Interhemispheric appraisal of the value of alkenone indices as temperature and salinity proxies in high-latitude locations

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Abstract. Alkenone sediment data from the Nordic seas and North Atlantic are compared to those from *Sikes et al.* [1997] for the Southern Ocean to evaluate further U^K_{37} and $U^K_{37}{'}$ as proxies to estimate cold temperatures ($<10°C$) and the effect of salinity and temperature in the relative abundance of 37:4 to the total abundance of C_{37} alkenones (37:4%). U^K_{37} and $U^K_{37}{'}$ are found to be equally viable as proxies, but there are significant regional differences in their cold temperature dependence. The measurement of 37:4% in cores from the North Atlantic region can be used to identify situations when $U^K_{37}{'}$ is not a reliable paleothermometer. Variations in salinity are probably responsible for changes in the sedimentary record of 37:4%, and a preliminary calibration has been obtained for 37:4%=f(salinity). This new relationship should be further confirmed through field or laboratory experiments, but it paves the way to derive a molecular proxy to reconstruct paleosalinity in surface waters.

1. Introduction

The U^K_{37} and $U^K_{37}{'}$ indexes are used as proxies to reconstruct absolute sea surface temperature (SST) variations in the photic zone [*Brassell et al.*, 1986; *Prahl and Wakeham*, 1987]. These indices are derived from the relative abundance of methyl alkenones with 37 carbon atoms and 2, 3, or 4 double bonds (e.g., 37:4 stands for C_{37} alkenone with 4 double bonds). Alkenones are biosynthesized by some algae of the class Haptophyceae/Prymnesiophyceae, such as *Emiliania huxleyi* which is the most abundant and widespread coccolithophorid in the oceans and is most probably the main producer of alkenones found in recent sediments [*Volkman et al.*, 1980a, b]. In comparison to other sedimentary organic components (i.e., biomarkers), alkenones are more resilient to degradation, abundant in sediments, and widespread in all oceans. The U^K_{37} index was initially defined as [*Brassell et al.*, 1986]:

$$U^K_{37} = \frac{37:2 - 37:4}{37:2 + 37:3 + 37:4}$$

U^K_{37} stands for unsaturated ketones with 37 carbon atoms. To the author's knowledge, most published alkenone stratigraphic data derives from sediment cores from low or "warm" latitudes and upwelling areas, which are mostly devoid of 37:4. Thus, except in a few cases [*Madureira et al.*, 1997; *Rosell-Melé et al.*, 1997], data are generally shown in the literature using the simplified index $U^K_{37}{'}$ without the tetraunsaturated component [*Brassell et al.*, 1986; *Prahl and Wakeham*, 1987]:

$$U^K_{37-} = \frac{37:2}{37:2 + 37:3}$$

In contrast to paleotemperature proxies based on abundance or isotopic composition of microfossil taxa, U^K_{37} and $U^K_{37}{'}$ have not been found to be affected by carbonate dissolution or salinity [*Sikes* *et al.*, 1991; *Rosell-Melé et al.*, 1994; *Sonzogni et al.*, 1997]. U^K_{37} and $U^K_{37}{'}$ are potentially the only option to reconstruct paleo-SST in locations lacking carbonate and silica tests or when not enough species diversity is available to calculate meaningful temperature transfer functions. Since the inception of U^K_{37} and $U^K_{37}{'}$ the application of this alkenone-based proxy in paleoceanography has been steadily increasing, and it is gradually becoming established in this field of research. However, U^K_{37} and $U^K_{37}{'}$ are not devoid of uncertainties, which may constrain their reliability as an accurate paleothermometer or even as SST proxies in certain locations, for example, fjords [*Conte et al.*, 1994; *Ficken and Farrimond*, 1995].

At the moment the $U^K_{37}{'}$ growth temperature calibration equation derived from a culture of a NE Pacific strain of *E. huxleyi* is generally applied to convert downhole variations of $U^K_{37}{'}$ to SST values [*Prahl and Wakeham*, 1987; *Prahl et al.*, 1988]. However, *E. huxleyi* is of late appearance in the fossil record, dated between ~285 and 230 kyr ago [*Hills and Thierstein*, 1989; *Ahagon et al.*, 1993], although the stratigraphic occurrence of alkenones has been documented in continuous stratigraphic sequences from present to the Miocene [*Haug*, 1996], and in isolated shales from the Miocene, Oligocene, Eocene, and the Cretaceous [*de Leeuw et al.*, 1980; *Marlowe et al.*, 1984; *Farrimond et al.*, 1986]. Remarkably, the *E. huxleyi* equation provides "reasonable" estimates even in sediments preceding this algae existence [e.g., *Müller et al.*, 1997], and it is comparable to a global calibration derived from $U^K_{37}{'}$ values in 370 core tops from all oceans [*Müller et al.*, 1998]. This is quite remarkable because work following that of *Prahl et al.* [1988] has revealed a lot of variability in the calibration equations of $U^K_{37}{'}$ using algal cultures [*Conte et al.*, 1995, 1998; *Volkman et al.*, 1995; *Sawada et al.*, 1996] or water column particulate matter [*Conte et al.*, 1992, 1993, 1994]. There is also some ambiguity on the temperature range over which U^K_{37} and $U^K_{37}{'}$ are valid and the linearity of the proxies against temperature [*Conte et al.*, 1992; *Conte and Eglinton*, 1993; *Conte et al.*, 1998; *Sikes et al.*, 1993; *Rosell-Melé et al.*, 1995a; *Sonzogni et al.*, 1997]. However, recently, *Müller et al.* [1998] have obtained a global calibration of $U^K_{37}{'}$ in the temperature range of $0°-29°C$ and across five biogeographic zones, indicating that the $U^K_{37}{'}=f$(SST) relationship is robust and linear despite the regional differences observed by other authors.

The purpose of this paper is to provide new insights into the values of U^K_{37} and U^K_{37}', to estimate SST at low temperatures (<10°C), and to examine the information contained in 37:4 for paleoceanographic reconstruction. Alkenone sediment data from the Nordic Seas are compared to that from *Sikes et al.* [1997] for the Southern Ocean to appraise the global significance of regional data from the northern and southern hemispheres. Sea surface temperatures used for the comparison correspond to caloric summer in both hemispheres.

2. Sampling and Analytical Methods

The surface sediments from the North Atlantic samples used in this study were mainly obtained by successive cruises of the German vessel *Meteor* between 1987 and 1989. Other samples were collected during the British Biogeochemical Ocean Flux Study (BOFS; 1989 and 1990) and the French Paleocinat I (1990 and 1992) programs. The sediments were retrieved from the seafloor using box corers or multicorers. A subsample from the top 1 cm was taken onboard the ship or in the laboratory and stored frozen or in a cool room until analysis. Alkenone data from these samples have been partially discussed previously [*Rosell-Melé et al.*, 1994, 1995a].

Cores HM79-6 (62°.58'N, 02°42'E; 850 m depth) and HM79-4 (63°06'N, 02°33'E; 850-983 m depth) were retrieved from the high sedimentation rate area off southeastern Norway, and data were correlated and combined to produce a composite record (HM79-6/4) as shown by *Koç-Karpuz and Jansen* [1992]. The correlation was based on the comparison of data from sedimentological, geochemical, isotopical and diatom floral analyses, and the chronology of the cores was established by using two ash layers and ^{14}C ages [*Koç-Karpuz and Jansen*, 1992]. Core 19K (41°5'N, 20°47'W; 3742 m) was retrieved during the cruise of the British vessel *Charles Darwin* [*McCave*, 1989], as part of the BOFS program. The age model of the core has been established by comparing its planktonic foraminifera_$\delta^{18}O$ record (N.J. Shackleton, unpublished data, 1993) with the SPECMAP curve [*Martinson et al.* 1987]. Core M23415 (~55°N, 19°W; 2500 m) was retrieved by the German vessel *Meteor*. The age model was established with 11 control points from ^{14}C accelerator mass spectrometry (AMS) measurements of *Globoratalia bulloides* and *Neoquadrina pachyderma* (s) and converted to calendar ages using the approach of *Bard et al.* [1993]. All the cores were stored in a cool room after retrieval and were subsampled at 5 cm intervals for alkenone analyses.

The procedures employed to determine alkenone indices have been described in detail elsewhere [*Rosell-Melé*, 1994; *Rosell-Melé et al.*, 1995b]. Briefly, sediment samples were freeze-dried, weighed (1-2 g), solvent-extracted (dichloromethane/methanol, 3:1 x 3), and the total extracts cleaned up using solid phase extraction on silica. After derivatization with bis-(trimethyl-silyl)-trifluoroacetamide (BSTFA) the C_{37} alkenones were quantified by gas chromatography ammonia chemical ionization mass spectrometry (GC-CIMS) [*Rosell-Melé et al.*, 1995b].

3. Results and Discussion

3.1. Significance of 37:4 to Reconstruct Paleotemperatures

In northeastern Atlantic surface sediments, 37:4 represents <10% of the combined amount of all the C_{37} components, and it is only apparent or easily detectable by conventional analytical methods in sediments deposited below waters colder than ≈ 10°C [*Rosell-Melé et al.*, 1994]. In fact, in such sediments the difference between U^K_{37} and U^K_{37}' is only potentially climatically significant (difference between estimates >1°C) when U^K_{37} < 0.4, which approximately equals a SST of 10°C [*Rosell-Melé et al.*, 1994]. However, in sediments from the western side of the Nordic seas the relative abundance of 37:4 of the total of C_{37} alkenones (37:4%) is much larger, close to 20% (Figure 1 and data in Table 1). In the data shown in Figures 1 and 2, from Nordic seas surface sediments, two groups of points can be distinguished. When 37:4% in sediments is > 5%, there is no correlation of such percentage with temperature. In contrast, below 5%, there is a linear correlation with overlying sea surface temperatures ([37:4%] = 6.46 - 0.45T (°C), r = 0.64, and n = 25). Such temperature dependence of 37:4% in sediments is not unexpected as this component was identified as temperature sensitive since the first studies on alkenones were carried out [*Marlowe*, 1984; *Brassell et al.*, 1986]. Furthermore, *Conte et al.* [1995] showed that when the 37:4 component is relatively abundant in algal strains, their biochemical response to temperature involves tetraunsaturated, triunsaturated and diunsaturated alkenones. In Figure 2, 37:4% = 5 can be considered a cutoff point or threshold which marks the lack of correlation between U^K_{37} and U^K_{37}' with SST for the same set of samples (Figures 3 and 4) [*Rosell-Melé et al.*, 1994, 1995a]. Hence, when 37:4% > 5, the temperature dependence of U^K_{37} and U^K_{37}' fail, at least in Nordic seas sediments, and the value of these proxies as paleothermometers to estimate cold temperatures is in doubt in this region. In coastal environments, where high 37:4% occur, for example, fjords, the temperature dependence of U^K_{37} and U^K_{37}' is also arguable [*Conte et al.*, 1994]. In principle, these are regional observations, and they do not challenge the global temperature range throughout which U^K_{37}' is known to be linear [e.g., *Müller et al.*, 1998].

Higher percentages of 37:4% downhole a core could then be used to identify situations when U^K_{37} and U^K_{37}' are not providing reliable SST estimates. In the North Atlantic and the Nordic seas, high occurrence of 37:4 seas could have been quite common. SST in the northeastern Atlantic during the last glacial maximum (LGM) were probably lower than today, below 10°C, from 50° northward [*Climate: Long-Range Investigation, Mapping and Prediction (CLIMAP) Project Members*, 1981; *Sarnthein et al.*, 1995]. Hence, in high-latitude locations, U^K_{37} values from glacial sediments could be expected to be < 0.4, and the 37:4 component could be expected to be abundant in such sediments, by analogy to distributions from modern surface sediments in the Nordic seas. As an example, shown in Figure 5 are 37:4% data and SST estimates from U^K_{37}' and diatom transfer function (DTF) from the analysis of composite cores HM79-6/4, located off southeastern Norway [*Koç-Karpuz et al.*, 1992]. The drop in temperature clearly marked by DTF estimates at the bottom of the record is not matched by an equivalent drop in the U^K_{37}' values. Part of the lowest SST section in the DTF curve coincides with the Younger Dryas episode (YD; ~10,000-11,000 ^{14}C yrs or 11,500-12,700 calendar years), which is widely acknowledged to be a cold climatic spell both in land and the oceans [cf. *Crowley and North*, 1991]. However, U^K_{37}' does not reflect such a cold episode and YD SSTs are even warmer than Holocene values. It is difficult to justify such a difference between both sets of estimates from a climatic or even ecological standpoint. Alkenone concentrations during the YD core section are quite low, but an analytical bias (i.e., irreversible adsorption) [*Rosell-Melé*, 1994; *Rosell-Melé et al.*, 1995b] would not shift values several
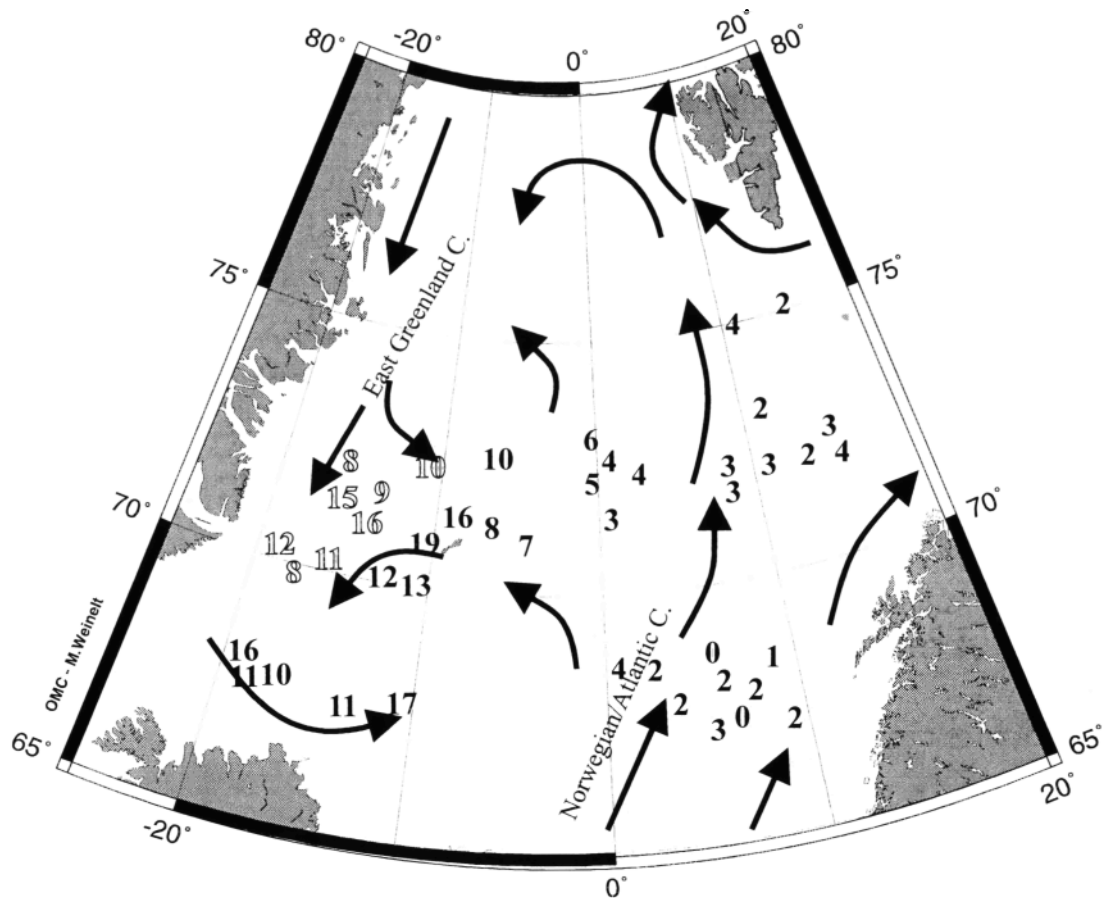
Figure 1. Map of the Nordic seas showing the location of the core tops whose data are discussed in the text and their values of 37:4% (relative abundance of 37:4 alkenone to the total abundance of C$_{37}$ alkenones, in percentage). Solid numbers indicate those samples that have been used to appraise the salinity dependence of 37:4%. The other samples (marked with open numbers) are referred to in the text as the "East Greenland Current" data set.

degrees, and such a bias was not noticed during the analyses. Remarkably, 37:4% values peak during the YD, at values higher than 5%, and descend to values between 0% and 1% for the Holocene (Figure 5). Hence, using the plot in Figure 2 as a modern analog, the high values of 37:4% in HM79-6/4 indicate that the U$^K_{37}$' estimates in the YD core section are not reliable. In Figure 5, the fact that 37:4% increases precisely during the YD episode suggests that 37:4% is capable of providing paleocenographic information. From the data in Figure 2 it should be expected that sediments deposited during glacial conditions would contain higher 37:4% percentages, as the abundance of 37:4 increases under cold water conditions. Hence the high 37:4% values in Figure 5 during the YD may indicate a cooling of the core location during such an episode, in agreement with the DTF data, but in contradiction to the U$^K_{37}$' values.

To investigate further the stratigraphic significance of 37:4%, this index was quantified downhole core 19K from the northeastern Atlantic (Figure 6). Throughout the core, 37:4% is not higher than 5%; thus the equation derived from Figure 2 can be used in principle to reconstruct paleo-SST. The SST estimates from 37:4% parallel the changes in U$^K_{37}$ and U$^K_{37}$' in the same core (Figure 6). Hence stratigraphic 37:4% variations provide a record of changes in paleo-SST. However, the magnitude of the oscillations and the

absolute value of the estimates are different. It should be noted that the calibration in Figure 2 for 37:4% spans a narrower range of temperatures than those covered by the U$^K_{37}$ and U$^K_{37}$' calibrations used to convert SST values in Figure 6. In fact, using the same sample set as in Figure 2, the scatter of U$^K_{37}$ and U$^K_{37}$' versus temperature is considerable between 5° and 11°C (Figures 3 and 4), and the coefficient of correlation (r) is zero for U$^K_{37}$' and 0.48 for U$^K_{37}$. Hence calibration of 37:4% against temperature would benefit from an enlarged data set to overcome the scatter obtained in field calibrations over narrow temperature ranges. Thus the difference between the value of the SST estimates for 37:4% versus U$^K_{37}$ and U$^K_{37}$' could be argued to be due to the calibration obtained for 37:4%, rather than a problem with 37:4% as a proxy for SST. Using data from *Prahl et al.* [1988], an approximate culture calibration of 37:4% can be obtained ([37:4%] ≈ 23.2 - 1.5*T*(°C), r = 0.98, and n = 3, only the data points where U$^K_{37}$'≠ U$^K_{37}$ have been considered), which if applied to core 19K data yields a very good agreement between 37:4% and U$^K_{37}$ and U$^K_{37}$'estimates (Figure 6). However, 37:4% values for the culture data of *Prahl et al.* [1988] are much higher than those in the Nordic seas for equivalent temperatures (e.g., at 8°C, 37:4% ≈ 12; Figure 7), which correspond to the group of points in Figure 2 where 37:4% ≠ *f*(SST). Such differences in both values of 37:4% and correlation coefficients pose an apparent

Table 1. Data for the Samples From the Nordic Seas

Core Meteor	Latitude, °N	Longitude, °E	37:4,[a] P.A.	37:3,[a] P.A.	37:2,[a] P.A.	37:4%	Salinity,[b] psu	Temperature,[b] °C
23304-1	67.8	6.0	-	96,889	100,635	0.0	34.82	10.1
23300-2	67.0	5.8	-	180,304	132,727	0.0	34.72	10.2
23321-1	67.8	6.0	10,368	397,288	370,281	1.3	34.82	10.1
23261-1	72.2	13.1	18,233	366,226	797,363	1.5	34.86	8.1
23331-2	66.9	7.8	37,331	994,916	691,554	2.2	34.44	10.8
23312-2	66.9	7.7	18,028	427,988	368,538	2.2	34.46	10.8
23024-2	67.7	5.8	19,556	539,129	382,329	2.1	34.83	10.2
23303-1	67.7	5.8	25,350	681,990	500,855	2.1	34.83	10 2
23001-1	67.7	3.7	46,697	1,534,016	1,224,315	1.7	35	9.9
23321-4	67.8	6.0	26,90	58,230	47,867	2.5	34.82	10.1
23003-1	67.9	2.9	41,846	1,085,507	875,627	2.1	35.06	9.7
23266-1	72.0	7.5	50,970	1,070,651	848,927	2.6	35.01	7.8
23260-1	72.1	11.5	49,730	1,021,947	829,677	2.6	34.91	7.3
23254-1	73.1	9.7	7,530	161,712	156,273	2.3	34.92	7.3
23258-3	75.0	14.0	25,785	540,530	469,161	2.5	34.62	5.7
23262-2	72.2	14.4	24,383	424,891	411,404	2.8	34.82	8.2
23301-2	67.0	7.8	3,829	59,853	66,960	2.9	34.46	10.8
23259-3	72.0	9.3	32,337	586,980	470,797	3.0	34.97	8.0
23269-2	71.4	0.7	6,890	114,816	101,199	3.1	34.95	7.2
23298-2	68.3	1.5	379,205	6,483,539	3,388,118	3.7	35.12	9.3
23262-3	72.2	14.4	90,077	1,246,979	924,267	4.0	34.82	8.2
23257-3	74.9	11.1	32,477	384,756	338,861	4.3	34.69	6.0
23289-2	72.4	1.8	166,166	2,145,809	1,465,233	4.4	34.84	6.7
23291-1	72.4	1.5	38,599	472,830	349,633	4.5	34.83	5.7
23279-1	72.1	-0.2	22,784	261,251	205,353	4.7	34.76	6.5
23270-2	73.2	-0.8	8,671	67,732	59,059	6.4	34.43	5.3
23477-1	71.0	-5.6	50,765	400,547	254,951	7.2	34.48	5.5
23351-4	70.4	-18.4	347,268	2,400,852	1,866,139	7.5	32.06	2.3
23343-4	72.2	-13.0	169,774	1,193,371	754,545	8.0	32.43	2.1
23295-4	71.2	-5.9	4,991	29,661	27,198	8.1	34.38	5.4
23345-2	71.7	-14.3	13,782	71,690	66,814	9.1	32.37	2.1
23483-2	67.9	-18.6	21,030	125,560	73,943	9.5	33.51	5.0
23293-2	72.6	-6.6	9,032	38,245	41,517	10.2	33.75	3.9
23294-3	72.4	-10.6	24,199	109,406	101,150	10.3	32.96	2.7
23347-4	70.4	-16.1	18,617	72,767	84,744	10.6	32.7	2.7
23487-1	67.3	-14.2	37,632	199,626	108,688	10.9	34.29	6.5
23486-3	67.9	-18.1	323,200	1,690,822	856,642	11.3	33.59	5.2
23348-2	70.4	-18.9	82,353	345,778	267,122	11.8	31.92	2.2
23353-2	70.0	-12.4	405,991	1,919,813	1,069,982	12.0	33.76	4.1
23480-2	69.4	-10.8	23,531	111,224	53,335	12.5	34.26	5.0
23344-4	71.7	-15.6	47,602	168,409	96,386	15.2	31.98	1.6
23482-2	67.9	-18.8	20,110	72,007	35,052	15.8	33.48	5.0
23346-3	71.3	-14.1	283,951	925,350	584,845	15.8	32.67	2.5
23478-2	71.6	-8.5	271,715	858,370	533,362	16.3	33.82	4.2
23489-2	67.5	-12.5	244,263	868,186	338,378	16.8	34.51	6.2
23352-2	70.0	-12.4	534,010	1,707,188	584,053	18.9	33.76	3.2

[a] P.A. stands for chromatographic peak areas.
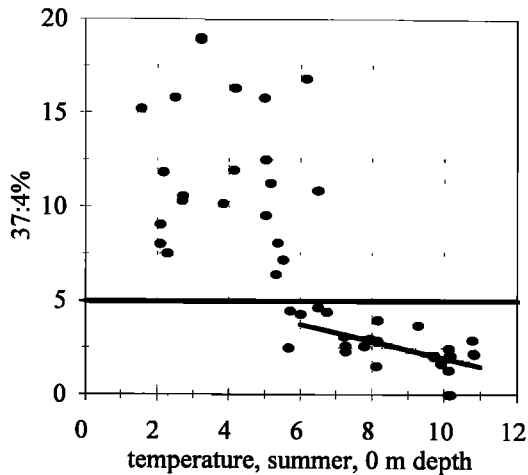[b] Data for summer at 0 m depth [Levitus and Boyer, 1994].

Figure 2. Core-top sediment data from the Nordic seas. The percentage of 37:4 alkenone to the total abundance of C_{37} alkenones (37:4%) is plotted against sea surface temperature (degrees centigrade) for summer at 0 m depth [*Levitus and Boyer*, 1994]. The line across the points below 5% corresponds to the regression line discussed in the text.
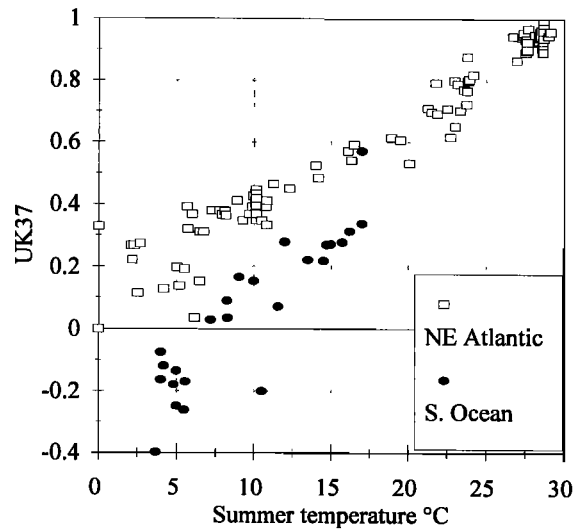

Figure 4. U^K_{37} data (sediment data) for the Southern Ocean [*Sikes et al.*, 1997] and the northeastern Atlantic [*Rosell-Melé et al.*, 1994] plotted against sea surface temperature (degrees celsius).

conundrum that could be related to various factors. For instance,

1. The difference in absolute 37:4% values between culture and sediment data may indicate that the variability of 37:4% depends on other parameters than SST. Thus the growth conditions in which the alkenones were biosynthesized both in the culture and the water column were not equivalent.

2. The strain of *E. huxleyi* used by *Prahl et al.* [1988], from the northwest Pacific, is not equivalent in terms of its 37:4% signal to the strains or species of dominant alkenone producers in the Nordic seas. However, it provides a more representative 37:4 to SST transfer equation (as it does for U^K_{37}') than the sediment data in Figures 1 and 2 and thus is best suited for paleotemperature reconstruction even in the North Atlantic.

3. The 37:4 alkenone is degraded preferentially through diagenesis in modern sediments [*Freeman and Wakeham*, 1992; *Rosell-Melé et al.*, 1995a; *Flügge*, 1997], so 37:4% in sediments appear lower than in cultures.
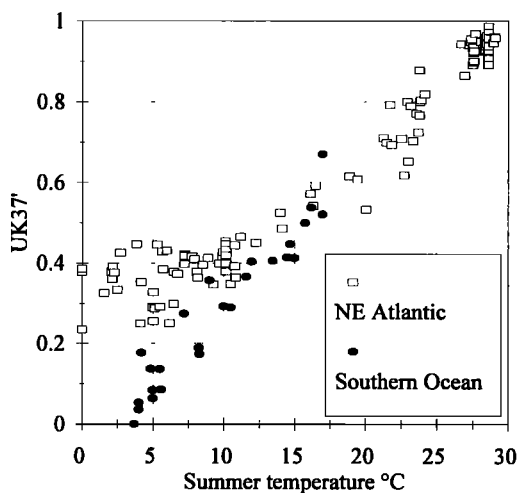
3.2. 37:4 in the Southern Ocean Versus Nordic Seas

The demonstration of the value of 37:4% as a temperature proxy is in apparent contradiction with data from the Southern Ocean, where no relationship with temperature was found for the relative abundance of 37:4, and thus U^K_{37}' appeared to be a better parameter than U^K_{37} for use in paleotemperature reconstruction even in cold
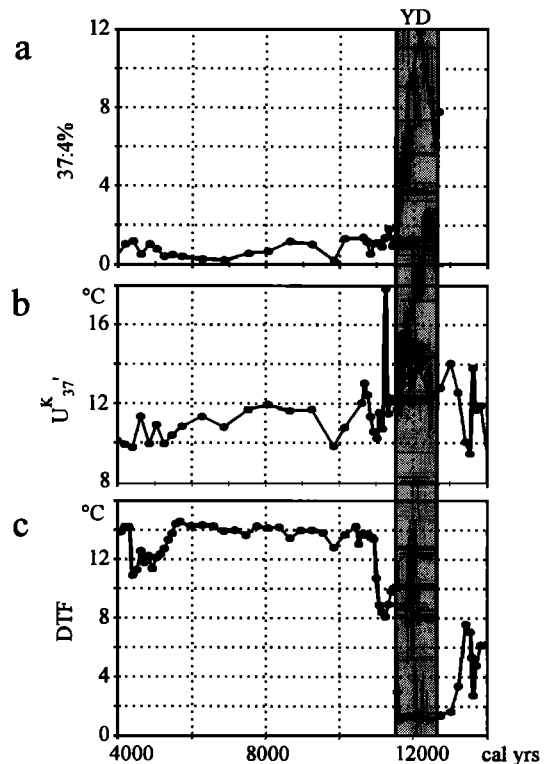

Figure 3. U^K_{37}' data (sediment data) for the Southern Ocean [*Sikes et al.*, 1997] and the northeastern Atlantic [*Rosell-Melé et al.*, 1994] plotted against sea surface temperature (degrees celsius).

Figure 5. Combined data from cores HM79-6/4, off southeastern Norway (~ 63°N, 3°E; 850-983 m depth), plotted against calendar years: (a) 37:4%, horizontal solid line marks the point at 37:4% = 5, (b) U^K_{37}' estimates, using the equation of *Prahl et al.* [1988], and (c) diatom transfer function sea surface temperature estimates, summer [*Koç-Karpuz et al.*, 1992].
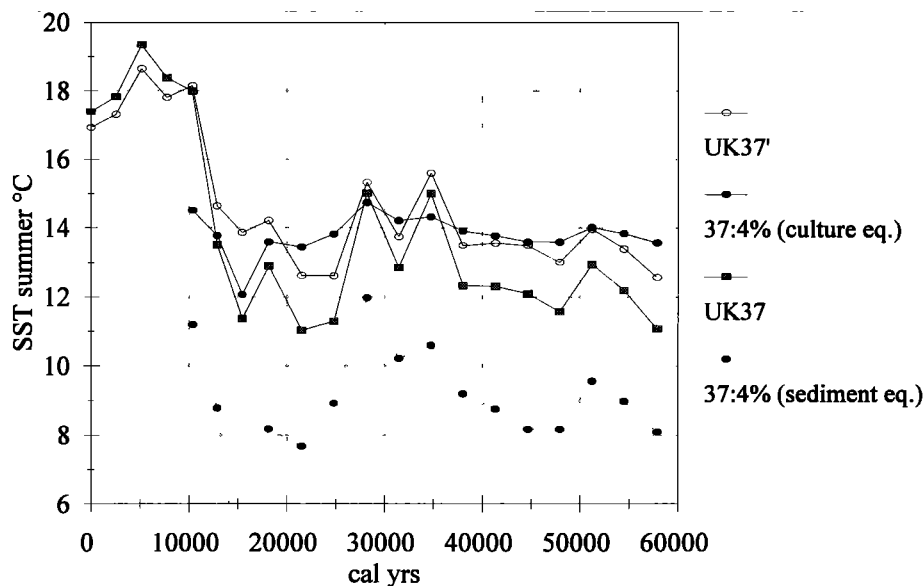
Figure 6. Data for core 19K in the northeastern Atlantic, (41°5'N, 20°47'W; 3742 m) [McCave, 1989]. U$^K_{37}$' and U$^K_{37}$ estimates have been obtained using the equation of Prahl et al. [1988] and Rosell-Melé et al. [1995a], respectively. The equations to convert 37:4% to SST are discussed in the text.

locations [Sikes et al., 1997]. Comparison of the alkenone data from the Southern Ocean and the North Atlantic (Figures 3, 4, 7, and 8) suggests that extrapolation of conclusions drawn from the Southern Ocean results to other oceanic locations is not straightforward. It should be noted that the comparison discussed in this paper has been done using only sediment data, unlike the original paper of Sikes et al. [1997] where both sediment and water column data were combined. Both types of samples are representative of natural inputs of alkenones, but over different time spans, i.e., hours to a few months for water column samples, compared to several tens or hundreds of years for core-top sediments, and are thus affected to a different extent by diagenetic processes. In addition, water column data contain an unknown degree of interannual and

intraannual variability, which is averaged out in sediments over a long period of time. Hence the combination of both sets of data, for the purpose of a calibration of a proxy, may complicate the analysis because the intercomparability of the data sets is not assured.

Sediments in the Southern Ocean contain much higher percentages of 37:4 than in the Nordic seas (Figure 7). In the former, 37:4% values occur between ~ 10% and 40% in a wide temperature range (3°-17°C) [Sikes et al., 1997], whereas in the latter most 37:4% data is between 0% and 20%. Such large variability in 37:4% must relate to distinct environmental conditions between both regions, in relation to a different 37:4% = f(SST) relationship perhaps affected by an additional oceanographic parameter and/or interhemispheric species/strains variability so that the north versus south alkenone producers respond to equivalent oceanographic conditions in a different manner. Conte et al. [1995] have shown that in cultures of E. huxleyi, 37:4% is strongly strain-dependent. Hence such 37:4% interhemispheric variations indicate that both data sets are not strictly comparable and probably cannot be assumed to be representative outside their immediate geographical regions. This can also be seen for U$^K_{37}$', which correlates with SST in the Southern Ocean [Sikes and Volkman, 1993; Sikes et al., 1997] with a similar relationship to the equation of Prahl et al. [1988], but this is not the case in other regions with very high 37:4%, i.e., Norwegian fjords [Conte et al., 1994; Ficken and Farrimond, 1995], the western Nordic seas (this paper), and in freshwater lakes [e.g., Thiel et al., 1997].

There are also some interregional similarities, and in the Southern Ocean, 37:4% increases strongly in SSTs colder than 5.5°-6°C (Figure 7), as noted previously for the Nordic Seas (Figure 2). This temperature threshold was noted by Sikes et al. [1997] for the change in the degree of unsaturation of alkenes versus alkenones and by Sikes and Volkman [1993] for an increase in the scatter of U$^K_{37}$'. Such change in response also occurs to alkenone producers in the Nordic seas at temperatures close to 5°-6°C [Rosell-Melé et al., 1994]. Hence, as also argued by Sikes et al. [1997], below
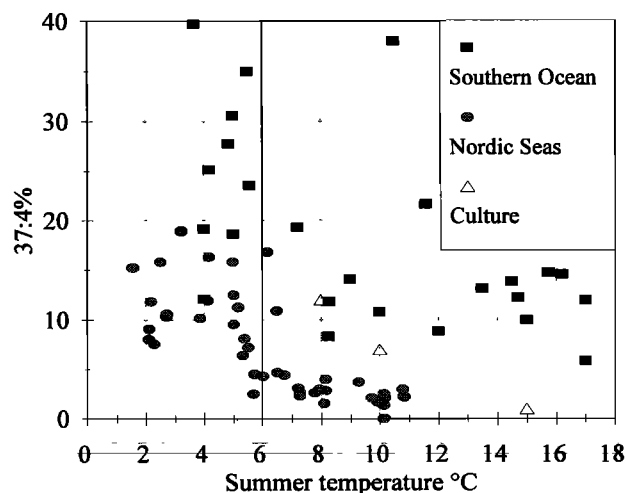

Figure 7. The 37:4% data (sediment data) for the Southern Ocean [Sikes et al., 1997] and the northeastern Atlantic plotted against sea surface temperature (degrees celsius). The 37:4% culture data have been estimated from Prahl et al. [1988] by subtracting U$^K_{37}$ from U$^K_{37}$', which is equivalent to 37:4% if the abundance of 37:4 is small compared to 37:3 + 37:2.
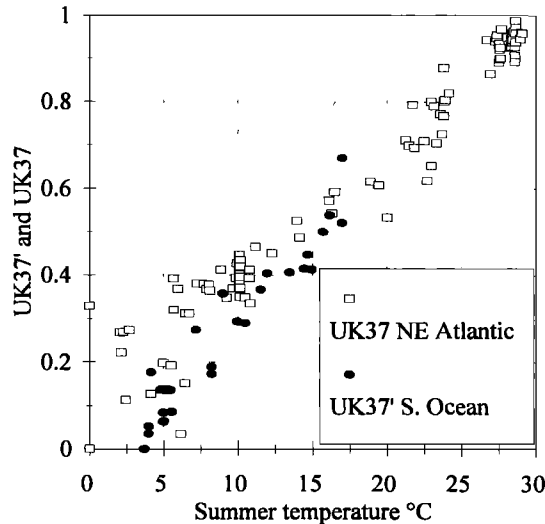
Figure 8. Sediment data of U^K_{37}' from the Southern Ocean [Sikes et al., 1997] and U^K_{37} from the northeastern Atlantic [Rosell-Melé et al., 1994] plotted against sea surface temperature (degrees celsius).

certain temperatures the alkenone producers seem to vary their biochemical response to changes in SST (~ 6°C, caloric summer). This could be taken as a lower threshold below which U^K_{37}' may not be reliable except that Müller et al. [1998] did not recognize it in a sample set from the South Atlantic. Unfortunately, no data are available on 37:4% from this area to find out if 37:4% increases at 6°C, but such interregional differences in alkenone data in areas with similar SST ranges indicate that at low temperatures, alkenone relationships in open ocean locations are affected by parameters other than SST even after the signal has been averaged out in the sediment.

Certainly, such differences clearly question the wisdom of extrapolating on the general applicability of U^K_{37} and U^K_{37} ' and other alkenone proxies to all areas of the oceans from conclusions drawn on data from a single regional study. For instance, in the Southern Ocean it is clear that both U^K_{37} and U^K_{37}' provide linear correlation with SST over a wide range of temperatures (Figures 3 and 4), with equations having similar slopes but different intercepts: U^K_{37} = -0.39 + 0.046T (r = 0.89) and U^K_{37} ' = -0.08 + 0.038T (r = 0.96). As shown in Figure 4, the large values of 37:4% strongly shift U^K_{37} in the Southern Ocean, so for a given range of temperatures, U^K_{37} in the Southern Ocean are ~ 0.4 units lower than in the NE Atlantic. In fact, despite the different behavior of 37:4 versus temperature from both high-latitude data sets, U^K_{37} correlates linearly with temperature over the same range of values both in the Southern Ocean and the NE Atlantic (Figure 4). In comparison, U^K_{37}' data from the Southern Ocean are somewhat closer to U^K_{37} from the North Atlantic except at lower temperatures (Figure 8). Differences are even larger at low temperatures between both high-latitude U^K_{37}' data sets (Figure 3). The deviation from Southern Ocean U^K_{37}' for both Nordic seas U^K_{37} and U^K_{37} ' takes place at ~ 10°C, but for U^K_{37}' in the Nordic Seas, there is no correlation with SST [Rosell-Melé et al., 1994] .

3.3. Value of 37:4% as a Salinity Proxy

The difference in the magnitude of 37:4% has been linked to types and origin of strains of E. huxleyi as higher percentages of 37:4

prevail in cultured strains of E. huxleyi from neritic compared to those from open ocean locations [Conte et al., 1995]. High percentages of 37:4 are also present in environments that are fresher than low and middle latitude oceans, where 37:4% are zero or close to zero. For instance, 37:4% values higher than 5% have been reported in Norwegian Fjords [Conte et al., 1994; Ficken and Farrimond, 1995], low-salinity marine environments such as the Baltic Sea [Schulz et al., 1997] and the western part of the Nordic seas (Figure 1), and notably, in lakes [Cranwell, 1985; Li et al., 1996; Thiel et al., 1997]. These data suggest a possible link between 37:4% and salinity and/or with other unknown variable(s) that may be common to all these environments. The Nordic seas data set is specially suitable to investigate the effect of salinity on 37:4% because of the marked SE-NW salinity gradient in the area, and the lack of a correlation between SST and values of 37:4% above 5%. The consideration of nutrient concentration having an effect on the degree of unsaturation of alkenones [Epstein et al., 1998; Popp et al., 1998] can also be discarded as having an influence on 37:4% because over the range of concentrations of nitrate investigated, no correlation was found with 37:4% (Figure 9).

The assignment of oceanographic variables to sediment data samples, as in the type of study discussed here, is certainly one of the more uncertain steps of the investigation. In this study, to investigate the role of salinity, this difficulty arises because of the very strong halocline in some of the sample locations and the difficulty of assigning the correct salinity value according to the average depth at which the alkenones were produced, which in any case is unknown. Thus samples collected from locations under the direct influence of the East Greenland Current (low-salinity current), marked with open symbols in Figures 1 and 10, have a salinity gradient of 1.5-2.5 psu between 0 and 50 m depth, whereas all the other sample locations show a gradient of < 1 psu (Figure 10). The choice of the water depth at which the alkenones were biosynthesized is more important in the former than in the latter groups of samples, and of course, such depth does not necessarily have to be the same throughout the Nordic seas as the extent of water column stratification, nutrients, and floral and faunal assemblages are different throughout the area [e.g., Samtleben et al., 1995]. Therefore, because of the distinct oceanographic features
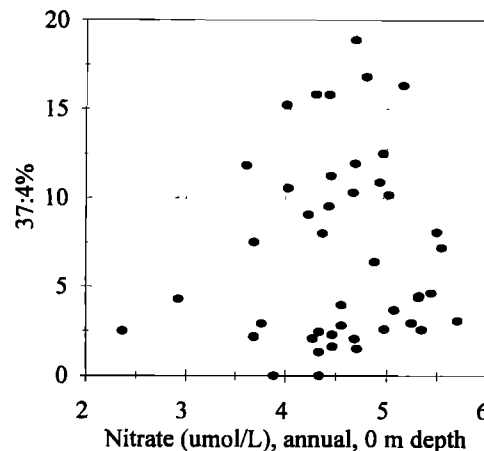

Figure 9. The 37:4% values from all samples in the Nordic sea (see Figure 1) plotted against nitrate concentrations (μmolL^{-1}; annual averages at 0 m depth) [Levitus and Boyer, 1994].
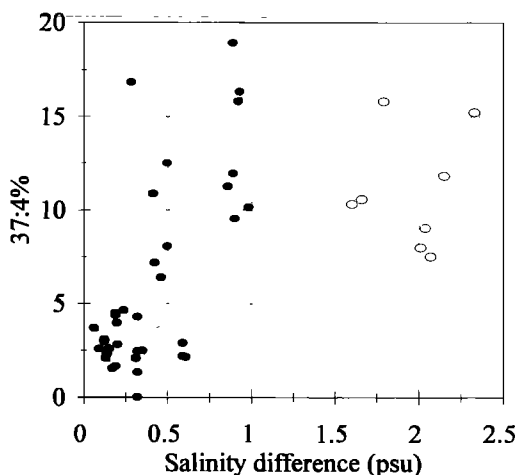
Figure 10. The 37:4% values from the Nordic seas data set (see Figure 1) plotted against the difference in salinity values between the depths of 0 and 50 m (summer average) [Levitus and Boyer, 1994]. The open symbols correspond to the East Greenland Current data set (see Figure 1).

of the East Greenland Current area where some of the samples were retrieved, i.e., directly affected by Arctic currents and sea ice [*Swift*, 1986], with a strong halocline [*Levitus and Boyer*, 1994], and distinct Haptophyte assemblages [*Samtleben et al.*, 1995], this group of samples is not comparable to the rest of samples available from the Nordic seas, and its inclusion in the analysis would introduce a large bias. Thus, to reduce some uncertainty in the analysis, the "East Greenland Current sample set," composed of eight samples (clustered together in Figure 1 and 10), has not been taken into account to address the effect of salinity on 37:4%.

In Figure 11, values of 37:4% are plotted against salinity at 0 m depth, showing a linear correlation between both parameters (37:4% = 304.95 - 8.66S (psu), r = 0.80, and n=38). Such a relationship should be further investigated in culture and field experiments, but it shows that 37:4% can probably be used as a proxy for paleosalinity in the Nordic seas, and, by extension, to the
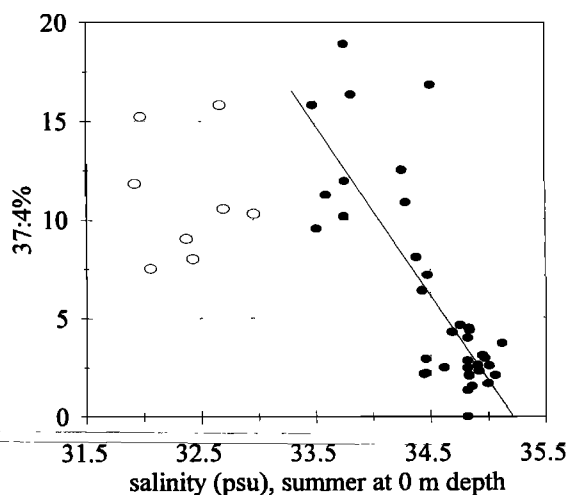
North Atlantic during the last glacial maximum to estimate salinity values below 35 psu. Its application to other environments must certainly be confirmed before the use of the proxy can be generalized, especially as when using the data of *Sikes et al.* [1997], no relationship was found between salinity and 37:4% in the Southern Ocean. Once the relationship between 37:4% and salinity is further studied, it may eventually be found not to be linear in nature. This is anticipated because the biological functions of the cell membrane, where alkenones may be found, are strongly dependent on the chemical potential or its gradient across the cell wall, which in turn depends on temperature and the logarithm of the molar fraction of solutes (e.g., salinity) in the media.

In the light of this new relationship the 37:4% record in Figure 5a can be interpreted as a paleosalinity record. The increase of 37:4% during the YD in core HM79-6/4 indicates a decrease in salinity of ~ 1.5 psu in the surface waters during that period, most probably induced by the melting of sea ice during summer off southwest Norway [*Koç et al.*, 1993]. The value of 37:4% can be further demonstrated with the data obtained from core M23415 in the northeastern Atlantic (Figure 12), which shows that the stratigraphic variability of 37:4% is indeed influenced by paleoceanographic changes. The 37:4% values show large variations (0%-11%) during the last 50,000 years, which parallel those in the $\delta^{18}O$ curve, with the lowest values of 37:4% occurring during the Holocene and the highest occurring during isotopic stages 2 and 3. These changes must be related to SST (37:4% < 5) and in a large part to salinity as demonstrated in Figures 2, 9 and 11 and also by
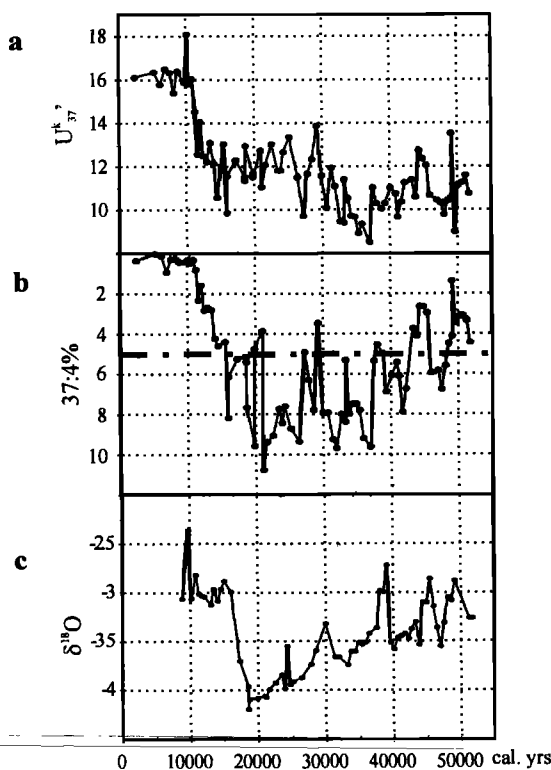

Figure 11. The 37:4% values from Figure 1 plotted against salinity (summer average at 0 m depth) [Levitus and Boyer, 1994]. The open symbols show those samples not considered to calculate the regression line (solid line).

Figure 12. Data from core M23415 (~ 55°N, 19°W; 2500 m) plotted against calendar years: (a) U$^K_{37}$' estimates, using the equation of *Prahl et al.* [1988], (b) 37:4%, horizontal dotted line marks the point at 37:4% = 5, and (c) oxygen isotopic data from *Neogloquadrina pachyderma* [S. Jung and M. Sarnthein, unpublished, 1997]

analogy with the $\delta^{18}O$ curve, which is controlled by the same parameters (T and S) in addition to ice sheet volume. The $U^K_{37}{}'$ and 37:4% records contain some marked differences (Figures 12a and 12b), which show that both variables are not interdependent and provide different proxy information. In fact, the $U^K_{37}{}'$ curve from M23415 (Figure 12a) is difficult to interpret if previous work on SST reconstructions in the area, using foraminifera transfer functions, are taken into account. For instance, *CLIMAP Project Members* [1981] and *Sarnthein et al.* [1995] have estimated for the LGM in the core location a SST of ~ 5°-6°C, whereas $U^K_{37}{}'$ provides values of ~ 12°C. To understand this large difference, one should consider that high inputs of reworked coccoliths to glacial sediments have been observed during stages 2 and 3, i.e., as ice-rafted debris [*Rahman*, 1995]. Hence, higher than expected SST $U^K_{37}{}'$ estimates could be attributed to the alkenone signal being biased because of allochthonous alkenones being associated with these inputs, which would contain a warmer signal because they would have been presumably produced during earlier, warmer geological periods than the late Quaternary. The high abundance of 37:4 in the sediments does not support such an argument as this component is associated to cold and fresh environments and has not been reported in pre-Quaternary sediments which contain a warm alkenone signal with a very high abundance of 37:2 [*de Leeuw et al.*, 1980; *Marlowe et al.*, 1984; *Farrimond et al.*, 1986]. Hence the substantial values of 37:4% in M23415 prove that autochthonous alkenones predominate in the sediment and, in fact, that the SST $U^K_{37}{}'$ estimates are not reliable in large parts of the record. The similarity of the oxygen isotopic and 37:4% records suggests that the latter can be considered a more reliable molecular proxy than $U^K_{37}{}'$ to reconstruct paleoceanographic changes at high latitudes, at least in the North Atlantic region, and, furthermore, that it can be used to derive paleosalinity information during glacial episodes.

4. Conclusions

There are significant regional or interhemispheric differences in the temperature dependence of the relative abundance of sedimentary alkenone data at the cold end of the temperature range. U^K_{37} can be used to estimate SST both in the Southern Ocean and the Nordic seas, albeit with different calibration equations. In contrast, $U^K_{37}{}'$ in the Nordic seas below 10°C shows no correlation with SST although $U^K_{37}{}'$ does correlate with SST in the Southern Ocean. Leaving practical or analytical considerations aside, the application

of U^K_{37} and $U^K_{37}{}'$ as climate proxies is thus equally justified but constrained by regional differences regarding their calibration equations and temperature ranges.

Such geographical variations are especially significant in relation to the relative abundance of the tetraunsaturated alkenone to the total abundance of C_{37} alkenones (37:4%), which show marked differences between data sets from the northern and southern hemispheres. In the Nordic seas, 37:4% correlates with SST when 37:4% < 5, but it does not when 37:4% > 5. On the basis of the latter the measurement of 37:4% downhole a core is proposed to identify situations when $U^K_{37}{}'$ is not a reliable paleothermometer in the North Atlantic, i.e., when 37:4% > 5 so $U^K_{37}{}' \neq f(SST)$. This relationship is tested in two cores and used to explain the anomalously high SST estimates obtained from $U^K_{37}{}'$ during the Younger Dryas and the last glacial maximum. Variations in salinity are argued to be responsible for changes in the sedimentary record of 37:4%, and a preliminary calibration has been obtained for 37:4% $= f(Salinity)$. This relationship may explain why 37:4% increases off southwestern Norway during the Younger Dryas, i.e., because of a decrease in surface salinity, which could be associated to the seasonal, summer melting of sea ice. However, the new relationship found between 37:4% and salinity should be further confirmed through field or laboratory experiments before its use can be generalized, especially outside the North Atlantic region.

The results presented do not question the value of $U^K_{37}{}'$ as a general paleotemperature proxy but are intended as a contribution toward constraining its use to estimating reliable low SST. Future studies or consideration of published investigations into the SST dependence of alkenones should take into account that observations or recommendations based on regional databases cannot be extended beyond their geographical coverage unless their global significance is proven. However, more regional studies are needed to understand the environmental variability of alkenone proxies and constrain their application as effective oceanographic proxies.

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