such as dam construction. However, the way in which the sediment geochemistry of the river has responded to the altered sediment routing processes is not well known. This study investigates the sediment source-to-sink routing regime of the Huanghe River using Nd isotope fingerprinting. Four major source terranes, namely the Songpan-Ganzi (SG) Block, Ordos Desert (OD), Chinese Loess Plateau (CLP) and North China Craton (NCC) are recognized according to their distinct Nd isotopes. The gradual downstream decrease in εNd values in sediments of the upper Huanghe indicates a decreasing contribution of material from the SG Block and a corresponding increase contribution of local underlying basement rocks, which is inferred to be related to sediment capture by a cascade of hydroelectric dams. A gradual increase in εNd from Yinchuan to Tongguan suggests an increasing contribution from the CLP under intense erosion. Relatively low εNd in the downstream sediments suggest a contribution from proximal NCC basement, consistent with the shift from deposition to erosion in the lower channel in recent years. The marked heterogeneity in Nd isotopes in the Huanghe sediments corresponds well to sediment source-to-sink processes in response to increasing human impacts. In a setting of global rivers facing strong anthropogenic impacts, the ways in which altered sediment routing systems affect river sediment geochemistry deserve more research attention.

Plain Language Summary The sediment flux of the Huanghe River (Yellow River) once exceeded 1,000 Mt/yr, but has decreased by ~90% over the past 60 years owing mainly to the nature and intensity of human activity, during which river sediment transport processes have also changed. Here, we present Nd isotopic data for sediments from the Huanghe mainstream to investigate sediment provenances and the river sediment routing system under anthropogenic influences. Nd isotopes of potential source terrane materials allow the sediment provenances to be discriminated. The construction of cascade hydroelectric engineering projects in the upper reaches might have caused a marked decrease in sediment contribution from the Songpan–Ganzi Block, as evidenced from the gradual downstream decrease in sediment εNd in the upper reaches. The gradual increase in εNd values from Yinchuan to Tongguan suggests an increasing sediment contribution from the China Loess Plateau by intense erosion. However, relatively low εNd values in the lower reaches suggest an increased contribution of proximal sediment derived from basement of the North China Craton, with a shift from depositional to erosional processes in the lower reaches. The heterogeneity in Nd isotopes in the river sediments reveals sediment routing processes under the impact of anthropogenic activities and interventions.

1. Introduction

Over the last century, most of the world's large rivers in densely populated areas have been strongly influenced by engineering projects such as the construction of hydroelectric dams and reservoirs, and only a few of these rivers remain in their natural free-flowing states. As a result of these anthropogenic activities and interventions, the natural source-to-sink transport processes of river sediments have been profoundly altered (Best, 2019). With respect to the detrital sedimentary provenance analysis, some studies attempt to disengage the short-timescale alterations of sediment mixing process under anthropogenic perturbations. For example, basin wide Sr-Nd isotopes systematics in the Mississippi River show that dam construction can significantly affect the sediment geochemistry via trapping and sorting of the sediment loads (Munoz et al., 2019).

The Huanghe River (Yellow River) in China has long been regarded as a typical large river influenced by intense catchment erosion and dam construction (Milliman et al., 1987). The Huanghe River is globally known for its previously very high sediment flux (>1 billion ton/yr) sourced mainly from the China Loess Plateau (CLP) in the middle reaches of the river; however, its sediment discharge has decreased by ∼90% over the past 60 years (H. Wang et al., 2007; Wang et al., 2015). Many studies have investigated the spatial–temporal changes in water and sediment discharges in response to various human activities in the Huanghe River catchment, using statistics on hydrological and sedimentological budgets (e.g., Chu, 2014; Chu et al., 2006; H. Wang et al., 2007; Wang et al., 2015). Other studies have focused on the Holocene sedimentation in the Huanghe delta and marginal seas to investigate how climate and human activities have influenced river sediment flux (e.g., Best, 2019; Ren, 2015; Wu et al., 2020). However, few studies have investigated the responses of river sediment geochemistry and provenance to changes in sediment transport/routing patterns related to increasing human activities and interventions in river catchments and mainstreams (Hu et al., 2015). We suggest that meticulous provenance analysis of Huanghe sediments has the potential to capture the anthropogenic perturbation signals and further to better understand the human impacts on the sediment dispersal systems of large rivers.

With respect to studies of sediment provenance on the continental margin of East China, sediment of the Huanghe River has been considered as either relatively homogeneous (over a time scale of 10^5–10^6 years) because of the dominant CLP source (Yao et al., 2017; Zhang et al., 2019), or a simple binary mixture of materials from the Loess and Ordos plateaus (for a time scale of 10^3–10^4 year) (Hu et al., 2012). Sediments from the middle–lower Huanghe reaches or its estuary have often been used to assess the average composition of Huanghe sediment discharged into the sea. However, the sediment discharge of the Huanghe River has increased only since 3,000 years ago (Best, 2019; Wu et al., 2020). Prior to that time, with weaker erosion in the CLP, the natural transport processes and chemical composition of Huanghe sediment discharged into the sea were likely much different from those of the present day (Wu et al., 2020). Thus, the geochemical heterogeneity of river sediments and the major provenances of sediment in different parts of the river basin need to be explored further to better constrain the sediment source-to-sink processes of the Huanghe River drainage basin.

The Huanghe River drainage basin covers several major source terranes with different geochemical geneses (Figures 1a and 1b), making it possible to discriminate sediment provenances provided that appropriate provenance indicators are used. In this study, overbank sediment samples along the course of the Huanghe River were collected and measured for trace elements and Sr–Nd isotopic compositions in detrital sediment fractions. We then verified whether these geochemical proxies can be used to identify and discriminate the mixed provenances for sediments carried by the Huanghe River. The overall aims of the study were to determine the heterogeneity of sediment sources in the modern Huanghe River basin and to investigate the relationship between sediment provenance and the sediment routing system in the context of the impacts of human activity in this large river basin.

2. Characteristics and Sediment Flux of the Huanghe River Basin

As the river has the highest total sediment flux among the world rivers before the significant impoundment, the Huanghe River has a length of 5,464 km and a catchment area of 7.5×10^5 km^2. The river has recorded an extremely high sediment load, averaging 16×10^8 t/yr between 1800 and 1970 (Ren, 2015). The Huanghe River originates from the Bayan Har Mountains in the eastern Tibetan Plateau and flows eastward across the Loess Plateau and Ordos region, to the North China Plain, and finally discharges its sediment load into the Bohai Sea (Figure 1).

The erodible CLP covers 44% of the area of the Huanghe River basin (Figure 1b). The river basin is characterized by diverse lithologies, including Archean metamorphic rocks, Paleozoic carbonates, Cenozoic clastic rocks, and Quaternary sediments. The basin has an arid to semi-arid continental climate, being more arid in the upper and middle reaches, and more humid and temperate in the lower reaches.

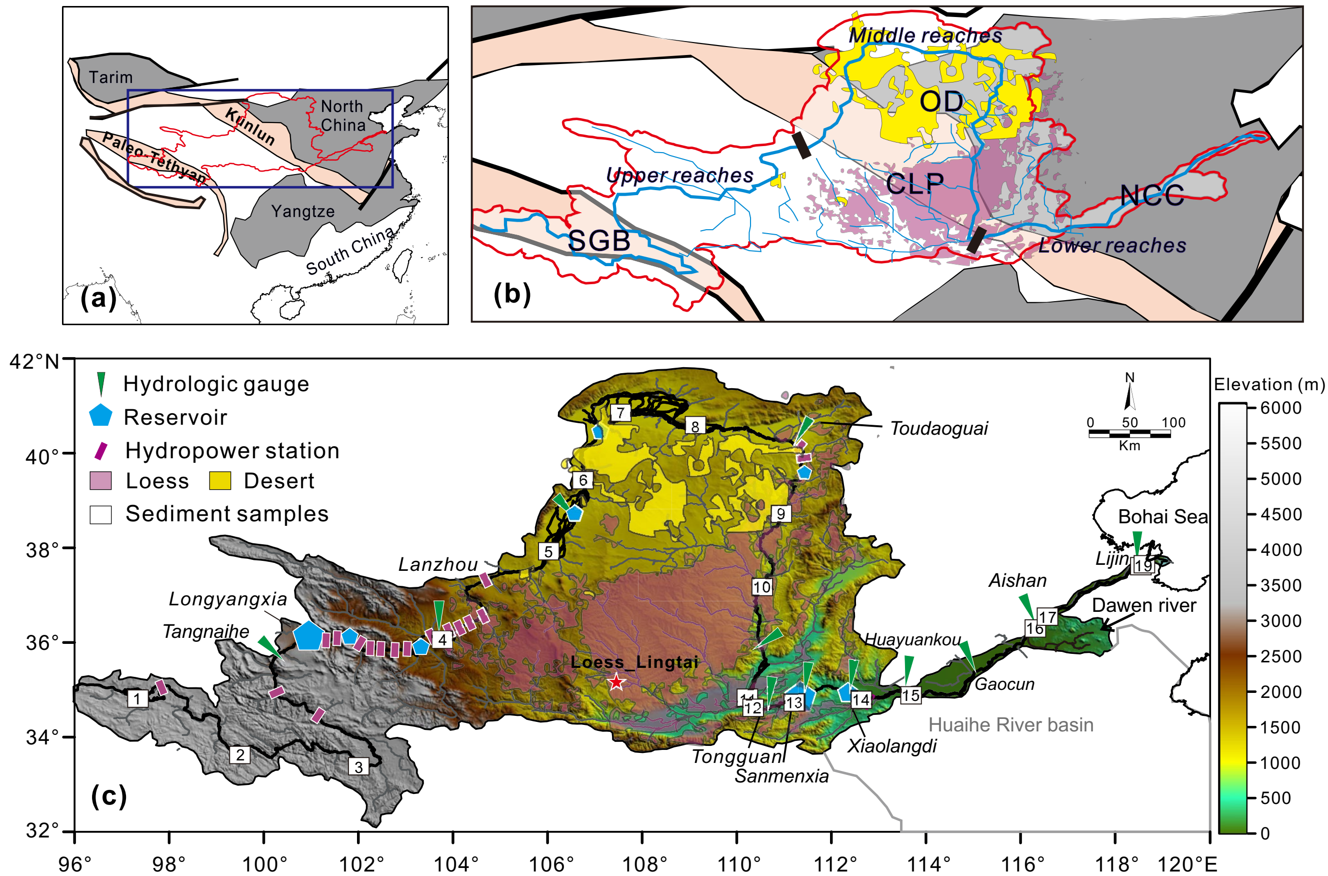

Figure 1. (a) Schematic map showing the major tectonic blocks (in gray) and orogenic belts (in pale pink) in China (after Zhang et al., 2008) and the Huanghe River basin (red lines). (b) Enlargement of the area indicated by the rectangle in (a), showing the four major source terranes of Huanghe River basin sediment, SGB= Songpan-Ganzi Block, OD= Ordos Desert, CLP= Chinese Loess Plateau, NCC= North China Craton (modified from the 1:2500000 Geological Map of China, http://dcc.ngac.org.cn/geologicalData/cn//geologicalData/details/doi/10.23650/data.H.2017.NGA121474.K1.1.1). (c) Map of the study area, showing the mainstream, major tributaries, major hydrologic gauges, sampling localities (numbers 1 to 19), and artificial impoundments. Loess samples were taken from the Lingtai section. Digital mapping data is from NOAA (ETOPO1).

The Huanghe River basin is recognized as the birthplace of ancient Chinese civilization and was the most prosperous region in early Chinese history. Changes in sediment flux estimated by deposition rates in the delta indicate that the anthropogenic impact on the Huanghe dates back to ~3,000 years ago (Wu et al., 2020). Over the last 1,000 years, human activities such as deforestation and agricultural development have caused the sediment flux to increase sharply by an order of magnitude, peaking at 1.6 Gt/yr. during the 1950s (Best, 2019; Wu et al., 2020). In recent 20 years, the sediment flux of the Huanghe River has dropped to the level that existed prior to intense human activity, meaning that the source-to-sink transport processes of Huanghe River sediments have undergone profound changes since the 1950s.

Here, we present sediment fluxes recorded by major hydrologic gauges in the Huanghe River basin for the periods 1950–1999, 2000–2006, and 2009–2016 (Figure 2). The records for 2009–2016 and 2000–2006 reveal a major change in the sediment source-to-sink routing regime of the Huanghe River compared with the record for 1950–1999, which provides a background state for comparison.

For 1950–1999, sediment fluxes display wide spatial variability along the river course, showing a slight increase from Tangnaihe to Toudaoguai in the upper reaches, with extremely high values at Tongguan, Sanmenxia, and Xiaolangdi stations (Figure 2), which reflect extremely high sediment yields from the CLP in the middle reaches. The sediment discharge from Sanmenxia to Lijin decreases gradually, reflecting large amounts of sediment being deposited in the lower river channel. The sediment fluxes at these hydrologic

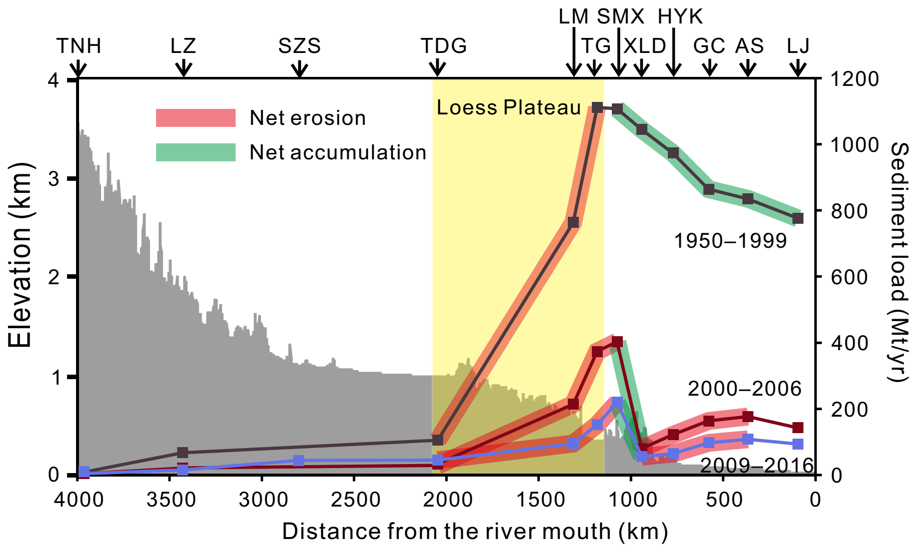

Figure 2. Spatial and temporal variations in sediment discharge of the Huanghe River. From upstream to downstream, major hydrologic gauges are Tangnaihe (TNH), Lanzhou (LZ), Shizuishan (SZS), Toudaoguai (TDG), Longmen (LM), Tongguan (TG), Sanmenxia (SMX), Xiaolangdi (XLD), Huayuankou (HYK), Gaocun (GC), Aishan (AS) and Lijin (LJ). The yellow-shaded area indicates the distribution of the CLP, and the gray area indicates altitude variation along the Huanghe River mainstream. The data of annual sediment discharges at different gauging stations in 1950–1999 and 2000–2006 are from Wang et al. (2010). The data of 2009–2016 are from the annual Yellow River Sediment Bulletin published by the Yellow River Conservancy Commission (YRCC, http://www.yrcc.gov.cn/nishagonggao/).

gauges show a gradual decrease from 1950 to 1999 under the combined effects of climate change and anthropogenic activity and interventions (H. Wang et al., 2007; Wang et al., 2010, 2015).

Sediment fluxes measured at different gauge stations during the periods 2000–2006 and 2009–2016 show similar patterns, but for most stations, sediment discharges during 2009–2016 were lower than those during 2000–2006. Sediment fluxes at major gauge stations in the upper reaches over the last two decades were ~40% lower than those for 1950–1999. From Toudaoguai to Sanmenxia, although the sediment flux increased markedly over 2000–2006 and 2009–2016, it only accounted for less than 30% of the corresponding values for the period 1950–1999. During 2000–2006 and 2009–2016, sediment discharges show a pronounced decrease at Xiaolangdi Reservoir. From Huayuankou to Aishan, sediment discharge gradually increased, suggesting that the lower Huanghe mainstream gained suspended load from channel erosion (Bi et al., 2014; Chu, 2014). This transformation from channel deposition to erosion was caused primarily by a reduction in the concentration of sediment in the Xiaolangdi Reservoir and by activities associated with the Water–Sediment Regulation (WSR) Project (Chu, 2014). Since 2002, annual WSR scheme activities have been conducted to reduce the amount of sedimentation within the Xiaolangdi and Sanmenxia reservoirs and to alleviate the pressing situation of a raised riverbed (Chu, 2014). These anthropogenic interventions have resulted in significant scouring of the downstream channel (Chu, 2014) and have led to changes in the river sediment routing regime in the lower basin.

3. Samples and Methods

The sampling locations of this study are shown in Figure 1. A total of 19 loess samples were taken from the Lingtai section, for which the detailed sampling method has been reported by Zhou et al. (2007). Nineteen fine-grained sediment samples were collected from the floodplain surface close to the Huanghe mainstream in both 2009 and 2016 (Table A1). The sampling locations were selected to avoid potential contamination from industrial areas, roads, ports, and residential areas.

Surface floodplain sediments comprise fine-grained suspended materials as well as coarser particles lifted from the riverbed by the high energy of water during flooding (Bølviken et al., 2004; Singh & Rajamani, 2001). A high proportion of the total sediment load in the Huanghe is transported as suspended load during floods, involving rapid morphological changes with sedimentation and erosion in the main channel and persistent sedimentation on the floodplain (Zhang et al., 2017). The geomorphology of this channel–floodplain system is sensitive to changes in sediment deposition and erosion processes (Yu, 2006; Zhang et al., 2017). The sediment source-to-sink routing regime of the Huanghe River during the sampling years (2009 and 2016) was relatively stable (Figure 2). Replicate samples were collected at Tongguan and Lijin to test the representativeness of the samples.

Previous studies have shown that the coarse- and fine-sized fractions of the loess and desert samples taken from northern China may have different provenances and transport processes, whereby the coarse fractions saltate or roll from proximal sources, whereas the fine fractions are transported suspended in the air from more distant upwind source areas (Rao et al., 2008). To reduce this grain size bias, the <50 μm fine fraction was used in this work and was separated by wet sieving. The results show that >91 wt.% of the loess and floodplain samples is composed of grain sizes of <50 μm. Moreover, given that the modal grain size interval of typical loess samples is 16–32 μm (Sun, 2004), the <50 μm fraction represents the main body of bulk aeolian sediment from the CLP and of sediments from the middle and lower reaches of the Huanghe River.

As important reservoirs for trace elements such as rare earth elements (REEs), the authigenic fractions of sediments (e.g., Fe–Mn oxyhydroxide fractions and organic matter) may have distinct REE patterns and Nd isotopic signatures that can contaminate the provenance signal carried by the detrital silicate fraction (Bayon et al., 2004; Freslon et al., 2014). To remove the non-detrital components but with minimal dissolution of detrital materials, the <50 μm sediment was treated following the procedure of Dosseto et al. (2010). The powdered samples were ignited at 550°C overnight (~8 h) to eliminate organic matter, and then leached with 1.5 N HCl for 6 h to remove the organic-bound matter, the exchangeable fraction, and carbonates. The residues were finally leached using 0.04 M NH_2OH–HCl at pH = 2 for 5 h to dissolve Fe–Mn oxides before measurements of trace element and Nd isotopic compositions. Approximately 0.1 g of each pretreated sample was digested using ultra-pure distilled HF + HNO_3 acid mix in high-pressure Teflon bombs at 190°C.

The contents of trace elements were determined by inductively coupled plasma–mass spectrometry (ICP–MS) using an Agilent 7,900 instrument at the State Key Laboratory of Marine Geology, Tongji University, Shanghai, China. The analytical precision and accuracy were monitored using the geostandards BCR-2 and GSD-9, yielding an analytical uncertainty of better than 5% (Table A1). The Sr and Nd isotopic compositions of loess samples and floodplain sediments sampled in 2009 were analyzed at the Radiogenic Isotope Facility (RIF) at the University of Queensland, Brisbane, Australia, using a Nu Plasma multicollector (MC)–ICP–MS instrument, and floodplain sediments sampled in 2016 were analyzed at the State Key Laboratory of Marine Geology at Tongji University using a Neptune Plus MC–ICP–MS instrument. Nd isotopic data in this study are expressed as εNd, calculated as $[(^{143}Nd/^{144}Nd)_{measured}/(^{143}Nd/^{144}Nd)_{CHUR} - 1] \times 10^4$, with the CHUR (Chondritic Uniform Reservoir) $^{143}Nd/^{144}Nd$ value of 0.512638 (Jacobsen & Wasserburg, 1980). The analytical precision and accuracy for Sr-Nd isotopes were monitored by the standards BCR-2 and BHVO-2 (Table A1). To constrain the average Nd isotopic composition of potential sources, this study also collected the available literature data for Nd isotopes of various source rocks and sediments in the Huanghe catchment (Table A3).

The grain size for the <50 μm fraction of each sample was measured using a Beckman Coulter LS230 particle size analyzer at the State Key Laboratory of Marine Geology, Tongji University. Repeated testing showed that the uncertainty on grain-size determinations is less than 3%.

4. Results

4.1. Trace Element Chemistry

Measured trace-element contents of the sampled Huanghe sediments were normalized with respect to the average composition of upper continental crust (UCC; Rudnick & Gao, 2003) (Figure 3a). The trace-element compositions of Huanghe river sediments are similar to those of the loess samples but with greater variation among samples. The relative standard deviation for contents of different elements is 16.6% for the Huanghe sediments and 7.1% for the loess samples. The Huanghe river and loess sediments are generally depleted in most trace elements relative to UCC, especially in Sr and Co. No obvious fractionation of REEs is observed in either the river or loess sediments, with all showing flat patterns.

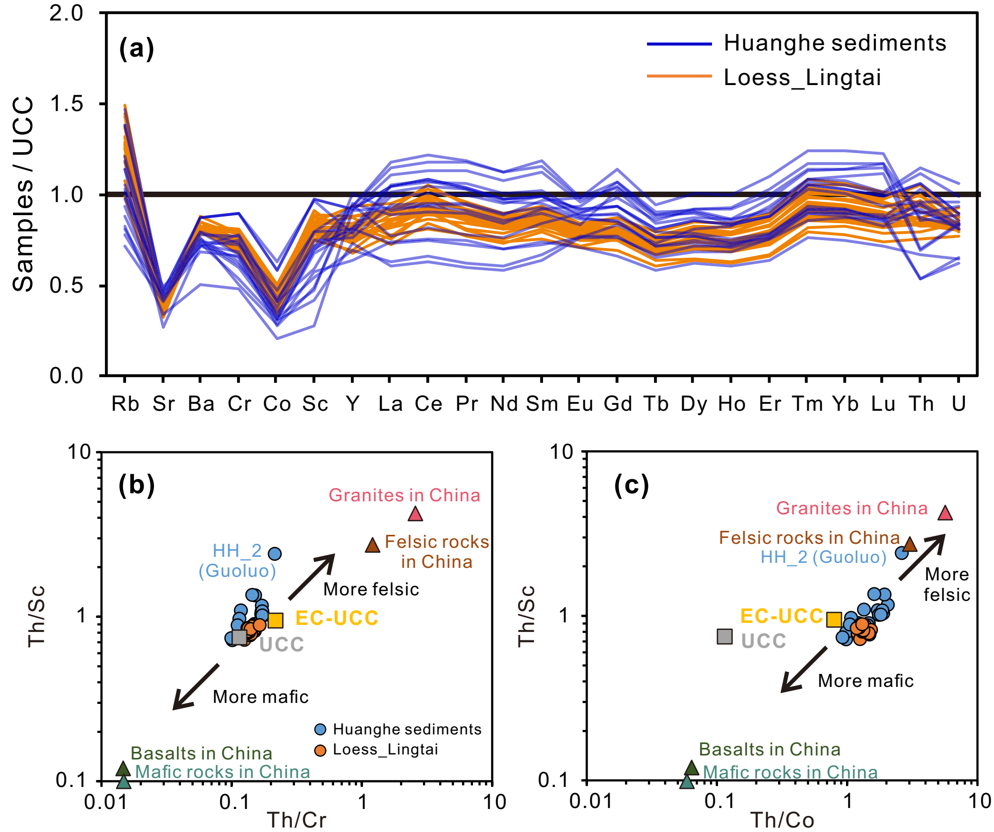

Figure 3. (a) Element contents of Huanghe sediments normalized to respective UCC averages (Rudnick & Gao, 2003). (b) Th/Sc versus Th/Cr and (c) Th/Sc versus Th/Co diagrams. Geochemical data for the granites, felsic rocks, basalts and mafic rocks in China are from Yan and Chi (2005).

The element Th is immobile and generally more enriched in felsic rocks than in mafic rocks, whereas Sc, Cr, and Co are more compatible and are enriched in mafic rocks (Taylor & McLennan, 1995). Therefore, the Th/Sc, Th/Cr, and Th/Co ratios are useful indicators of felsic versus mafic sources (Cullers et al., 1988). The loess samples display narrow ranges of Th/Sc, Th/Cr, and Th/Co values (mean values of 0.84 ± 0.04, 0.14 ± 0.01, and 1.32 ± 0.10, respectively; 1σ). The Huanghe sediments show wider ranges of values (mean values of 1.07 ± 0.37, 0.15 ± 0.03, and 1.52 ± 0.46, respectively; 1σ). Most of the samples are close to the average ratios of East China UCC (EC–UCC), except for the sample collected at Guoluo (HH_2), which has Th/Sc and Th/Co values closer to those of felsic rocks in China (Figures 3b and 3c).

4.2. Sr–Nd Isotopes

$^{87}Sr/^{86}Sr$ ratios of loess samples and Huanghe river sediment samples range from 0.719298 to 0.721172 and from 0.716866 to 0.720533, with mean values of 0.719916 and 0.718342, respectively. εNd values of loess samples and Huanghe sediments vary from −11.0 to −10.5 and from −12.4 to −10.2, with mean values of −10.7 and −11.4, respectively (Table A1). Loess samples have narrower ranges of $^{87}Sr/^{86}Sr$ and εNd compared with Huanghe sediments, showing more radiogenic values overall.

There are no obvious correlations of $^{87}Sr/^{86}Sr$ and εNd with mean grain size for the loess samples, possibly because these samples have a more restricted variation in mean grain size compared with the Huanghe sediments. Significant correlations between $^{87}Sr/^{86}Sr$ and mean grain size are observed for the Huanghe sediments, but no obvious correlation exists between εNd and mean grain size (Figure 4).

Meng et al. (2008) reported a wider range of εNd (from −13.9 to −9.6, mean of −11.5) for Huanghe sediments compared with the data measured in the present study. This difference probably arose because most

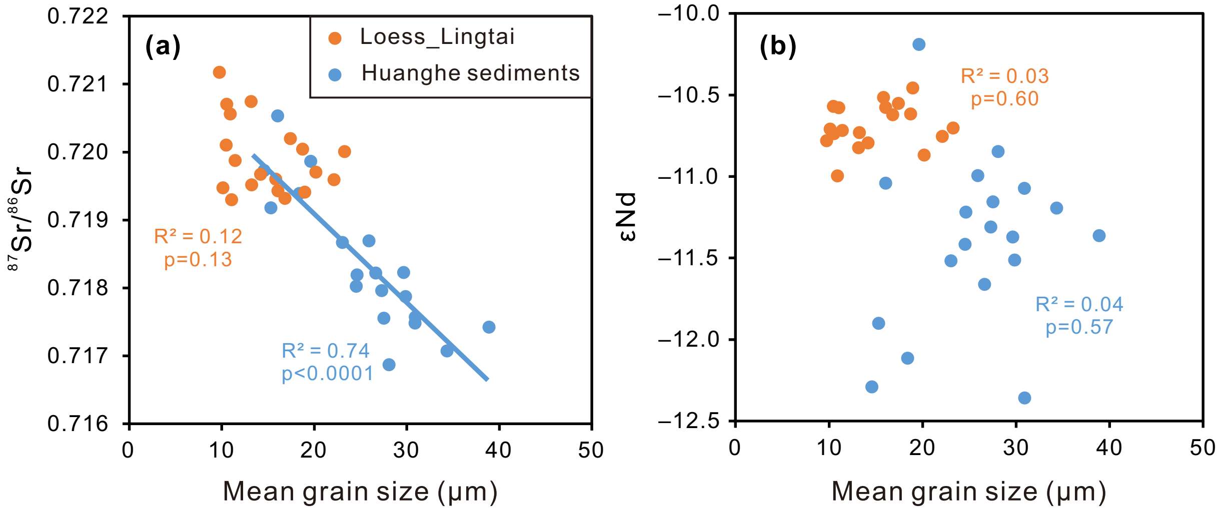

Figure 4. (a) $^{87}Sr/^{86}Sr$ versus mean grain size and (b) εNd versus mean grain size diagrams for Huanghe river sediments and loess sediments.

samples analyzed by those authors were taken from major tributaries and second-order drainage basins and were not subjected to chemical treatment, meaning that secondary components in their samples might have caused the wide variation in εNd values. Further, the more scattered εNd values reported by Meng et al. (2008) may also have been caused by sampling of overbank samples at a depth of 80 cm. This mode of sampling could have introduced vertical variations in data (Bølviken et al., 2004). The siliciclastic fractions of 20–25 μm size for Huanghe sediments have εNd values ranging from −12.5 and −10.0, averaging at −11.6 (Li et al., 2018; Zheng, 2018), which are overall comparable with result of this study. Sediment samples reported by Meng et al. (2008) have $^{87}Sr/^{86}Sr$ values of 0.712868–0.718860 (mean of 0.715475), generally lower than those of this study. We infer that the difference arose because those authors measured a wider grain size range (<100 μm) and/or because carbonates with low $^{87}Sr/^{86}Sr$ (e.g., 0.71036–0.71176 for calcites in Chinese loess [Yang et al., 2000]) were not removed from their samples before isotopic analysis.

5. Discussion

5.1. Geochemical Variability of Sediments and Provenance Fingerprinting Using Trace Elements and Sr–Nd Isotopes

Similar trace element compositions of the studied river and loess sediment samples do not enable the sediment sources to be identified (Figure 3). Previous studies have also found no significant variation in element geochemistry between sediments taken from the mainstream and major tributaries of the Huanghe River, and the river sediments have a mixed felsic–mafic source with an affinity toward average EC–UCC (Pang et al., 2018; Yang et al., 2002). Hence, other reliable provenance indicators must be applied for effective discrimination of sediment provenance.

Radiogenic isotopes of Sr ($^{87}Sr/^{86}Sr$) and Nd ($^{143}Nd/^{144}Nd$) in terrigenous detritus are largely controlled by the Rb/Sr and Sm/Nd ratios of source rocks and average crustal residence ages (DePaolo, 1981; O'nions et al., 1983). Therefore, these ratios have been widely used to decipher the provenance of fine-grained siliciclastic sediments in terrestrial and marine environments. Differences in $^{87}Sr/^{86}Sr$ and εNd between the Huanghe river sediments and loess samples may imply a sediment contribution from non-loess sources to the Huanghe River basin, indicating the usefulness of Nd–Sr isotopes of the Huanghe sediments for tracing their provenance (Figure 4).

For the Huanghe river sediments, Sr isotopes generally increase with decreasing grain size, mainly because finer sediments contain more K-rich clay minerals and micas with high Rb/Sr and $^{87}Sr/^{86}Sr$ ratios (Garçon et al., 2014). The poor correlation between εNd and mean grain size confirms the weak influence of grain size on Nd isotopes.

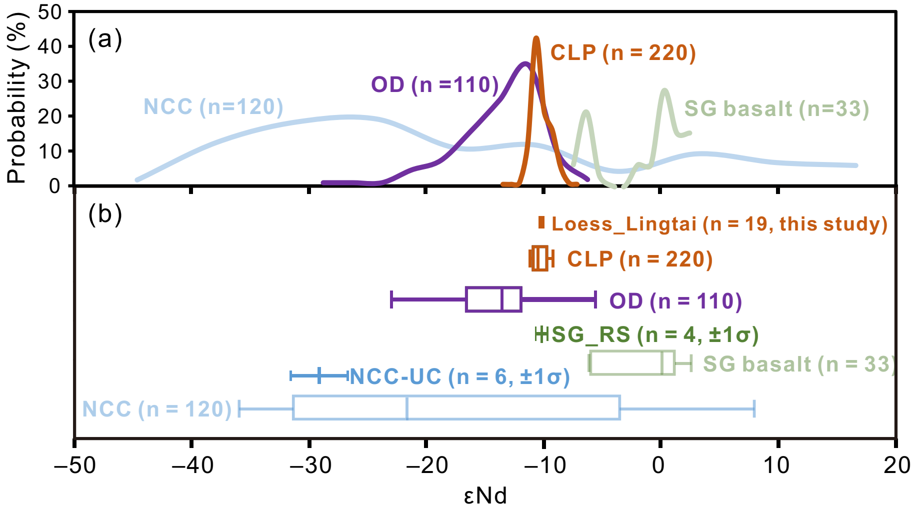

Figure 5. Distributions of Nd isotopes in major tectonic terranes and source areas in the Huanghe River basin: (a) Smoothed histogram curves with 10 bins; (b) boxplot visualization. CLP=China Loess Plateau; OD=Ordos Desert; SG=Songpan-Ganzi; SG_RS = River sediment in Songpan-Ganzi Block; NCC=North China Craton. NCC-UC= Inferred average composition of crystalline basement rocks in the upper crust of North China Craton, see Figure 6 for the details. Data of rocks from Songpan-Ganzi Block and North China Craton are from GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/; Sarbas and Nohl [2008]); data of river sediment in Songpan-Ganzi Block from (Wu et al., 2010); data of Ordos Desert from Chen et al. (2007), Rao et al. (2008), Rao et al. (2011) and Li et al. (2018); Loess Plateau data from Liu et al. (1994), Jahn et al. (2001), Yokoo et al. (2004), Sun (2005), Chen et al. (2007), Rao et al. (2008), Chauvel et al. (2014), Zhang et al. (2015), Li et al. (2018) and Y. Wang et al. (2007).

The Nd isotope compositions of sediments and potential sources are discussed in detail below in Sections 5.2 and 5.3, as these compositions can be used to discriminate continental crustal materials of different ages (DePaolo, 1981; O'nions et al., 1983) and are less influenced by hydrodynamic sorting (Figure 4) and chemical weathering compared with $^{87}Sr/^{86}Sr$ ratios (Garçon et al., 2014).

5.2. Nd Isotopic Compositions of Major Source Terranes

Hu et al. (2012) argued that the CLP and Ordos Desert are the two major sources of Huanghe river sediments. However, the sampling locations of the present study include coverage of the eastern part of the Tibetan Plateau, so the contribution of source rocks in these upper catchments should also be considered. The Huanghe River originates in the Songpan–Ganzi Block on the northeastern Qinghai–Tibet Plateau. With the extremely high uplift and denudation rates in this tectonically active region, the Songpan–Ganzi Block supplies large amounts of clastic material to the upper reaches of the Huanghe River (Nie et al., 2015). In the context of channel erosion in the lower reaches, some proximal materials (e.g., from the North China Craton through which the river flows) may be transported to the mainstream. Therefore, four characteristic provenances (source terranes) for Huanghe sediments are identified in this study: Songpan–Ganzi (SG) Block, Ordos Desert (OD), CLP, and North China Craton (NCC) (Figure 1b).

Compiling published data for sediments and rocks is an effective approach for validating Nd isotopic fingerprinting for source areas (Clift et al., 2013; Wu et al., 2010). Nd isotopic compositions of loess and desert sediments from the CLP and OD yield relatively small variations, indicating that these materials were well mixed prior to deposition. The Nd isotopes of rock outcrops from the SG Block and NCC, however, show large variations, suggesting heterogeneity in source rock compositions of these regions (Figure 5a).

As the dominant sediment supply for the Huanghe River basin, the CLP in North China is characterized by a huge accumulation of loess measuring hundreds of meters in depth and has a depositional history dating back to the Paleogene (Guo et al., 2002). Nd isotopes of the CLP loess are relatively uniform and yield small temporal and spatial variations (Chen et al., 2007; Rao et al., 2008, 2011; Yokoo et al., 2004). The loess samples taken from the Lingtai section in the present study have a mean εNd value of −10.7 ± 0.2 (1σ), consistent with literature data (−10.3 ± 0.8 [1σ]; Figures 5a and 5b).

The OD, which lies in the central North China Block, has a basement of Archean and Paleoproterozoic metamorphic crystalline rocks overlain by thick sedimentary rocks of Proterozoic, Paleozoic, and Mesozoic ages (Zhang et al., 2008). Generally, the old basement-derived materials are characterized by low εNd (e.g., the Ordos sandstones with εNd values of −15.2 to −21.8, $n = 14$), whereas the exotic dust from the northern Tibet Plateau and the Huanghe River basin contributes a considerable amount of fine-grained sediment with more radiogenic Nd isotopes (Rao et al., 2008, 2011).

It is challenging to directly compare the Nd isotopes between river sediments and the compiled data from rock outcrops because of the high geochemical heterogeneity of source rocks and the resulting overlaps in isotopic spectra for different sources (Clift et al., 2013). Hence, a robust constraint for the average Nd isotopic compositions of eroded upper crust in the source areas is required for provenance tracing of river sediments. A simple compilation of literature data for exposed rocks of the SG Block and NCC cannot faithfully mirror the actual distribution of outcrops and lithologies. This is apparent from the high variability of reported Nd isotopes from outcrops in the SG block and NCC, which would hamper the discrimination of provenances for Huanghe River sediments.

The SG massif is composed of a giant Triassic polymorphic complex and Permian–Cenozoic felsic rock masses. Permian basalts in the SG block with affinity to the Emeishan large igneous province have high εNd values (Zi et al., 2010). Most of the collected data are from basalts, with a lack of data for other lithologies, making it impossible to infer the average composition of eroded upper crust in the SG block. Instead, we collected published εNd data for sediments that were obtained from small rivers draining only the SG Block, to constrain the composition of weathered upper crust in this area (Figure 5b). These sediments yield εNd values of −10.8 to −9.0, with a mean of −9.9 (Wu et al., 2010), which is in good agreement with the sediment samples taken from upstream of Maduo (HH_1, −10.2) and Guoluo (HH_2, −10.8) in the present study.

There are numerous published Nd isotopic data for outcrops with different lithologies in the NCC. To estimate the average Nd isotopes of NCC basement upper crust (NCC–UC), we plotted the relevant literature data in a diagram of εNd versus Th/Sc, as proposed by McLennan et al. (1993), to distinguish the tectonic affinity of sediment provenance types. The Th/Sc ratio is a well-established indicator for distinguishing mafic from felsic sources. The good correlations between literature εNd values and this indicator suggest that εNd values of NCC rocks are determined mainly by the end-member compositions of their source materials; that is, crustal sources with low εNd and high Th/Sc values, and mantle sources with high εNd and low Th/Sc values. Thus, the Th/Sc value can be used for a range of materials from the NCC–UC (data from Yan and Chi [2005]) to constrain the corresponding Nd isotopes. By using this approach, we obtained a mean εNd of −29.2 ± 2.5 (1σ) for the NCC–UC (Figure 6) and a corresponding mean Nd model (T_{DM}) age of 2.73 ± 0.18 Ga (1σ) (see Table A4 and Text A1), which is consistent with the major period of crustal growth (~2.7 Ga) for this old craton (Jiang et al., 2010).

In summary, the distinct Nd isotopes of upper crust in the four major provenances of Huanghe River sediments can be recognized using literature data, allowing us to discriminate the provenances of the Huanghe sediments investigated in this study.

5.3. Variations in Nd Isotopes of the Huanghe Sediments and Provenance Discrimination

The sediment sample taken from Maduo, the most upstream site in this study, has the highest measured εNd value (Figure 7), suggesting a significant sediment contribution from the SG Block. Downstream to Yinchuan, sediment εNd values decrease gradually, indicating a decrease in the contribution of high-εNd material from the SG block source region and an increase in low εNd material from basins along the river course, such as sediments from the OD and the high-grade metamorphic crystalline basement of the piedmonts of the Qilian mountains, which is also characterized by low εNd (many samples yield εNd values as low as −20) (Zhang et al., 2007). Since the completion of hydroelectric engineering projects such as the Liujiaxia and Longyangxia reservoirs (constructed in 1968 and 1985, respectively) in the upper reaches of the Huanghe River, most of the sediments supplied from above Lanzhou have been trapped in these reservoirs (H. Wang et al., 2007), which can be indirectly confirmed by the lower sediment discharges in 2000–2006

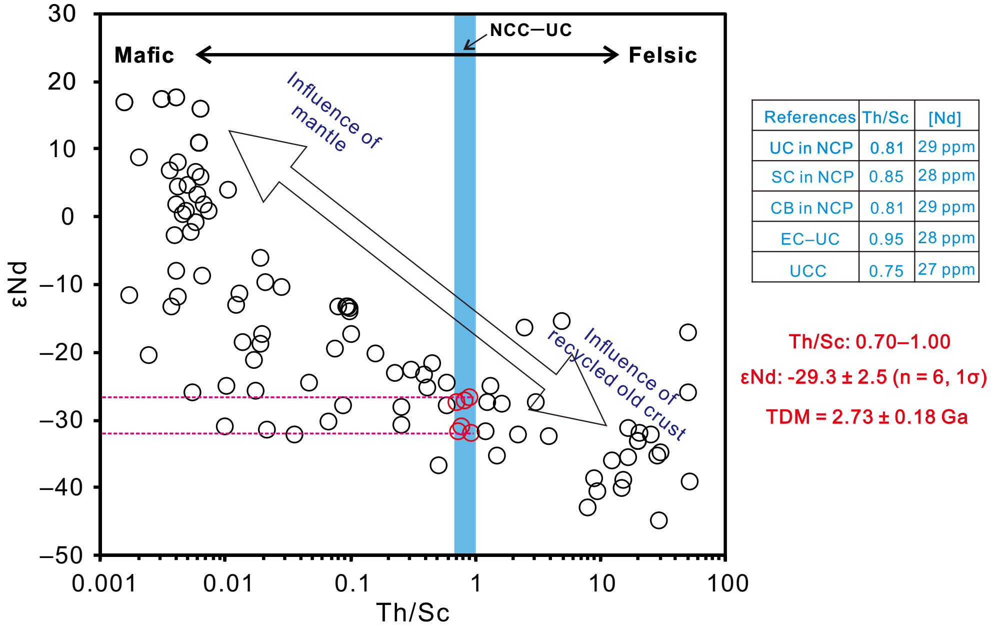

Figure 6. εNd versus Th/Sc diagram for rocks from the NCC. This data set was obtained from the GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/). Red circles and the data within the dotted red lines indicate the estimated averages of Th/Sc and εNd for the NCC–UC, respectively. NCP=North China Platform, SC=Sediment Cover, CB=Crystalline Basement, EC-UCC = the average of East China Upper Continental Crust. The Th/Sc and [Nd] values of UC, SC and CB in NCP, and EC-UCC are from (Yan & Chi, 2005), Th/Sc and [Nd] values of UCC from (Rudnick & Gao, 2003).

and 2009–2016 compared with 1950–1999 (Figure 2). We infer that the cascade hydroelectric engineering projects in upstream reaches have retained sediment from the source areas and disproportionately reduced the contribution of SG materials.

Sediment εNd values increase gradually from Yinchuan to Tongguan in the middle reaches (Figure 7), suggesting a gradual increase in sediment contribution from the CLP with higher εNd. However, εNd values for

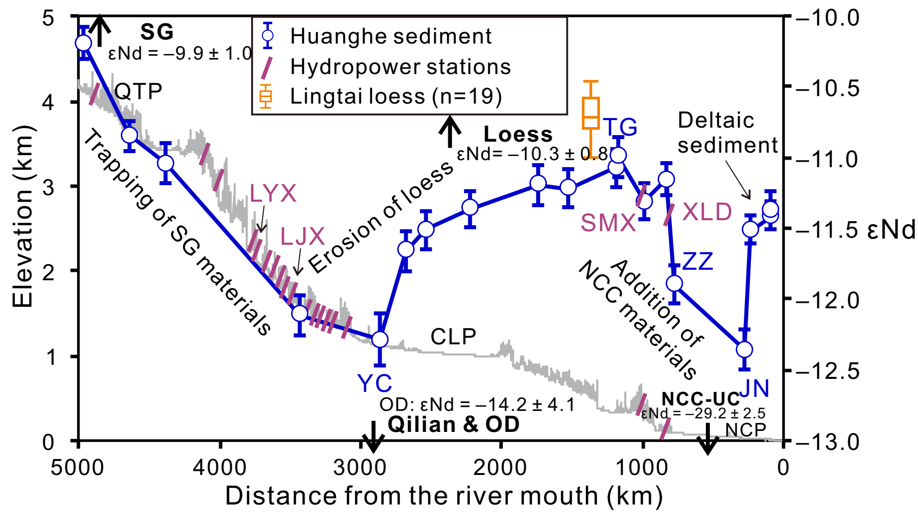

Figure 7. Downstream variation in εNd in the studied Huanghe sediments. River channel elevations and the locations of the main hydropower stations on the Huanghe River are also shown. Error bars represent the analytical uncertainties (±2σ). QTP = Qinghai-Tibet Plateau, LYX = Longyangxia, LJX = Liujiaxia, NCP= North China Plain, YC=Yinchuan, TG = Tongguan, ZZ = Zhengzhou, JN = Jinan, SMX=Sanmenxia, XLD = Xiaolangdi.

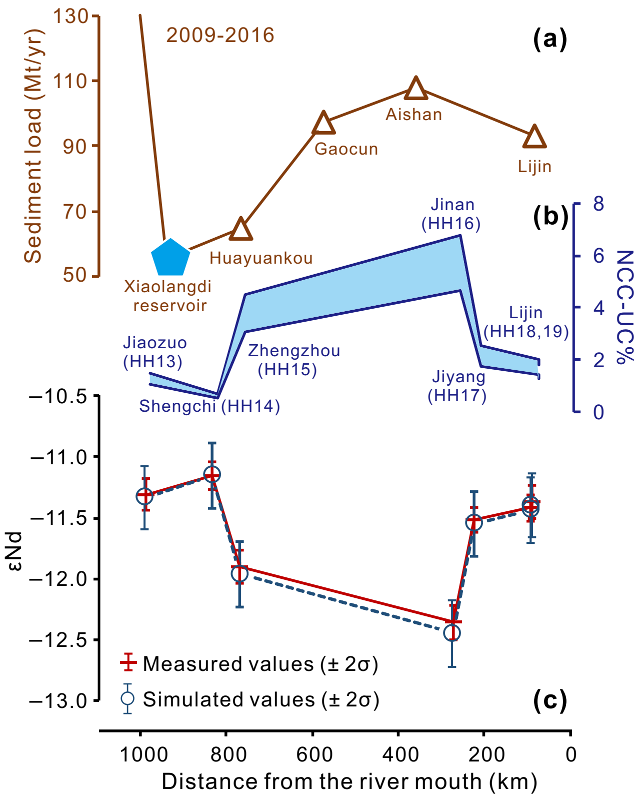

Figure 8. (a) Annual sediment load at different gauging stations in the lower Huanghe reaches. (b) Estimated percentage of NCC–UC-derived materials in sediments from the lower Huanghe (Text A2 for details of the calculation). (c) Comparison of measured and simulated Nd isotopes for sediments from the lower Huanghe. Each simulated value and its uncertainty were obtained from statistics of 10^4 modeled mixtures generated by Monte Carlo simulation. The framework of the Nd isotopic mixing model and the characterization of parameters using the Monte-Carlo modeling are described in detail in Supporting Information S1.

the lower mainstream sediments taken from Tongguan to Jinan decrease sharply downstream, yielding values as low as −11.9 and −12.3 at Zhengzhou and Jinan, respectively (Figure 7).

Considering the overwhelming dominance of CLP-derived sediment in the lower Huanghe mainstream, the clearly lower εNd values in the lower reaches (Zhengzhou and Jinan), which deviate from those of typical loess, are unusual. This difference may be due to the considerable contribution of proximal sediments that have low εNd values, such as those from the NCC–UC. Considering the very low mean εNd value of −29.2 ± 2.5 (1σ), even a 5% addition of NCC–UC would induce a decrease in εNd of 1.0–1.5 units in sediments of the lower Huanghe reaches (Text A2). In recent years, the suspended sediment concentration in the lower reaches has decreased sharply, mainly because of the construction of several dams, soil conservation measures in the CLP, and the use of river water in upstream areas (Wang et al., 2015). Thus, the relative contribution of CLP-derived sediments to the lower mainstream has been decreasing (e.g., the sediment loads at Xiaolangdi have decreased to 55.6 Mt/yr during 2009–2016, only ~5.3% of the 1950–1999 level), making the contribution of proximal sources more noticeable. In particular, since 2002 the WSR Project has caused a mean erosion rate of 6 cm/yr. in the lower reaches downstream of the Xiaolangdi Reservoir, which has resulted in pronounced river-channel scouring (Chu, 2014).

Sediment εNd values at Jiyang and Lijin near the estuary are higher than those upstream, up to −11.5 (Figure 7), which suggests the addition of "old" CLP-derived sediments from the Huanghe River delta. In recent 20 years, the delta and shoreline of the Huanghe River mouth have been retreating because of a change from deposition to erosion (Bi et al., 2014; Chu et al., 2006). This human-induced channel and delta erosion is discussed in more detail in Section 5.4 with respect to its influence on the natural source-to-sink transport processes and chemical compositions of river sediment.

5.4. Influence of River Channel and Delta Erosion on Chemical Signals of Huanghe Sediments

During 2009–2016, the mean sediment flux at Xiaolangdi Station (~942 km from the river mouth) was only 56 Mt/yr, much lower than the mean value for 1950–1999 of 1045 Mt/yr, suggesting significant retention of sediment in reservoirs. The sediment flux downstream of Xiaolangdi station increases gradually with decreasing distance from the river mouth, reaching a maximum of 108 Mt/yr at Aishan Station (~367 km from the river mouth) and then decreasing slightly to about 93 Mt/yr at Lijin Station (~96 km from the river mouth) (Figure 8a). It is inferred that this downstream variation in sediment flux reflects the strong river channel erosion between Xiaolangdi and Aishan (Chu, 2014).

The contribution of the NCC–UC source to sediment in the lower reaches of the Huanghe River is calculated using a two-component mixture model of Nd isotopes. The two sediment samples collected at Tongguan in 2009 and 2016 have different Nd contents (25.7 and 16.4 ppm, respectively) but similar εNd (−11.1 and −11.0, respectively). These two samples were used as references for the Huanghe_TG component. The average Nd content of the NCC–UC has been reported by Yan and Chi (2005), and its Nd isotopic composition was estimated by using a statistical method based on the GEOROC database (Figure 6; $\overline{[\mathrm{Nd}]} = 28$, $\overline{\varepsilon\mathrm{Nd}} = -29.2$). The adopted two-component mixture model (Text A2) reveals that the NCC–UC contribution of sediment to the Huanghe River between Jiaozuo and Sanmenxia is minor, whereas the contribution is much higher in the river section between Zhengzhou and Jinan (Figure 8b). At Jiyang and Lijin stations near the river mouth, the NCC–UC contribution is much lower than at Jinan (Figure 8b). We performed a Monte Carlo simulation to validate our estimations of the NCC–UC contribution (Text A2). The uncertainties of the simulated values are ~0.25 εNd units, meaning that the downstream trend is clear and corresponds well with the pattern shown by the measured samples (Figure 8c).

The riverbed of the lower Huanghe lies above the land surface beyond its banks or levees, and thus it is unlikely that the Huanghe mainstream can erode the basement rocks. However, sediments previously eroded from the NCC–UC and transported by tributaries of the Huanghe or other local rivers (e.g., the Dawen and Huaihe rivers) can accumulate in the alluvial plain proximal to the mainstream of the lower Huanghe channel. This proximal source may supply considerable amounts of material with low εNd to the lower Huanghe River. The so-called proximal materials generated by channel erosion consist of recycled sediments that were previously deposited in the North China Plain and the NCC–UC component. The recycled sediments have similar εNd values to those of typical Huanghe sediments owing to their provenance inheritance, whereas the NCC–UC-derived proximal material has more negative εNd values.

In summary, Nd isotopes of Huanghe sediments document the influences of the interception of sediment from source areas by dam cascades and the erosion of NCC–UC-derived proximal materials. Our findings confirm that sediment provenance and geochemistry can respond to sediment routing processes altered by the impacts of anthropogenic influences such as dam construction and water regulations. The data and analytical method used in this study should provide guidance for future comprehensive investigations of erosion and sedimentation processes in large river systems that are subject to strong anthropogenic impact. The quantification of Nd isotopes of modern Huanghe River basin sediments should also shed new light on sediment provenance discrimination and paleoenvironmental reconstruction in East China marginal seas.

6. Concluding Remarks

The natural sediment routing of the Huanghe River has been substantially altered by human activities since the mid-twentieth century. This study investigated how river sediment geochemistry has responded to changes in the river sediment source-to-sink routing regime, based on Nd isotopic fingerprinting. The main conclusions of the study are as follows.

Nd isotopic compositions of four major source terranes of Huanghe River basin sediments are well constrained, which allows us to discriminate the sedimentary provenances. The Nd isotopes of sediments collected along the Huanghe River course show systematic variations, reflecting the mixing of detrital materials from four major provenances with distinct Nd isotope signatures. The gradual downstream decrease in εNd values of sediments in the upper Huanghe indicates a decreasing sediment contribution from the SG Block, which has been partly induced by sediment retention in cascade hydroelectric engineering/dam projects. The gradual downstream increase in sediment εNd values in the middle reaches suggests an increase in the contribution of sediment from the CLP, caused by intense erosion. The marked decrease in εNd values in the downstream sediments is a result of the input of NCC–UC derived proximal materials from enhanced channel scouring in recent 20 years.

The Huanghe River presents a prime example of a large river that first underwent a rapid increase in sediment flux owing to changes in catchment land use, followed by a dramatic decrease as a result of cascade dams/reservoirs, water use practices, and other anthropogenic activities and interventions. At present, the sediment discharge of the Huanghe River has been reduced to its former natural level on account of these activities and interventions, which have significantly altered the natural sediment routing processes of the river and generated marked changes in the provenance proportions and geochemistry of the river sediments. As the Huanghe once discharged huge amounts of terrigenous sediments into the continental margin of East China, investigations of sediment source-to-sink in the marginal seas need to consider changes in the routing of sediment through the Huanghe River basin as presented here. In addition, sediment fingerprinting using Nd isotopes can be used to trace sediment provenance and establish the influence of anthropogenic activity and interventions on river sediment flux and routing, as well as related processes.

Data Availability Statement

The data in this study are available in the ZENODO https://zenodo.org/record/4974266#.YMtfNUy-uUk.

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