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Neodymium isotope evolution of NW Tethyan upper ocean waters throughout the Cretaceous

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Abstract

Neodymium isotope compositions of twenty-four fish teeth, nineteen from the NW Tethys and five from different locations within the Tethys, are interpreted to reflect the evolution of Tethyan upper ocean water composition during the Cretaceous and used to track changes in erosional inputs to the NW Tethys and in oceanic circulation throughout the Cretaceous. The rather high ε_{Nd} (up to -7.6) of the NW Tethyan upper ocean waters recorded from the Late Berriasian to the Early Aptian and the absence of negative excursions during this interval support the presence of a permanent westward flowing Tethys Circumglobal Current (TCC). This implies that temperature variations during this time period, inferred from the oxygen isotope analysis of fish tooth enamel, were not driven by changes in surface oceanic currents, but rather by global climatic changes. The results presented here represent a significant advance over previously published Cretaceous seawater Nd isotope records. Our newly acquired data now allow the identification of two stages of low ε_{Nd} values in the NW Tethys, during the Early Albian–Middle Albian interval (down to -10) and the Santonian–Early Campanian (down to -11.4), which alternate with two stages of higher ε_{Nd} values (up to -9) during the Late Albian–Turonian interval and the Maastrichtian. Used in conjunction with the oxygen isotope record, the fluctuations of ε_{Nd} values can be related to major climatic, oceanographic, and tectonic events that appeared in the western Tethyan domain.

Keywords: neodymium; cretaceous; tethys; oceanic circulation

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1. Introduction

The Cretaceous was a period of major climatic [1–3] and paleogeographic changes, such as the widening of the North Atlantic Ocean, the opening of the South Atlantic Ocean, and the establishment of marine connections between the North and South Atlantic basins [4,5]. These global changes are likely to have resulted in a major reorganisation of the oceanic circulation pattern. Two of the main features of the Cretaceous paleogeography were the presence of the Tethys Ocean and the absence of the Panama isthmus, which may have contributed to the equable climate (weak pole-to-equator surface-temperature gradient) that characterised the main part of the Cretaceous by providing a marine circumglobal passage at low paleolatitudes [6]. The paleocirculation in the Tethys is therefore of considerable interest.

The neodymium isotope composition ($\varepsilon_{Nd}(0)=[(^{143}Nd/^{144}Nd)_{sample}/^{143}Nd/^{144}Nd)_{CHUR}-1]\times 10^4$ expressed in ε-units) of a wide variety of marine minerals (ferromanganese nodules, carbonates, phosphates, glauconites) has been used to trace past oceanic circulation patterns [7–10]. The Nd isotope composition of seawater is not uniform in the present-day oceans (e.g. [11]) because the oceanic residence time of Nd (500 yr; [12]) is shorter than the mixing time of the oceans (~1500 yr; [13]) and the relative contributions of Nd from ancient continental versus young volcanogenic materials differ in the various basins. Due to the discharge of very unradiogenic Nd from ancient continental land masses adjacent to the North Atlantic basin (e.g. Greenland: $\varepsilon_{Nd}(0)=\sim-19$; Canadian shield: $\varepsilon_{Nd}(0)=\sim-14/-32$), the deep water generated in the North Atlantic has fairly unradiogenic $\varepsilon_{Nd}(0)$ values (about -13) [14–17]. In contrast, the more radiogenic values ($\varepsilon_{Nd}(0)=\sim-4$) that characterize the North Pacific reflect the contribution of Nd weathered from volcanic rocks surrounding the basin (e.g. Japan: $\varepsilon_{Nd}(0)=\sim0$; Indonesia: $\varepsilon_{Nd}(0)=\sim+2$) [18–20]. Therefore, the Nd isotope system can allow reconstitution of water exchange between isotopically distinct water masses, in both modern and ancient oceans [8,10,21]. In addition, as continental weathering is the principal source of Nd to the oceans (e.g. [20,22,23], the Nd isotope system also has the potential to provide information on erosional inputs to the oceans.

Due to the very high Nd concentration in apatite and to the better resistance of phosphates to diagenesis relative to that of carbonates, Nd isotope ratios in fish teeth are preserved over geologic timescales (Martin and Haley [24]). Fish teeth are therefore particularly useful for such paleoceanographic reconstructions [25]. The Nd isotope composition of apatites is acquired quite rapidly at or near the sediment-water interface by adsorption of seawater Nd on the apatite after the death of the organism [26,27]. The Nd isotope composition of fish teeth will thus reflect that of seawater at the bottom of the water-column.

The purpose of this study is to reconstruct the Nd isotope evolution of NW Tethyan upper ocean water throughout the Cretaceous by analysing fish tooth enamel collected from open-platform environments. The recorded variations will then be related to possible changes in erosional inputs to the ocean and in the global oceanic circulation pattern. Previous reconstructions of the evolution of the Tethyan Nd isotope composition have been realised over longer time periods, but the data concerning the Cretaceous remain quite scarce (e.g. [8,9]). In this study dedicated to the Cretaceous, we present twenty-four neodymium

Notes to Table 1:
Measured $^{143}Nd/^{144}Nd$ of the La Jolla standard = 0.512799 ± 21, 2 std. dev. external reproducibility. $^{143}Nd/^{144}Nd$ values listed in the table have been corrected by +0.000060, assuming a true value of 0.512859 for this standard.
Listed uncertainties are in-run 2 standard errors.
[a] Te (tooth enamel and enameloid), Tw (whole tooth), b (bone).
[b] L (Lower), M (Middle), U (Upper).
[c] Absolute ages are derived from the time scale of Gradstein et al. [28].
[d] Available Nd and Sm elemental concentrations have been analysed by Lécuyer et al. [38].
[e] $\varepsilon_{Nd}(T)=[[(^{143}Nd/^{144}Nd)_{sample}(T)/(^{143}Nd/^{144}Nd)_{CHUR}(T)]-1]\times 10^4$, where CHUR is the Chondritic Uniform Reservoir. The initial $(^{143}Nd/^{144}Nd)_{CHUR}$ was calculated using a $^{147}Sm/^{144}Nd$ ratio of 0.1967, and the initial $(^{143}Nd/^{144}Nd)$ or each sample was calculated using either their Sm/Nd ratios when available, or the mean Sm/Nd value of 0.2058 determined from the available ratios of the Cretaceous samples.
* duplicates.

Table 1

Descriptions, locations, ages, Nd (elemental and isotopic) and Sm (elemental) compositions of Cretaceous fish samples analysed in this work

Sample	Location	Fauna, remains[a]	Stratigraphic age (ammonite zone or horizon)[b]	Age (Ma)[c]	Nd (ppm)[d]	Quantity Nd analysed (ng)	Sm (ppm)[d]	$^{143}Nd/^{144}Nd$	$\varepsilon_{Nd}(T)$[e]
V40	Bonvillars, Switzerland	Undetermined, Te	U. Berriasian (Alpillensis zone)	137.5	162.01	9800	36.38	0.512116 ± 26	−9.1 ± 0.5
V39	Ponte du Suchet, Switzerland	Undetermined, b	L. Valanginian	136.2	809.73	48600	191.87	0.512143 ± 14	−8.7 ± 0.3
V39*	Ponte du Suchet, Switzerland	Undetermined, b	L. Valanginian	136.2	809.73	27500	191.87	0.512155 ± 09	−8.5 ± 0.2
V42	Auberson, Switzerland	Undetermined, Tw	U. Valanginian (Pronecostatum horizon)	134.8	495.25	5900	107.73	0.512160 ± 13	−8.2 ± 0.3
A4	Cenoran (Jura), SE France	Undetermined, Te	U. Valanginian (Callidiscus zone)	132.5				0.512182 ± 15	−7.7 ± 0.3
D1	Bleigny-le-Carreau (Yonne), N France	*Sphaerodus neocomiensis*, Te	Hauterivian	129.5	15.68	900	3.27	0.512145 ± 13	−8.5 ± 0.3
Cot1	Sisteron (Alpes-de-Haute Provence), SE France	Undetermined, Te	Upper part of the L. Aptian	118.0				0.512190 ± 24	−7.7 ± 0.5
C1	La Tuilière (Vaucluse), SE France	*Protolamna sokolovi*, Tw	Gargasian	116.1				0.512165 ± 08	−8.2 ± 0.2
A7	Lancrans (Ain), SE France	Undetermined, Te	L. Albian (Tardefurcatus Mamillatum zone)	110.7				0.512109 ± 22	−9.3 ± 0.4
PS25	La Houpette (Meuse), N France	*Otodus sp.*, Te	upper part of the L. Albian	107.7	86.21	2400	17.85	0.512058 ± 16	−10.3 ± 0.3
PS25*	La Houpette (Meuse), N France	*Otodus sp.*, Te	upper part of the L. Albian	107.7	86.21	6100	17.85	0.512067 ± 09	−10.2 ± 0.2
D5	Ardennes, N France	*Odontaspis*, Tw	M. Albian (Lyelli to Intermedius zone)	105.5	28.62	700	6.39	0.512124 ± 20	−9.2 ± 0.4
D5*	Ardennes, N France	*Odontaspis*, Tw	M. Albian (Lyelli to Intermedius zone)	105.5	28.62	700	6.39	0.512089 ± 22	−9.9 ± 0.4
Vr1	Salazac (Gard), SE France	Undetermined, Te	Vraconian	101.5				0.512142 ± 10	−8.7 ± 0.2
A5	Entrèves (Savoie), SE France	Undetermined, Te	U. Albian (Mortoniceras inflatum zone)	100.6				0.512138 ± 10	−8.8 ± 0.2
H1	Cap de la Hève (Seine-inférieure), N France	Undetermined, Te	L. Cenomanian	97.4				0.512139 ± 12	−8.8 ± 0.2
G3	Le Teil (Ardèche), SE France	Lamniforme, Te	Turonian	91.2				0.512140 ± 08	−8.9 ± 0.2
PI27	Vallée de la Maye (Somme), N France	Undetermined, Te	Coniacian	87.4	7.45	1000	1.27	0.512086 ± 08	−9.7 ± 0.2
n1an	Beauval (Somme), N France	*Anomotodon sp.*, Te	basal Santonian	85.6				0.512020 ± 10	−11.3 ± 0.2
D10	Sens (Yonne), N France	*Squalicorax pristodontus*, Te	L. Campanian	82.1	10.39	700	1.62	0.512000 ± 32	−11.4 ± 0.6
H2	Malakoff (Charentes Maritimes), W France	Undetermined, Te	U. Campanian	73.8	33.13	900	6.79	0.512142 ± 17	−9.0 ± 0.3
C3	Eben-Emael, Belgium	*Squalicorax pristodontus*, Te	U. Maastrichtian	67.2				0.512170 ± 15	−8.5 ± 0.3
S2d1	Mokrzesz, Poland	*Otodus appendiculatus*, Te	Lower Cenomanian	98.1				0.512127 ± 14	−9.1 ± 0.3
Kaz2	Kazakhstan	*Squalicorax kaupi*, Te	Upper Santonian	84.0				0.512239 ± 14	−7.0 ± 0.3
S9d1	Nasilov, Poland	*Odontaspis sp.*	Maastrichtian/Danian boundary	65.0				0.512137 ± 20	−9.2 ± 0.4
At1	Tamaloukt, Morocco	Undetermined	Coniacian	87.4				0.512247 ± 14	−6.8 ± 0.3
Siv1	Asen, Sweden	*Squalicorax kaupi*, Te	Uppermost L. Campanian	81.4				0.511726 ± 19	−17.0 ± 0.4

isotope analyses of stratigraphically well constrained fish tooth enamel collected in western Europe (nineteen samples) and other areas within the Tethyan realm (Morocco, Kazakhstan, Poland; four samples), and in Sweden (one sample). The oxygen isotope compositions of the teeth from western Europe, analysed in this study for their Nd isotope composition, have been previously determined [3]. The combination of oxygen and neodymium isotope analyses allows discussion of the possible changes in the NW Tethyan upper ocean water mass sources in the context of the concomitant evolution of surface temperatures. This study therefore contributes to a better understanding of the Cretaceous ocean-climate system.

2. Samples and analytical procedures

Except for the sample "Cot1", the fish teeth from France and Switzerland, analysed in this study for their Nd isotope compositions, have been described previously by Pucéat et al. [3]. Available information concerning sampling localities, genus and species, and stratigraphic ages are given in Table 1. Sample locations are shown in Fig. 1. Correspondence with absolute ages is from Gradstein et al. [28]. Teeth from France and Switzerland analysed in this study were sampled from open platform depositional environments, with the exception of sample Cot1 from southeast France which was deposited closer to the basin in a hemi-pelagic environment. Bathymetry in the Vocontian Trough (southeast France) remains a matter of debate. However, although this basin was deeper than the Paris Basin, it almost certainly did not exceed a thousand meters [29]. More recent estimates based on dinoflagellate cyst distribution patterns argue in favour of at most a few hundred meters [30]. Therefore, the teeth from western Europe are expected to record the Nd isotope compositions of upper ocean waters (<1000 m). The depth of the depositional environment for the remainder of the sample collection is not known with precision, although it is known that these samples were deposited in epicontinental seas that most probably were not very deep.

The chemistry and isotopic analyses were performed at the CRPG/CNRS laboratory in Nancy, France. To avoid contamination by detrital particles, enamel was preferred to dentine, because of its lower porosity. When possible, enamel was separated from the whole tooth. After cleaning in an ultrasonic bath to remove sediment particles, the samples were leached in 2 N acetic acid for approximately 20 min and rinsed twice in tridistilled water. Once dried, the samples were ground to a fine powder with an agate mortar and pestle. From 12 to 135 mg of powdered samples were dissolved in 6 N HCl, then evaporated to dryness and dissolved again with 1 ml of 2.5 N HCl. Nd was separated by standard cation exchange techniques. Samples were loaded in 2.5 N HCl onto columns containing Bio-Rad AG 50 W X8 resin. The columns were rinsed with 2.5 N HCl and 2.9 N HNO_3, then the REE (Rare Earth Element) fractions were eluted with 4.4 N HNO_3. These fractions were evaporated to dryness, then redissolved in 0.5 ml of 0.16 N HCl. Columns containing HDEHP resin on teflon powder were then used to separate Nd from the other REE, with loading and rinsing in 0.16 N HCl and Nd elution in 0.27 N HCl. Nd isotope analyses were performed on a Finnigan MAT 262 mass spectrometer in static multicollection mode. Nd was run as metal on a double Re-Ta filament with P_2O_5 activator. Typically, 240 isotopic ratios were collected for a given sample. Nd isotope ratios were corrected for mass discrimination by normalizing to $^{146}Nd/^{144}Nd = 0.7219$. In-run uncertainties on the $^{143}Nd/^{144}Nd$ ratios were, on average, 0.000016, and were always less than or equal to 0.000032 (2 standard errors, see Table 1). Six analyses of the La Jolla standard obtained during the period of analysis yielded a $^{143}Nd/^{144}Nd$ ratio of 0.512799 ± 0.000021 (external reproducibility, 2 standard deviations). This value is somewhat lower than that obtained in most other laboratories for the La Jolla standard, so 0.000060 has been added to all values in Table 1 to facilitate interlaboratory comparison (ie, a "true" value of 0.512859 was assumed for the La Jolla standard). A gradual drift of similar magnitude has been observed in the $^{143}Nd/^{144}Nd$ ratio of the CRPG internal J-M Nd standard over the course of the past 7 years. The total procedural Nd blank at the time of analysis was ~1 ng. Blanks were determined by adding a ^{150}Nd spike at the beginning of the total chemical procedure. Total blank concentrations were ~1 ng, which is negligible considering the amount of Nd (700–48,600 ng) processed for each sample. Duplicate analyses were performed (indicated by

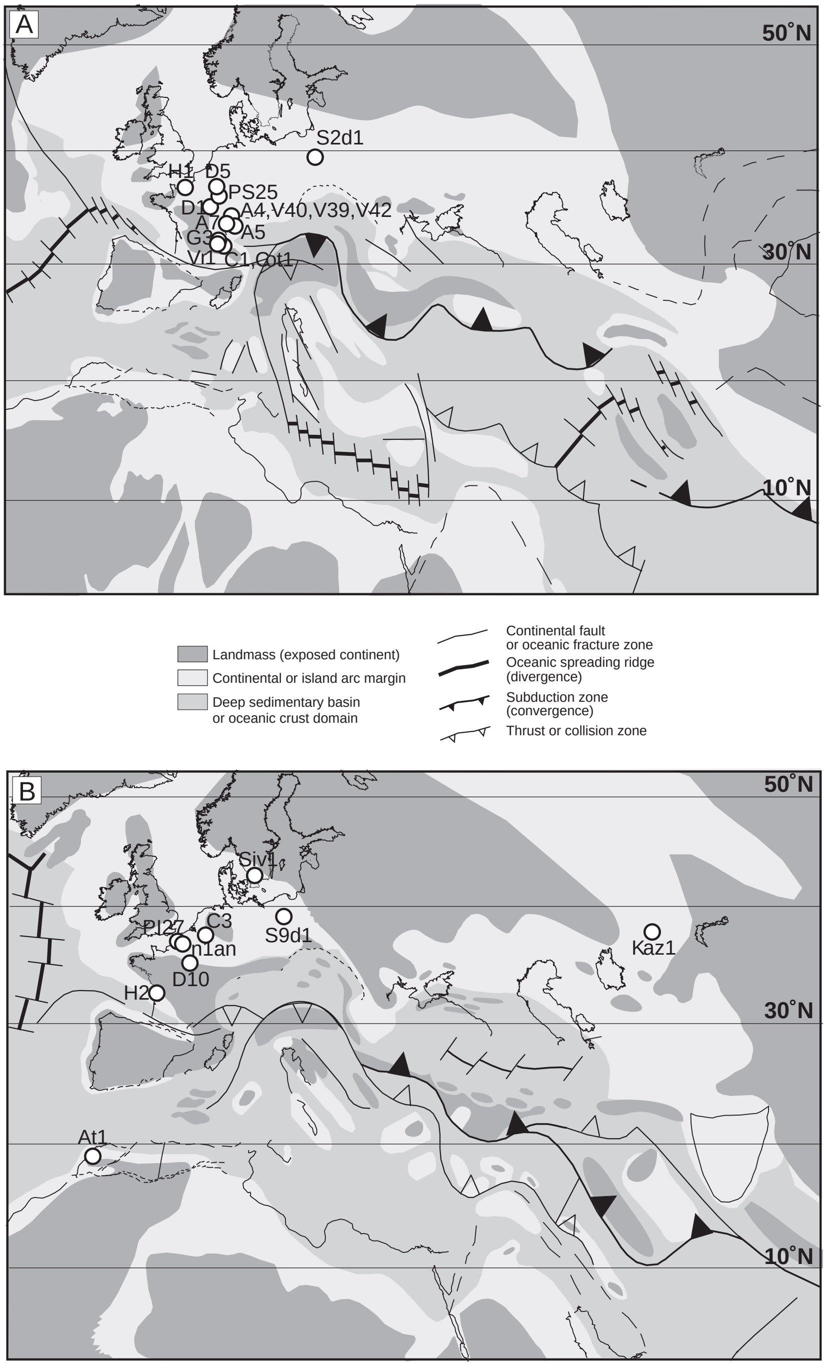
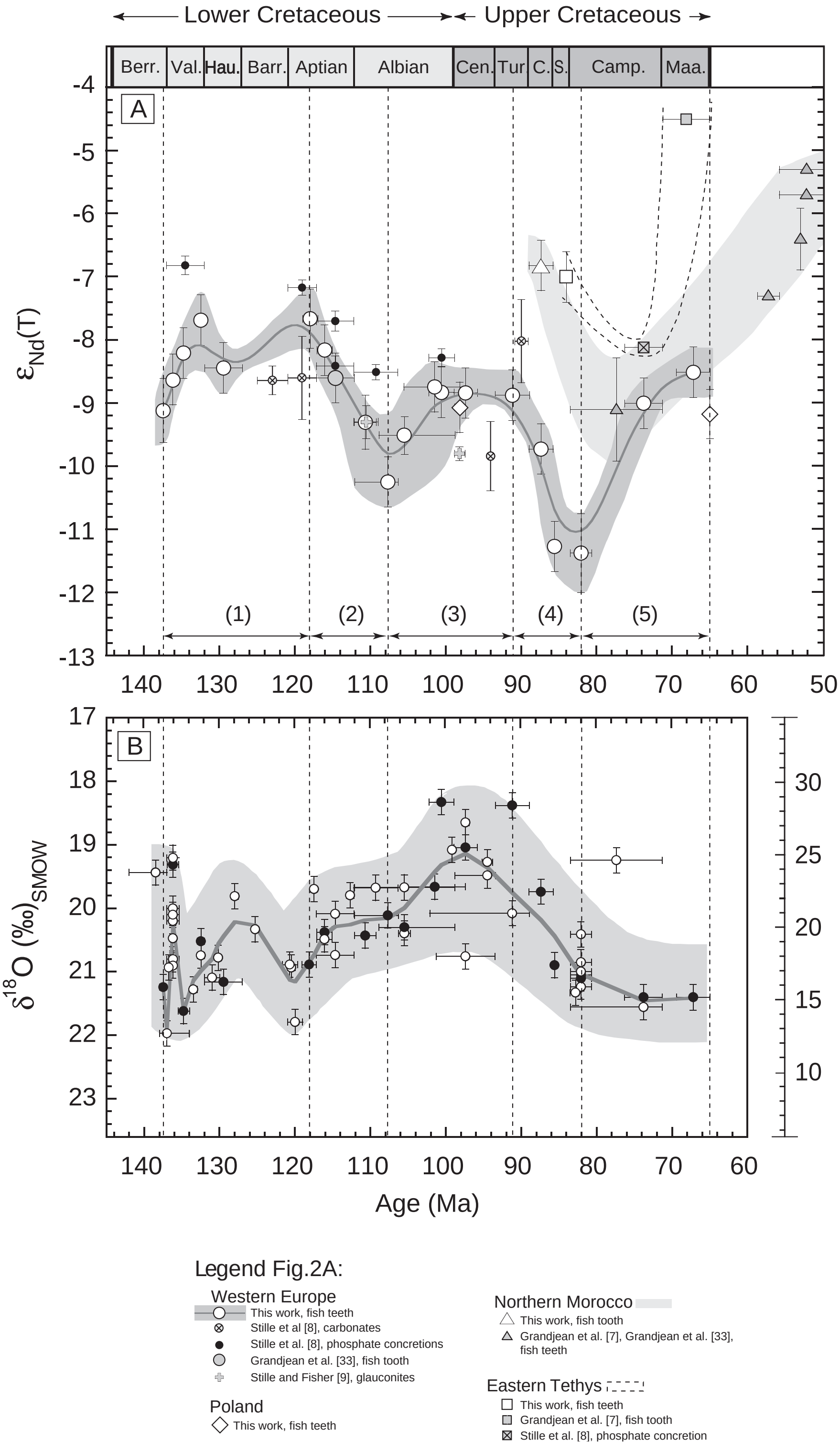


Fig. 1. Location of fish tooth samples analysed in this study for their Nd isotope compositions. Paleogeographic maps are modified from Vrielynck and Bouysse [72]. A) Cenomanian; B) Maastrichtian. All the samples older than the Coniacian are located in Fig. 1A. All the Coniacian–Maastrichtian samples are located in Fig. 1B.


Lower Cretaceous
Upper Cretaceous
Berr.
Val.
Hau.
Barr.
Aptian
Albian
Cen.
Tur
C.
S.
Camp.
Maa.
A
εNd(T)
(1)
(2)
(3)
(4)
(5)
B
δ18O (‰)SMOW
Temperature (°C)
Age (Ma)
Legend Fig.2A:
Western Europe
This work, fish teeth
Stille et al [8], carbonates
Stille et al. [8], phosphate concretions
Grandjean et al. [33], fish tooth
Stille and Fisher [9], glauconites
Poland
This work, fish teeth
Northern Morocco
This work, fish tooth
Grandjean et al. [7], Grandjean et al. [33], fish tooth
Eastern Tethys
This work, fish teeth
Grandjean et al. [7], fish tooth
Stille et al. [8], phosphate concretion

asterisks in Table 1), using separate aliquots of sample powder. All three duplicates yielded results reproducible within the 2σ external uncertainty determined from repeated analysis of the La Jolla standard.

3. Results

3.1. Western Europe and Poland

The initial ε_{Nd} values of the fish teeth from western Europe analysed in this study fluctuate between -11.4 and -7.6 throughout the Cretaceous period (Fig. 2A). Contemporaneous samples from Poland yield Nd isotope compositions similar to those of the samples from western Europe (Table 1, Fig. 2A). The data exhibit little scatter for a given time, and therefore allow definition of a smooth curve. The envelope shown in Fig. 2A includes all of the western European data including uncertainties. This envelope should however be considered to represent the minimum variability in the ε_{Nd} of the NW Tethyan upper ocean waters because variations of at least 1 ε-unit may occur in water columns of more than 500 ms [31].

The earliest Cretaceous is characterised by the highest ε_{Nd} values of the Cretaceous period with an average value of about -8.3 from the Berriasian to the Early Aptian. Within this time interval, the data show a slight increase from -9.1 in the Late Berriasian to -7.7 in the Late Valanginian. The ε_{Nd} values then remained around -8 until the Early Aptian. After the Early Aptian, the ε_{Nd} values began to decrease, reaching a minimum of -10.3 in the Early Albian interval. This minimum was followed by an increase of about 1.5 ε-units towards more radiogenic values that prevailed from the Late Albian to the Turonian. After the Turonian, ε_{Nd} values decreased to about -11.4 in the Santonian–Early Campanian interval. Note that this minimum is defined by only two samples and must be confirmed by further studies. Northwestern Tethyan upper ocean water at the end of the Cretaceous was characterised by an increase towards more radiogenic values, up to -8.5.

3.2. Northern Europe

One sample from the Lower Campanian of Sweden yielded a very unradiogenic value of -17. This sample comes from the margin of the Kristianstad Basin [32] which was part of the boreal realm during the Cretaceous.

3.3. Eastern Tethys

The samples from the eastern Tethys (this study and literature data) have ε_{Nd} values about 4 units higher than those from western Europe during the Santonian and the Maastrichtian, although this gap diminished temporarily in the Late Campanian (Fig. 2A). Like the western European samples, the fish teeth from the eastern Tethys have more radiogenic values in the Maastrichtian (-4.5) than in the Santonian (-7). In contrast with the western European samples, ε_{Nd} values from the eastern Tethys show a small decrease from -7 in the Late Santonian to about -8 in the Late Campanian.

3.4. Northern Morocco

The samples from northern Morocco (this study and literature data) have ε_{Nd} values of around -7 in the Coniacian decreasing to a minimum of about -9

Fig. 2. Evolution of initial ε_{Nd} values of Cretaceous phosphates, carbonates and glauconites deposited in Tethyan upper ocean waters. A) Initial ε_{Nd} values. Note that the uncertainty associated with the age of the samples is quite variable, reflecting a variability in the quality of the biostratigraphic control that reaches a resolution of less than 0.3 Myr for the Berriasian–Valanginian interval. A minimum error of 0.4 ε-unit (corresponding to the external error associated with Nd isotope analyses—see Section 2) has been applied to all samples, except for those having higher internal errors. Carbonate, phosphate and glauconite Nd isotope values from the literature are also presented with error bars when available. Correspondence between stratigraphic and absolute ages is derived from the time scale of Gradstein et al. [28]. The vertical dotted lines delimit time intervals numbered from 1 to 5, which are discussed in the text. The samples from western Europe come from open platform environments and therefore are expected to record the Nd isotope compositions of upper ocean waters (<1000 m). Abbreviations are: Berr=Berriasian; Val=Valanginian; Haut=Hauterivian; Barr=Barremian; Cen=Cenomanian; Tur=Turonian; C=Coniacian; S=Santonian; Camp=Campanian; Maa=Maastrichtian. B) $\delta^{18}O$ values recorded by fish teeth from western Europe (mainly France), from Pucéat et al. [3]. The fish teeth analysed in this study for their Nd isotope compositions are represented by plain black circles. Isotopic temperatures are calculated using the equation of Kolodny et al. [73] with $\delta^{18}O_{seawater}$ of -1‰.

in the Campanian. A trend towards more radiogenic ε_{Nd} values is suggested by the less negative (between −5.3 and −7.3 ε-units) ε_{Nd} values of Late Paleocene–Early Eocene age (Fig. 2A). It is not possible to precisely determine the timing of the ε_{Nd} minimum within the Campanian stage, given the limited number of samples and the large uncertainty on the stratigraphic age of the sample yielding the lowest ε_{Nd} value. Consequently, the Moroccan ε_{Nd} evolution may resemble either that of western Europe or that of the eastern Tethys. Additional data from the Campanian and Maastrichtian are needed to choose between these two possibilities.

4. Discussion

4.1. Comparison of the Nd isotope records obtained from various mineral deposits

Our Nd isotopic data, obtained from fish teeth, are in good agreement with the Nd isotope composition of a fish tooth analysed by Grandjean et al. [33] and with those inferred from phosphate concretions reported by Stille et al. [8] for the same location (Fig. 2A). The Nd isotopic trend displayed by the phosphate concretions is parallel to that defined by our data. This demonstrates the reliability of phosphatic materials, which include both fish teeth and concretions. Note however that the Nd isotope compositions of phosphate concretions tend to be slightly more radiogenic than those of fish teeth of equivalent age.

In contrast, Nd isotopic data obtained from carbonate samples plot either above or below the envelope defined by the fish teeth samples, even if part of the apparent scatter could result from the relatively large associated uncertainties. The significance of the Nd isotope signature of carbonate samples remains the subject of discussion (ex. [34,35]). More specifically, significant changes in the Nd content and isotopic composition of foraminifera after their death have been observed. These changes most likely reflect Nd addition from deeper waters. Therefore, the Nd in biogenic carbonates may represent a mixed signal derived from surface, bottom and pore waters [35]. This may introduce some variability to carbonate samples. In addition, it is not clear from Stille et al. [8] whether Fe–Mn oxyhydroxide coatings have been removed from the carbonates prior to analysis. Fe–Mn coatings acquire their Nd signature from the bottom or pore waters and generally have a significantly higher Nd concentration than biogenic calcite. Therefore, failure to remove these coatings may significantly modify the measured Nd isotope composition of bulk carbonates [34].

To avoid the scatter of the ε_{Nd} values displayed by the carbonate samples, phosphatic materials such as fish teeth should be preferred for paleoceanographic reconstructions.

4.2. Significance of Nd isotope ratios recorded in fish teeth

Several studies have demonstrated that the original Nd isotope composition of fish teeth is resistant to diagenetic alteration [24,27,33]. In addition, the La/Sm ratio of fish teeth can be used to assess the preservation state of biogenic apatites [36]. Reynard et al. [36] have shown that marine biogenic apatites with 'bell-shaped' REE patterns, defined by a La/Sm ratio (normalised to the North American Shale Composite; [37]) lower than 0.3, have been affected by recrystallization processes associated with REE substitution during a stage of "extensive" or "late" diagenesis. The REE concentrations of about half of the samples from western Europe were analysed in a previous study [38]. None of these samples has a La/Sm ratio (normalised to the North American Shale Composite) lower than 0.3, which tends to indicate that their original REE contents were not modified by extensive diagenetic processes [36].

The fish teeth from western Europe analysed in this study are therefore expected to record the Nd isotope composition of upper ocean waters (Table 1). Grandjean et al. [33] emphasized that biogenic phosphates from platform environments should be used with caution for paleoceanographic reconstructions, as their Nd isotope compositions may not always be indicative of an open-sea environment. Only two samples, D1 and D5, have shale-like REE patterns that may reflect a major detrital contribution according to Lécuyer et al. [38]. Note that the Nd isotope curve (Fig. 2A) would be unchanged if these two samples (D1 and D5) were excluded.

Contemporaneous samples from north and southeast France yield similar Nd isotope signatures

(Table 1), suggesting that they shared the same water mass or a similar Nd source. This observation allows use of the samples from both north and southeast France to reconstruct the evolution of the Nd isotope composition of the NW Tethyan upper ocean water. Samples from Poland also have ε_{Nd} values similar to those from western Europe, suggesting again that either these two areas received Nd from continental sources with similar isotopic compositions or they belonged to the same water mass. The latter scenario would imply that oceanic circulation was vigorous enough to ensure a well-mixed water mass in the western Tethyan epicontinental area.

4.3. Significance of Nd isotope variations of seawater

The Nd isotope evolution of a given water mass reflects changes in two factors:

- the amount and/or isotopic composition of Nd discharged into the ocean during continental weathering including Nd exchange between dissolved and particulate phases [39,40].
- the oceanic circulation pattern.

Very little is known about the Nd isotope composition of the oceans during the Cretaceous period. However, the available data suggest that the Nd isotope composition of the Pacific Ocean has remained more radiogenic than that of the Atlantic and Tethys Oceans over the last 200 Ma [41,42]. Therefore, enhanced inputs of Pacific radiogenic waters into the Tethys should result in an increase of the Tethyan ε_{Nd} values. In contrast, boreal waters are expected to have very low ε_{Nd} values due to the input of unradiogenic Nd derived from the erosion of Archean and Proterozoic continental land masses (northern Canada, Greenland, Scandinavia) surrounding the shallow water separating Greenland from Norway and Scotland. This seaway connected the Arctic Ocean with the Atlantic and occasionally the western Tethys [5,43]. The assumed unradiogenic nature of boreal seawater in the Cretaceous is supported by the very low ε_{Nd} value displayed by the tooth from Sweden (about -17; Table 1). Therefore, an increase in the Tethys of the proportion of boreal waters relative to Pacific waters would result in a decrease of the Tethyan ε_{Nd} values.

4.4. Nd isotope evolution of NW Tethyan seawater during the Cretaceous (Fig. 2A)

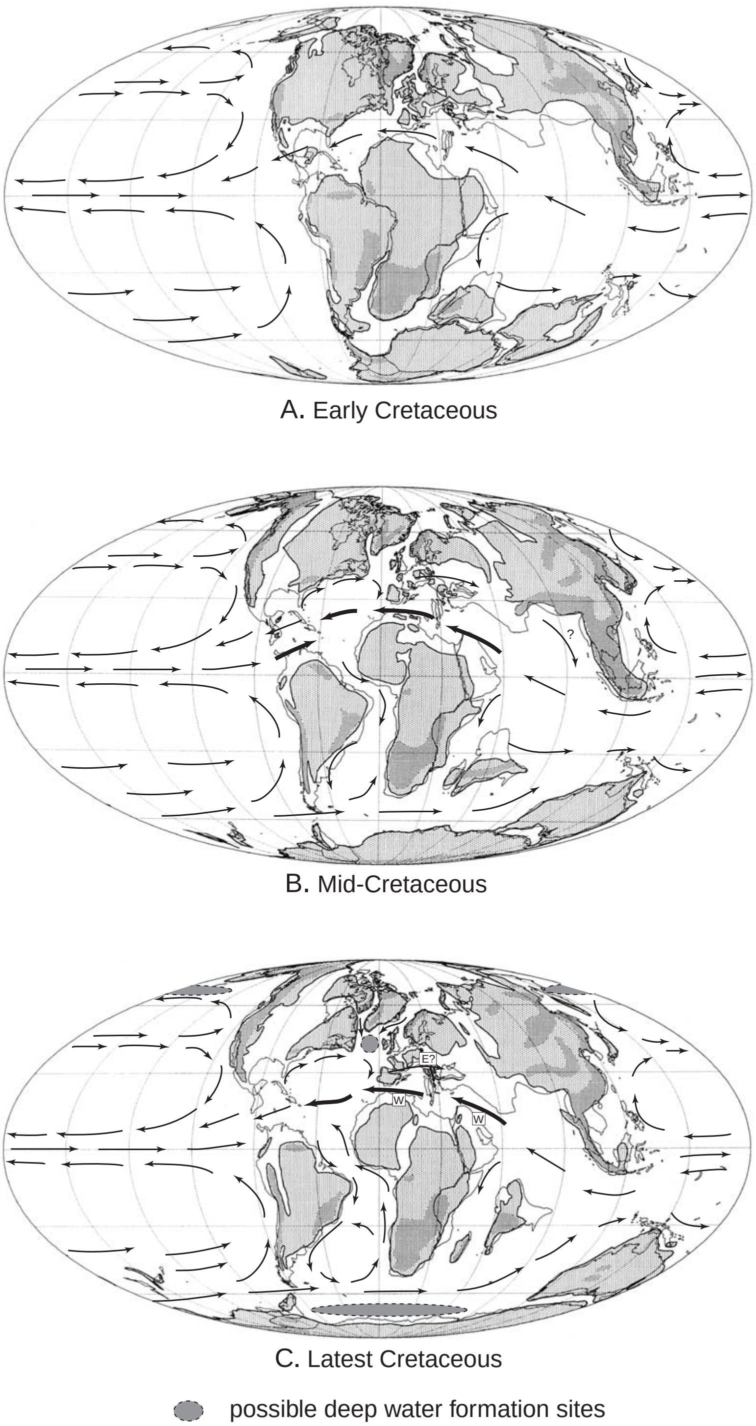
4.4.1. From the Berriasian to the Early Aptian (interval 1)

The initial ε_{Nd} values were relatively high (above -9.1 and up to -7.6) during the Berriasian–Early Aptian, compared to those of the rest of the Cretaceous, although lower than those (close to -7) reported by Stille et al. [44] for the Late Jurassic (Fig. 2A). During the Berriasian–Valanginian interval, the ε_{Nd} increase may point to increasing influence of Pacific seawater in the northwestern Tethyan realm. Stille et al. [8] also suggested that large masses of radiogenic water entered the Tethys during the earliest Cretaceous, which would support the existence of a westward flowing Tethys Circumglobal Current (TCC). This surface current would have allowed large masses of radiogenic Pacific waters to flow through the Indian-Tethys seaway into the NW Tethys (Fig. 3A). Although the TCC has been presumed to exist since the Late Jurassic [8,45,46], its presence throughout the Cretaceous has been recently questioned [47,48]. Our data support a transport of western Pacific upper ocean waters to the Mediterranean area (western Tethyan upper ocean waters), and therefore are in agreement with the existence of a permanent westward flowing Tethys Circumglobal Current from the Late Valanginian until at least the Early Aptian.

The samples characterized by the maximum $\delta^{18}O$ values do not have the minimum ε_{Nd} values. This observation argues against the scenario of intermittent surface currents entering the NW Tethys from the Arctic Ocean [5,43] because boreal waters most likely had very low ε_{Nd} values (see Section 4.3). This result suggests that the rapid cooling events recorded by the oxygen isotope compositions of fish tooth enamel ([3], Fig. 2B) do not result from the influence of boreal oceanic currents but most likely reflect global climatic changes.

4.4.2. From the Early Aptian to the Early Albian (interval 2)

The observed decrease in ε_{Nd} from the Early Aptian to the Early Albian could have either been driven by local or regional changes in the Nd continental supply (weathering of older continental material or increase of continental inputs) or by changes in the oceanic


A. Early Cretaceous
?
B. Mid-Cretaceous
E?
W
W
C. Latest Cretaceous
possible deep water formation sites
surface paleocurrents

circulation pattern. Oxygen isotope compositions recorded in the fish teeth do not increase during this period (Fig. 2B). Therefore it is unlikely that the decrease observed in the ε_{Nd} values was induced by unradiogenic boreal water masses entering the western Tethys. Despite the weak latitudinal temperature gradient that characterised the Cretaceous period (e.g. [1,3]), the occurrence of such southward currents would most probably have been associated with a decrease of the water temperatures in the NW Tethys and therefore by an increase of the fish tooth $\delta^{18}O$.

Since the oxygen isotope compositions of fish teeth do not suggest a large temperature increase from the Early Aptian to the Early Albian, enhanced continental weathering linked to the possible onset of greenhouse conditions is unlikely during this period. Therefore, the large decrease of the ε_{Nd} value is interpreted to result from changes in the sources of Nd. Alteration of older and less radiogenic continental crust would have lowered the ε_{Nd} of seawater.

4.4.3. From the Early Albian to the Turonian (interval 3)

Late Albian to Turonian fish teeth from western Europe record more radiogenic Nd values than those from the Early Albian (Fig. 2A). Although changes in the Nd input from the western European continental masses (alteration of younger continental material or decreased continental inputs) cannot be excluded as a possible cause for the shift towards more radiogenic Nd isotopic values, there are several arguments in favour of a change in the oceanic circulation pattern. The positive shift in Nd isotope ratios toward more radiogenic values coincides with the opening of marine connections between the North Atlantic and South Atlantic basins. General Circulation Model (GCM) experiments performed for the Albian and Turonian suggest that the opening of a gateway between the North and South Atlantic had two important effects on the North Atlantic shallow circulation [49,50]: 1) the westward flow of warm tropical Pacific waters through the Tethys was strengthened, and 2) part of the westward flow of North Atlantic waters into the eastern Pacific through the Caribbean area was reversed (Fig. 3B). Moreover, Poulsen et al. [50] predicted that the opening of the equatorial Atlantic gateway would result in a slight increase of sea surface temperatures of the western Tethyan domain, as documented by the concomitant decrease of the fish tooth $\delta^{18}O$ values interpreted as reflecting sea surface warming (Fig. 2B; [3]). We therefore argue that the positive shift in Nd isotope ratios towards more radiogenic values could result from a larger input of radiogenic shallow Pacific water into the NW Tethys, linked to the opening of marine connections between the North and South Atlantic basins.

4.4.4. From the Turonian to the end of the Cretaceous (intervals 4 and 5)

Fish teeth Nd isotopic data from the Turonian to the end of the Cretaceous display two striking features. Firstly, the Nd isotopic compositions of upper ocean waters from the three different Tethyan areas are distinguishable (northwestern Tethys, eastern Tethys, and northern Morocco). Secondly, there is an overall decrease in ε_{Nd} values of NW Tethyan upper ocean waters after the Turonian that is followed by a recovery of relatively radiogenic values during the Late Campanian and Maastrichtian.

The discrepancy between Nd isotopic data from the various Tethyan areas can arise from:

(1) Changes in erosional inputs at a local or regional scale. This would imply that either the erosion rate in the eastern Tethys was lower than in the NW Tethys or the weathered crust was younger in the eastern Tethys, releasing more radiogenic Nd. At present, particulates from the Nile river that are carried to the Mediterranean sea, have high ε_{Nd} values (~$-3/-1$; [51–53]), presumably reflecting erosion of relatively juvenile Pan-African crust in NE Africa. In contrast, partic-

Fig. 3. Paleogeographic maps of three Cretaceous intervals discussed in the text (from Smith et al. [5]) along with the main surface palaeocurrents (after Poulsen et al. [49,50], Otto-Bliesner et al. [67], Lloyd [74], Roth [75], Bush and Philander [76], Neraudeau and Mathey [77]). Continents are represented by the shaded areas, with higher elevations in darker shades. A. Early Cretaceous (Barremian-Hauterivian map), B. Mid-Cretaceous (Turonian map), C. Latest Cretaceous (Maastrichtian map). The bold arrows indicate intensified surface currents. For the Latest Cretaceous map, the possible sites for deep water formation are represented by grey ellipses (see text for discussion and references). The arrows labelled “E” and “W” are discussed in the text.

ulates from the Po and Rhône rivers, which drain southern Europe, have lower ε_{Nd} values (-10.8 and -9.7, respectively; [54]). Prior to or during the Cretaceous, both East Africa–Arabia and southern Asia experienced episodes of basaltic volcanism [55,56]. Weathering of such radiogenic rocks from either one or both of these areas could have contributed to releasing radiogenic Nd into the eastern Tethys through river discharges during the Cretaceous. Therefore, regional isotopic differences in erosional Nd inputs may partly or totally account for the more radiogenic values of the eastern Tethyan seawater.

(2) The presence of a strong westward surface flow located along the southern Tethyan margin (arrow noted "W" in Fig. 3C) that brought radiogenic Pacific waters into the eastern Tethys and northern Morocco areas. We propose that besides this strong westward surface flow, a weaker eastward surface flow was still active along the northern margin of the Tethys (arrow noted "E" in Fig. 3C) but this flow no longer reached the Indian Ocean. The influence of North Atlantic waters would be then stronger in the NW area of the Tethys. The closer ε_{Nd} values of eastern and western Tethyan sea waters in the Late Campanian may then arise from a temporary eastward extension of this surface current, resulting in more efficient mixing of the Tethyan upper ocean waters.

However, neither changes in erosional inputs nor distinct local water masses in the three areas can explain the overall ε_{Nd} pattern observed in the NW Tethyan upper ocean waters. Indeed, model results based on the evolution of the strontium isotope composition of seawater suggest that variations in continental weathering were limited during the Late Cretaceous (e.g. [57]). Another explanation involves the input of deep waters to the Tethyan realm coming from the North Atlantic during the Coniacian and Early Campanian, and from the Southern Ocean or the North Pacific during the Late Campanian and Maastrichtian. The progressive narrowing of the Tethyan circumglobal passage that occurred during the Senonian (Coniacian–Maastrichtian interval) [55], along with the global cooling which may have intensified the Trade Winds and therefore strengthened the TCC, could have favoured the development of extensive upwellings in the Tethyan area [58–61] (Fig. 3C). Several arguments support an intensification of low-latitude upwellings during the Coniacian–Maastrichtian interval: 1) deposition of giant phosphorites along the Tethyan margins during the Late Cretaceous, peaking in the Coniacian–Maastrichtian period [58,60,61], 2) microfossil assemblage compositions representative of highly productive paleoenvironments along the southern Tethyan margins [62–65]. Therefore during the Senonian, deep waters likely contributed to the Nd isotopic signature of the Tethyan upper ocean waters, the ε_{Nd} values of individual deep water masses being largely influenced by the composition of Nd discharged into their source regions [18,20,23]. Most studies suggest that high latitudes were the dominant sites of deep water formation during the Late Cretaceous [49,66,67]. Source of deep water formation at high latitudes could have been the Southern Ocean [49,66,67], the North Pacific [67,68], or the North Atlantic [69,70]. A North Atlantic source for the Tethyan deep waters would result in a contribution of very unradiogenic Nd to the upper ocean Tethyan waters through the upwellings. In contrast, a North Pacific or a Southern Ocean source would result in a contribution of more radiogenic Nd [71].

Both the occurrence of upwellings and the $\delta^{18}O$ evolution of the fish teeth lead us to propose the following scenario for Nd isotope evolution of the upper ocean Tethyan waters throughout the Coniacian–Maastrichtian interval. From the Coniacian to the Early Campanian (interval (4) in Fig. 2A), the correlation of the ε_{Nd} drop with a simultaneous increase of fish teeth $\delta^{18}O$ values (Fig. 2) could reflect the intensification of upwellings. This isotopic pattern is linked to the global cooling and an increased contribution of deep water sourced in the North Atlantic Ocean, providing cool and unradiogenic waters. From the Early Campanian to the Maastrichtian (interval (5) in Fig. 2A), the recorded ε_{Nd} increase in the Tethys could then result from a change in the Tethyan deep water supply, from a dominant North Atlantic to a dominant North Pacific or Southern Ocean source. This change in the Tethyan deep water supply could be linked to the progressive narrowing of the Tethyan seaway leading to a shutdown or at least a severe confinement of the North Atlantic source. Consequently, Pacific waters or Southern Ocean waters could have become predominant again at the end of the Cretaceous. We emphasize that this scenario is based on the limited existing Nd isotope data set for the different oceans and that additional fish teeth data are needed to document more precisely the evolution of Cretaceous North Atlantic and Pacific upper water masses.

5. Conclusions

The rather high ε_{Nd} (up to -7.6) of the NW Tethyan upper ocean waters recorded from the Late Berriasian to the Early Aptian and the absence of negative excursions during this interval are consistent with the presence of a permanent westward flowing Tethys Circumglobal Current. This result suggests that the rapid temperature variations, inferred from the $\delta^{18}O$ values of fish tooth enamel during the same interval, were not driven by episodic influxes of boreal waters but rather by global climate change.

The Early Aptian–Early Albian interval is marked by a decrease of the ε_{Nd} values. We suggest that local or regional changes in the Nd continental supply (weathering of older continental material) may have driven the evolution of the NW Tethyan upper ocean waters.

The subsequent shift towards more radiogenic ε_{Nd} values during the Late Albian–Turonian interval is interpreted to reflect a change in the oceanic circulation pattern. This shift coincides with the opening of marine connections between the North and South Atlantic basins that may have strengthened the westward flow of radiogenic Pacific waters in the Tethys. Therefore, the influence of Pacific water masses upon the western Tethyan upper ocean waters became more significant.

The Coniacian–Maastrichtian interval is characterized by a fairly large decrease of the NW Tethyan ε_{Nd} values until the Santonian–Early Campanian, followed by an increase towards more radiogenic values. The progressive narrowing of the Tethyan seaway combined with the climatic cooling documented by the $\delta^{18}O$ values of fish teeth could have produced an intensification of upwelling in the Tethys and a more important contribution of deep waters to the upper ocean waters. Likely formed at high latitudes, these deep waters would have amplified the decrease of the tropical temperatures. The Nd isotope variations may then be interpreted in terms of changes in the deep water sources, from high latitude North Atlantic (producing unradiogenic deep waters) to high latitude North Pacific or Southern Ocean (producing more radiogenic deep waters). We emphasize however that this scenario must be verified by additional fish tooth Nd isotope data from various marine areas and by numerical simulations obtained from a coupled ocean-climate model.

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