



ELSEVIER

Earth and Planetary Science Letters 144 (1996) 9–19

EPSL

Nd isotope evidence for the evolution of the paleocurrents in the Atlantic and Tethys Oceans during the past 180 Ma

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Received 3 March 1996; revised 18 July 1996; accepted 30 July 1996

Abstract

The Nd isotopic composition of Atlantic and Tethys seawater, as deduced from marine phosphorites, varied considerably during the past 180 Ma. The early Tethys and Central Atlantic seawater from 180 to 160 Ma ago (Early–Middle Jurassic) had a Nd isotopic signature identical to that of the Pacific ($\epsilon_{Nd} \approx -6$) suggesting that Pacific seawater entered the newly forming Tethys basin. However, with time continental runoff draining into the young basin became more important and led to a decrease in the Nd isotopic composition and, finally, a decoupling from the Pacific Ocean. During the late Early Cretaceous (120–90 Ma ago) Atlantic and Tethys seawater reached continental crust-like Nd isotopic composition values ($\epsilon_{Nd} \approx -10$) which could reflect high weathering rates induced by the warm and humid climate at that time. In the time span between 80 and 50 Ma (Late Cretaceous–Early Tertiary) the Nd isotopic composition of both Tethys and Atlantic seawater strongly increased towards Pacific seawater values. In the case of the Atlantic Ocean, this change has been correlated with the opening of the South Atlantic, which enabled the more radiogenic Pacific seawater to travel westwards around the southern edge of Africa into the South and finally the North Atlantic. In the case of Tethys seawater we have to assume that, during this period, large masses of Pacific seawater entered the Tethys again directly through the Indian–Tethys seaway. The renewed importance of this seaway might be related to the Late Cretaceous–Early Tertiary first-order, global sea level highstand. The most abrupt change towards lower, more crust-like values occurred in the early Miocene (25–17 Ma ago) in the South and North Atlantic, as well as in the Tethys. During this period, the $^{143}\text{Nd}/^{144}\text{Nd}$ isotope ratios dropped from ≈ 0.5124 ($\epsilon_{Nd} \approx -4.2$) down to ≈ 0.5122 ($\epsilon_{Nd} \approx -8$). This change indicates a diminishing influx of Pacific seawater into the Atlantic and Tethys Oceans and can be correlated with the breakdown of the circum-equatorial circulation patterns of the world oceans, due to plate tectonics. From 20 to 17 Ma ago (Miocene) the Nd isotopic composition of the Atlantic Ocean increased again and reached values as high as 0.5123 ($\epsilon_{Nd} \approx -6.6$), whereas Tethys seawater remained at low values. This decoupling can be correlated with the incipient complete isolation of the Tethys; the Nd isotopic composition was henceforth controlled by the continental runoff from surrounding land masses. The major increase in the Nd isotopic composition observed in the Atlantic can be related to the opening of the Drake Passage and establishment of the circum-Antarctic current system. This allowed Pacific seawater to enter directly the South Atlantic around Antarctica. A

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renewed drop in the Nd isotopic composition of Atlantic seawater during the late Miocene can be related to the onset of North Atlantic Bottom Water formation and its subsequent upwelling.

Keywords: Atlantic Ocean; Tethys; paleocurrents; Nd-144/Nd-143

1. Introduction

Various studies have shown that the present-day waters of the major oceans differ in their Nd isotopic compositions and that, even within a single basin, measurable differences in Nd isotopic values exist [1–3]. A recent isotope study of hydrogenous ferromanganese encrustations and nodules demonstrated that their region-dependent Nd isotopic variations display a marked similarity to the broad patterns of present-day deep-ocean circulation [4]. This suggests that the Nd isotope system is a powerful tool for distinguishing between different water masses and for tracing the water exchange between different basins in both the modern and ancient oceans.

These intra-oceanic isotopic variations are not observable in the Sr isotope system because seawater is well mixed with respect to Sr but poorly mixed with respect to Nd. This is due to the markedly different residence times; Sr has a residence time of 2×10^6 yr [5], which is much longer than the mixing time of the oceans (1×10^3 yr), whereas the dissolved Nd resides less than 1×10^3 yr in the oceans [1,6].

The Sr and Nd isotopic compositions of seawater have varied considerably over time [7,8]. However, the Nd isotopic composition of the Pacific Ocean has remained more or less stable during the past 200 Ma and always been significantly higher (more radiogenic) than in the Atlantic and Tethys Ocean ([9] and references therein). Rapid fluctuations in the Nd isotopic composition can be induced by changes in: (1) the relative erosion rate of very young to very old crustal material; (2) the input of Nd during times of high volcanic and hydrothermal activity at oceanic spreading ridges; and (3) paleocurrents between isotopically different paleoceans induced by changes in plate tectonics and sea level.

Stille [10] observed major changes in the Nd isotopic composition of Atlantic seawater towards more radiogenic, Pacific-like values sometime between 75 and 35 Ma (Late Cretaceous–Early Ter-

tiary) and 20 Ma (Miocene) ago. However, during this time the Sr isotopic composition of seawater is generally constantly increasing. Thus, Sr and Nd are geochemically decoupled; Sr reflects the increasing influence of a ‘crust-type’ source, whereas Nd reflects a ‘mantle-type’ source. Therefore, it has been suggested that this isotopic decoupling and the abrupt changes in Nd isotopic compositions reflect mixing of different water masses rather than changes in weathering and runoff. The opening of the South Atlantic, northward drift of India and Australia, and opening of the Drake Passage probably affected the circulation patterns in the Atlantic, allowing communication with the Pacific Ocean, which affected the Nd isotopic composition of these water masses [10]. In the present study, new data confirm the changes in Nd isotopic composition 75–35 Ma and 20 Ma ago in the Atlantic Ocean and elucidate the paleoceanic evolution of the Tethys Ocean.

2. Sampling

Different materials, such as metalliferous sediments, Fe–Mn ores, carbonate fossils, marine phosphates, authigenic argillites, clay fractions and glauconites have been used to estimate Nd isotopic variations in ancient ocean waters [1,7,11–21]. Each of these components might have integrated the Nd and Sr isotopic composition of seawater but, because they originate in very different depositional environments, their Nd isotopic compositions are quite variable and reflect the broad environments in which they formed. Therefore, to minimize the scatter of the Nd secular variation, Stille [10] avoided compilation of isotope data from different mineralogies that formed in very different depositional environments. Fe–Mn deposits were totally excluded because they formed slowly at the bottom of the oceanic basins and integrate the Nd isotopic composition of seawater over millions of years.

Nevertheless, the abrupt change observed in Nd

isotopic composition during the Miocene (20 Ma ago [10]) remained questionable. This change becomes mainly visible when the Nd isotope data of Fe–Mn coatings on planktonic foraminifera tests from the South Atlantic are combined with data of phosphate peloids from the North Atlantic (Carolina phosphogenic province). The phosphate peloids were deposited in an pericontinental environment, whereas Fe–Mn coatings of planktonic foraminifera have adsorbed rare earth elements during sedimentation at 2000 m depth and consequently represent the Nd isotopic composition of deeper bottom water [15]. Stille [10] used carbonates and fish skeletal remains in his compilation, which raises the question as to whether the data of these very different materials really represent similar water masses.

In order to resolve the question concerning the abrupt change in Nd isotopic composition during the Miocene (20 Ma ago) in the Atlantic, we analyzed individual phosphate peloids coming from precisely dated Miocene phosphate units of the Carolina phosphogenic province that formed during the period between 22 and 18 Ma. Very detailed Sr isotope studies have been performed on the same section [20,22]. These investigations concluded that, of the five phosphate grain types (brachiopods, vertebrae, teeth, bone splinters and peloidal grains), phosphate

peloids are the most suitable to determine the Sr and Nd isotopic composition of shallow shelf seawater.

In order to reconstruct the water exchanges of the Tethys with the Atlantic and Indian oceans, the Nd isotopic composition of phosphate peloids from a Miocene section in Malta (Mediterranean) have been analyzed. A detailed Sr, K–Ar and C isotope study has recently been performed on the Malta section in order to date and characterize the age of phosphogenesis in this region. The results demonstrate that the formation ages of the Malta phosphate horizons correlate well with those of the Carolina phosphogenic province and with similar sections in Florida, and they manifest a positive $\delta^{13}\text{C}$ excursion typical of the Monterey Formation [23]. The initial Sr isotopic compositions of the phosphate peloids from both the Carolina and Malta sections are, with one exception (Messinian sample from Malta section), identical with those of contemporaneous seawaters, as suggested by Hodell et al. [24].

3. Analytical procedures

Peloidal grains were hand-picked to 99% clean under a binocular microscope. Samples were ground to a fine powder by agate mortar and pestle. Pow-

Table 1
Nd isotopic compositions of carbonates and phosphate concretions from the Tethys

Sample	Stage	Age ^a (Ma)	Origin ^b	Lithology	¹⁴³ Nd/ ¹⁴⁴ Nd ^c	¹⁴⁷ Sm/ ¹⁴⁴ Nd	¹⁴³ Nd/ ¹⁴⁴ Nd (T)	ϵ_{Nd} ^d (T)
IP 22	Bajocian	168	VT	carbonate	0.512277(31)	0.1198	0.512145	–5.40
IP 25	Late Barremian	126	VT	carbonate	0.512135(12)	0.1232	0.512033	–8.64
IP 26	Early Aptian	120	VT	carbonate	0.512136(34)	~ 0.118	0.512043	–8.60
IP 27	Late Cenomanian	93	VT	carbonate	0.512086(28)	~ 0.118	0.512014	–9.84
IP 28	Early Turonian	90	VT	carbonate	0.512179(34)	0.1156	0.512111	–8.02
3144	Late Albian	100	VT	phosphate	0.512166(7)	0.1231	0.512085	–8.28
2469	Early Albian	108	VT	phosphate	0.512147(6)	0.1192	0.512063	–8.51
3181	Late Aptian	112	VT	phosphate	0.512195(8)	0.1313	0.512099	–7.70
3012	Late Aptian	115	VT	phosphate	0.512164(8)	0.1401	0.512059	–8.41
2950	Early Aptian	120	VT	phosphate	0.512222(6)	0.1348	0.512116	–7.17
BC 1	Valanginian	137	BC	phosphate	0.512210(8)	0.1099	0.512111	–6.83
Pho 3904	Late Campanian	75	Egypt	phosphate	0.512185(14)	0.1215	0.512125	–8.12
Pho 3903	Lutetian	53	HR	phosphate	0.512188(10)	0.1209	0.512148	–8.29

^a Absolute ages according to [45]. ^b VT = Vocontian Trough, BC = Bethic Cordillera, HR = Helvetic realm. ^c Given uncertainties of the present-day Nd isotopic compositions are 2σ of the mean values. ^d $\epsilon_{\text{Nd}}^{\text{(T)}} = 10^4 \times [({}^{143}\text{Nd}/{}^{144}\text{Nd})_{\text{sample}}^{\text{(T)}} - {}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{CHUR}}^{\text{(T)}}] / ({}^{143}\text{Nd}/{}^{144}\text{Nd}_{\text{CHUR}}^{\text{(T)}})$, CHUR = Chondritic Uniform Reservoir.

Table 2

Sr ages and present-day Nd isotopic compositions of Miocene phosphates from the North Atlantic (Carolina phosphogenic province) and the Mediterranean sea (Malta)

Sample ^a	Sr age ^b	¹⁴³ Nd/ ¹⁴⁴ Nd ^c	ε _{Nd}
<i>Aurora section (Carolina phosphogenic province)</i>			
GH8,5R (-96 ft)	17.1	0.512209(7)	-8.37
GH8,5R (-108 ft)	12.1	0.512272(31)	-7.14
GH8,5R (-109 ft)	17.8	0.512213(7)	-8.29
GH8,5R (-119 ft)	17.5	0.512262(16)	-7.33
GH8,5R (-126 ft)	17.7	0.512266(7)	-7.26
GH8,5R (-133 ft)	18.0	0.512243(13)	-7.71
GH8,5R (-144 ft)	18.5	0.512232(11)	-7.92
GH8,5R (-162 ft)	21.4	0.512209(12)	-8.37
GH8,5R (-170 ft)	21.5	0.512188(9)	-8.78
GH8,5R (-174 ft)	22.8	0.512218(8)	-8.19
GH8,5R (-178 ft)	22.9	0.512230(6)	-7.96
GH8,5R (-182 ft)	22.9	0.512220(7)	-8.15
<i>Onslow Bay</i>			
OB3-7	10.2	0.512246(7)	-7.65
OB3-14	10.8	0.512262(8)	-7.33
OB24-5	17.8	0.512319(7)	-6.22
OB24-6	17.8	0.512333(12)	-5.95
OB24-9	19.7	0.512270(13)	-7.18
OB24-11	19.2	0.512304(15)	-6.52
OB29B-9	18.4	0.512318(21)	-6.24
OB29B-21	18.6	0.512307(22)	-6.46
OB29B-29	19.0	0.512336(15)	-5.89
OB45-1	9.6	0.512217(8)	-8.21
OB45-15	16.8	0.512242(8)	-7.72
OB53-14	8.4	0.512283(7)	-6.92
OB53-24	8.0	0.512272(6)	-7.14
OB64-14	17.8	0.512329(6)	-6.03
OB64-18	18.3	0.512322(7)	-6.16
OB64-21	18.8	0.512330(11)	-6.01
OB64-26	19.1	0.512322(9)	-6.16
<i>Malta (Mediterranean Sea)</i>			
QAB28	17.2	0.512199(13)	-8.56
QAB29	17.3	0.512207(11)	-8.41
QAB30	17.0	0.512175(17)	-9.03
QAB31	17.0	0.512242(19)	-7.72
IG8	6.0	0.512145(18)	-9.62
FG5	24.5	0.512370(11)	-5.23
FG8	24.3	0.512399(16)	-4.66
FG17	22.0	0.512277(10)	-7.04
FG41	17.1	0.512177(10)	-8.93
FG43	16.1	0.512215(8)	-8.25

QAB = Qammieh section; IG = Il Gelmus; FG = Fomm Ir Rih Bay section. ^a Sedimentary unconformity in Aurora section at 109 ft. ^b Sr ages according to [20,22] (Carolina) and [23] (Malta). ^c Given uncertainties of the Nd isotopic compositions are 2σ of the mean values.

dered samples were treated with 0.5 N acetic acid in order to remove Sr-rich calcite overgrowths and rinsed with distilled water prior to dissolution in 6 N HCl. Chemical separations were performed using ammonium citrate and 4 and 6 N HCl as eluents through AG50W-X12 (200–400) ion-exchange resin in 1 ml quartz columns for bulk rare earth element separation. Nd was separated using the same type of resin and column and α-hydroxyisobutyric acid as eluent. The blank at the time of analysis was < 180 pg for Nd. Nd was measured using triple Ta–Re–Ta filament assemblies. Isotopic measurements was performed on a fully automatic VG Sector mass spectrometer with a 5 cup multicollector. The ratio ¹⁴⁶Nd/¹⁴⁴Nd = 0.7219 was used for fractionation correction. The average ¹⁴³Nd/¹⁴⁴Nd for 16 La Jolla standard measurements was 0.511840 ± 6 (2 standard deviation of the mean). Typically, 80–150 ratios for Nd were collected to achieve adequate precision. The isotope data are compiled in Tables 1 and 2.

4. Results and discussion

4.1. The Nd isotopic evolution of Tethys seawater from 180 to 90 Ma

In this section we will focus on the early evolution of Tethys seawater. The early Tethys was an appendix of the Pacific or Panthalassa (Fig. 1A). From 170 to 150 Ma ago (Early–Middle Jurassic), Tethys seawater had Nd isotopic signatures close to the Pacific (¹⁴³Nd/¹⁴⁴Nd ≈ 0.51211; ε_{Nd} ≈ -6; Fig. 2). However, with the breakup of Pangea and the subsequent plate movement, continental runoff draining into the young basin became more important and led to a decrease in Nd isotopic composition values. Consequently, the Nd isotopic evolution suggests that Tethys seawater became increasingly decoupled from the Pacific Ocean, as previously observed by Shaw and Wasserburg [7]. Measurements on authigenic phosphate concretions from the Vcontian Trough, located in the northern part of Tethys (Table 1), indicate that Tethys seawater reached its

lowest Nd isotopic composition values, almost continental crust-like, during the late Early Cretaceous (120–90 Ma ago; $\epsilon_{\text{Nd}} = -8.8$ and -10 , respectively). These low values probably result from mechanical erosion of continental crust in combination with the very special climatic conditions at that time. During this period worldwide basaltic volcanism and associated outgassing of CO_2 was extensive and led to the establishment of greenhouse conditions ([25] and references therein) which accelerated weathering and flux rates of continental detritus into the oceans [26].

From 180 to 90 Ma the Nd isotopic evolution is geochemically not decoupled from Sr, with both suggesting an increasing continental runoff. In the Aptian, the Sr seawater curve suddenly dips downward, suggesting an increased mid-ocean ridge hydrothermal flux which recovers gradually through the Albian [27]. Volcanism, seafloor spreading, and hydrothermal activities have frequently been invoked to explain short-term excursions towards lower $^{87}\text{Sr}/^{86}\text{Sr}$. Similarly, the few Nd isotope data presently available also suggest an increased hydrothermal flux between 120 and 90 Ma ago.

4.2. The global paleocirculation and Nd isotopic evolution of seawater from 90 to 25 Ma

From 100 to 80 Ma ago, the counter-clockwise rotation of the African plate lead to the opening of the South Atlantic. This in turn, lead to compressional plate movements in the Tethys region between the European and African plates (Fig. 1B [28]). However, this did not lead to an immediate narrowing of the Tethys seaway because global sea level was rising at the same time. Only in the Tertiary, when this late Cretaceous first-order sea level highstand was over [29], did the new plate kinematic framework lead to a complete change in the general global current pattern. The E–W currents in the former Indian–Tethys–Caribbean seaway were replaced by N–S currents in the northern and southern Atlantic ([30] and references therein).

Between 90 and 25 Ma ago (Late Cretaceous–Early Tertiary) the Nd isotopic composition of Atlantic and Tethys seawater increased significantly, from $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of 0.51205 ($\epsilon_{\text{Nd}} \approx -10$) to 0.5123 ($\epsilon_{\text{Nd}} \approx -6$; Fig. 2 and Fig. 3) which is

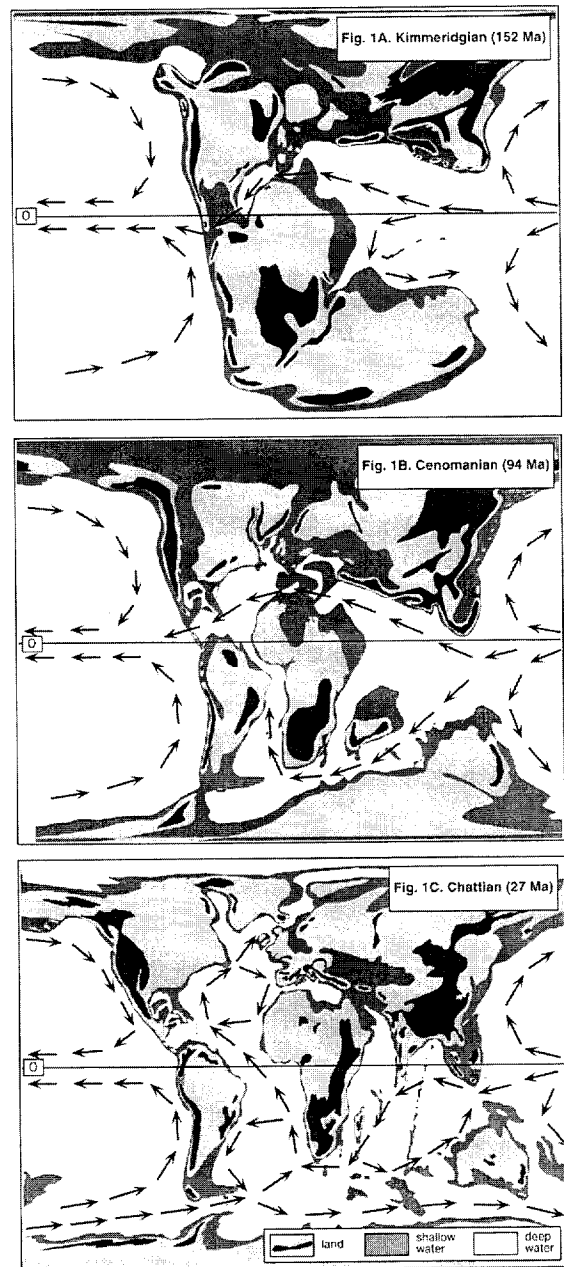


Fig. 1. Evolution of the Tethys basin during the Mesozoic and Cenozoic according to Scotese et al. [28]. (A) Jurassic: breakup of Pangea and formation of Tethys with its appendix-like shape. (B) Cretaceous: opening of the South Atlantic and northward drift of India. (C) Early to middle Tertiary: separation of Australia from Antarctica and its northward drift and collision with Indonesia during the Middle Miocene.

close to the lowermost values observed for the Pacific Ocean at that time. New data on phosphate concretions from northern Tethys (Helvetic realm) and the southern margin of Tethys (Egypt) support this change towards higher, more mantle-like isotopic composition values for Tethys seawater (Fig. 3; Table 1). However, the Sr isotopic composition increased constantly towards crustal compositions in both basins, indicating that Sr and Nd isotope systems were, in contrast to the period from 180 to 90 Ma, geochemically decoupled at this time. In the Atlantic, this decoupling from 80 to 25 Ma was explained by the exchange and mixing of isotopically different water masses rather than the increasing influence of a mantle or crustal source [10]. The advent of isotopically different water masses in the North Atlantic was probably due to the opening of the South Atlantic and the northward drift of India at about 90–80 Ma ago (Fig. 1B). This enabled the more radiogenic Pacific surface waters ($\epsilon_{\text{Nd}} > -4.7$; $^{143}\text{Nd}/^{144}\text{Nd} > 0.5124$) to travel westwards around southern Africa into the South Atlantic and, finally,

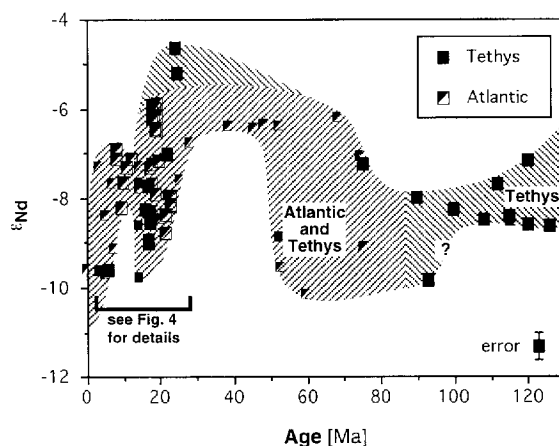


Fig. 3. Variations in ϵ_{Nd} of Atlantic and Tethys seawater during the past 120 Ma. Symbols as in Fig. 2.

the North Atlantic, where it caused the observed rise in Nd isotopic compositions.

In the case of Tethys seawater we observe an even stronger increase in the Nd isotopic composi-

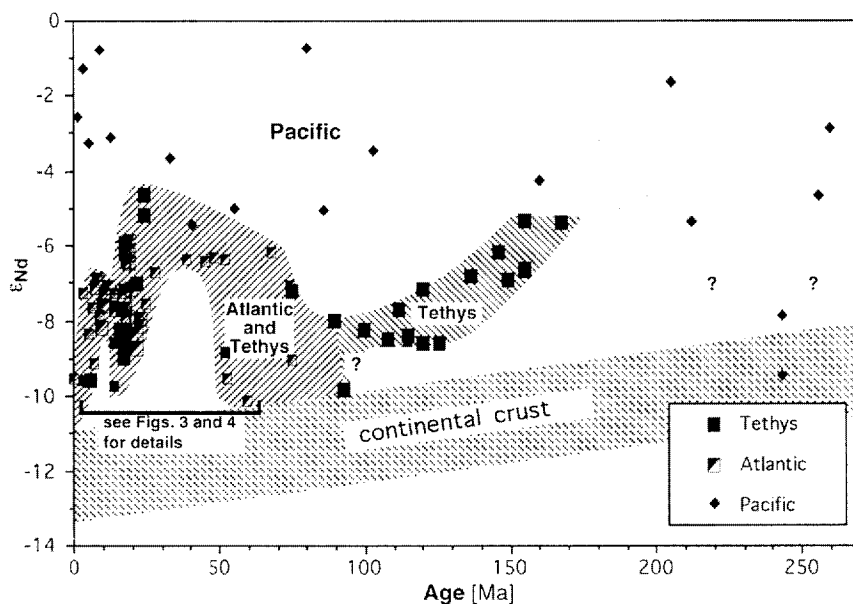


Fig. 2. Variations of ϵ_{Nd} of Pacific, Atlantic and Tethys seawater during the past 250 Ma. Large symbols are from this study, Stille and Fischer [19] and Stille et al. [18]. Small symbols represent data from the literature: Chyi et al. [43] for cherts, Shaw and Wasserburg [7] for carbonates and phosphates, Staudigel et al. [14] for phosphates, Palmer and Elderfield [15] for Fe–Mn coatings of foraminifera, Grandjean et al. [16] and Keto and Jacobsen [44] for phosphates. The errors in ages are equal to or smaller than the symbols. Absolute ages according to Harland et al. [45].

tion at 25 Ma (Late Oligocene–Early Miocene) reaching values above 0.51237 ($\epsilon_{\text{Nd}} \approx -4.6$; Fig. 3), which is higher than observed for contemporaneous Atlantic seawater. This indicates that, during this period, large masses of Pacific surface waters entered the Tethys basin again through the Indian–Tethys seaway. This increasing inflow of Pacific seawater into Tethys was probably related to a first-order sea-level highstand some 80–50 Ma ago (Late Cretaceous–Early Tertiary [29]). This highstand apparently could completely compensate the plate tectonic induced narrowing of Tethys seaway which started in Late Cretaceous. Erosional base-level was higher during this highstand, which diminished continental runoff. Furthermore, the extensive growth of carbonate margins during the Middle and Late Cretaceous might have sealed many siliciclastic shorelines, which also led to a reduction in continental runoff. Therefore, the combination of a higher input of Pacific seawater and a reduction in continental runoff may have led to the increase in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios observed in the Tethys at that time.

During the Eocene (55–35 Ma) the Indian–Tethys seaway became very narrow between the Arabian plate to the south and the Eurasian plate to the north [30] but the Nd isotopic composition of Tethys seawater remained elevated. This indicates that considerable amounts of Pacific seawater could still enter Tethys. In the Early Miocene (25 Ma) the Nd isotopic composition of Tethys seawater was very close to Pacific seawater (Fig. 2). This supports the hypothesis that Pacific seawater still fed the Tethys basin, in spite of the narrow seaway. Based on stable isotope data of benthic foraminifera and sedimentological and faunal evidence, Woodruff and Savin [31] suggested that, during the Early Miocene, a warm saline plume of bottom water flowed from eastern Tethys across the present day Persian Gulf region into the Indian Ocean. According to these authors this outflow probably triggered a surface counter-current of Indian seawater into Tethys, which is in good agreement with our Nd isotope data.

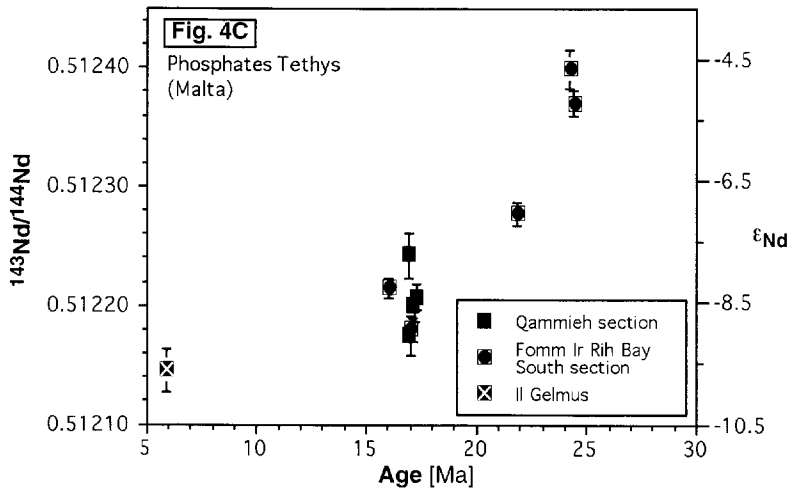
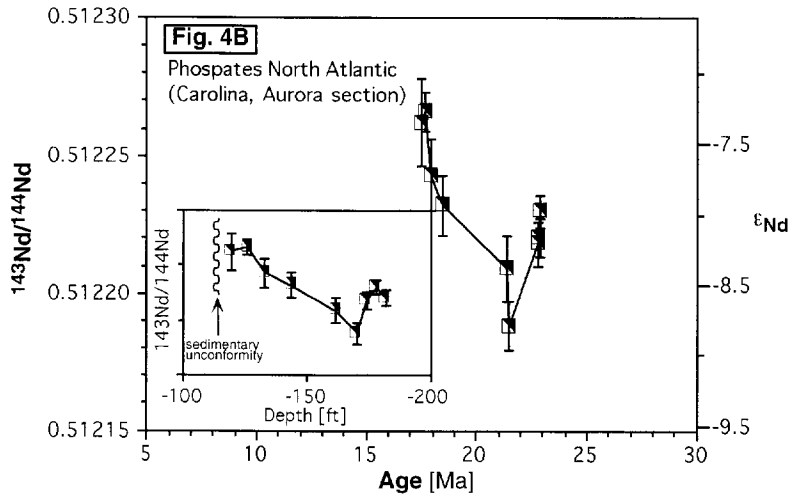
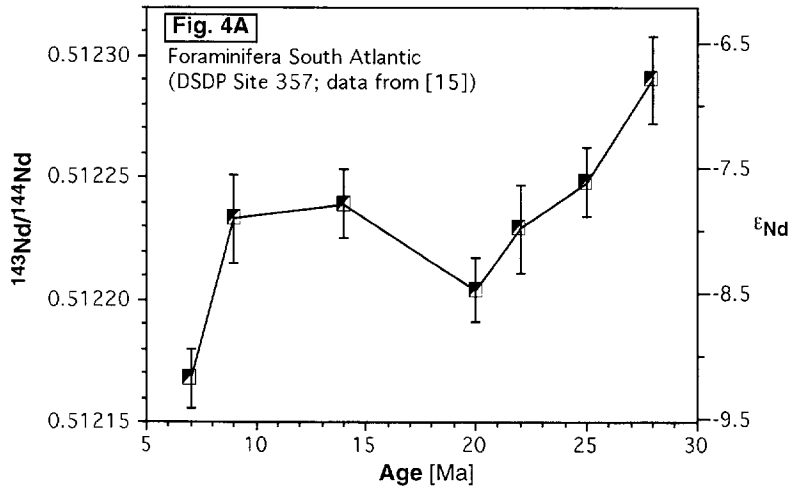
4.3. The paleocirculation and Nd isotopic evolution of Pacific and Tethys seawater from 25 to 10 Ma

A strong decrease in Nd isotopic ratios in Atlantic seawater 25–20 Ma ago was first described by Palmer

and Elderfield [15] using only Nd isotope data from Fe–Mn coatings of foraminifera from the South Atlantic (Fig. 4A). In the present study we use new data for phosphate peloids from two different areas: one in North Carolina (Fig. 4B), representing North Atlantic seawater, and another on Malta (Mediterranean), representing Tethys seawater (Fig. 4C). These new data confirm the major change towards less radiogenic Nd isotopic composition values 20 Ma ago and suggest the establishment of a new circulation pattern in the Atlantic and Tethys oceans. During this period, from 180 to 100 Ma, Sr and Nd were probably not geochemically decoupled and suggest increasing continental runoff.

The largest decrease in Nd isotopic ratios is observed in Tethys seawater (Fig. 4C). The phosphate peloids from 3 different and well dated stratigraphic sections from Malta [23] show the highest $^{143}\text{Nd}/^{144}\text{Nd}$ isotopic composition values of 0.5124 ($\epsilon_{\text{Nd}} = -4.1$) at approximately 25 Ma ago. This Nd value is close to contemporaneous Pacific Ocean water. The isotopic composition decreased to values lower than 0.5122 ($\epsilon_{\text{Nd}} = -8.2$) about 17 Ma ago. At this time the Nd isotopic ratios ranged between 0.5122 and 0.51224, corresponding to a variation of 1.3 ϵ units. This major change in Nd isotopic composition may reflect the collision of the Arabian and Eurasian plates. Thus, the Indian–Tethys seaway started to close about 25 Ma ago leading to the diminution of radiogenic Pacific seawater and the breakdown of the circum-equatorial circulation pattern of the world oceans approximately 20 Ma ago (Early Miocene; Fig. 1C). The lowest $^{143}\text{Nd}/^{144}\text{Nd}$ value of 0.51215 ($\epsilon_{\text{Nd}} = -9.4$) has been found in the youngest phosphate sample, which formed about 6 Ma ago (Messinian). During the Messinian the seaway between Atlantic and Tethys closed in response to a worldwide regression [32]. The Mediterranean Sea dried up and its basinal parts were filled by thick evaporites. Fresh waters overprinted the geochemical composition of the evaporites, leading to the development of nonmarine $^{87}\text{Sr}/^{86}\text{Sr}$ values and precluding their use for stratigraphic correlation and dating [33]. This complete isolation of the Tethys could also explain the very low crust-like Nd isotopic composition found in the Messinian of the Malta section.

Data from phosphate peloids in the lowermost



part of the North Carolina Miocene section suggest an isotopic evolution in the North Atlantic Ocean about 23–20 Ma ago that is similar to that of the Tethys (Fig. 4B). However, it is not likely that closing of the small Indian–Tethys seaway alone caused this strong Nd shift. Rather, we suggest that less radiogenic seawater entered the Atlantic from the south. A possible explanation for this could be the separation of Australia from Antarctica about 45 Ma ago and its northward drift and collision with Indonesia during the Middle Miocene (Fig. 1C, [34]). This plate tectonic event led to the evolution of today's circulation pattern, with individual current systems for the Indian and Pacific oceans [35]. Consequently, it was no longer radiogenic Pacific water that entered the Atlantic directly from the south around Africa 23–20 Ma ago but, rather, Indian Ocean water whose present-day Nd isotopic composition is known to be less radiogenic [4]. In addition, Sr isotopic composition values of seawater indicate that, during the Miocene, continental weathering and runoff increased mainly due to the collision of India with Asia and denudation of the Himalayan–Tibetan orogen [36,37]. Thus, continental weathering probably also became a dominant process for the oceanic Nd budget.

Beginning about 20–18 Ma ago, the Nd isotopic compositions of the Atlantic and Tethys oceans evolved differently. Whereas the Nd isotopic composition of Tethys seawater continued to decrease, the isotopic composition of the North Atlantic Ocean water started to increase again and reached values as high as 0.51226 ($\epsilon_{\text{Nd}} \approx -7.4$) at about 18 Ma ago (Fig. 2 and Fig. 3 and 4B). Phosphate peloids from the Aurora section in the Carolina phosphogenic province have $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of higher than 0.5123 ($\epsilon_{\text{Nd}} \approx -6.6$) which are close to values found for contemporaneous Pacific seawater. At this time Sr and Nd are again decoupled from 80 to 25 Ma. Therefore, the significant increase in Nd isotopic composition in the Atlantic might be caused by major changes in the circulation pattern within the

Atlantic Ocean itself between 25 and 20 Ma. The change in currents might have been in response to the opening of Drake Passage, which led to establishment of the circum-Antarctic current system (Fig. 1C, [38]). Opening of the Drake Passage enabled Pacific seawater to travel around Antarctica and to enter the South Atlantic and to flow northwards to join the Gulf Stream portion of the North Atlantic gyre. This new admixture of more radiogenic Pacific seawater would explain the increase in Nd isotopic composition of the Atlantic between 20 and 18 Ma.

Since about 18 Ma, the Nd and Sr isotopic composition of the Atlantic Ocean have no longer been decoupled and have evolved to present-day seawater values (Fig. 3). This might indicate that the influx of Pacific seawater into the Atlantic diminished soon after the Drake Passage opened. However, it is more probable that the continued opening of the North Atlantic between Greenland and Scandinavia at the Reykjanes Ridge and subsidence of the Iceland–Faeroe Ridge about 16 Ma ago [39–41] allowed the Arctic Norwegian Overflow Water to enter the North Atlantic basin and form the North Atlantic Bottom Water. No data exist for the isotopic composition of the paleo-North Atlantic Bottom Water. However, investigations of the present-day current demonstrate that it has the lowest Nd isotopic composition (≈ 0.51195 ; $\epsilon_{\text{Nd}} \approx -13.5$) ever measured in the open ocean [3]. Because upwelling of cold, nutrient-enriched bottom water became a widespread phenomenon during the Miocene that led to deposition of primary phosphate sediments [42], we suggest that exchange of these non-radiogenic bottom waters with surface waters caused the decreasing isotopic composition values in the youngest history of the Atlantic Ocean.

5. Conclusions

The combination of Sr and Nd isotope data from marine phosphates allows reconstruction of the paleo-

Fig. 4. (A) Variations in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios in Fe–Mn coatings of foraminifera from the South Atlantic (DSDP Site 357) during the past 30 Ma. Data from Palmer and Elderfield [15]. (B) and (C) Variations in $^{143}\text{Nd}/^{144}\text{Nd}$ ratios in phosphate peloids from Carolina (North Atlantic) and Malta (Tethys), respectively. In (B) (Aurora section, Table 2) only samples below the sedimentary unconformity and depth-increasing Sr ages have been used. Insert in (B) shows the same data plotted vs. core depth.

oceanic evolution of the Atlantic and Tethys oceans during the past 180 Ma. Changes in circulation patterns, especially connected with the increasing input of Pacific seawater, caused a decoupling of the Sr and Nd isotope systems. However, with diminishing input of Pacific seawater a coupled evolution of Sr and Nd is observable for the Atlantic and Tethys oceans which suggests increasing continental runoff.

During two major phases the input of Pacific seawater was important and caused the decoupling of the Sr and Nd isotope systems: (1) in the Late Cretaceous–Early Tertiary (90–25 Ma ago), due to the opening of the South Atlantic and a global sea level highstand; and (2) in the Miocene from about 20 to 18 Ma ago, due to the opening of the Drake Passage. The Nd isotopic evolution is not geochemically decoupled from Sr from 180 to 90 Ma, from 25 to 20 Ma and from 18 Ma to the present day.

Acknowledgements

We thank B. Kiefel, D. Tisserand, and D. Ames for their technical assistance in the laboratory and S.L. Goldstein and two anonymous reviewers for critical reviews of the manuscript that helped clarify several points. E. Jacobs furnished the Malta samples and J.G. Br  h  ret the phosphate samples from the Vocontian Trough series. *[UC]*

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