Thermal evolution of Cretaceous Tethyan marine waters inferred from oxygen isotope composition of fish tooth enamels

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[1] The evolution of subtropical (30–35°N) upper ocean temperatures through the Cretaceous is inferred from the oxygen isotope compositions of 64 fish teeth (enamel) coming from the western Tethyan platform. Mean $\delta^{18}O$ values of 22‰ at the Berriasian-Valanginian boundary decrease, with oscillations to 18.5‰ around the Cenomanian-Turonian boundary, and progressively increase to 21.5‰ by the end of the Cretaceous. The similarity of this oxygen isotope curve for bioapatites from platform environments with those for foraminifera and bulk carbonates that were deposited in deeper waters and at other paleolatitudes indicates that they record global climatic signals. Major cooling events at the million-year scale can be distinguished: (1) at the Berriasian-Valanginian boundary and (2) during the earliest Late Valanginian. A third cooling event is detected during the earliest Aptian. These events, already proposed as icehouse interludes during the lower Cretaceous, are also recorded at subtropical latitudes. A progressive warming is identified from the Aptian to the Cenomanian-Turonian interval that corresponds to a thermal optimum, and then upper ocean temperatures decreased to the Maastrichtian. Minimum isotopic temperatures range from 15°C to 28°C, assuming a $\delta^{18}O_{seawater}$ of −1‰, for an ice-free world. Taking more realistic $\delta^{18}O_{seawater}$ values of ~0‰ for tropical waters, during glacial periods (within the Berriasian-Valanginian interval, and earliest Aptian) or with above average salinities (possibly the Maastrichtian), temperatures are increased by 4–5°C. Temperature differences between climatic extremes of the Valanginian and Cenomanian-Turonian are estimated to have been 10°C. Latitudinal thermal gradients for the Albian-Cenomanian, Turonian, and Maastrichtian were 0.2–0.3°C/° latitude and thus weaker than modern oceanic values at about 0.4°C/° latitude. *INDEX TERMS:* 1620 Global Change: Climate dynamics (3309); 4267 Oceanography: General: Paleoceanography; 4825 Oceanography: Biological and Chemical: Geochemistry; 4870 Oceanography: Biological and Chemical: Stable isotopes; *KEYWORDS:* Cretaceous, climate, oxygen isotopes, fish teeth

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1. Introduction

[2] During the past decades, the Cretaceous period has largely been studied for its anoxic events, biological crises, and climatic modes. The Cretaceous is considered as the warmest and relatively equable period of the Phanerozoic [*Frakes*, 1979; *Barron*, 1983; *Hallam*, 1985], although recent studies have suggested the existence of cool periods during the Early Cretaceous [*Frakes and Francis*, 1988; *Gregory et al.*, 1989; *De Lurio and Frakes*, 1999; *Price*, 1999; *van de Schootbrugge et al.*, 2000]. Stable isotope proxies, mainly derived from carbonate-skeletons fauna, have already been used to quantify the evolution of marine temperatures [*Savin*, 1977; *Barrera et al.*, 1987; *Pirrie and Marshall*, 1990; *Ditchfield et al.*, 1994; *D'Hont and Arthur*, 1996]. However, climatic reconstructions have been limited to certain Cretaceous intervals [*Kolodny and Raab*, 1988; *Huber et al.*, 1995; *Barrera et al.*, 1997; *Li and Keller*, 1998; *Norris and Wilson*, 1998; *Clarke and Jenkyns*, 1999; *Voigt and Wiese*, 2000]. Attempts to establish a global climatic trend throughout the Cretaceous [*Ditchfield et al.*, 1994; *Frakes*, 1999] have been based on compilations that integrate oxygen isotope data from various fauna (foraminifera, belemnites, ammonites), but they generally lack documentation for the Early Cretaceous. This period is of interest because of the possible occurrence of icehouse interludes.

[3] The oxygen isotope composition of PO_4 from fish remains has widely been used for paleotemperature reconstructions of past seawater [*Kolodny and Luz*, 1991; *Lécuyer et al.*, 1993; *Picard et al.*, 1998; *Vennemann and Hegner*, 1998]. The phosphate-water oxygen isotope system is characterized by at least three interesting properties for estimating marine paleotemperatures that are (1) the good resistance of tooth enamel relative to dentine or bone to diagenetic alteration [*Kolodny et al.*, 1983, 1996], (2) a

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unique fractionation equation applicable to all fish species [*Kolodny et al.*, 1983; *Lécuyer et al.*, 1996; *Venneman et al.*, 2001], and (3) the possibility to collect fish remains from open platform environments throughout the Cretaceous.

[4] The purpose of this study is to establish for a given paleolatitude a long-term $\delta^{18}O$ curve for the Cretaceous (upper Berriasian to Maastrichtian) by analyzing fish tooth enamel. Our study gathers $\delta^{18}O$ values from 64 Cretaceous fish remains, mostly tooth enamel, whose stratigraphic positions give an average time resolution of 4 Myr for the whole Cretaceous, and down to 0.25 Myr for the earliest Cretaceous. Sixty samples come from the western Tethyan open marine platform environments (France and Switzerland) that remained at low latitudes (30°N–35°N) [*Scotese et al.*, 1988] throughout the Cretaceous. The $\delta^{18}O$ curve will be compared to previously published data obtained from fish remains [*Kolodny and Raab*, 1988], planktonic foraminifera [*Huber et al.*, 1995], and bulk carbonates [*Clarke and Jenkyns*, 1999] of various paleolatitudes. The comparisons will be used to discuss the relevance of our data in terms of global or regional climatic variations. Finally, we combined four new $\delta^{18}O$ data from additional low-latitude sites (Morocco and Israel) with published data to discuss both the evolution of low-latitude marine temperatures and latitudinal thermal gradients.

2. Samples and Techniques

[5] Sixty-four teeth and one bone from 28 selachians (sharks), 5 teleosteans (pycnodonts), and 31 undetermined fish have been analyzed for their PO_4 oxygen isotope compositions (Table 1). Enamel has been separated from the dentine except in the case of the smallest teeth for which only bulk analyses are available (see Table 1). Phosphate from biogenic apatites was isolated as Ag_3PO_4 crystals following the procedure of *Lécuyer et al.* [1993], which was adapted from *Crowson et al.* [1991]. CO_2 was extracted from silver phosphate using the graphite method [*O'Neil et al.*, 1994; *Lécuyer et al.*, 1998], and analyzed with a VG Prism™ mass spectrometer at the Ecole Normale Supérieure of Lyon. Oxygen isotope compositions are reported in the δ notation relative to SMOW. Repeated analyses of phosphorite NBS120c gave an average $\delta^{18}O$ value of 21.7‰ and reproducibility is better than ±0.2‰.

[6] Rare earth element (REE) compositions of a selection of samples have been used in order to detect and assess the importance of possible diagenetic processes. *Reynard et al.* [1999] have shown that biogenic apatites with bell-shaped REE patterns and La/Sm ratios (normalized to the North American Shale Composite) lower than 0.3 recrystalized in the presence of water during a stage of "extensive" or "late" diagenesis. The REE contents of 28 samples among those presented in this work were analyzed by *Lécuyer et al.* [2001], and the pertinent ratio is reported in Table 1. Three of these 28 samples show REE patterns with La/Sm ratios lower than 0.3 and therefore they should be considered with caution.

3. Results

[7] The $\delta^{18}O$ values of PO_4 from Cretaceous fish remains of the western Tethys range between 18.5‰ and 22‰ SMOW (Table 1). The absolute ages reported in Table 1 are derived from the timescale of *Gradstein et al.* [1994]. The range of $\delta^{18}O$ values with time is shown on Figure 1. Note that the uncertainty associated with the age of the samples is quite variable. It reflects variability in the quality of the biostratigraphic control, with the best, particularly for the Berriasian-Valanginian, being better than 0.3 Myr. The range of values for a given age is typically around 1.5‰, but invariably less than 2.8‰. The highest range of 2.8‰ is obtained for the Valanginian period. Because of these variations, a tendency curve is shown on Figure 1. This tendency curve was computed using a fitting function based on the locally weighted least squared error method, with a smoothing factor of 20%. The earliest Cretaceous interval investigated in this study (from upper Berriasian-Valanginian to lower Hauterivian) shows rapid oscillations of about 2.5‰ on a million-year scale, around a mean $\delta^{18}O$ value of 20.6‰. Two 'positive excursions' of the $\delta^{18}O$ values are identified: (1) possibly at the Berriasian/Valanginian boundary, and (2) in the lowermost upper Valanginian (Verrucosum zone). A small $\delta^{18}O$ increase can also be detected during the earliest Aptian. Oxygen isotope compositions progressively decreased throughout the lower Cretaceous for 30 Myr reaching a minimum value close to 18.5‰ near the Cenomanian-Turonian interval. The $\delta^{18}O$ values steadily increased from the Turonian until the upper Campanian, and are then followed by apparently uniform values close to 21.5‰ during the Maastrichtian. This curve confirms that the latest Albian-Turonian period is characterized by the lowest $\delta^{18}O$ values documented throughout the Cretaceous.

4. Discussion

[8] The $^{18}O/^{16}O$ ratio recorded in fish tooth phosphate during its formation depends on both temperature and the isotopic composition of the environmental water. Knowledge of the $\delta^{18}O_{seawater}$ is therefore crucial for the temperature calculation. Variations in the oxygen isotope composition of seawater are governed by several factors including (1) the evolution of the mass of continental ice that modifies the $\delta^{18}O$ of seawater by storing preferentially ^{16}O in ice; (2) local evaporation/precipitation ratio and continental runoff that influence both the salinity and the local $\delta^{18}O$ of seawater.

[9] The Cretaceous period has often been considered to be 'ice-free' because of the absence of glacial deposits during its major part. This would correspond to a $\delta^{18}O$ for ocean water close to -1‰ SMOW, as suggested for an ice-free Earth by *Shackleton and Kennett* [1975]. However, ice caps (though probably smaller than today) may have been episodically present during the earliest Cretaceous [*Frakes and Francis*, 1988; *Gregory et al.*, 1989; *Stoll and Schrag*, 1996; *De Lurio and Frakes*, 1999; *Price*, 1999] and even during the mid-Cretaceous [*Blattner et al.*, 1997; *Ramstein et al.*, 1997; *Stoll and Schrag*, 2000]. We must also keep in mind that at tropical latitudes, the evaporation/precipitation ratio tends to be higher than 1 resulting in increased salinity and $\delta^{18}O$ values of surface waters. We thus propose to calculate marine paleotemperatures for $\delta^{18}O_{seawater}$ values of -1‰ and 0‰. Because of the mobility of fish through the water column, these temperatures should be considered as upper ocean and not sea surface temperatures. Figure 1

Table 1. Description, Location, Age, and Oxygen Isotope Compositions of Cretaceous Fish Samples[a]

Samples	Location	Fauna, remains	Stratigraphic Age (Ammonite Zone or Horizon)	Age, My	$\delta^{18}O$, ‰ SMOW	Temperature a, °C	Temperature b, °C	REE Analyses
C3	Eben-Emael, Belgium	*Squalicorax pristodontus*, Te	U. Maastrichtian	67.2	21.4	15.2	19.6	(3)
H2	Malakoff (Charentes Maritimes)	undetermined, Te	U. Campanian	73.8	21.4	15.2	19.6	(1)
H5	Meudon (Hauts de Seine)	undetermined, Tw	U. Campanian	73.8	21.6	14.5	18.9	(3)
D12	Beauval (Somme)	undetermined, Te	Campanian	77.4	19.2	24.6	29.0	(1)
N3an	Beauval (Somme)	*Anomotodon* sp., Te	L. Campanian	82.1	21.2	15.9	20.3	(3)
N3sq	Hallencourt (Somme)	*Squalicorax kaupi*, Te	L. Campanian	82.1	20.4	19.5	23.9	(3)
PC21	Puchevillers (Somme)	undetermined, Te	L. Campanian	82.1	20.9	17.6	22.0	(1)
M3	Hallencourt (Somme)	*Scapanorhynchus* sp., Tw	L. Campanian	82.1	21.0	16.9	21.3	(1)
D10	Sens (Yonne)	*Squalicorax pristodontus*, Te	L. Campanian	82.1	21.1	16.5	20.9	(1)
D11	Sens (Yonne)	*Cretolamna appendiculata*, Te	L. Campanian	82.1	21.0	16.7	21.1	(3)
N2an	Beauval (Somme)	*Anomotodon* sp., Te	basal Campanian	82.8	21.3	15.5	19.9	(3)
N1an	Beauval (Somme)	*Anomotodon* sp., Te	basal Santonian	85.6	20.9	17.4	21.8	(3)
PI27	Vallée de la Maye (Somme)	undetermined, Te	Coniacian	87.4	19.7	22.5	26.9	(1)
G3	Le Teil (Ardèche)	Lamniforme, Te	Turonian	91.2	18.4	28.4	32.8	(3)
PI23	bassin de Vivier et du Teil (Ardèche)	undetermined, Te	Turonian	91.2	20.1	21.0	25.4	(1)
M2	Le Mans (Sarthe)	*Squalicorax falcatus*, Tw	U. Cenomanian (Guerangeri zone)	94.5	19.3	24.5	28.9	(1)
M1	Les Renardières (Charentes Maritimes)	*Carcharias amonensis*, Tw	U. Cenomanian (Guerangeri zone)	94.5	19.5	23.6	28.0	(1)
PS29	La Hèvre (Meuse)	undetermined, Tw	L. Cenomanian	97.4	20.8	18.0	22.4	(3)
H1	Cap de la Hève (Seine-inférieure)	undetermined, Te	L. Cenomanian	97.4	19.0	25.5	29.9	(3)
D9	Neuvy-Sautour (Yonne)	*Lamna acuminata*, Te	L. Cenomanian	97.4	18.6	27.3	31.6	(1)
Bl1	Blieux (Alpes de Haute Provence)	undetermined, Te	U. Albian (Dispar zone, Perinflatum horizon)	99.2	19.1	25.4	29.8	(3)
A5	Entrèves (Savoie)	undetermined, Te	U. Albian (Mortoniceras inflatum zone)	100.6	18.3	28.7	33.0	(3)
Vr1	Salazac (Gard)	undetermined, Te	Vraconian	101.5	19.7	22.8	27.2	(3)
PI26	Viry (Jura)	undetermined, Te	Albian	105.5	19.7	22.8	27.2	(1)
D7	Courcelles (Aube)	Lamniforme, Te	M. Albian (Lyelli to Intermedius zone)	105.5	20.4	19.6	24.0	(1)
D5	Ardennes	*Odontaspis*, Tw	M. Albian (Lyelli to Intermedius zone)	105.5	20.3	20.0	24.4	(1)
PS25	La Houpette (Meuse)	*Otodus* sp., Te	upper part of the L. Albian	107.7	20.1	20.9	25.2	(1)
D4	Grusse (Jura)	Lamniforme,Te	L. Albian	109.3	19.7	22.7	27.1	(2)
A7	Lancrans (Ain)	undetermined, Te	L. Albian (Tardefurcatus Mamillatum zone)	110.7	20.4	19.5	23.8	(3)
Apt1	Arnayon (Rhône-Alpes)	undetermined, Tw	U. Aptian (Jacobi zone)	112.7	19.8	22.2	26.6	(3)
D3	Trou du Mège, Allan (Ardèche)	*Pycnodus* sp., Tw	U. Aptian	114.7	20.7	18.1	22.5	(1)
D2	Trou du Mège, Allan (Ardèche)	*Otodus* sp., Te	U. Aptian	114.7	20.1	20.9	25.3	(1)
C1	La Tuilière (Vaucluse)	*Protolamna sokolovi*, Tw	Gargasian	116.1	20.4	19.7	24.1	(3)
D13	Martigues (Bouches-du-Rhône)	undetermined, Te	Gargasian	116.1	20.5	19.2	23.6	(1)
A1	La Lance, Switzerland	undetermined, Te	U. Bedoulian (Furcata zone)	117.5	19.7	22.7	27.0	(3)
D6	Bellegarde (Ain)	Odontaspididae *gracilis*, Te	L. Bedoulian	120	21.8	13.5	17.9	(1)
A2	Gorges du Frou (Chartreuse)	undetermined, Te	L. Bedoulian (Tuarkyricus zone)	120.5	20.9	17.2	21.6	(3)
Bd1	St Jean de Conz (Savoie)	undetermined, Tw	Bedoulian (lower part of the Tuarkyricus zone)	120.7	20.9	17.4	21.8	(3)
Ba2	La Béguère (Vercors)	undetermined, Tw	L. Barremian (lower part of the Caillaudianus zone)	125.3	20.3	19.9	24.3	(3)
Ha6	Vaulion, Switzerland	undetermined, Te	L. Hauterivian (Balearis zone to the lower part of the Angulicostata *Auct.*)	128	19.8	22.2	26.5	(3)
D1	Bleigny-le-Carreau (Yonne)	*Sphaerodus neocomiensis*, Te	Hauterivian	129.5	21.2	16.2	20.6	(1)
A3	Grand Essert (Jura)	undetermined, Tw	L. Hauterivian (Nodosoplicatum zone)	130.2	20.8	17.9	22.3	(3)
H4	St Pierre de Cherennes (Isère)	undetermined, Te	L. Hauterivian	131	21.1	16.5	20.9	(3)
A4	Cenoran (Jura)	undetermined, Te	U. Valanginian (Callidiscus zone)	132.5	20.5	19.1	23.4	(3)

Table 1. (continued)

Samples	Location	Fauna, remains	Stratigraphic Age (Ammonite Zone or Horizon)	Age, My	$\delta^{18}O$, ‰ SMOW	Temperature a, °C	Temperature b, °C	REE Analyses
A6	Ste Croix, Switzerland	undetermined, Tw	U. Valanginian (Callidiscus zone)	132.5	20.7	18.1	22.5	(3)
Tr1	Les Jouvencelles (Jura)	*Pycnodus couloni*, Te	U. Valanginian (Trinodosum zone)	133.5	21.3	15.7	20.1	(3)
V42	Auberson, Switzerland	undetermined, Tw	U. Valanginian (Pronecostatum horizon)	134.8	21.6	14.2	18.6	(1)
Pe1	St Laurent sous Coirons (Ardèche)	undetermined, Te	L. Valanginian (Pertransiens zone)	136.2	20.5	19.3	23.6	(3)
V5	St Symphorien (Gard)	*Sphenodus* sp., Te	L. Valanginian	136.2	20.8	17.8	22.2	(3)
V4a	Moules et Baucels (Gard)	*Paraorthacodus* sp., Te	L. Valanginian	136.2	20.1	20.9	25.3	(3)
V2a	Moules et Baucels (Gard)	*Sphenodus* sp., Te	L. Valanginian	136.2	20.0	21.3	25.7	(3)
V39	Ponte du Suchet, Switzerland	undetermined, b	L. Valanginian	136.2	19.3	24.4	28.7	(1)
V2b	Moules et Baucels (Gard)	*Sphenodus* sp., Te	L. Valanginian	136.2	20.2	20.4	24.8	(3)
V3a	Moules et Baucels (Gard)	*Welcomia bodeuri,* Te	L. Valanginian	136.2	20.2	20.4	24.8	(3)
V1a	Moules et Baucels (Gard)	*Sphenodus* sp., Te	L. Valanginian	136.2	20.9	17.4	21.8	(3)
V1b	Moules et Baucels (Gard)	*Sphenodus* sp., Te	L. Valanginian	136.2	19.2	24.8	29.2	(3)
VSR	La Cabane (Gard)	undetermined, Te	L. Valanginian (Otopeta zone)	136.75	20.93	17.3	21.6	(2)
G2	Val de Fier (Haute Savoie)	*Pycnodus*, Tw	U. Berriasian - L. Valanginian (Alpillensis to Pertransiens zone)	137	22.0	12.7	12.7	(1)
V40	Bonvillars, Switzerland	undetermined, Te	U. Berriasian (Alpillensis zone)	137.5	21.2	15.9	15.9	(1)
G1	Beaulieu (Ardèche)	undetermined, Tw	U. Berriasian	138.5	19.43	23.8	28.2	(2)
H3	Youssoufa, Morocco	undetermined, Te	Maastrichtian	68	20.4	19.6	19.6	(3)
Mmsel	Oued Zem, Morocco	Lamniforme, Te	Maastrichtian	68	20.2	20.4	20.4	(3)
Cav1	Goulmina, Morocco	Pachyrhizodontidae, Tw	L. Turonian	92.6	17.2	33.6	33.6	(3)
PI11	Giv'at Mador, Israel	*Cretolamna* sp., Te	Maastrichtian	68	20.0	21.3	21.3	(3)

[a]Fauna remains: Te (tooth enamel and enameloid), Tw (whole tooth), b (bone). Age: L (lower), M (middle), U (upper). Absolute ages are derived from the timescale of *Gradstein et al.* [1994]. Temperatures have been calculated using the fractionation equation given by *Kolodny et al.* [1983]. Temperatures "a" and "b" are calculated using a $\delta^{18}O_{seawater}$ of −1‰ and 0‰ (SMOW), respectively. REE analyses: (1): samples which have a La/Sm higher than 0.3 (see text and *Reynard et al.* [1999] for discussion); (2) samples which have a La/Sm ratio lower than 0.3; (3) no REE analyses available.

presents the δ^{18}O evolution through the Cretaceous along with the corresponding isotopic marine temperatures calculated using the equation of *Kolodny et al.* [1983]:

$$T(^\circ C) = 113.3 - 4.38\left(\delta^{18}O_{measured} - \delta^{18}O_{seawater}\right).$$

[10] An envelope of ±0.8‰ around the δ^{18}O-tendency curve includes essentially all of the data set. This isotopic range is comparable to the one measured by *Vennemann et al.* [2001] on recent fish teeth, which is typically between 0.6 and 1.1‰ for different teeth from a single shark. This naturally occurring variation in oxygen isotope compositions recorded in fish teeth may reflect the diversity of the living environments which includes: (1) thermal gradients within the water column; (2) seasonal thermal variations since the growth of a fish tooth represents less than a year (several weeks to several months depending on species); (3) coastline proximity and the influence of freshwater inputs; and (4) the possible occurrence of surficial oceanic currents.

[11] In addition, it cannot be excluded that a component of the variation in the oxygen isotope compositions of fish teeth results from limited diagenetic perturbations. However, we note that two of the three teeth with possible REE evidence for diagenetic alteration (Figure 1) plot quite close to the tendency curve, suggesting that diagenesis is not a significant problem even though the third sample (from the Berriasian) might have suffered slight diagenetic overprint, as diagenetic alteration normally tends to lower δ^{18}O values [*O'Neil*, 1987].

[12] Taking into account the above constraints, the significance of the oxygen isotope temperature curve is discussed in terms of regional versus global climatic variations. We compare our curve with those inferred from previous studies [*Kolodny and Raab*, 1988; *Huber et al.*, 1995; *Clarke and Jenkyns*, 1999] that were performed on different biogenic materials sampled at various paleolatitudes (Figure 2). The Cretaceous curve of *Kolodny and Raab* [1988] is based on oxygen isotope analyses of fossil fish teeth and bones from Israel and Sinai that were located from 5°N of paleolatitude for the Early Cretaceous to 20°N for the Late Cretaceous, according to *Scotese et al.* [1988]. Note that there are only two data for the lower Cretaceous. The curve proposed by *Clarke and Jenkyns* [1999] for the Aptian–Maastrichtian interval was established by using both fine fractions (<63 μm)

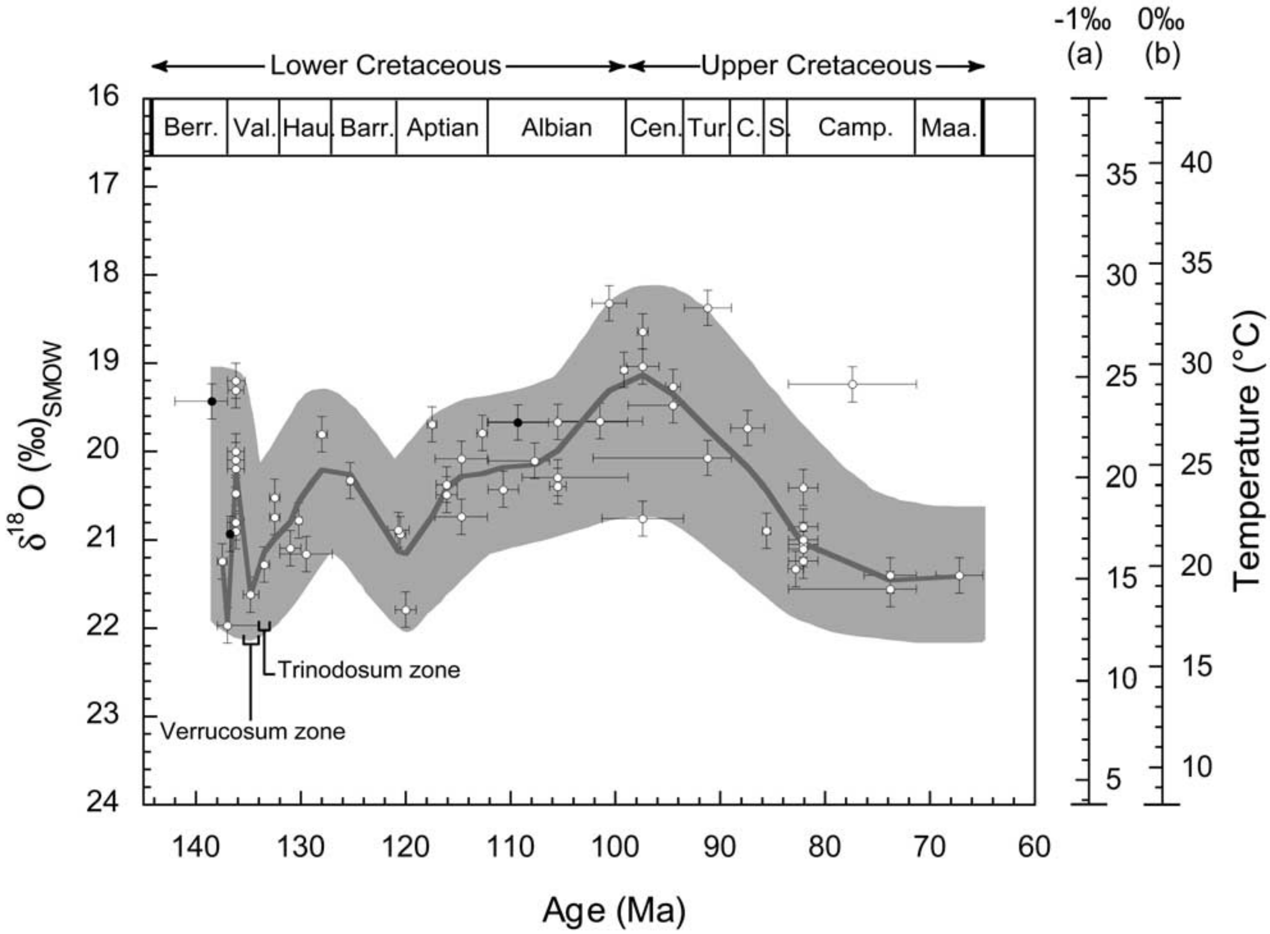

Figure 1. Evolution of $\delta^{18}O$ values of fish teeth from the western Tethys during the Cretaceous (Table 1). Open circles mark samples with a La/Sm ratio higher than 0.3 and solid circles mark samples with a La/Sm ratio lower than 0.3 (see the text for more details). Isotopic temperatures are calculated using the equation of *Kolodny et al.* [1983] with $\delta^{18}O_{seawater}$ of (a) −1‰ and (b) 0‰. Absolute ages are derived from the timescale of *Gradstein et al.* [1994]. The solid curve represents the variation of the mean value with time, computed using a fitting function based on the locally weighted least squared error method with a smoothing factor of 20%, and its envelope at ±1‰. The position of the sample on its temporal error bar is determined by its most probable stratigraphic position. Two important ammonite zones are given for reference. Abbreviations are: Berr = Berriasian; Val = Valanginian; Haut = Hauterivian; Barr = Barremian; Cen = Cenomanian; Tur = Turonian; C = Coniacian; S = Santonian; Camp = Campanian; Maa = Maastrichtian.

and bulk samples of pelagic and hemipelagic chalk and calcareous claystones from the Southern Hemisphere (Exmouth Plateau, from 54°S of paleolatitude for the Aptian to 43°S for the Late Cretaceous). Data from *Huber et al.* [1995] were obtained on planktonic foraminifera from the Falkland Plateau and the Naturalist Plateau (both at about 60°S) from the Albian through to the Maastrichtian.

[13] Two main observations can be inferred from Figure 2: (1) $\delta^{18}O$ values increase from low to high paleolatitudes except for the curve given by *Huber et al.* [1995] who do not exclude "invisible diagenetic perturbations" lowering the pristine oxygen isotope ratios of foraminifera [see, e.g., *Schrag et al.*, 1992]; and (2) The four oxygen isotope curves present, very broadly, a similar evolution with minimal $\delta^{18}O$ values around the latest Albian-Turonian interval. The timing of the minimal $\delta^{18}O$ values, interpreted as a stage of maximal climatic warming, however, does not appear to be finely synchronous over all latitudes (Figure 2). This may be due to variability of the temporal resolution of the sampling. In particular, note that the curve from *Kolodny and Raab* [1988] lacks mid-Albian-Early Cenomanian samples, and that the curve from *Huber et al.* [1995] is interrupted by several discontinuities, especially around the Albian/Cenomanian boundary (for example, Early and mid-Cenomanian is missing at site 511). The Late Albian-Turonian interval may actually be composed of successive and rapid oxygen isotope oscillations giving two temperature maxima: a first one in the latest Albian, and the second one in the Turonian [*Clarke and Jenkyns*, 1999; *Stoll and Schrag*, 2000; *Wilson and Norris*, 2001]. In addition, *Huber*

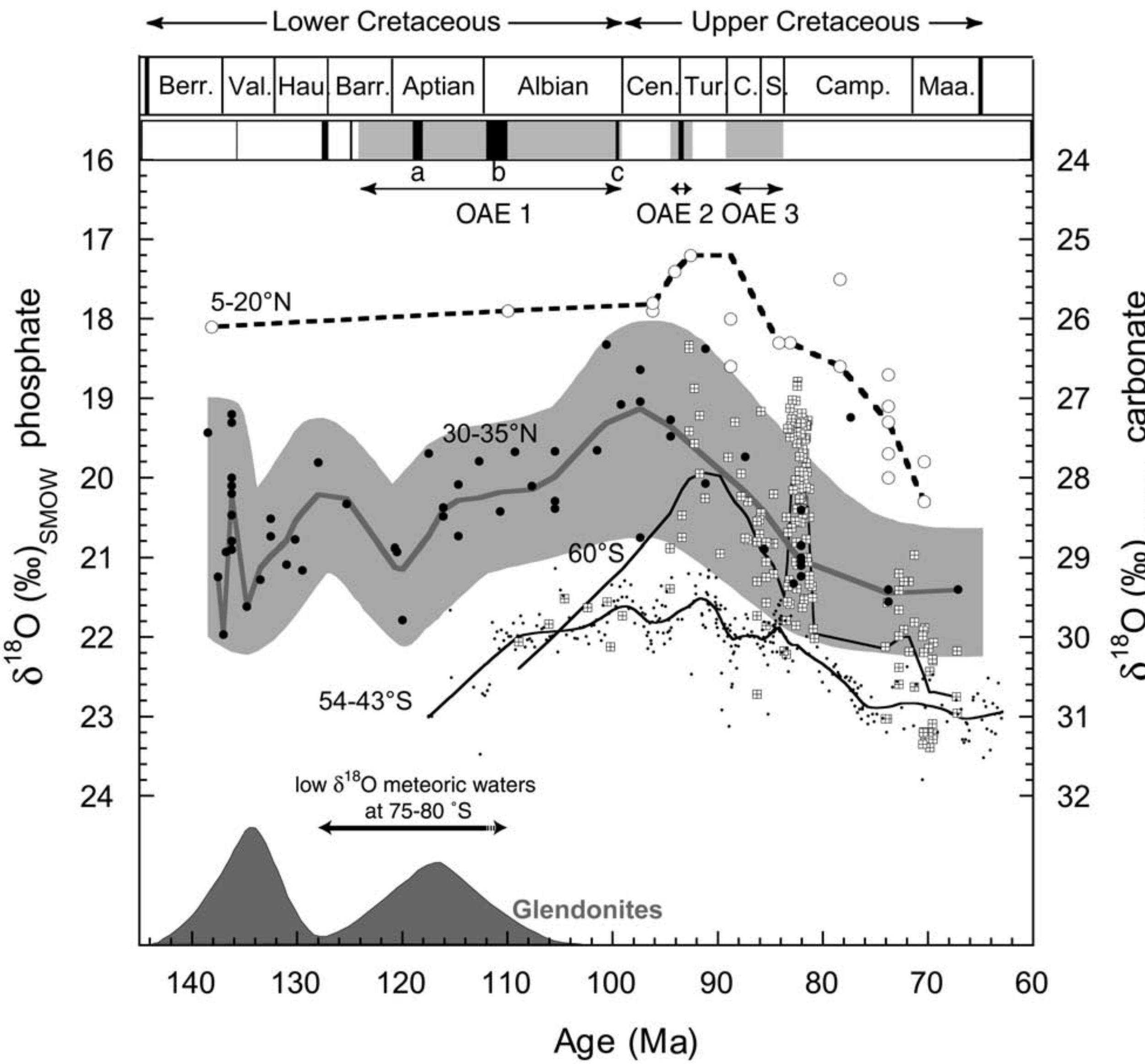

Figure 2. Comparison of $\delta^{18}O$ variations between fish remains (open circles: *Kolodny and Raab* [1988], bones, teeth, and vertebra; and solid circles: this work, teeth), planktonic foraminifera (crossed squares: *Huber et al.* [1995]), and fine fraction and bulk carbonates (dots: *Clarke and Jenkyns* [1999]). Absolute ages are derived from the timescale of *Gradstein et al.* [1994]. The curves for oxygen isotope data are based on the locally weighted least squared error method using a smoothing coefficient of 20%. Based on fractionation equations, two isotopic scales are given to facilitate the comparison of $\delta^{18}O$ values of both carbonates and phosphates [*Epstein et al.*, 1953; *Longinelli and Nuti*, 1973; *Kolodny et al.*, 1983]. Paleolatitudes are estimated from the maps proposed by *Scotese et al.* [1988]. Glendonite events, indicated with an arbitrary scale, are from *Price* [1999]. Oceanic anoxic events, OAE, are from *Breheret* [1994], *Crumière et al.* [1990], *Baudin et al.* [1999], and *Reboulet et al.* [2000] for the southeast of France (black rectangles) and from *Jenkyns* [1980] for worldwide OAE (gray rectangles). Note that the gray rectangles represent time intervals including the cumulative occurrence of OAEs around the globe. Low-$\delta^{18}O$ meteoric waters at 75–80°S within the lower Cretaceous are from *Gregory et al.* [1989]. See Figure 1 for other abbreviations.

et al. [2002] $\delta^{18}O$ record from Late Albian to Early Turonian, inferred from planktonic foraminifera from Blake Nose (about 30°N of paleolatitude in the mid-Cretaceous), shows that minimal $\delta^{18}O$ values are recorded in the Late Albian. This record does not include data from Late Turonian.

[14] The similarity of the form of the $\delta^{18}O$ curves (Figure 2) established from foraminifera, fish teeth and bulk carbo-

nates deposited at various paleolatitudes suggests that possible local $\delta^{18}O_{seawater}$ variations only contributed a minor part to the evolution of the $\delta^{18}O$ recorded in the different marine organisms with time. In addition, the carbonate-skeletal secreted materials (bulk carbonates and foraminifera) were deposited in pelagic or hemipelagic environments, whereas our fish teeth come from platform environments. Importantly, this indicates that surface seawater signals recorded in platform and more pelagic environments are equivalent. Therefore we propose that the oxygen isotope ratios of fish teeth from the Cretaceous Tethyan open platform provide reliable data for large-scale regional or global paleoclimatic reconstructions.

4.1. Long-Term Paleotemperature Trend

[15] Because of both poor temporal resolution and a certain variability in our $\delta^{18}O$ data, only long-term western Tethyan paleotemperature trends will be discussed, except for the earliest Cretaceous for which the chronostratigraphic resolution is tightly constrained. The global evolution of $\delta^{18}O$ values of fish teeth reveals that the Cretaceous underwent significant climatic variations on a 10 Myr scale when considering the whole stage, and down to the Myr scale for the earliest Cretaceous. Assuming a $\delta^{18}O_{seawater}$ of −1‰ for a continental ice-free Earth, marine temperatures within the subtropical zone (i.e., 30–35°N) range from 13–14°C in the Valanginian and Campanian-Maastrichtian interval to 28–29°C in the Cenomanian-Turonian period.

[16] The earliest lower Cretaceous was characterized by relatively short-term (1 Myr) temperature fluctuations of several degrees (Figure 1). A cooling event can be distinguished during the lowermost upper Valanginian (Ammonite Standard Verrucosum zone), as well as another possible one at the Berrasian/Valanginian boundary, both presenting minimum temperatures of 13–14°C. Previous studies [*Ditchfield*, 1997; *Podlaha et al.*, 1998; *van de Schootbrugge et al.*, 2000; *Price et al.*, 2000] have also suggested thermal oscillations with the occurrence of low temperatures for the earliest lower Cretaceous, although this was inferred from isotopic analyses of belemnites whose living environment is still debated [*Pirrie and Marshall*, 1990; *van de Schootbrugge et al.*, 2000].

[17] The significant climatic fluctuations detected during the Valanginian times are supported by paleontological data. The migrations of lower Cretaceous (pre-Aptian) ammonites between the Boreal and Tethyan realms were episodic and controlled by sea level changes according to *Rawson* [1993]. However, the simple opening of seaways, such as the Danish-Polish corridor and the Atlantic way, was a necessary but not sufficient condition. A climatic control has been suggested to explain the migrations of Boreal faunas observed in western Europe [*Kemper*, 1987; *Rawson*, 1994; *Reboulet and Atrops*, 1995]. During the Valanginian, the southward migrations of typical boreal ammonites into the Tethyan areas were limited to the southern margin of the European plate and mainly occurred during the upper Valanginian [*Thieuloy*, 1977; *Rawson*, 1994]. The southeast France basin, located at the palaeolatitude of 30°N, was probably the most southern area where boreal ammonites are recorded [*Cecca*, 1998]. The first immigration wave of typical boreal ammonites has been described at the lower-upper Valanginian boundary [*Rawson*, 1994]. They are relatively frequent in the Verrucosum zone (lowermost upper Valanginian) where several horizons of boreal genera such as *Prodichotomites* and *Dichotomites* have been distinguished [*Besse et al.*, 1986; *Thieuloy et al.*, 1990; *Bulot et al.*, 1992; *Reboulet and Atrops*, 1995]. Boreal ammonites are always rare in the Trinodosum zone (mid to upper Valanginian), and their migration ended during the uppermost Valanginian (Callidiscus zone;) [*Thieuloy*, 1977; *Bulot et al.*, 1992; *Reboulet*, 1996]. The migration pattern of the Boreal ammonite faunas supports the cooling event recorded during the Verrucosum zone (Figure 1).

[18] In addition, *Walter* [1989, 1991] observed a very important decrease in the species richness of the Valanginian bryozoan fauna in the ''calcaire à Alectryonia'' and ''Grande Lumachelle'' formations of the Jura and Provence platforms, respectively. Both formations were correlated [*Walter*, 1991] and dated to the Peregrinus horizon (top of the Verrucosum zone) [*Reboulet*, 1996]. *Walter* [1989] interpreted the crisis of the bryozoan fauna as the result of the coldest period of the Valanginian. The identification of a *Dichotomites* horizon both at the top of the ''Grande Lumachelle'' formation [*Thieuloy et al.*, 1990] and above the ''Faisceau Médian'' in the Vocontian basin [*Reboulet*, 1996] seems to confirm that the cooling event was probably stronger during the Peregrinus horizon, which belong to the Verrucocum zone (Figure 1). All these results suggest that global cooling events affected the subtropics and were not limited to higher latitudes, as previously proposed by *Frakes et al.* [1992].

[19] We emphasize that upper ocean temperatures close to 15°C may be considered to be surprisingly low for subtropical latitudes. For example, tropical fauna such as crocodilians require average temperatures of at least 20°C [e.g., *Adams et al.*, 1990; *Markwick*, 1998; *Voigt et al.*, 1999]. This apparent contradiction between low isotopic temperatures and the occurrence of tropical fauna may be partly reconciled if low-^{18}O continental ice was stored at high latitudes. The presence of icehouse interludes has been previously proposed for the earliest Cretaceous [*Stoll and Schrag*, 1996; *Price*, 1999]. *Price* [1999] took into account sedimentological, paleontological, paleobotanical, and geochemical data as well as results from General Circulation Model simulations that imply the presence of cold or subfreezing polar conditions during the Mesozoic. We have marked on Figure 2 the occurrence of glendonites at high paleolatitudes (simplified from *Price* [1999]), which are considered to have been formed under subfreezing aqueous depositional conditions [*Francis and Frakes*, 1993; *De Lurio and Frakes*, 1999]. Our temperature curve suggests substantial cooling events coinciding in time with the major occurrences of glendonites during the Valanginian and Aptian. Subglacial temperatures (mean annual continental temperatures of no more than 5°C, and probably below 0°C) at 75–80°S during the Aptian are also strongly supported by the evidence for the presence of meteoric waters depleted in ^{18}O ($\delta^{18}O \sim -20$‰) both during the formation of sedimentary carbonate concretions [*Gregory et al.*, 1989]

and in meteoric-hydrothermal systems around Cretaceous intrusions [*Blattner et al.*, 1997] in the south polar regions. The Cretaceous icehouse simulation of *Price et al.* [1998] predicts that polar ice may have covered approximately one third of the surface of the current permanent ice sheet. Moreover, during the Cretaceous Western Europe was located in a subtropical zone, which typically is associated with an evaporation/precipitation ratio above 1. The present-day dominance of evaporation in the tropics results in both higher salinity and a $\delta^{18}O_{seawater}$ increase of 0.5–1‰ compared to the mean ocean value [*Bigg and Rohling*, 2000]. Therefore, for the earliest Cretaceous, both glaciation and local excess evaporation may have been responsible for a $\delta^{18}O_{seawater}$ of at least 0‰ for surface waters.

[20] A global warming stage began in the Aptian and lasted throughout a period of about 20 Myr. It was particularly marked in the upper Albian-lower Cenomanian leading to temperatures up to 29°C during the Cenomanian and Turonian (Figure 1). These temperatures are similar to those obtained from planktonic foraminifera from Blake Nose (around 30°N of paleolatitude in the mid-Cretaceous), which yield temperatures from about 22.5°C to 30°C in the Late Albian-Early Turonian interval (temperatures recalculated from *Norris and Wilson* [1998], *Wilson and Norris* [2001], and *Huber et al.* [2002]; with the equation of *Erez and Luz* [1983]; and a $\delta^{18}O_{seawater}$ of −1‰ SMOW), although *Fassell and Bralower* [1999] report temperatures as low as of 20°C for the same area. All these temperatures must be considered as minimal estimates if we consider possible regional ^{18}O-enrichment of seawater above −1‰, as discussed above. The high Cenomanian-Turonian temperatures of 4–5°C above present-day values were not restricted to the subtropical zone. They also affected polar regions, as suggested by paleontological and paleobotanical evidence [*Tarduno et al.*, 1998; *Herman and Spicer*, 1996]. Limited ice caps during episodic cooling might however have been present even during this mid-Cretaceous climatic optimum, as proposed by *Stoll and Schrag* [2000] whose high-resolution $\delta^{18}O$ record shows positive excursions well correlated with sea level falls, during the lower Cenomanian and upper Turonian-Coniacian. In addition, *Ramstein et al.* [1997] using mid-Cretaceous AGCM (Atmospheric Global Circulation Model) simulations showed that the increase of temperature resulting from three times the modern amount of CO_2 in the atmosphere is too weak to prevent ice sheet formation on the high-latitude continents. Such coolings and ice sheet buildups must however have been limited in time (lasting about several hundred thousand years, as estimated by *Stoll and Schrag* [2000]) in a period of global warmth; they cannot be characterized with our time resolution.

[21] Following the Turonian, a progressive cooling over the next 25 Myr is recorded in the $^{18}O/^{16}O$ ratios of the fish teeth until the upper Campanian and Maastrichtian, and they yield temperatures similar to those of earliest Cretaceous seawaters. The upper Campanian and Maastrichtian stages in the western Tethyan domain are characterized by rather low temperatures, about 5°C lower than average modern values in subtropical zones. Rapid climatic variations have also been suggested for the Maastrichtian [*Saito and Van Donk*, 1974; *Li and Keller*, 1998], but there are too few data for them to be detectable in our study. Such low temperatures in the subtropical zone as early as the Campanian are somewhat puzzling, considering the tropical fauna assemblages such as rudist bivalves that prevailed until the Middle Maastrichtian [*Johnson et al.*, 1996; *Voigt et al.*, 1999]. This apparent disagreement between oxygen-isotope data and the paleontological record may be resolved if the oxygen isotope composition of the ocean was higher than −1‰. As previously stated, two main mechanisms may be involved: ^{16}O storage in high-latitude ice or evaporation dominating over precipitation at low latitudes. In contrast to the Early Cretaceous, there is no evidence for glaciation in the Late Cretaceous geological record (such as glendonites or ice-rafted debris). *Spicer and Parrish* [1990], based on paleobotanical data from the North Slope of Alaska, point to a climatic deterioration from the Cenomanian to the Campanian-Maastrichtian. It is thus conceivable that there was a limited increase in ^{16}O-storage in high-latitude ice from the mid to Late Cretaceous, contributing in part to the increase of the $\delta^{18}O$ measured in the organisms (Figures 1 and 2). Despite the evidence for this slight climatic deterioration recorded in the upper Cretaceous of northern Alaska, high-latitude temperatures remained considerably warmer than today, based on both paleobotanical studies [*Parrish and Spicer*, 1988; *Spicer and Parrish*, 1990] and geochemical analyses of high paleolatitude planktonic foraminifera [*Stott and Kennett*, 1990; *Barrera et al.*, 1997] and macrofossils [*Pirrie and Marshall*, 1990]. In addition, these Maastrichtian high-latitude temperatures (above 60°) do not differ very strongly from those recorded in high-latitude Turonian sediments, compared to the more pronounced decrease observed over the same interval for low and midlatitude temperatures (Figures 1 and 2). It has been shown, however, that high latitudes experienced a consequent cooling in the upper Maastrichtian [*Ditchfield et al.*, 1994; *Frank and Arthur*, 1999]. These observations suggest that the "global cooling trend" may be primarily restricted to low and midlatitudes, at least until the upper Maastrichtian.

[22] Despite the possible presence of high-latitude mountain glaciers and winter snow, or even of limited ice caps, it appears unlikely that the occurrence of polar ice could be responsible for a $\delta^{18}O_{seawater}$ significantly higher than −1‰ during the Late Cretaceous. Therefore it is proposed that a $\delta^{18}O_{seawater}$ higher than −1‰ is mainly the result of the dominance of evaporation over precipitation at low latitudes. Thus either the temperatures have been underestimated for the whole of the Late Cretaceous, or the climatic conditions have changed, from subtropical humid in the mid-Cretaceous to subtropical dry in the latest Cretaceous. Intuitively, it could be expected for evaporation in middle and low latitude to decrease during cooler time periods. However, variations in evaporation/precipitation ratio involve beside temperature other parameters like wind force, intensity of moisture transport, and the position of the Hadley circulation cells. Particularly, changes in global temperature affect the relative sizes of the atmospheric circulation cells (Polar, Ferrel and Hadley), causing them to shift in latitudinal position. This shift results in latitudinal changes in humidity and runoff [*Lockwood*, 1979]. For example, following the scheme of *Matthews and Perlmutter*

[1994], the humid belt during a climatic minimum is reduced to 10° around the equator, whereas the tropical and subtropical belts (from 10 to 30°) encounter dry to arid conditions. Even if this scenario is proposed in the case of a climatic minimum like a Quaternary ice age, it reveals that a climatic cooling may induce drier conditions in the tropics. In addition, the Maastrichtian is thought to have been a time of major reorganization in oceanic circulation patterns, with the opening of the North Atlantic and the possible initiation of a thermohaline circulation similar to today's [*Barrera et al.*, 1997; *Frank and Arthur*, 1999]. Such a change in oceanic circulation patterns would be associated with major reorganization in atmospheric circulation.

[23] Thus, after the relatively rapid climatic fluctuations and the occurrence of glendonites during the Early Cretaceous, the period is characterized by an overall warming to a thermal optimum in the latest Aptian-Turonian interval that is then followed by a cooling and/or a transition to more arid conditions until the Maastrichtian. No glendonites have been recorded from the Late Cretaceous. These data are consistent with the Oceanic Anoxic Events (OAE) distribution throughout the Cretaceous (Figure 2). The factors controlling the OAE distribution are believed to be an increased oceanic productivity in concert with a relative sea level rise and global warming. High temperatures lower the solubility of oxygen throughout the water column, and an ice-free Earth would be associated with a sluggish bottom water circulation with poor-oxygenated bottom waters, favoring anoxia [*Jenkyns*, 1980]. The OAE occurrences through the Cretaceous appear indeed in good correspondence with the overall warming through the Early until the mid-Cretaceous, as shown in Figure 2. Particularly, there is a fine correlation of the organic-rich shales in southeast France with thermal optima recorded by the fish teeth from France: "Barrande" level [*Reboulet et al.*, 2000] in the uppermost lower Valanginian; Faraoni level [*Baudin et al.*, 1999] in the uppermost Hauterivian; OAE1a-b [*Breheret*, 1994] in the Aptian-Albian; OAE1c and Thomel level (Cenomanian/Turonian boundary) [*Crumière et al.*, 1990] in the Cenomanian-Turonian. Also, it should be noted that glendonites and OAEs do not occur at the same time, except in the lower Aptian, which marks the beginning of the OAEs. Neither glendonite nor OAE have been recorded in the latest Cretaceous (Campanian and Maastrichtian).

4.2. Latitudinal Temperature Gradient

[24] In order to investigate the evolution of the latitudinal temperature gradient of surface waters, data from the literature have been combined with our data set, assuming uniform $\delta^{18}O_{seawater}$ of -1‰ SMOW at all sites (Figure 3). Temperature gradients have been thus established for the Albian-Cenomanian (during the progressive warming), the Turonian (around the thermal maximum), and for the Maastrichtian (after the subsequent cooling), and they are compared to the present-day gradient. These curves are mainly based on isotopic temperatures inferred from near-surface living organisms, but also include data from paleobotanical studies (mean annual temperatures, mainly inferred from leaf physiognomy), especially for high latitudes. Because of the lack of data with latitude for the lower Cretaceous, latitudinal temperature gradients could not be determined for the Berriasian-Valanginian and lower Aptian, the periods with probable significant volumes of continental ice.

[25] The latitudinal temperature gradients are substantially weaker than the modern one. They are about 0.2°C/°latitude between 20° and 80° for the Albian-Cenomanian and Maastrichtian intervals, and slightly stronger (about 0.3°C/°latitude) for the Turonian, whereas the present-day gradient is about 0.4°C/°latitude. These Cretaceous temperature gradients are probably underestimated since variations in $\delta^{18}O_{seawater}$ with latitude are not taken into account. If the modern latitudinal $\delta^{18}O_{seawater}$ distribution [*Bigg and Rohling*, 2000] is comparable to the Cretaceous one, as presumed by *D'Hont and Arthur* [1996] for the sea surface salinity gradient, then the low-latitude temperatures are underestimated by 2 to 4°C, and high-latitude temperatures are overestimated by 1 to 1.5°C. Consequently, the latitudinal temperature gradients should be corrected by about 0.05 to 0.09°C per degree of latitude. Overall these gradients are similar to those previously proposed [*Barron*, 1983; *Sellwood et al.*, 1994; *D'Hondt and Arthur*, 1996; *Li and Keller*, 1999; *Frakes*, 1999; *Huber et al.*, 2002].

[26] For the Albian-Cenomanian and Turonian intervals, the low-latitude paleotemperatures are comparable to modern temperatures. In contrast, high-latitude temperatures are 5°C to 10°C higher, suggesting the absence of permanent ice sheets. Compared to the present day, the latitudinal temperature gradient for the Maastrichtian is surprisingly weak and characterized by warm high-latitude and cool low-latitude temperatures assuming a $\delta^{18}O_{seawater}$ of -1‰. Global Circulation Models for the Late Cretaceous could be very useful to identify the required conditions for satisfying such a low thermal gradient. The mid-Cretaceous period, considered as very warm and equable, has been interpreted by *Barron et al.* [1995] as resulting from a combination of the Cretaceous paleogeography, an increased partial pressure of atmospheric CO_2, and an increased oceanic heat transport.

5. Conclusion

[27] Oxygen isotope compositions of fish tooth enamel from platform environments provide a useful and viable proxy for the quantification of upper ocean temperature variations that are of global climatic significance. For the subtropical Cretaceous western Tethyan seas, a number of short-term (million years) oscillations are observed within the 3‰ variation of the bioapatites. There was a progressive warming throughout the lower Cretaceous until the thermal maximum of the latest Albian-Turonian interval, followed by a cooling prevailing in the Campanian and Maastrichtian. Major cold or significant cool periods are observed at the Berriasian-Valanginian boundary, during the lowermost upper Valanginian, the earliest Aptian, and during the Maastrichtian.

[28] Minimum isotopic temperatures within the marine subtropical zone (30–35°N paleolatitude) range from 13–14°C during the Valanginian and Late Campanian–Maastrichtian and up to 28–29°C during the Cenomanian-

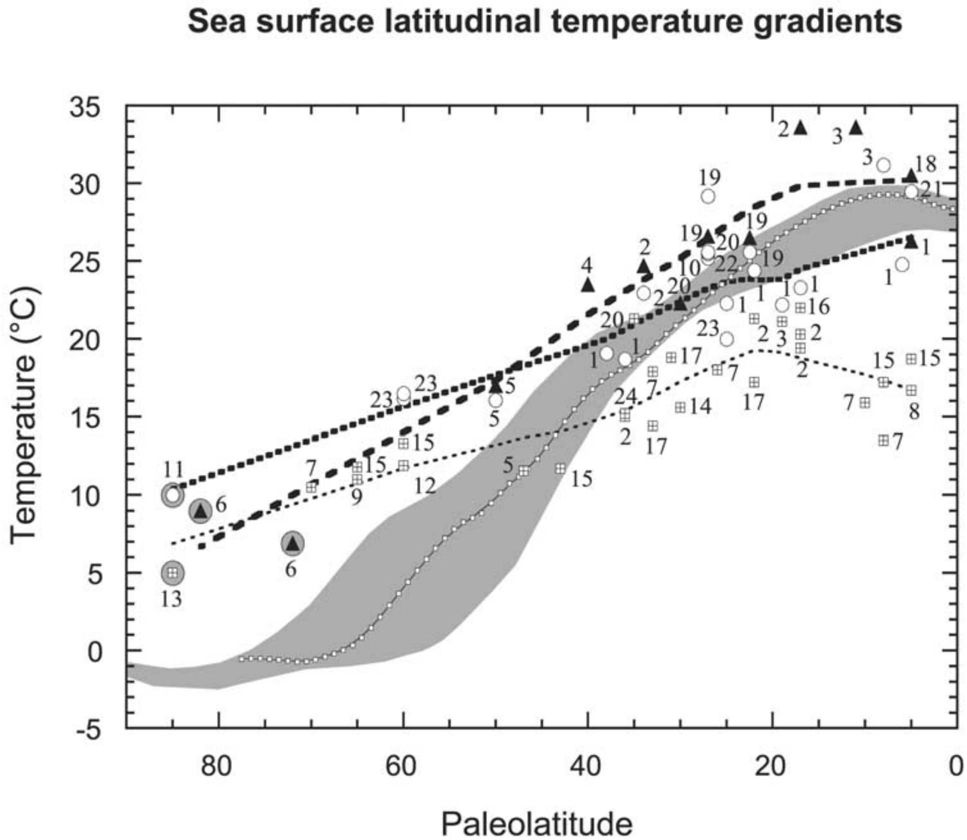

Figure 3. Sea surface latitudinal temperature gradients. Open squares: present day; open circles: Albian-Cenomanian; solid triangles: Turonian, and crossed squares: Maastrichtian. The present-day annual sea surface temperature gradient has been established using the data library LEVITUS94 for the south Pacific Ocean. The gray envelope marks the extension of the modern gradient, if including North Pacific, South Atlantic, and North Atlantic surface temperatures. Sources for the Cretaceous gradients are: 1. *Sellwood et al.* [1994], 2.This work, 3. *Kolodny and Raab* [1988], 4. *Jenkyns et al.* [1994], 5. *Clarke and Jenkyns* [1999], 6. *Herman and Spicer* [1996], 7. *D'Hondt and Arthur* [1996], 8. *Li and Keller* [1999], 9. *Stott and Kennett* [1990], 10. *Norris and Wilson* [1998], 11. *Spicer and Parrish* [1986], 12. *Pirrie and Marshall* [1990], 13. *Parrish and Spicer* [1988], 14. *Saito and Van Donk* [1974], 15. *Barrera et al.* [1997], 16. *Lécuyer et al.* [1993], 17. *Frank and Arthur* [1999]. 18. *Wilson et al.* [2002], 19. *Stoll and Schrag* [2000], 20. *Huber et al.* [2002], 21. *Norris et al.* [2002], 22. *Wilson and Norris* [2001], 23. *Fassell and Bralower* [1999], 24. *Li and Keller* [1998]. The temperatures have been averaged when several data were available for the different time slices. Isotopic temperatures for planktonic foraminifera are calculated from the published $\delta^{18}O$ values by using the equation of *Erez and Luz* [1983]. For bulk carbonates and fish remains, temperatures were calculated using equations given by *Anderson and Arthur* [1983], and *Kolodny et al.* [1983], respectively, using a $\delta^{18}O_{seawater}$ of -1‰ SMOW. For sources 6, 11, and 13, temperature estimates (mean annual temperature) are provided by paleobotanical studies, mainly from leaf physiognomy, and are distinguished from the rest of the data by a gray shaped circle.

Turonian. However, surface seawater $\delta^{18}O$ values may have been variable and not always around -1‰, as consequence of significant continental ice sheets during parts of the Berriasian and Valanginian, and the effects of high evaporation to precipitation ratios for some tropical waters, in relation to a dry climate (e.g. Maastrichtian). Considering the variation in fish teeth $\delta^{18}O$ values of 2‰ as resulting from thermal effects alone, a temperature difference of 10°C between the climatic extremes (Valanginian versus Cenomanian-Turonian) at subtropical latitudes is deduced. Latitudinal thermal gradients for the mid and late Cretaceous were generally weaker (0.2–0.3°C/° latitude) than today (0.4°C/° latitude). With recorded high-latitude temperatures of 5–10°C above modern ones, the presence of polar ice caps during the upper Cretaceous is improbable.

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