

27. THE LATE NEOGENE $^{87}\text{Sr}/^{86}\text{Sr}$ ISOTOPIC RECORD IN THE WESTERN ARABIAN SEA, SITE 722¹

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ABSTRACT

Continuous magnetostratigraphy and biostratigraphy made it possible to construct a detailed late Neogene record of $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios of ocean water, as measured in the tests of planktonic foraminifers. Sediments recovered during Leg 117, in the western Arabian Sea, provide a continuous, high resolution sedimentary record from the early Miocene to present. The late Miocene to Recent is marked by rapidly increasing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in seawater, which results in a chronostratigraphical resolution varying from 0.2 Ma to 1.5 Ma. The $^{87}\text{Sr}/^{86}\text{Sr}$ seawater curve has a stepwise character similar to the one determined by DePaolo on Site 590B, in the Tasman Sea, and to the one determined by McKenzie on Site 653A, in the Mediterranean, confirming its use as a chronostratigraphic tool for this time span. Periods of rapid increase in the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio of seawater are correlated with important changes in tectonic and climatic conditions.

Experiments showed that bulk carbonate sediment samples have differing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from those of planktonic and benthic foraminifers from the same depth.

INTRODUCTION

The evolution of strontium isotope ratios in seawater, as recorded in marine carbonates is a major stratigraphic tool for dating sediments within certain periods of geological time (Burke et al., 1982; DePaolo and Ingram, 1985; Hess et al., 1986; Palmer and Elderfield, 1985; Elderfield, 1986). Various studies (DePaolo and Ingram, 1985; DePaolo, 1986; Koepnick et al., 1988) indicate that the Neogene can be dated with relative high precision (0.1–0.5 Ma), during those periods with rapidly increasing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in seawater.

The strontium isotopic ratio has been shown to be constant throughout the oceans at any one time, because of the long residence time of Sr (4–5 Ma) relative to the mixing time of the oceans (1000 yr; Broecker and Peng 1982). Variations of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in seawater through time are caused by Sr input from different sources, each with a specific isotopic ratio. These are: continental radiogenic input (Average $^{87}\text{Sr}/^{86}\text{Sr} = 0.711$), mid-oceanic hydrothermal input (Average $^{87}\text{Sr}/^{86}\text{Sr} = 0.703$) and mid-oceanic hydrothermal input 0.708 (Brass, 1976; Elderfield, 1986). The resulting Sr isotopic signal of seawater is thus dependent on a complexity of factors, which affect source as well as input: including plate tectonics, rate of seafloor spreading, orogenic activity, climate, sea-level changes, glacial activity, and (paleo)oceanography. For a full discussion on the interaction of these factors one is referred to Veizer (1989), Koepnick et al. (1988), Elderfield (1986), DePaolo (1986), Armstrong (1971), and Raymo et al. (1988).

The Sr isotopic record for the Neogene as determined by DePaolo (1986) on DSDP Hole 590B bulk carbonate samples demonstrated the potential of Sr isotopes as a chronostratigraphical tool, especially for the late Miocene–early Pliocene and late Pliocene–Pleistocene intervals. This paper focuses on the same time intervals with the aim to obtain a $^{87}\text{Sr}/^{86}\text{Sr}$ isotope stratigraphy with a higher resolution for the last 6 Ma.

This paper reports the $^{87}\text{Sr}/^{86}\text{Sr}$ seawater curve from a middle Miocene–Pleistocene section of Site 722 (Fig. 1) measured on the calcareous tests of cleaned planktonic foraminifers.

Site 722 in the western Arabian Sea drilled a detailed late Neogene sedimentary record with abundant planktonic foraminifers and continuous biostratigraphic and magnetostratigraphic age control (Shipboard Scientific, Party 1989; Berggren et al., 1985). The western Arabian sea is a high-productivity upwelling area under direct influence of monsoonal circulation and climate. The high sediment accumulation rates (up to 4.9 cm/k.y.) provide a detailed sedimentary record for the middle Miocene–Pleistocene interval. Therefore, these sediments are suitable for using the variation of Sr isotopic ratios in seawater through time, both to establish a fine-tuned dating tool and to better understand the mechanisms which may cause these variations.

METHODOLOGY

Samples and Preparation

Planktonic foraminifers from Holes 722A and B (Table 3) were separated from the bulk sediment using standard washing and sieving methods. The foraminifers were handpicked from the $>250\ \mu\text{m}$ fraction, and inspected under the light microscope for preservation quality.

The first 231.70 mbsf were sampled about every 10 m (core catchers). Below 231.70 mbsf in Holes 722A and 722B, only intervals with abundant planktonic foraminifers were sampled. The foraminifers were cleaned for 10 min in an ultrasonic bath and centrifuged over a $30\ \mu\text{m}$ sieve to remove clay and other fine material. After drying and weighing, the samples (5–10 mg) were dissolved in 5 molal acetic acid for 4–10 hr at room temperature. After complete dissolution of the calcium carbonate, the liquid was pipetted and centrifuged to remove remaining clay particles (5–10 min, 6000 rev./min). Again the liquid was pipetted, and evaporated at 110°C . Next step before concentrating the Sr fraction on cation exchange columns is the conversion of the acetates into chlorides by adding 1–2 mL 3.25 M HCl and drying at 110°C . The samples were diluted to $50\ \mu\text{L}$ 1.50 M HCl sample solutions, with a maximal concentration of 4 mg Ca. These were loaded onto high-pressure (0.6–0.8 bar) cation exchange columns; the columns dimensions are $150 \times 3\ \text{mm}$ with a content of 1 mL. The resin used is Aminex Q155. To obtain a

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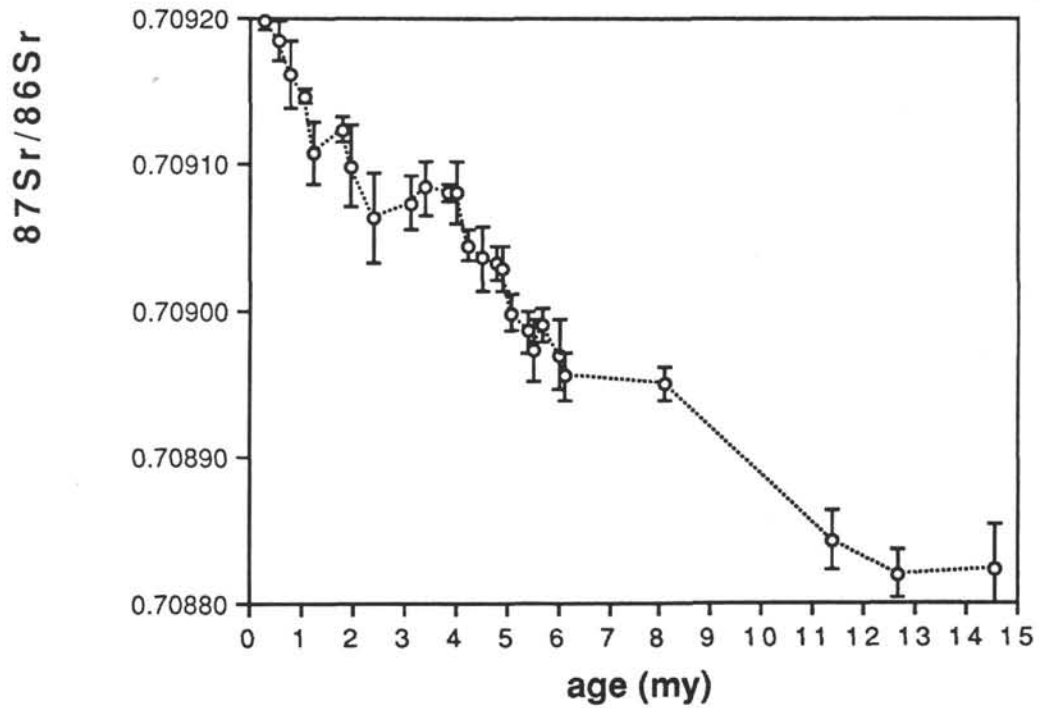
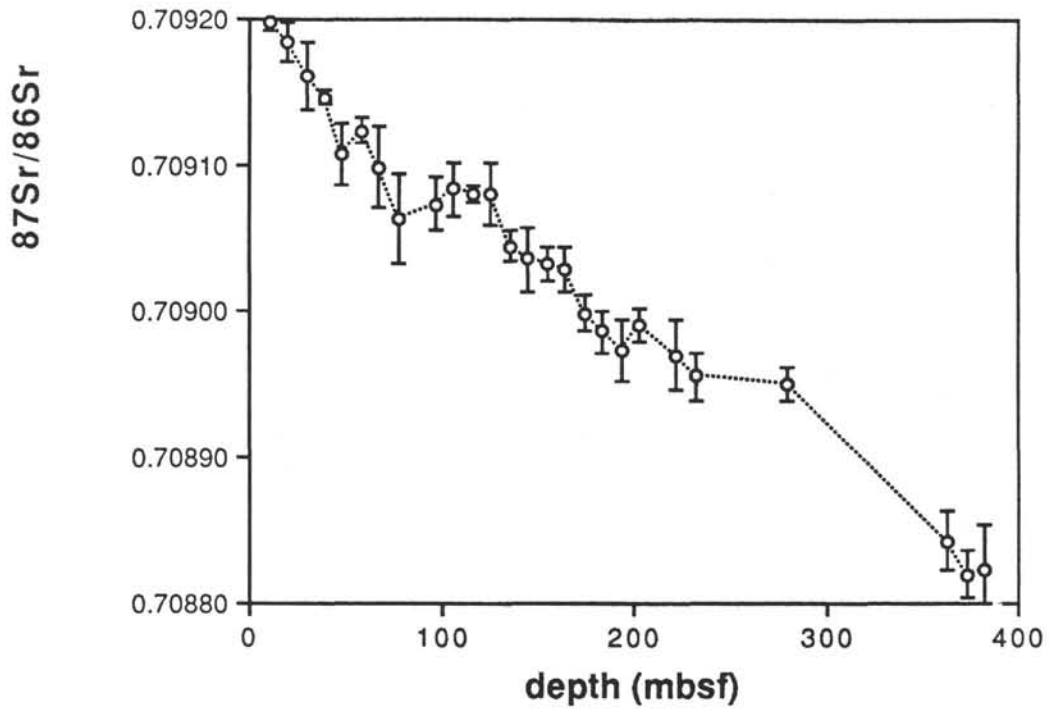


Figure 1. Graph of the measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios vs. depth (mbsf) at Site 722 and vs. age (m.y.) based on the Hole 722A and B biostratigraphy and magnetostratigraphy. From ~ 5.5 Ma to ~ 4 Ma and from 2.4 Ma to the late Pleistocene, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio increases rapidly. Between ~ 8.5 Ma and ~ 5.5 Ma, the ratio is nearly constant around 0.708955 and between ~ 4 Ma and 2.4 Ma it varies only slightly around 0.709080 (NBS 987 = 0.701258).

good separation of Sr from Ca and Y, 1.50 M HCl is used as eluent. Only 2 mL 1.50 M HCl is required, because of the capacity of the columns. Hence the blank for the chemical procedure is very low (0.012 ng strontium). After collection of the Sr fraction, the samples are dried and converted to nitrates.

The Sr prepares were loaded manually with teflon tube micropipet onto zone-refined rhenium filaments. The samples were mounted on a sample turret which contains 12 samples and a NBS 987 SrCO₃ standard, and measured using a Finnigan MAT 261 fixed multicollector (nine Faraday cups) mass spectrometer (Isotope Geology Laboratory, Amsterdam). The Sr isotopic ratios were determined using a Sr double-jump experiment, measuring 80 scans (intergration time of 16 s per peak/scan) at a current of approximately $4-6 \times 10^{-11}$ A for mass 88. Data were corrected for any Rb interference and normalized to an ⁸⁸Sr/⁸⁶Sr ratio of 8.37521. NBS 987 SrCO₃ standard was measured as 0.710258 ($n = 10$) with a standard deviation of 0.000015 as an reproductive external precision during the course of the analyses. For a single analysis the average in-run precision for the standard was 0.000015 and for the samples 0.000016 ($n = 26$).

Experimental

Several workers have used different kinds of sample material, for example, Burke et al. (1982) and DePaolo (1986) took bulk samples of foraminifer-nannofossil ooze and chalk, whereas, Elderfield et al. (1982), Palmer and Elderfield (1985), and Hess et al. (1986) used carefully checked planktonic foraminifers. The use of bulk sample material holds the risk of including uncertain, diagenetic effects. In addition to the diagenetic components, bulk sediments can contain clay- and silt-size eolian, detrital calcite (for some sites of Leg 117 more than 20%, Shipboard Scientific Party, 1989) which carries a different Sr isotopic signature from that of the seawater in which it was deposited.

To describe these different contributions to the Sr isotopic ratio of the bulk (carbonate) sediments, the various Sr isotope carriers were investigated. Section 722B-7H-CC was divided into six fractions (Table 1).

There are distinct differences between foraminifers and bulk sample Sr isotopic ratios and also between the sieved bulk fraction and the foraminifers. It is evident that the Sr isotopic ratios of both the benthic and the planktonic foraminifers are more radiogenic than those of the bulk sample and the sieved bulk sample. Furthermore, there seems to be no variation in the Sr isotopic ratios between benthic and planktonic foraminifers.

The minor variations in Sr isotope ratios obtained from the same sample, using different acids for dissolution of the bulk sample calcium carbonate (1 N HCl and 5 N HOAc), indicate that there is no leaching of clay minerals. The leaching of clay minerals with 1 N HCl and 5 N HOAc was investigated in another experiment (Table 2).

Again, the foraminifers have a higher ⁸⁷Sr/⁸⁶Sr ratio than the bulk sample. The bulk sample has the same ratio for 1N HCl and 5N HOAc dissolved fractions. Because of the high Sr isotopic ratio of land-derived clay minerals (see residue of the bulk sample = 0.712202), the bulk sample Sr isotopic ratios would have been higher than the foraminifer Sr isotopic ratio, if clay minerals had been leached with the chloric and acetic acid. However, because it actually is lower, it seems safe to conclude that virtually no other material than CaCO₃ is dissolved with both the 1 N HCl and the 5 N HOAc. Moreover, it is not the contribution of clay minerals in the bulk sample that influences the ⁸⁷Sr/⁸⁶Sr ratio of the contemporaneous seawater as recorded in the carbonate, but rather it is the detrital calcite.

Although there is no significant difference in Sr isotope ratios between benthic and planktonic foraminifers, planktonic

Table 1. Comparison of the Sr isotopic ratios of the different constituents of sample from Section 722B-7H-CC; results from experiments with different sample fractions and acids. The measured ⁸⁷Sr/⁸⁶Sr show a distinct difference between bulk sample and foraminifers.

Fraction	Sample 722B-7H-CC		
	Acid	⁸⁷ Sr/ ⁸⁶ Sr	2σ (× 10 ⁻⁶)
1) total bulk	1 N HCl	0.709061	13
2) total bulk	5 N HOAc	0.709075	42
total bulk	5 N HOAc	0.709072	10
3) < 20 μm bulk	1 N HCl	0.709092	24
4) < 20 μm bulk	5 N HOAc	0.709071	26
5) Benthic foram.	5 N HOAc	0.709104	20
6) Plankt. foram.	5 N HOAc	0.709105	18
Plankt. foram.	5 N HOAc	0.709108	9

Table 2. Comparison of results from experiments in which the chemical methods are checked for possible leaching of silicate materials; the bulk samples show no sign of contributions by leached clay minerals. The difference between bulk sample and foraminifers Sr isotopic signal is caused by the presence of detrital calcite with a lower ⁸⁷Sr/⁸⁶Sr ratio in the bulk sample.

Fraction	Sample 722B-3H-CC		
	Acid	⁸⁷ Sr/ ⁸⁶ Sr	2σ (× 10 ⁻⁶)
1) mixed forams	1 N HCl	0.709163	16
2) < 40 μm bulk	1 N HCl	0.709097	21
3) < 40 μm bulk	5 N HOAc	0.709091	18
4) residue	HF/HNO ₃	0.712202	60

species were used in conformity with other investigators (Hess et al., 1986; Palmer and Elderfield, 1985).

RESULTS

The ⁸⁷Sr/⁸⁶Sr ratio shows a stepwise evolution from 15 Ma toward the present (Fig. 1). This confirms earlier observations of, among others, DePaolo (1986) and Hodell et al. (1989). The curve shows intervals of rapidly increasing ⁸⁷Sr/⁸⁶Sr ratios with alternating intervals of more or less constant ratio (plateaus). The denser sampling, 22 sample points over the last 6 m.y., makes it possible to distinguish more variations in the ⁸⁷Sr/⁸⁶Sr ratio record in the late Miocene-Pleistocene interval (Table 3).

The last 6.15 m.y. are characterized by three distinct episodes in the evolution of the Sr isotope curve of seawater. First, a steady increase from 6.15 Ma until 4.0 Ma. Second, a stabilization and/or decrease from 4.0 Ma until 2.41 Ma. Third, a steep two-step increase from 2.41 Ma until 0.28 Ma. Within this main trend finer subdivisions can be distinguished: the periods with rapid increasing ⁸⁷Sr/⁸⁶Sr ratios are 6.15-5.68 Ma, 5.53-4.89 Ma, 4.23-3.83 Ma, 2.41-1.76 Ma, and 1.21-0.28 Ma. Periods with slowly increasing and/or stable ratios are: 4.89-4.23 Ma, 3.83-3.38 Ma, 3.38-2.41 Ma, and 1.76-1.21 Ma.

DISCUSSION

The ⁸⁷Sr/⁸⁶Sr seawater curves for the late Neogene as determined by DePaolo (1986), Hodell et al. (1990), and McKenzie et al. (1990) all show a similar (averaged) main trend for the last ~6 Ma. Rapid increases between ~6 and ~4.4 Ma and ~2.4 Ma and the present, constant ratios between ~4.4 and 2.4 Ma. The curve described in this paper (Fig. 1), which is based on sediments from the Arabian Sea, shows the same main features (see "Results"). The sediments on which

Table 3. Measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the middle Miocene to Pleistocene planktonic foraminifers from Site 722 (Shipboard Scientific Party, 1989).

Corecatcher	Depth (m)	Age (my)	$^{87}\text{Sr}/^{86}\text{Sr}$	2σ ($\times 10^{-6}$)
722A-1	9.8	0.28	0.709199	7
722A-2	19.4	0.55	0.709185	14
722A-3	29.0	0.76	0.709162	23
722A-4	38.6	1.05	0.709147	4
722A-5	48.2	1.21	0.709108	21
722A-6	57.5	1.76	0.709124	9
722B-7	67.2	1.95	0.709099	28
722A-8	76.8	2.41	0.709063	31
722A-10	96.2	3.13	0.709074	18
722A-11	105.9	3.38	0.709084	18
722A-12	115.6	3.83	0.709080	6
722A-13	125.3	4.00	0.709080	21
722A-14	134.9	4.23	0.709045	10
722A-15	144.6	4.49	0.709036	22
722A-16	154.3	4.80	0.709033	11
722A-17	163.9	4.89	0.709029	15
722A-18	173.6	5.08	0.708999	13
722A-19	183.3	5.39	0.708986	14
722A-20	193.0	5.53	0.708973	21
722A-21	202.7	5.68	0.708990	11
722A-23	222.0	6.00	0.708970	24
722A-24	231.7	6.15	0.708955	16
722B-29	280.0	8.10	0.708950	12
722B-38	362.8	11.40	0.708843	20
722B-39	372.5	12.66	0.708820	16
722B-41	391.8	14.58	0.708823	31

DePaolo (1986) determined the $^{87}\text{Sr}/^{86}\text{Sr}$ seawater ratios were derived from the southwest Pacific (DSDP Hole 590B), those on which Hodell et al. (1990) determined their curve also, and from the Caribbean and the South Atlantic (DSDP Sites 590, 588, 502, and 519). McKenzie et al. (1990) used sediments from the Mediterranean (ODP Hole 653A). All these different geographic locations provide the same (global) $^{87}\text{Sr}/^{86}\text{Sr}$ seawater evolution curve.

Variations in the nature of the riverine Sr flux to the oceans determine the changes in $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of seawater in time (Palmer and Elderfield, 1985; Wadleigh et al., 1985; and Palmer and Edmond, 1989). This means that either the continental flux of Sr must change or the isotopic composition of this flux or a combination of both.

The importance of Cenozoic mountain building, and in particular the uplift of the Himalayas, as a primary cause of changes in the geochemical budget of the oceans is stressed by Raymo et al., 1988. This would increase the radiogenic Sr flux to the oceans because of intensified weathering due to tectonic uplift. Moreover, the onset of the monsoonal circulation would increase the chemical weathering in the Himalayas.

Hodell et al. (1989) illustrated the stepwise character of the Sr isotope curve on the rapid increase between 5.5 and 4.5 Ma and suggested an evolution from one steady state in the oceans to another, with disturbances caused by "sudden" increases in residence time of Sr in the oceans: 4–5 m.y.). They used the "pulse-like" increase between 5.5 and 4.5 Ma, of 1×10^{-4} in 1 m.y., to model changes in Sr input and oceanographic Sr output, necessary to cause such a rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio.

Distinct pulses would occur at about 12 Ma, 5.53 Ma, and 2.41 Ma (Fig. 1) which resulted in steep increases in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio. As mentioned earlier, it is difficult to attribute these shifts in ^{87}Sr input to one single factor, especially because of the magnitude of the change needed for such a global increase in seawater Sr isotopic ratios.

Several authors noticed a covariation between $^{87}\text{Sr}/^{86}\text{Sr}$ seawater isotopic ratios and eustasy (Spooner, 1976; DePaolo,

1986; Koepnick et al., 1988; and Ludwig et al., 1988). DePaolo (1986) correlated sharp increases in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio to sea-level lowstands and periods with stable ratios to periods of sea-level highstands. This indeed matches quite well, e.g., the sharp increases at 5.5 Ma and at 2.41 Ma (Fig. 1) coincide with the eustatic falls related to increased glaciation in both Northern and Southern hemispheres (Haq et al., 1987; van Hinte, 1982; DePaolo, 1986; Shackleton and Kennett, 1975). This correlation is not perfect, however and other mechanisms must effect the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of seawater.

From 2.41 Ma until Recent the intensified frequency of glacial-interglacial sea-level cycles (Ruddiman, 1989; Raymo et al., 1989; and Shackleton et al., 1984), is superimposed on the increased rates of uplift of various mountain belts around the globe (Himalayas, Andean region, Rocky Mountains) resulting in a higher flux from the continents due to enhanced weathering (Raymo et al., 1988).

Tectonic uplift was probably the driving force for increased runoff during the past 15 Ma, but certainly major modifications were caused by the changes in climate, glacial-interglacial sea-level cycles, monsoonal circulation, and paleoceanography (e.g., changes in the calcite compensation depth (CCD)). As discussed below, the sharp changes at about 12 Ma, 5.6 Ma, and 2.4 Ma probably all three reflect different combinations of global geological changes.

During the middle Miocene the uplift of the Himalayas triggered the monsoonal circulation in the northwestern Indian Ocean (Gansser, 1964). The effects would be twofold. First, due to the uplift enhanced mechanical weathering. Second, due to the onset of the monsoons more intense chemical weathering, together resulting in a much higher amount of oceans and a rapid increase in the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratio.

The increase at 5.5 Ma which is discussed and modeled by Hodell et al. (1989) can also be attributed to a change in the nature of the Sr riverine flux due to glacio-eustasy: a phase of ice-growth in the Antarctic (Shackleton and Kennett, 1975) and arctic ice buildup together with a sea-level fall (Haq et al., 1987).

The late Pliocene (2.6–2.4 Ma) is a time of increased Northern and Southern Hemisphere glaciation. Also, the frequency and magnitude of sea-level change increase (Ruddiman et al., 1989; Raymo et al., 1989). According to Raymo et al. (1988), this was accompanied, or caused, by global tectonism (increased uplift of the Himalayas, Andes, and Rockies). The result is an increase in continental Sr flux to the oceans and a rapid increase in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of seawater.

CONCLUSIONS

The $^{87}\text{Sr}/^{86}\text{Sr}$ curve measured on planktonic foraminifers from late Neogene Indian Ocean sediments confirms the curve of DePaolo (1986) based on DSDP Hole 590B in the Tasman Sea, and shows a higher resolution during the last 6 Ma. It is very similar to the Pliocene-Pleistocene curve of MacKenzie et al. (1990) from Hole 653A in the Tyrrhenian Sea and the averaged 9–2 Ma curve of Hodell et al. (1989) representing various DSDP sites in the Caribbean (Site 502), South Atlantic (519), and southwest Pacific (588, 590, and 593). The fact that these four $^{87}\text{Sr}/^{86}\text{Sr}$ curves are so alike confirms its value as a chronostratigraphical tool for the Neogene.

The general $^{87}\text{Sr}/^{86}\text{Sr}$ seawater trend during the last 15 m.y. is one of stepwise increase, with periods of steep increase, suited for high resolution chronostratigraphy, and periods with stable ratios, where the chronologic resolution is low.

The more detailed analyzed period from 6 Ma to Recent shows pulse-like increases on a scale of 200 k.y. (for instance, 5.08–4.89 Ma). However, it is difficult to explain these events by single mechanisms like glacial-interglacial sea-level variations. Probably a combination of tectonism, climate changes, and gla-

cial-interglacial sea-level cycles controls the changes in the nature and the quantity of riverine input and thus the geochemical cycles of the oceans (Weizer, 1989; Hodell et al., 1989; Raymo et al., 1988; and Delaney and Boyle, 1988). Correlations between glacial-interglacial sea-level cycles and rapid $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio increases in seawater can only be tested on detailed, well dated sections (McKenzie et al., 1990).

The importance of checking the sample quality needs to be stressed. Not only diagenetic effects can change the $^{87}\text{Sr}/^{86}\text{Sr}$ composition of the bulk sample, but also the presence of detrital calcite can decrease the ratio of the bulk carbonate samples compared to the signal obtained from the cleaned foraminiferal separates.

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