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C_{37} alkenone measurements of sea surface temperature in the Gulf of Lions (NW Mediterranean)

Isabel Cacho[a,b], Carles Pelejero[a], Joan O. Grimalt[a,*], Antoni Calafat[b], Miquel Canals[b]

[a]*Department of Environmental Chemistry (C.I.D.-C.S.I.C.), Jordi Girona 18, 08034 Barcelona, Catalonia, Spain*
[b]*G.R.C. Marine Geosciences, Department of Stratigraphy and Paleontology, University of Barcelona, 08071 Barcelona, Catalonia, Spain*

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Abstract

A study of the C_{37} alkenone compositions in suspended particulate matter in the northwestern Mediterranean Sea has shown a correspondence between $U^{K'}_{37}$ and sea surface temperature that significantly deviates from the general equation regularly observed in most marine world areas (Müller et al., 1998). However, the temperatures measured in the core top sediments using the general equation are in agreement with the annual average water column temperatures between 0–40 m depth. These discrepancies suggest that despite the rather constant correlation between $U^{K'}_{37}$ and seawater temperature throughout the world oceans, specific calibrations should be developed for each new area of application of the C_{37} alkenones for paleotemperature determination.

Keywords: Sea surface temperatures; Alkenones; $U^{K'}_{37}$ index; Paleoceanography; Mediterranean Sea; Suspended particulate matter; Recent sediments

1. Introduction

The reconstruction of marine sea surface temperatures along the glacial and interglacial cycles of the Quaternary provides a fundamental parameter for the description of the air–sea interactions during these periods. These reconstructions are based on the study of the sedimentary record using indirect thermometers which fossilized their thermal signal.

Among the various methodologies available for this purpose, the C_{37} di-, tri-, and tetraunsaturated alkenones have received widespread attention. In open marine waters these compounds are known to be synthesized by a few species of the Haptophyceae, including *Emiliania huxleyi* and *Gephyrocapsa oceanica* (Volkman et al., 1980, 1995; Marlowe et al., 1984, 1990; Conte et al., 1995; Sawada et al., 1996). The relative content of alkenones having different degrees of unsaturation is related to growth temperature (Brassell et al., 1986; Prahl and Wakeham, 1987).

The correspondence between temperature and relative composition of C_{37} alkenones has been calculated using the following ratios: $U^{K}_{37} = (C_{37:2} - C_{37:4})/(C_{37:2} + C_{37:3} + C_{37:4})$ and $U^{K'}_{37} = C_{37:2}/(C_{37:2} + C_{37:3})$. Usually, the second ratio is used, since the relationship

* Corresponding author. Tel.: +34-3-400-6122; fax: +34-3-204-5904.
E-mail address: jgoqam@cid.csic.es (Joan O. Grimalt)

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Table 1
Water particulate and sediment samples collected in the Gulf of Lions

Station	Latitude	Longitude	Water depth (m)	Date	Sediment	Suspended particles	
						Depth (m)	Volume (m)
5	42°54′N	4°58′E	600	14-4-97	✓	5	182.1
						40	124.9
						110	128.1
6	42°48′N	5°02′E	1200	15-4-97	✓	5	50.6
						40	58.9
						110	88
8	42°48′N	4°44′E	600	16-4-97	✓	5	32.3
						40	34.2
						110	80.4
9	42°42′N	4°49′E	1200	17-4-97	✓		

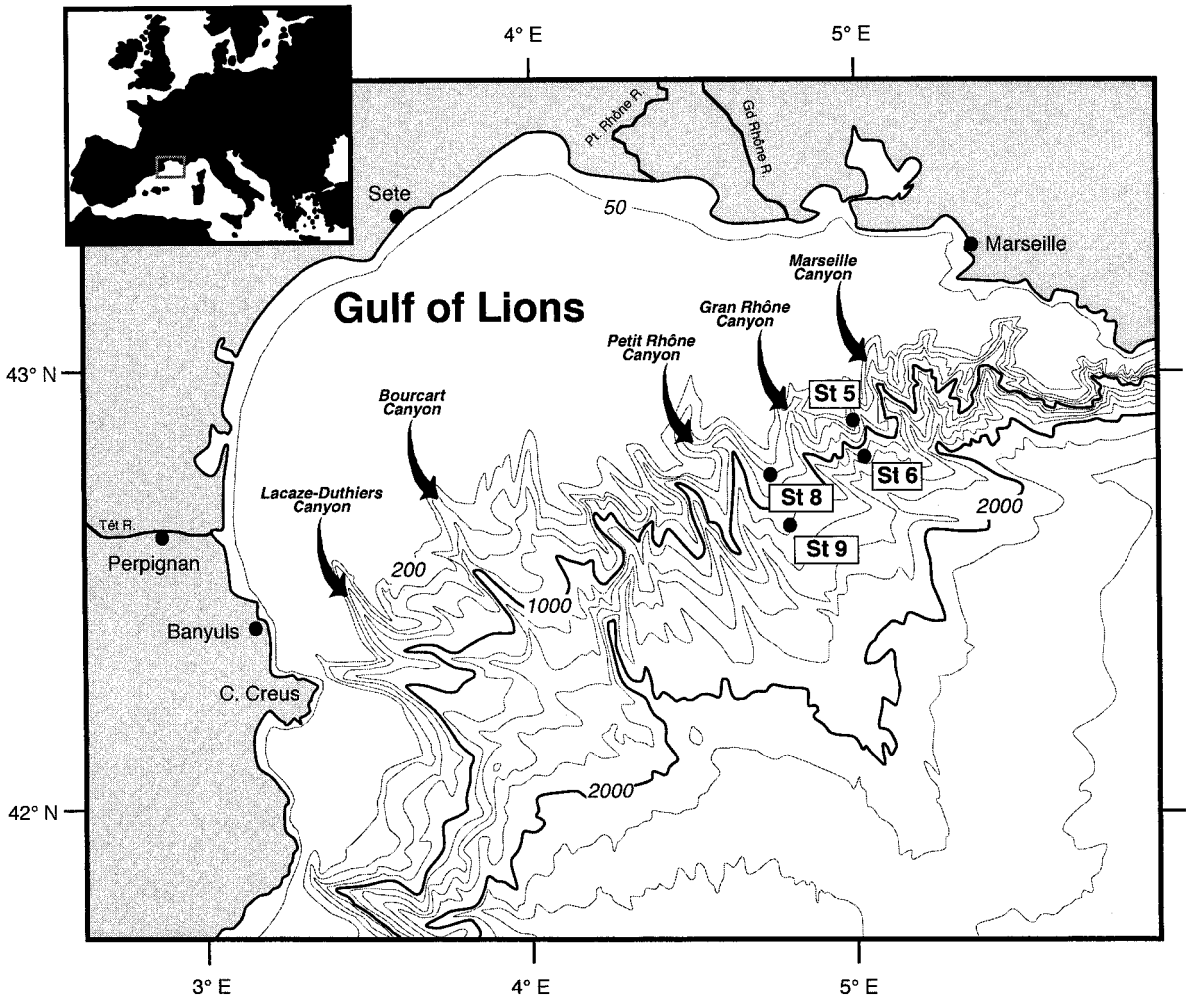

Fig. 1. Bathymetric chart of the Gulf of Lions showing the location of the stations selected for study.

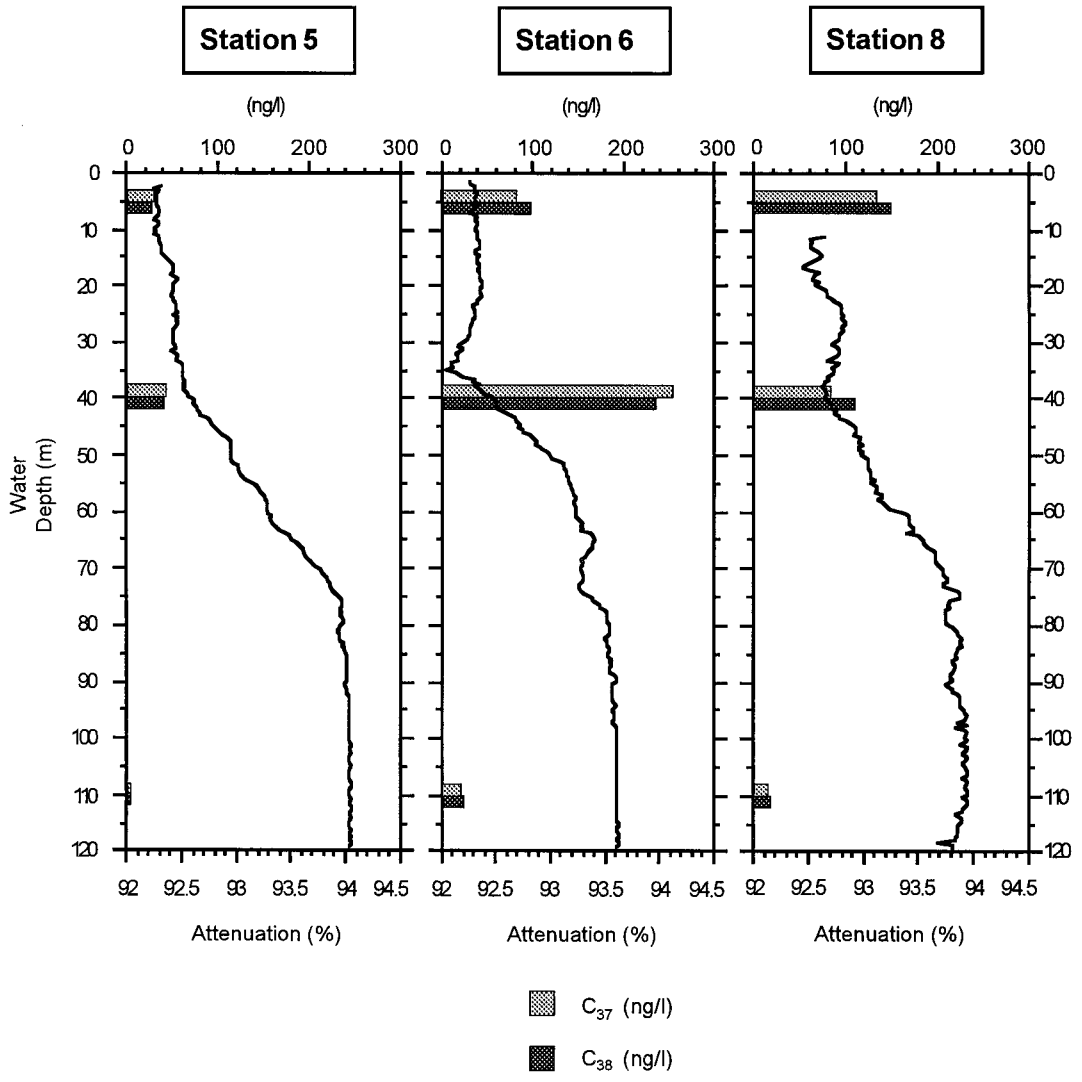

Fig. 2. Total C_{37} and C_{38} alkenone concentrations (bars) at the water depths considered for study (5, 40 and 110 m). The continuous line indicates the CTD measurements of light transmission which are related to the turbulence and particle concentration in the water column.

between $C_{37:4}$ and water temperature is still under debate (Sikes et al., 1997). Laboratory cultures have shown that these ratios are linearly related to temperature (Prahl and Wakeham, 1987; Prahl et al., 1988; Sawada et al., 1996). Calibrations with water column particles (Prahl and Wakeham, 1987; Sikes and Volkman, 1993; Ternois et al., 1997) and sediment core tops (Sikes et al., 1991, 1997; Rosell-Melé et al., 1994, 1995; Sonzogni et al., 1997a, 1997b; Pelejero and Grimalt, 1997; Müller et al., 1998) have confirmed this linear relationship, at least over the temperature range 5–28°C.

One interesting aspect of this linear correspondence is the rather constant slope between $U^{K'}_{37}$ and water column temperatures which shows similar values in culture experiments, water particles and sediment core top data (Müller et al., 1998). From a geographical perspective, this uniformity indicates that specific differences in the relationship between alkenone composition and seawater temperature, such as changes in precursor strains, are not significant even when different locations are considered. This aspect has been recently illustrated in a pooled calibration including core-top sediments from the Atlantic, Pacific and

Indian oceans located between 60°S–60°N (Müller et al., 1998). A recent calibration study in the South China Sea and the Indian Ocean has also shown that even at high water column temperature (25–29°C) the same slopes are observed (Pelejero and Grimalt, 1997; Sonzogni et al., 1997b). The relationship between $U^{K'}_{37}$ and temperature in waters colder than 5°C still requires further calibration studies (Sikes and Volkman, 1993; Rosell-Melé et al., 1994, 1995; Sikes and Keigwin, 1996; Sikes et al., 1997; Ternois et al., 1998).

In contrast with this uniform $U^{K'}_{37}$–temperature linear relationship, some alkenone studies in specific sites such as the Black Sea (Freeman and Wakeham, 1992), the northwestern Mediterranean Sea (Ternois et al., 1997), and Eastern North Atlantic sites (Conte et al., 1992) have shown major deviations from the general trend. In this respect, a calibration curve distinct from the general equation has been obtained in the northwestern Mediterranean (Ternois et al., 1997). These results encourage further studies in these locations for better appraisal of their singularity.

The present study is devoted to further assess the correspondence between $U^{K'}_{37}$ and water column temperatures in the Gulf of Lions (northwestern Mediterranean Sea). For this purpose, water particles and sediment core tops have been collected and analyzed. The results have been compared with both in situ temperature measurements and averaged data from the Levitus dataset (Levitus and Boyer, 1994).

2. Methods

2.1. Samples

Samples were collected in spring 1997 during the TRACEURS 2–BENTHOS 3 cruise onboard the oceanographic vessel Aegaeo. This cruise was part of the high-frequency flux (HFF) experiment carried out in the Gulf of Lions in the framework of the MTP II–MATER Project. Strong north winds (Mistral) were blowing during sampling which generated strong vertical water column mixing and an absence of a marked thermocline.

Water sampling for suspended particles was carried out at three stations: St. 5, 6 and 8 (Table 1; Fig. 1), and three different depths (5, 40 and 110 m). Depths were selected in order to obtain the maximum thermal gradient after examination of the CTD (conductivity, temperature and depth) profiles (Fig. 2). Suspended particulate matter was obtained by in situ pumping (Challenger Oceanic) through glass fiber filters (GFF Millipore, 142 mm diameter). Filters were kiln-fired at 400°C for 12 h prior to use. Surface sediment samples were collected at stations 5, 6, 8 and 9 Table 1; Fig. 1 with a multicore system affording sediment columns with an undisturbed water–sediment interface. All filter and sediment samples were frozen immediately after recovery.

2.2. Analysis

The procedures and equipment used for U^{K}_{37} index determinations are described elsewhere (Villanueva et al., 1997). Briefly, filters were cut into small pieces and sediment samples (2–3 g) were freeze-dried and manually ground for homogeneity. After addition of an internal standard mixture containing *n*-nonadecan-1-ol, *n*-hexatriacontane and *n*-tetracontane, samples were extracted in an ultrasonic bath with dichloromethane. The extracts were hydrolyzed with 6% potassium hydroxide in methanol to eliminate interferences from wax esters. The hexane extracts were then fractionated by silica column chromatography to separate hydrocarbons and ketones (dichloromethane–hexane, 8:2) from the polar fraction (dichloromethane–methanol, 9:1). The extracts were derivatized with bis(trimethylsilyl)trifluoroacetamide before instrumental analysis.

Alkenones were analysed with a Varian gas chromatograph Model 3400 equipped with a septum programmable injector and a flame ionisation detector. Alkenones were separated in a CPSIL-5 CB column coated with 100% dimethylsiloxane (film thickness 0.12 μm). Hydrogen was the carrier gas (50 cm/s). The oven temperature was programmed from 90 to 140°C at 20°C/min, then to 280°C at 6°C/min (holding time 25 min), and finally to 320°C at 10°C/min (holding time of 6 min). The injector was programmed from 90°C (holding time 0.3 min) to 320°C at 200°C/min (final holding time 55 min). *n*-Hexatriacontane was used as internal standard for alkenone quantitation.

Selected samples were examined by gas chromatography/mass spectrometry for confirmation of compound identification and evaluation of possible coelutions. Spectra were obtained with a Fisons MD-800 instrument operated in the electron impact mode (70 eV) by scanning from mass 50 to 550 every second. The GC conditions were the same as described above.

3. Oceanographical setting and study sites

The oceanographic regime of the Gulf of Lions is mainly controlled by the Northern Current (or Liguro–Provençal Current: Millot, 1987, 1990), an almost cyclonic permanent current that flows along the slope from east to west. The Northern Current involves both Levantine Surface Water and Modified Atlantic Water (MAW). The Gulf of Lions is also affected by the fresh and nutrient-rich water discharge from the Rhône and smaller rivers. River inputs are

controlled by the local and regional rainfall which is especially strong during autumn (October–November) and winter (January–February) (Monaco et al., 1990).

The Gulf of Lions, like the entire Mediterranean Sea, is characterized by strong seasonal changes. During winter, the north–northwesterly winds (Tramontana and Mistral) are very strong. They cool and homogenize the MAW inducing the formation of Western Mediterranean Deep Water (WMDW; Millot, 1990; Durrieu de Madron et al., 1990). The Northern Current is also associated with the formation of WMDW, displaying maximum transport during this period of the year (Béthoux et al., 1988; Conan and Millot, 1995). In spring, when sea surface waters become calmer, but are still nutrient enriched as a consequence of winter vertical mixing, numerous blooms develop over the entire area. These blooms start in mid-April and last about six weeks with maximal extension in early May (Morel and André, 1991). Thermocline formation is initiated in April and progresses throughout summer leading to strong water stratification. In autumn, nutrients are again brought to the upper water layers by erosion of the seasonal thermocline thus inducing a secondary bloom period for two weeks in October (Morel and André, 1991; Lefevre et al., 1997).

The studied stations are located on the continental slope of the eastern Gulf of Lions which extends from 200 to 2000 m depth and is indented by a large number of submarine canyons. These canyons are the main transport pathways for suspended particulate matter from the shelf to the slope (Durrieu de Madron et al., 1990). As a consequence of the eastern component of the Northern Current there is a clear east–west gradient along the continental slope in the concentration of suspended particulate matter. The northeastern area where our stations are located is less influenced by the Rhône and other rivers, the canyons being located in the southwestern most part (Bourcart and Lacaze–Duthiers canyons) through which the suspended material originating from the whole shelf passes (Durrieu de Madron et al., 1990; Monaco et al., 1990). A large portion of the continental shelf organic matter (nearly 50%) is also exported from the shelf to the adjacent slope and open sea through these canyons (Buscail et al., 1990). The exported flow is highly variable and seasonally influenced. On the other hand, the distribution of organic carbon in surface sediments shows higher values in the canyons than in areas between them (Buscail et al., 1990). All our stations are placed in these interfluve locations: stations 5 and 6 are situated in the interfluve between the Marseille and the Grand Rhône canyons and stations 8 and 9 are in the interfluve between the Grand Rhône and the Petit Rhône canyons (Fig. 1). They are located at different

Table 2
C_{37} alkenone composition in the water particulate samples collected in the Gulf of Lions

Station	Water depth (m)	$C_{37:3}$ (ng/l)	$C_{37:2}$ (ng/l)	C_{37} (ng/l)	$C_{38:3Et}$ (ng/l)	$C_{38:3Me}$ (ng/l)	$C_{38:2Et}$ (ng/l)	$C_{38:2Me}$ (ng/l)	C_{38} (ng/l)	$U^{K'}_{37}$	Temperature		
											Eq. (1)[a]	Eq. (2)[b]	CTD
5	5	17	10	28	7.1	6.6	8.1	3.7	26	0.37	9.7	14.2	14.3
	40	26	14	41	11.4	9.6	12	4.7	37	0.36	9.4	13.8	13.8
	110	1	0.6	1.7	0.7	0.5	0.9	0.2	2.4	0.39	10.3	14.6	13.5
6	5	45	32	77	26	21	31	16	95	0.42	11.2	15.3	15.0
	40	152	97	250	62	61	73	36	230	0.39	10.3	14.6	14.2
	110	10	6.2	17	6.5	4.8	6.9	2.1	20	0.37	9.7	14.1	13.4
8	5	82	50	132	42	37	48	21	150	0.38	10.0	14.4	14.5
	40	57	24	81	38	26	36	8.9	110	0.30	7.6	12.4	13.6
	110	7.7	4.2	12	5.2	3.5	5.3	1.3	15	0.35	9.1	13.8	13.4

[a] Equation (1): $U^{K'}_{37} = 0.033T + 0.044$ (Müller et al., 1998).
[b] Equation (2): $U^{K'}_{37} = 0.041T - 0.21$ (Ternois et al., 1997).

Table 3
C_{37} alkenone composition in the sediments of the Gulf of Lions

Station	$C_{37:3}$ (ng/g)	$C_{37:2}$ (ng/g)	C_{37} (ng/g)	$C_{38:3Et}$ (ng/g)	$C_{38:3Me}$ (ng/g)	$C_{38:2Et}$ (ng/g)	$C_{38:2Me}$ (ng/g)	C_{38} (ng/g)	$U^{K'}_{37}$	Temperature	
										Eq. (1)[a]	Eq. (2)[b]
5	55	71	130	30	25	73	23	150	0.56	15.5	18.9
6	92	102	190	55	36	102	20	210	0.53	14.6	18.0
8	55	72	130	28	24	64	20	140	0.57	15.8	19.0
9	43	52	96	32	20	67	16	130	0.55	15.2	18.4

[a] Equation (1): $U^{K'}_{37} = 0.033T + 0.044$ (Müller et al., 1998).
[b] Equation (2): $U^{K'}_{37} = 0.041T - 0.21$ (Ternois et al., 1997).

depths, stations 5 and 8 in the upper slope (600 m), and stations 6 and 9 in the middle part (1200 m).

4. Results and discussion

4.1. Alkenone concentrations in the water column

In Fig. 2 the measured water column C_{37} and C_{38} alkenone concentrations are plotted together with the CTD profile of light transmission. This parameter is related to suspended particulate matter which depends on phytoplankton concentration and therefore vertical water column mixing. The vertical distribution of these alkenones exhibits a contrast between the first two depths (5 and 40 m) and 100 m depth. This contrast is concordant with the transmission profile indicating that suspended particles and therefore phytoplankton were in low concentrations below depths of 50 m (station 6), or 70 m (stations 5 and 8). The concentrations of both C_{37} and C_{38} alkenones show parallel trends in all filters which is in agreement with their common origin.

The concentrations of both C_{37} and C_{38} alkenones exhibit similar values at the shallower levels which is consistent with the vertical mixing due to the strong wind conditions present during the sampling period (Fig. 2). Station 6 displays the highest contrast between 5 and 40 m. This contrast is concordant with the transmission and temperature CTD profiles (Fig. 2 and Table 2), indicating more stratified water conditions for this station. The higher stratification in this station is likely due to meteorological conditions since sampling was carried out after a few days of calmer wind conditions than during the sampling at the other stations. In this case, the alkenone concentrations are greater at 40 m depth which is in agreement with previous observations of pigments (Barlow et al., 1997) and primary production (Lefevre et al., 1997) in this area under water stratification conditions.

4.2. Water column temperature measurements

In Table 2 the $U^{K'}_{37}$ temperature estimates obtained from the suspended particles filtered at various water column depths are compared with the CTD temperature measurements during sampling. These estimates have been calculated using the equation calibrated for the northwestern Mediterranean region from suspended particulate matter (Ternois et al., 1997), and have been compared with the ones obtained with the general equation (Müller et al., 1998) showing differences of 4.5–5.5°C. In all cases the Ternois et al. (1997) equation provides higher values, in better agreement with the CTD measurements.

Thus, at 5 m depth the differences between CTD and the $U^{K'}_{37}$ measurements calculated with this latter equation are smaller or equal to 0.25°C. At 40 m depth these differences account for 0, 0.5°C and for 1.2°C in stations 5, 6 and 8, respectively. At 100 m the $U^{K'}_{37}$ values reflect the temperatures at shallower levels than at this depth. This last difference is consistent with synthesis in the more illuminated upper waters and transport to deeper levels. The vertical distribution of alkenone concentrations is also consistent with this difference since the concentrations at 100 m depth are significantly lower than in the euphotic zone (Fig. 2).

4.3. Sediment temperature estimates

The temperatures resulting from the $U^{K'}_{37}$ measurements in the sediments have also been calculated using the general equation (Müller et al., 1998) and the specific equation for this area (Ternois et al., 1997; Table 3). A difference of 3.2–3.4°C is observed between the results of these two equations.

Comparison of the average sedimentary values of these temperature estimates with the average water column profile obtained from the Levitus data (Levitus and Boyer, 1994) for this region (Fig. 3) shows a better agreement with the results of the general equation (Müller et al., 1998). The values provided by this equation are coincident with the average temperatures

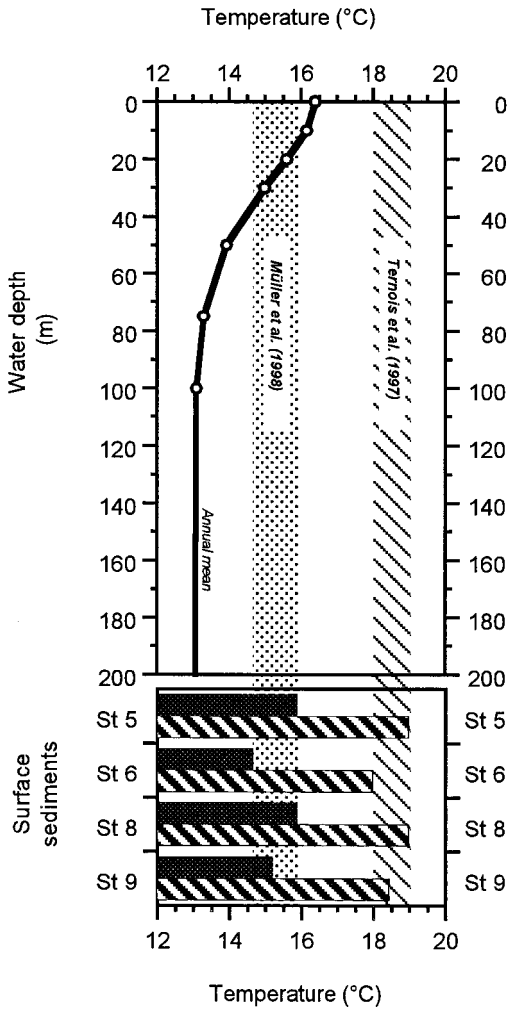

Fig. 3. Average water column temperature profile in the studied area (Levitus and Boyer, 1994) compared with the $U^{K'}_{37}$ temperature estimates from sediment measurements using the equations from Müller et al. (1998) (stippled bars) and Ternois et al. (1997) (striped bars).

between 10–40 m depth, the section where most of the alkenones are synthesized. In contrast, the sedimentary $U^{K'}_{37}$ temperatures estimated with the Ternois et al. (1997) equation are higher than all of the water column values. They are even 2°C higher than the surface temperatures.

Further assessment of these different estimates can be obtained by consideration of the seasonal temperature changes. Thus, in Fig. 4 the average sedimentary temperatures are compared with the monthly Levitus water column temperatures at 0, 30 and 100 m depth. In both cases, the temperatures recorded in the sediments are closer to those measured at shallower depths (0–30 m). The Levitus temperatures at 100 m depth are always less than these two estimates. However, whereas the Ternois et al. (1997) equation gives estimates in agreement with the warmer end (summer and autumn), the results from the general equation nearly correspond to the average annual temperature at these depths as already seen in Fig. 4. Sediment trap studies in the Ligurian Sea indicates that maximum alkenones flux occurred in October (Ternois et al., 1996). A smaller alkenone productivity maximum is detected in May (Ternois et al., 1997) corresponding with the spring bloom period typical of the area (Morel and André, 1991). Haptophytes are not the major phytoplanktonic species during the spring bloom (Ternois et al., 1997). Studies from nearby areas indicate that during cold months (including the spring bloom) the distribution of algal species is very homogeneous and dominated by diatoms. In contrast, in the warm months (including the autumn bloom) a larger variety of species is observed and coccolithophores are found in significant proportion (Travers, 1975; Estrada et al., 1985; Knappertsbusch, 1993).

All measured $U^{K'}_{37}$ values in the water column of this study and that of Ternois et al. (1997), fall in the range 0.3–0.5, and are lower than the sedimentary values (0.53–0.63). However, the time span of the water column samples represented in this study and that by Ternois et al. (1996) is short and do not include data from October. Lateral transport from the shelf related to fluvial input could account for the differences between water and sediment $U^{K'}_{37}$ indices, but as explained in the hydrographic setting section, the position of the stations, in the eastern part of the slope and outside the canyons, minimizes the influence of river input.

Furthermore, the distributions of C_{37} alkenones from coastal areas receiving significant freshwater inputs have been observed to contain $C_{37:4}$ in high proportion (Conte et al., 1994; Ficken and Farrimond, 1995) which is consistent with the distributions observed in lakes (Cranwell, 1985; Li et al., 1996; Thiel et al., 1997). In all these cases, the relationship between alkenone distributions and water temperatures is different from the general equation (Prahl and Wakeham, 1987; Müller et al., 1998). However, no traces of $C_{37:4}$ have been found in the water column and sediment samples examined from the Gulf of Lions. The lack of this alkenone suggests that the water column alkenone distributions in the northwestern Mediterranean Sea cannot be due to contributions from Haptophyceae species living in coastal or freshwater environments as in the other examples. In fact, no relationship between SST and U^{K}_{37} has been found in cases of high $C_{37:4}$ content, whereas in the Gulf of Lions there is a clear linear relationship between SST and alkenone distribution, albeit different from the general equation reported by Müller et al. (1998).

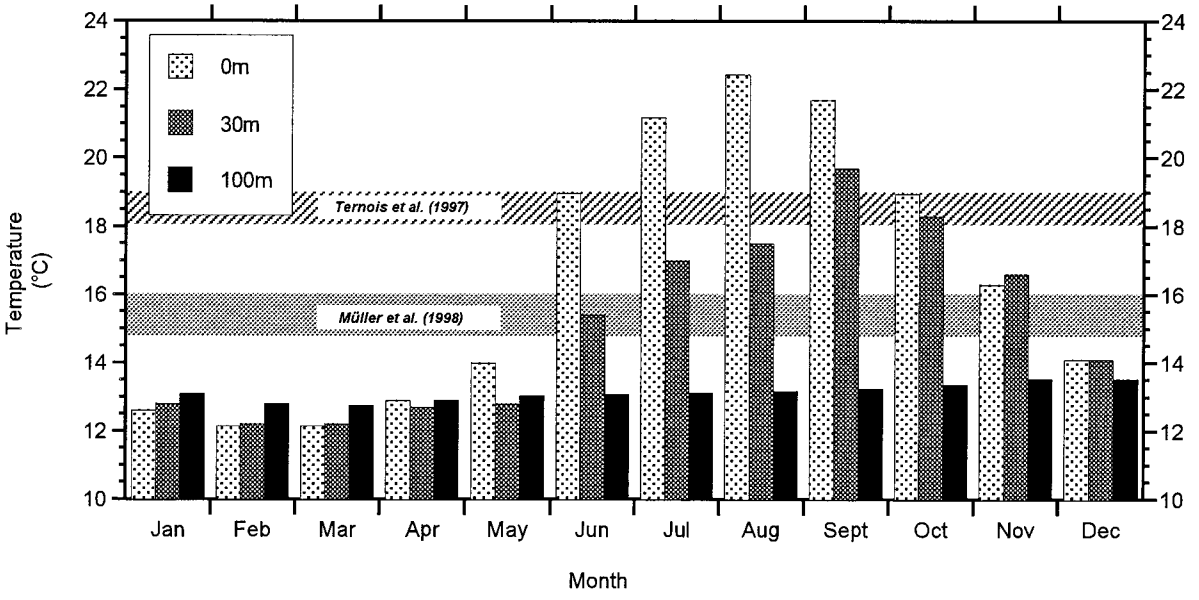

Fig. 4. Monthly mean water temperatures at different depths for the studied stations (0, 30 and 100 m; Levitus and Boyer, 1994). The shaded horizontal bars indicate the sedimentary $U^{K'}_{37}$ temperature estimates obtained with the Müller et al. (1998) and Ternois et al. (1997) equations.

In this case, the discrepancies between water column and sediments may be related to seasonal changes of Haptophyceae species in this area. However, the organic remains of the species or strains responsible for the SST deviations may not be efficiently transported to the underlying sediments since the sedimentary alkenone composition is reflecting the annual average temperatures recorded in the Levitus dataset (Levitus and Boyer, 1994).

5. Conclusions

A close agreement between spring suspended particulate C_{37} alkenone concentrations and water column temperatures is found in the northwestern zone of the Mediterranean Sea when the equation of Ternois et al. (1997) is used for calibration. This correspondence significantly deviates from the general equation regularly observed in most world-wide marine areas. However, the sedimentary C_{37} alkenones exhibit a relationship with annual average 0–40 m water column temperatures in agreement with the general equation (Müller et al., 1998). Application of the Ternois et al. (1997) equation to the C_{37} alkenone sedimentary composition provides temperature estimates that are in accordance with the "warm end" of the water column temperatures of this region.

This discrepancy may be related to seasonal changes of Haptophyte species or to other alkenone-synthesizing algae permanently living in the water column whose organic remains are not efficiently transported to the underlying sediments. Further insight into this question may be obtained from water column measurements covering the whole annual cycle, particularly in autumn which seems to be a main season for Haptophyte productivity.

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