

## Is strontium isotope record a strict proxy for chemical weathering rates during the late Miocene (Ocean Drilling Program Site 758A)?

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The strontium isotope ratios are examined in the shells of planktic foraminifer *Orbulina universa* in 20 samples from the late Miocene (8.03–6.36 Ma) sequence of Ocean Drilling Program (ODP) Site 758A, northeastern Indian Ocean. The Sr isotope ratios increased between 7.91 and 7.36 Ma and in the latest Miocene (6.48–6.36 Ma) reflecting high riverine flux from increased chemical weathering and high precipitation in the Himalayan region. The <sup>87</sup>Sr/<sup>86</sup>Sr values decreased during 7.36 to 6.48 Ma coincident with the main phase of Chron-6 negative carbon shift. The carbon shift reflects high delivery of nutrients to the oceans. This contradicts the use of Sr isotope record as a strict proxy for global chemical weathering rates. The Sr isotope ratio should be used with great care in understanding the climatic–tectonic connections.

THE climate of the Earth has fluctuated during the Neogene owing to tectonic<sup>1–3</sup> and/or non-tectonic disturbances<sup>4</sup>. Berger *et al.*<sup>5</sup> observed that the relationship between tectonics and climate is not simple because of the poor time constraint of these events. In spite of these ambiguities, the paleoceanographers continue to seek climate–tectonics connections on long-term changes. The tectonic uplift of the Himalayan–Tibetan Plateau has been linked to Miocene and Pliocene glaciations<sup>2,6</sup> and development of the Indian monsoon circulation<sup>7</sup>. Significant increase in upwelling intensity<sup>8–11</sup> and enhanced delivery of terrigenous sediment to the northern Indian Ocean<sup>12</sup> are clues to the Himalayan uplift-related climate change between 9.0 and 6.0 Ma. Chemical weathering associated with the rising mountain ranges has resulted from atmospheric CO<sub>2</sub> draw-down in the middle and late Miocene<sup>1,13–15</sup>, enhancing the cooling of the atmosphere. The vegetation cover in the Indian subcontinent changed from C<sub>3</sub> forests to C<sub>4</sub> grasses in the latest Miocene as is evident in *d*<sup>13</sup>C of pedogenic carbonate, which is related to a change in monsoonal precipitation and a decline in the atmospheric CO<sub>2</sub> (refs 16–18). The increased rates of mountainous uplift and resultant chemical weathering in the

late Miocene may have controlled changes in strontium isotopic composition of sea water<sup>19</sup>.

The use of strontium isotope ratios as a proxy to silicate chemical weathering rates<sup>13,20–22</sup> related with the major Himalayan uplift and erosion is the topic of recent debate. Hodell *et al.*<sup>19</sup> related the changes in strontium isotope ratios with the continental erosion accelerated by tectonic uplifts in the mountain ranges. In this study we used <sup>87</sup>Sr/<sup>86</sup>Sr ratios of planktic foraminifer *Orbulina universa*, combined with stable isotope record of benthic foraminifer *Cibicides wuellerstorfi*<sup>11</sup> from the late Miocene interval (8.03–6.36 Ma) at Ocean Drilling Program (ODP) Site 758A, north-eastern Indian Ocean to understand whether the Sr isotope record is a strict proxy for chemical weathering rates. This would help comprehend a link between Himalayan tectonics, monsoonal climate and delivery of weathering products to the marine realm.

ODP Site 758A is located on the western flank of the Ninetyeast Ridge at 5°23.05'N, 90°21.67'E, and a water depth of 2924 m in the northeastern Indian Ocean (Figure 1). At present the site lies below Equatorial Waters (EQW) with a seasonally changing current pattern<sup>23</sup>. During the northern winter (NE monsoon) the North Equatorial Current (NEC) flows to the west, while during the summer surface currents are controlled by the eastward flowing turbulent drift current of the SW monsoon<sup>23,24</sup>. The latter causes intense upwelling, leading to seasonally high productivity and a distinct fauna and flora<sup>25</sup>. Near the Miocene–Pliocene boundary the site was at ~3°N (ref. 26). The late Miocene section consists

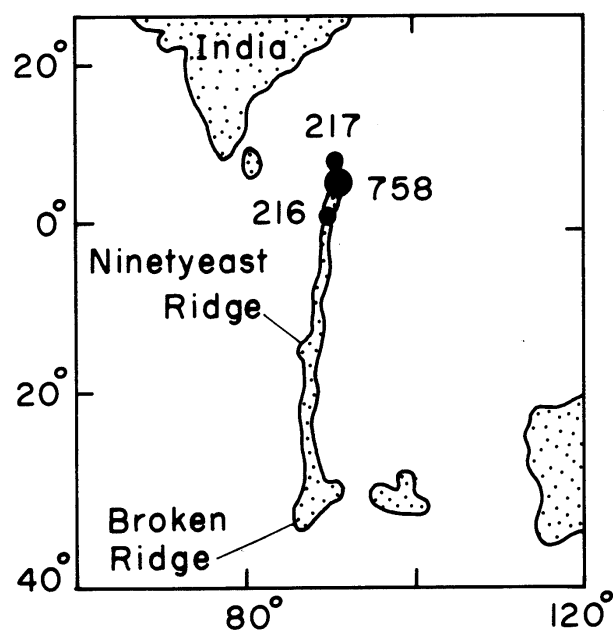


Figure 1. Location map of ODP Site 758A, northeastern Indian Ocean.

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of clay-rich, foraminiferal nannofossil ooze. The age model is based on nannofossil events<sup>26</sup> and interpolated numerical ages<sup>11</sup> are according to Berggren *et al.*<sup>27</sup>. The sedimentation rate was about 11.9 m/m.y. in the study interval<sup>11</sup>.

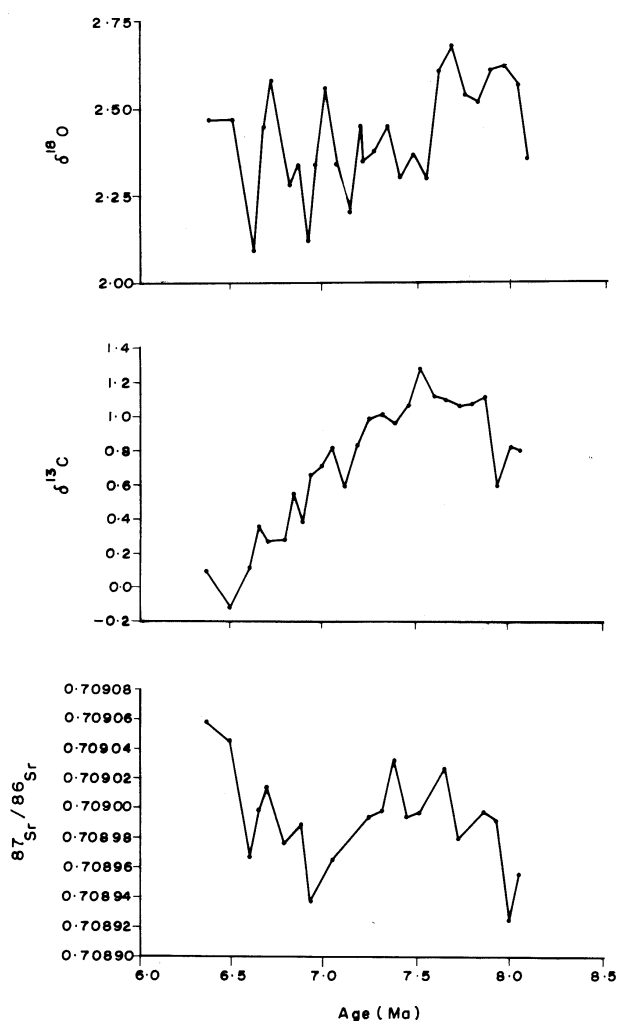
We analysed Sr isotope ratios in the shells of planktic foraminifer *O. universa* from 20 samples from the latest Miocene (8.03–6.36 Ma) sequence at ODP Site 758A. The test chambers of about 40 to 50 *O. universa* specimens were broken mildly using glass rods before cleaning them by methanol and ultrapure water (MQ-water) in an ultrasonic bath. The process was repeated three times to avoid contamination. About 1 to 2 mg of the cleaned sample was dissolved in 0.2 M hydrochloric acid and kept overnight. Weak acid was used to prevent dissolution of any non-carbonate material. Sr was separated using 1 ml (3 mm × 12 cm) column of Dowex 50 w × 8 cation exchange resin. Strontium isotopic measurements were performed on VG 354 TIMS in dynamic triple collector mode. Inter-run precision was estimated from 16 analyses of International Standard NBS-987. Each set of samples was accompanied by an NBS-987 standard. The mean value of NBS-987 obtained was 0.710221 with 2s SD of better than ± 0.000022. This mean value is very close to the reported value of NBS-987 (0.710235). The <sup>87</sup>Sr/<sup>86</sup>Sr values have been combined with stable oxygen and carbon isotope data of benthic foraminifer *Cibicides wuellerstorfi*<sup>11</sup> and plotted in Figure 2 to understand tectonic–climatic controls of Sr isotope ratios during the late Miocene.

The seasonal reversal of the Indian Monsoon System is one of the most fascinating features of the earth's climate. The long-term changes in monsoonal circulation have been linked with the presence of the Himalayan–Tibetan Plateau at least since the late Miocene<sup>28–30</sup>. The rapid uplift of the Himalayas has been suggested to intensify monsoonal circulation which accelerates the seasonal upwelling and oceanic productivity<sup>6,8–10,31,32</sup>. The intense monsoon would enhance precipitation rates and chemical weathering in the Himalayan region<sup>1,13</sup>. This would increase the riverine flux from the mountain terrain and high delivery of radiogenic strontium to the Indian Ocean<sup>19</sup>.

The late Miocene high <sup>87</sup>Sr/<sup>86</sup>Sr values in the Pacific and Indian Oceans have been attributed to greater rates of chemical weathering of the continents<sup>19,33,34</sup>. It has been presumed that anomalously high <sup>87</sup>Sr/<sup>86</sup>Sr values in Ganga and Brahmaputra rivers result from exceptionally rapid erosion rates in the Himalayas due to rapid uplift and increased monsoonal rainfall<sup>21,35</sup>. A major episode of erosion and weathering in the Himalayan–Tibetan region has been related with the rapid increase in *d*<sup>87</sup>Sr since the last 20 Ma (refs 13, 36). However, Pagani *et al.*<sup>37</sup> have recently found a lack of coherence between Sr and pCO<sub>2</sub>, and argued against the use of Sr isotope re-

cord as a proxy for global chemical weathering rates in the early and middle Miocene (21–17 Ma and 14–10 Ma). In this study, we made an effort to corroborate their argument.

At Site 758A <sup>87</sup>Sr/<sup>86</sup>Sr values show a long-term increase with fluctuations through 8.03 to 6.36 Ma, from a low of 0.708925 at 7.98 Ma to a high 0.709058 at 6.36 Ma (Table 1, Figure 2). Within this long-term increase a downturn is visible during 7.36 to 6.5 Ma (Figure 2). Farrell *et al.*<sup>34</sup> also observed a flat slope in their Sr isotope record in the interval 7 to 6.5 Ma. Our average Sr ratios are slightly higher than those of Farrell *et al.*<sup>34</sup>. This could either be due to interspecific differences, since Farrell *et al.* used mixed species of planktic foraminifera in this interval, or due to inter-laboratory analytical differences. A rapid increase in Sr ratio at 6.5 Ma observed by Farrell *et al.*<sup>34</sup> is also visible in our record. The benthic carbon isotope values show a continuous decrease from 7.5 to 6.5 Ma, the so-called



**Figure 2.** <sup>87</sup>Sr/<sup>86</sup>Sr values of *O. universa* combined with the benthic foraminiferal carbon and oxygen isotope values<sup>11</sup> for Site 758A plotted against ages during the late Miocene.

**Table 1.**  $^{87}\text{Sr}/^{86}\text{Sr}$  values of *O. universa* from ODP Site 758A during the late Miocene

Sample no.	Age (Ma)	$^{87}\text{Sr}/^{86}\text{Sr}$
10H-1, 06-08	6.357	0.709058
10H-1, 72-74	6.48	0.709045
10H-2, 01-03	6.587	0.708967
10H-2, 81-83	6.637	0.708998
10H-3, 01-03	6.681	0.709014
10H-4, 01-03	6.774	0.708976
10H-4, 73-75	6.868	0.708989
10H-5, 72-74	6.912	0.708937
10H-6, 01-03	7.028	0.708965
10H-7, 01-03	7.223	0.708994
11H-2, 01-03	7.294	0.708998
11H-2, 72-74	7.359	0.709033
11H-3, 02-04	7.432	0.708994
11H-3, 72-74	7.496	0.708997
11H-4, 01-03	7.633	0.709027
11H-5, 02-04	7.706	0.70898
11H-5, 73-75	7.843	0.708998
11H-6, 73-75	7.909	0.708992
11H-7, 02-04	7.981	0.708925
11H-CC, 01-03	8.032	0.708956

Chron-6 carbon shift. The benthic  $d^{18}\text{O}$  values are relatively low between 7.5 and 6.5 Ma and strongly fluctuate from 7.10 to 6.5 Ma, indicating highly unstable conditions marked by major glacial–interglacial cycles<sup>11</sup>.

Thus if we follow the model suggested by Raymo and coworkers<sup>1,6,13</sup>, then the high  $^{87}\text{Sr}/^{86}\text{Sr}$  values between 7.91 and 7.36 indicate increased rates of chemical weathering due to rapid Himalayan uplift,  $\text{CO}_2$  removal and consequent atmospheric cooling. The increased weathering in the Himalayan region would enhance nutrient flux causing low  $d^{13}\text{C}$  values rather than high  $d^{13}\text{C}$  values, as are visible in this interval at Site 758A (Figure 2). This relationship is also not visible during 7.36 to 6.5 Ma when Sr isotope ratios decrease along with the  $d^{13}\text{C}$  values (Figure 2). The interval 7.36 to 6.5 Ma coincides with two important events that support increased weathering rates in the Himalayas and intense monsoonal circulation. These are: (1) a major increase in mass accumulation rates (MAR) in the Indus and Bengal fans<sup>12</sup>, and (2) a change from  $\text{C}_3$  to  $\text{C}_4$  vegetation<sup>17</sup>. The dramatic expansion of  $\text{C}_4$  biomass at ca 7.4 Ma was caused by a decline in  $\text{pCO}_2$  in the latest Miocene<sup>17</sup>, which could be linked with the increased chemical weathering rates due to rapid Himalayan uplift<sup>13</sup>. This contradicts the positive relationship between Sr isotope record and chemical weathering rates. We thus support the argument of Pagani *et al.*<sup>37</sup> against the use of the Sr isotope record as a strict proxy for global chemical weathering rates. We should seek other factors to explain changes in strontium isotope ratio of sea water. However, our study is not intended to underestimate

the relationship of Sr isotopes with chemical weathering rates in other time intervals and other areas as has been suggested by Raymo and coworkers<sup>1,6</sup>. In any event, given the shortness of the interval under consideration and the very long residence time of Sr in the ocean, the decrease in  $^{87}\text{Sr}/^{86}\text{Sr}$  during 7.36 to 6.5 Ma must be authenticated by high-resolution replicate analysis at numerous sites. The late Miocene Chron-6 carbon shift may have been caused by many factors, including increased surface productivity, a change in vegetation and increased terrigenous flux.

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