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Seasonal variations in the Sr-Nd isotopic compositions of suspended particulate matter in the lower Changjiang River: Provenance and erosion constraints

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Suspended particulate matter samples were collected monthly for more than 2 years in Nanjing, China to examine seasonal changes in the Sr-Nd isotopic compositions of the lower Changjiang River (CR). The results indicate that the $^{87}Sr/^{86}Sr$ ratios of the samples ranges from 0.725352 to 0.738128, and the values of $\varepsilon_{Nd}(0)$ ranges from −10.55 to −12.29. The Sr-Nd isotopic compositions show distinct seasonal variations. The samples had lower $^{87}Sr/^{86}Sr$ ratios and higher $\varepsilon_{Nd}(0)$ values during the flood season than the dry seasons. The seasonal variations primarily reflect the controls of provenance rocks and erosion in different sub-catchments. The relative decrease in $^{87}Sr/^{86}Sr$ ratios and the increase in $\varepsilon_{Nd}(0)$ values during the flood season may reflect an increasing in the mechanical erosion rate in the upper basin and the contribution of more sediment from the upper reaches. The end member values of $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ of the samples were 0.728254 and −11.26, respectively.

Changjiang River, suspended particulate matter, Sr-Nd isotopic compositions, seasonality, provenance

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In recent years, major rivers from the Himalayan-Tibetan area have attracted increased interest in the weathering intensity and development history of their drainage basins, as well as changes in their chemical flux to the global ocean because they have recorded the uplift history of the Tibet and Asian monsoon evolution in the Cenozoic period [1–9].

The Changjiang (Yangtze) River (CR) is the third longest (6300 km) river in the world. The CR originates on the Tibetan Plateau at 5100 m and drains into the East China Sea, covering a total area of 181×10^4 km^2, or nearly 20% of the total terrestrial area of China. As the longest river in Asia, the CR runs through several tectonic systems and geomorphic units. The river is also the result of both the Cenozoic Asian topographic regime and the Asian monsoon regime [1,5,10].

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The CR is characterized by high elevation in the inner part of the catchment basin, a monsoon climate, and intense weathering in most parts of the basin, resulting in large freshwater and sediment discharges. As a result, the CR is one of the most important suspended particulate matter (SPM) transporting rivers in the world. The upper CR basin plays an important role in this process because it supplies most of the suspended sediments discharged to the ocean by the river [11–13]. Under the control of the Asian summer monsoon, water and sediment discharge from both the upper and lower reaches of the CR show seasonal patterns. The precipitation and runoff change seasonally, with 70%–80% occurring from May to October [12]. From 1950–1990 more than 70% of the water was discharged from the CR during summer (May–October), with the average peak occurring in July [14]. The modern Yangtze River discharges most of its annual sediment load between June and September [15]. In addition, a large portion of the CR Basin has a subtropical monsoon climate. Because water discharge is subject to strong seasonality, the suspended particulate matter of the CR varies drastically according to season and from one year to another. Accordingly, several studies have investigated seasonal variations in the geochemistry of dissolved and particulate matter [16–19].

Radiogenic neodymium and strontium isotopes are generally considered to be reliable indicators of the provenance of sediments, not only because geological bodies have different Sr-Nd isotopic compositions that depend on their origins and ages, but also because the Sr-Nd isotopes undergo limited alterations during surficial processes such as weathering and transportation [20–25]. As a result, the Nd-Sr isotopic signatures of the sediments have been useful for identifying their sources and studying erosion processes [2,26–30]. In addition, the Nd-Sr isotopic signatures of geological time scale sediments have been widely used to probe evolution of the Asian monsoon and the uplift history of Tibetan Plateau [31–39]. Several studies of the Nd-Sr isotopic compositions of the CR sediments have recently been conducted [40–42]. However, changes in the seasonal Nd-Sr isotopic compositions of the SPM have not yet been investigated in detail, although a study of modern Amazon Rivers showed that the Sr and Nd isotopic composition of the suspended sediments is seasonally controlled [43].

The present study was conducted to examine the temporal distribution of Sr-Nd isotopic compositions in the lower CR SPM in detail through seasonal sampling and to assess variations in east Asian monsoon rainfall controls based on their seasonal changes. The results of this study will contribute to the overall understanding of the developmental history of the CR, the continental weathering processes during the Cenozoic period, uplift of the Tibetan Plateau, evolution of the east Asian monsoon, and discrimination of sediment sources in East China and the marginal seas.

1 Materials and methods

Seasonal variations were examined by taking monthly SPM samples from the lower stream of the CR at Nanjing (Figure 1) for 2 years. The first year (2005) included only two samples, one from January (dry season), and one from August (flood season). During the second year, samples of SPM were collected monthly from October 28, 2006 through September 24, 2007. All samples were collected at the same location, 32°05′33.9″N, 118°43′27.6″E. Water samples were collected from a boat in the middle of the river channel at a water depth of about 30 cm.

Samples were collected in acid-cleaned containers and filtered through 0.45 μm Millipore membranes (herein defined as the size fraction ranging from 63 to 0.45 μm) to collect SPM. The authigenic carbonate minerals may change the Sr isotopic compositions of the detrital sediments of the CR [44]. In this study, all of the samples was selectively dissolved with purified acetic acid solution (0.5 mol/L) at room temperature for up to 24 h in order to remove carbonate minerals. The mineralogy of acid-insoluble samples was then determined using an X-ray diffractometer (XRD), and the dissolution of carbonate minerals in the samples after acetic acid treatment was evaluated based on the XRD analyses (Figure 2).

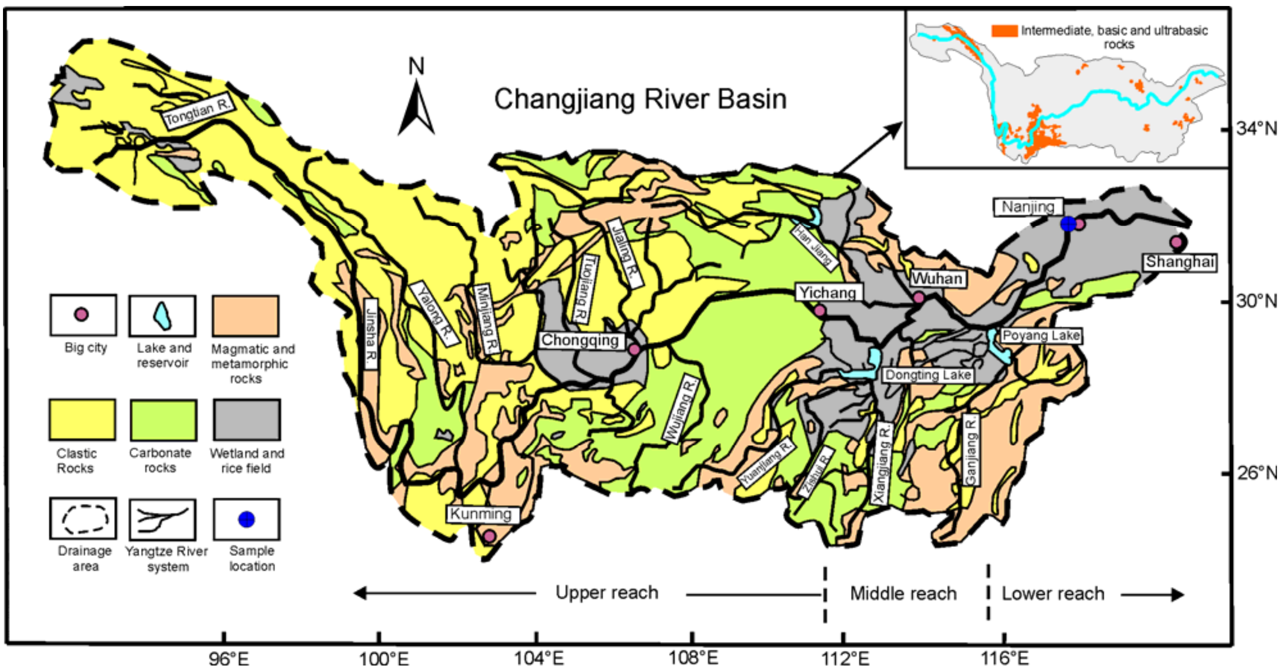

Figure 1 Geological map of the Changjiang River drainage basin and sampling site (modified from the map in Wing et al. [42]).

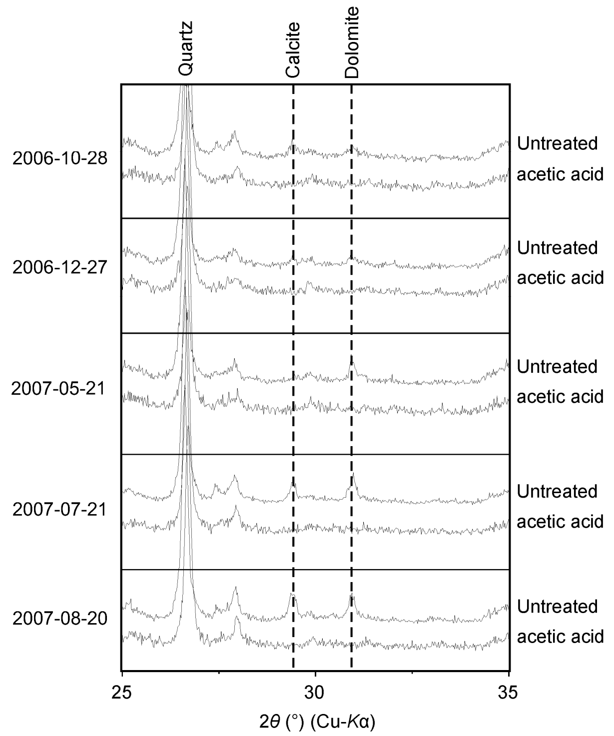

Figure 2 X-ray diffraction patterns of selected samples of SPM, bulk samples and acetic acid treated samples.

Table 1 Nd and Sr isotopic compositions of the SPM samples at the monitoring stations (Nanjing)

	$^{87}Sr/^{86}Sr$	2σ	$^{143}Nd/^{144}Nd$	2σ	$\varepsilon_{Nd}(0)$
2005-01-05	0.734557	16	0.512008	12	−12.29
2005-08-18	0.727226	6	0.512087	8	−10.75
2006-10-28	0.731997	2	0.512026	16	−11.94
2006-11-28	0.732020	2	0.512040	5	−11.67
2006-12-18	0.735263	2	0.512035	16	−11.76
2006-12-27	0.734579	2	0.512046	5	−11.55
2007-01-19	0.733670	9	0.512032	4	−11.82
2007-02-27	0.729762	2	0.512022	5	−12.02
2007-03-22	0.731698	2	0.512019	6	−12.07
2007-04-23	0.738128	2	0.512028	3	−11.90
2007-05-21	0.733041	2	0.512062	10	−11.24
2007-06-22	0.729996	6	0.512044	6	−11.59
2007-07-21	0.725352	1	0.512053	8	−11.41
2007-08-20	0.727994	2	0.512079	5	−10.90
2007-09-24	0.728456	2	0.512097	12	−10.55
Weighted average[a)]	0.728254		0.512060		−11.26

a) Calculated by averaging monthly Nd and Sr isotopic compositions of the samples according to their monthly fluxes (from 2006 to 2007).

The pretreated samples were cleaned in pure water, after which they were digested with a mixture of HNO_3 + HF solution. Sr and Nd were separated using the standard ion exchange techniques and their isotopic ratios were determined using a Finnigan Triton thermal ionization mass spectrometer at the Department of Earth Sciences, Nanjing University. $^{87}Sr/^{86}Sr$ was normalized to $^{86}Sr/^{88}Sr$=0.1194 and $^{143}Nd/^{144}Nd$ was normalized to $^{146}Nd/^{144}Nd$=0.7219. The analytical blank was <1 ng for Sr and <60 pg for Nd, respectively. The reproducibility and accuracy of the Sr and Nd isotopic analyses were periodically checked by running the Sr standard SRM987 and Nd standard La Jolla, with a mean $^{87}Sr/^{86}Sr$ value of 0.710268 ± 20 (2r external standard deviation, n=15) and a mean $^{143}Nd/^{144}Nd$ value of 0.511840 ± 8 (2r external standard deviation, n=6), respectively.

2 Results

The Sr and Nd isotopic compositions of the 15 SPM samples from the CR are given in Table 1 and Figure 3. During the sampling periods, the water discharge varied ranged from 10400 m^3/s (January) to 44200 m^3/s (July). From October 2006 to September 2007, the $^{87}Sr/^{86}Sr$ ranged from 0.725352 to 0.738128 and displayed a large seasonal variation, with the minimum value being observed during flood season (July 2007). The $\varepsilon_{Nd}(0)$ values in the suspended samples also showed a distinct seasonal variations (Figure 3), ranging from −12.07 to −10.55, with the maximum value

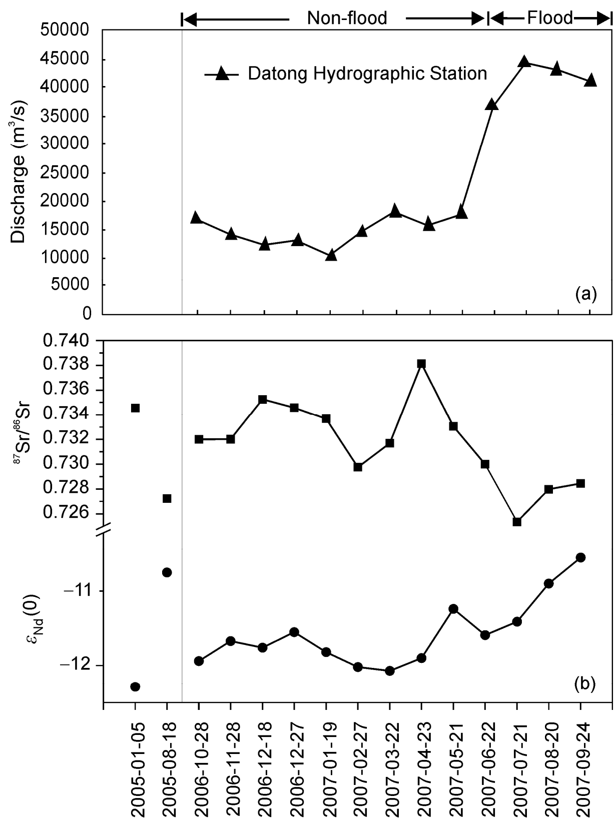

Figure 3 Monthly water discharge measured at the Datong Hydrographic Station during 2006–2007 (a), together with the monthly Nd and Sr isotopic compositions of suspended particulate matter (b). Hydrological data are taken from http://www.hydrodata.gov.cn. Note: rainy season or high flow season, from May to October; flood season, from June to September; non-flood season, from January to May and October to December.

being observed in September 2007 during the flood season. Moreover, the most consistent variations in the samples Sr-Nd isotopic compositions of the samples were observed in 2005 and 2006–2007, suggesting that the study years were representative and showed a regular seasonal patterns.

The suspended samples referred to in this paper were collected from the lower stream of the CR, which is considered one of the most efficiently mixed materials of the drainage basin. Thus, the Nd and Sr isotopic compositions of these samples likely represent the mean isotopic signature of the average drainage basin. The average value of the acid-insoluble residues $\varepsilon_{Nd}(0)$ of the CR was −11.26, and plot in intermediate position of world rivers (drainage area> 100000 km^2, $-4 < \varepsilon_{Nd}(0) < -20$). The $\varepsilon_{Nd}(0)$ values of the CR samples were close to the average value for weathered continental crust (−11.4±4) [20], but higher than the average but higher than the average value of Upper Continental Crust (−17) [21], These findings suggest that the $\varepsilon_{Nd}(0)$ values of suspended samples from the CR are a good indicator of the average value of the drainage basins.

The average $^{87}Sr/^{86}Sr$ ratio of the acid-insoluble residues for the CR samples was about 0.728, which is much higher than the average $^{87}Sr/^{86}Sr$ value of Upper Continental Crust (0.716) [21], and slightly higher than the values of bulk suspended phases (containing the carbonate fraction) observed by Wang et al. [42]. The acid-insoluble residues suspended loads in the CR had higher $^{87}Sr/^{86}Sr$ values (0.7178–0.7252) than the bulk suspended loads because marine carbonates of various periods are widespread in the CR Basin, having high concentrations of Sr and low $^{87}Sr/^{86}Sr$ isotopic ratios [44,45]. Wang et al. [42] found that a downriver increase in suspended $^{87}Sr/^{86}Sr$ ratios in the mainstream portion of the CR may reflect increased relative contributions of silicate particles in the suspended materials from the upper to lower reaches. Moreover, Ding et al. [46] reported that SiO_2 concentrations gradually increased downstream in the suspended sediments of the CR, suggesting the control of increasing clay minerals and decreasing carbonate mineral contents in the river suspended sediments.

3 Discussion

3.1 Causes of seasonal variations in riverine isotopic compositions

The radiogenic neodymium and strontium isotopes are generally considered to be reliable indicators of the provenance of sediments because the geological bodies have different Nd-Sr isotopic compositions that depend on their origins and ages [24–27]. Geologically, the CR Basin consists of variable strata from the Archean to the Quaternary period. However, different drainage basins in the CR Basin and its major tributaries consist of distinct tectonics and source rock types (Figure 1).

The upper Jinsha River Valley (Qinghai-Tibet Plateau) comprises metapsammite and metapelite, carbonate rocks and acidic igneous rocks, especially intermediate-acidic rocks formed during the Himalayan Stage. The sedimentary rocks in the drainage basins of the Jinsha, Yalong, Dadu and Minjiang rivers are characterized by Triassic low grade metamorphic clastic rocks to metamorphic rocks and carbonate rocks. Jurassic red sandstone is widely distributed in the Sichuan Basin. Paleozoic Carbonate rocks are widely spread throughout the basin and are particularly abundant in the southern parts of the upper reach (Yunnan, Guizhou and western Hunan provinces). The Emeishan basalt is considered the only large igneous province in China and one of the largest basalt provinces in the world. The Cenozoic felsic rocks, late Permian basalts and concomitant felsic rocks of the Emeishan large igneous province are typically distributed in the upper CR Basin.

The middle-lower basin primarily consists of Paleozoic marine and Quaternary fluviolacustrine sedimentary rocks together with intermediate to felsic igneous rocks that are common, but sporadic. Proterozoic metamorphic rocks are primarily distributed in the middle-lower CR drainage basin in areas such as the Qingling-Dabie Belt, Yuanjiang River, Xiangjiang River and the area to the east of Poyang Lake. The early Archean metamorphic rocks are scattered throughout the Qingling-Dabie belt [47]. Large amounts of intermediate, basic and ultrabasic rocks are widely distributed in the upper CR Basin resulting in isotopic compositions of this region having the highest $\varepsilon_{Nd}(0)$ values and the lowest $^{87}Sr/^{86}Sr$ ratios (Figure 1). In contrast, there are wide outcrops of Meso-Neoproterozoic low grade metamorphic rocks and Phanerozoic sedimentary rocks in the middle and lower reaches of the river, whereas intermediate, basic and ultrabasic rocks occur sporadically. Thus, materials derived from the middle and lower reaches produce the lowest $\varepsilon_{Nd}(0)$ values associated with the highest $^{87}Sr/^{86}Sr$ ratios.

The upper CR Basin plays an important role in this process because it supplies most of the suspended sediments to the ocean [16,17,48]. In particular, a large number of intermediate, basic and ultrabasic rocks in the upper CR Valley are especially vulnerable to weathering and erosion, and therefore make an important contribution to the Sr-Nd isotopic compositions of sediments in the CR. The collision of India and Asia resulted in the buildup of the Himalayas and the Tibetan Plateau, where incident solar heating in the summer drives strong atmospheric convection and rainfall associated with the Asian monsoon [49].

In June, there is usually a strong summer monsoon in the CR region. During this period, colder air is continuously transported southward by the westerly circulation, while warm and humid air is transported northward by the southwest monsoon. Thus, the cold and warm air converge in the CR-Huaihe River basins, which lead to heavy precipitation over the middle-lower CR Basin from mid-June to mid-July (known as the meiyu period). Conversely, rainfall in these regions is significantly reduced from the middle of July to mid-late August under the control of the western Pacific subtropical high. The rainband shifts toward the upper reaches of the CR, in northeast to southwest distribution, advances toward the Mingjiang, Tuojiang, Jialingjiang, lower Jinshajiang, and Hanjiang drainage area. At the same time, the middle-lower reaches of the CR and the eastern part of Sichuan Province receive little precipitation owing to the control of the subtropical high. In September, the rainband shifts toward the upper-middle reaches of CR [50]. Our study years were typical years of the long-term hydrological cycle. This is very similar to the long-term average water discharge and precipitation of the CR Basin [51]. During flood season (July to September) rainfall is concentrated in the upper CR, where there is local runoff generated and accompanying erosion [52]. There is a greater input of sediments, with the highest $\varepsilon_{Nd}(0)$ values and lowest $^{87}Sr/^{86}Sr$ ratios flowing from upstream into the lower CR. In contrast, physical erosion in the upper basin becomes weaker during the dry season. The association of the lowest $\varepsilon_{Nd}(0)$ values with the highest $^{87}Sr/^{86}Sr$ ratios of the suspended samples during the dry season is interpreted to reflect a decreasing supply of materials delivered from the upper drainage basin and increasing proportions of materials input from the middle-lower reaches of the CR to the suspended load. We suspect increased physical weathering of the upper basin during the rainy season to be the main cause of these seasonal changes. Furthermore, this is in good agreement with the results of our previous study in which clay minerals for suspended samples of the CR were investigated. The results of our previous study suggest that a strengthened physical erosion of source rocks in the upper basin during flood seasons results in an increased contribution of less weathered illite to the river system, while in the dry season, a decreasing supply of well-crystallized illite is delivered from the upper drainage basin and increasing proportions of poorly crystallized illite and kaolinite are input from the middle-lower reaches of the CR to the suspended load [53]. In addition, channel erosion of the lower reaches was enhanced after construction of the Three Gorges Dam, which increased the relative contribution of radiogenic Sr and non-radiogenic Nd to the suspended load during dry seasons [53].

Several investigations of the size fractions of the sediments have indicated that as the grain size decreases, the $^{87}Sr/^{86}Sr$ ratios increase in the acid-insoluble residual fraction and yield the highest value in the clay fraction [54]. The upper portion of the CR Basin primarily runs mainly through the Qinghai-Tibet Plateau and has a number of major tributaries, where the exposed rocks are clastic sedimentary (including Jurassic red sandstone), igneous and metamorphic. These rocks contain abundant micaceous, plagioclase minerals that easily produce abundant sediments with low $^{87}Sr/^{86}Sr$ ratios and coarse grain size by physical erosion and limited chemical weathering. Conversely, during the low flow season, the increased relative contribution of the clay fraction by the middle-lower reaches results in an increase in the $^{87}Sr/^{86}Sr$ ratios. We suggest that the grain size is controlled by the seasonal variations in water discharge and the origin of sediments. Conversely, the $^{87}Sr/^{86}Sr$ ratios of suspended samples in the CR ranged from 0.725352–0.738128 and presented wider variations, but relatively narrow ranges of grain size values (Figure 4). Suspended materials in the CR tend to be enriched with the fine fraction. Base on grain size analysis, the suspended samples were dominated by fine silts. The mean grain-size values of two typical samples were 10μm (August) for the flood season and 7.9 μm (December) for the dry season. Finally, the little variability of grain size in suspended sediments could not dominate the $^{87}Sr/^{86}Sr$ ratios varied significantly with season. Despite the different source rock composition being the first order control, the grain size may enhance the seasonal variability of the $^{87}Sr/^{86}Sr$ ratios of the suspended samples.

Yang et al. [41] measured Nd isotopes in SPM of the CR and found that the Nd isotopic compositions of the SPM samples demonstrated regular variations from the upper part to the estuary, showing decreasing $^{143}Nd/^{144}Nd$ ratios and $\varepsilon_{Nd}(0)$ values downstream (Figure 5) that correspond to $\varepsilon_{Nd}(0)$ values ranging from −9 to −11.5 and from −11.4 to −13 for the upper reaches and middle-lower reaches, respectively. While the $^{87}Sr/^{86}Sr$ ratios increased in a corresponding manner, the downriver increase of suspended $^{87}Sr/^{86}Sr$ ratios and decrease of suspended $\varepsilon_{Nd}(0)$ ratios in the CR mainstream water may reflect the increase of relative contributions of suspended materials from the upper reaches to the lower reaches [41]. The Nd and Sr isotopic compositions of the samples collected in this study are reported on a $\varepsilon_{Nd}(0)$ versus $^{87}Sr/^{86}Sr$ diagram (Figure 5) together with other relevant data, including the SPM of the CR from the upper to lower reaches [41] and modern sediments from a headwater region of the Changjiang, Huanghe, Mekong and Jinsha rivers [55]. As shown in Figure 5, modern

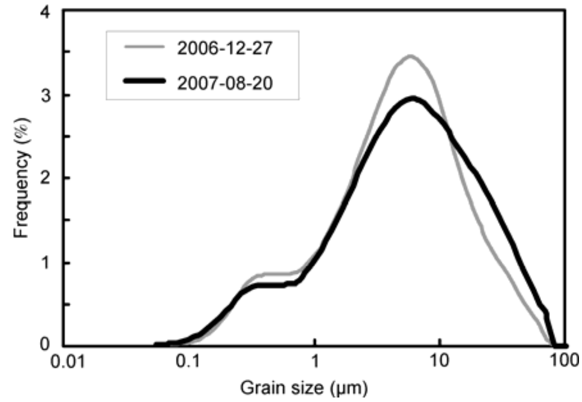

Figure 4 Grain size distributions of two typical samples selected from the SPM sample. The lighter-colored line is the dry season sample (27 Dec 2006) whereas the darker line is the flood season sample (20 Aug 2007). Grain size data from Mao et al. [53].

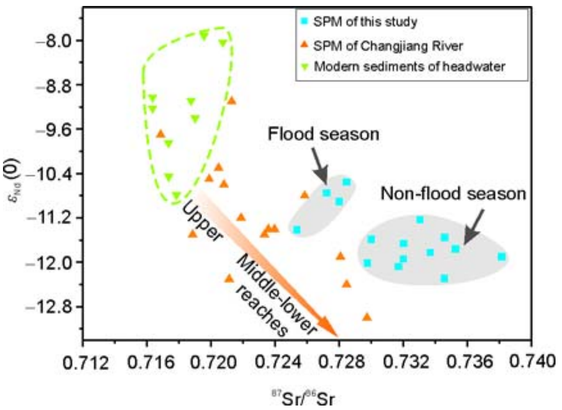

Figure 5 $^{87}Sr/^{86}Sr$ versus $\varepsilon_{Nd}(0)$ diagram of suspended loads observed in this study together with other relevant data: the SPM of the Changjiang River from the upper to lower reaches [41]; modern sediments from the headwater region of the Changjiang, Huanghe, Mekong and Jinsha rivers (rivers in the east Tibet Plateau) [55].

sediments from the headwater regions of these rivers and the SPM of the upper reaches of the CR had a high $\varepsilon_{Nd}(0)$ value and low $^{87}Sr/^{86}Sr$ ratio. The Nd and Sr isotopic compositions of the samples collected in this study of flood season plot closer to the SPM of the upper CR reaches observed by Yang et al. [41]. This observation is strongly correlated with our previous discussion, which suggests that there is strong physical erosion in the upper CR Basin during flood season that leads to the contribution of more sediments with the highest $\varepsilon_{Nd}(0)$ values and the lowest $^{87}Sr/^{86}Sr$ ratios from upstream into the lower CR.

3.2 Implications for tracing sediment sources

In the present study, seasonal variations in the Sr-Nd isotopic compositions of the suspended particulate matter of the lower CR were found to be controlled by erosion rates of the upper CR Basin, which are strongly affected by the monsoon climate. This is likely because a portion of the CR system drains regions of high relief, where greater runoff influenced by strong monsoon precipitation is the principal driver of physical erosion. Thus, the combination of high topographic relief and intense precipitation drives aggressive erosion. On geologic timescales, either an increase in monsoon strength or uplift of the Tibet Plateau could increase the erosion rates of the Upper CR. When the relief of the CR drainage basins was stable, changes in the strength of the Asian monsoon represented the first order force driving changes in the Sr-Nd isotopic compositions of the sediment deposited in the delta and offshore. Strong physical erosion caused by tectonic activities or monsoon climate along the eastern margin of the Tibetan Plateau is responsible for the high levels of sediments with the highest $\varepsilon_{Nd}(0)$ values and the lowest $^{87}Sr/^{86}Sr$ ratios being observed in the lowlands of the CR Basins or the marginal sea.

Furthermore, the CR transports a large number of sediments to the East China Sea (480×10^6 t/a), and several studies have suggested that the sediments in the CR could also be transported to the northern portion of the South China Sea [56,57]. Because the Nd-Sr isotopic signatures of the marine sediments have been useful for identifying their sources [29], we calculated the Sr and Nd isotopic composition end member values (weighted average) of the suspended matter exported by the CR to the ocean. The end member values of the CR SPM $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ were found to be 0.728254 and −11.26, respectively. As shown in Table 1, the end member values were close to the Sr-Nd isotopic values of the suspended samples collected from the CR during flood season.

4 Summary

This study provides insight into temporal variations in Sr and Nd cycling in suspended phase of the lower CR. The results revealed substantial seasonal variations in the Sr-Nd isotopic compositions of the suspended phase, which were correlated with discharge from the CR main stem. The samples collected had lower $^{87}Sr/^{86}Sr$ ratios and higher $\varepsilon_{Nd}(0)$ values during flood season than the dry seasons. The seasonal variations primarily reflect the controls of provenance rocks and erosion between different sub-catchments. The relative decrease in $^{87}Sr/^{86}Sr$ ratios and increase in $\varepsilon_{Nd}(0)$ values during the flood season can be interpreted to reflect an increase in the mechanical erosion rate in the upper basin and the contribution of more sediment from the upper reaches. We calculated the end member values of the $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ in the samples to be 0.728254 and −11.26, respectively.

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