INTEGRATED ORGANIC GEOCHEMICAL-PALYNOLOGICAL RESEARCH FOR THE STUDY OF ABRUPT CLIMATE VARIABILITY IN THE NORTH ATLANTIC:

Preliminary results from the Western Mediterranean Region

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1. Introduction

Organic molecules in marine sediments are widely applied as geochemical tools in paleoclimatic studies, since they revealed to be very useful proxies of past oceanic and climatic changes (e.g. Brassell *et al.,* 1986; Villanueva *et al.,* 1997a and 1998). The most successful application of these biomarkers is the use of C_{37} alkenones for the estimation of sea surface temperatures (SST) through the ratio between di and triunsaturated congeners that is compiled in the form of a $U^{K'}_{37}$ index (Brassell *et al.,* 1986; Prahl & Wakeham, 1987). High resolution SST records based on this alkenone method have provided significant information on climate variability in different oceanographic regions at glacial-interglacial time scales (Villanueva *et al.,* 1998; Pelejero *et al.,* 1999a; Calvo *et al.,* 2001a; Rostek *et al.,* 1997). Thus, $U^{K'}_{37}$-SST records showed the occurrence of abrupt climatic variability in the last glacial period at much lower latitudes and at different regions (Cacho *et al*., 1999; Pailler & Bard, 2002; Sachs & Lehman, 1999) than in Greenland ice and northern North Atlantic marine sediments (Dansgaard-Oeschger and Heinrich events; Dansgaard *et al*., 1993; Bond *et al*., 1993; Heinrich, 1988). Furthermore, the concentrations or fluxes of C_{37} alkenones have been proposed to be indicators of paleoproductivity of Haptophyceae (e.g. Schubert *et al.,* 1998; Villanueva *et al.,* 2001).

Nevertheless, there are many more organic molecules which can provide interesting information, such as C_{23}-C_{33} *n*-alkanes and C_{22}-C_{28} *n*-alkan-1-ols, which are major lipid components of epicuticular waxes of higher plants (Eglinton & Hamilton, 1967; Tulloch, 1976). These compounds were used as indicators of continental material inputs into the marine environments, either by fluvial discharges (Prahl *et al.,* 1994; Pelejero *et al.,* 1999b), aeolian transport (Calvo *et al.,* 2001b; Gagosian & Peltzer, 1986; Ohkouchi *et al.,* 1997; Poynter *et al.,* 1989) or ice-transported debris (Villanueva *et al.,* 1997a; Martrat *et al.,* 2003). Application of these terrestrial biomarkers to paleoceanographic studies is useful for a more comprehensive description than that only based on marine proxies, such as $U^{K'}_{37}$-SST and foraminifera-derived records (e.g. Calvo *et al.*, 2001b; Cacho *et al.,* 2000). In a previous study in the North Atlantic comparing $U^{K'}_{37}$-SST and *n*-alkane records at latitudes of 37ºN (MD95-2037 core) and 43ºN (SU90-08 core), changes in concentration of ice-transported *n*-alkanes provided a very important clue for the interpretation of the $U^{K'}_{37}$-SST differences between sites, showing that the polar front was displaced at lower latitudes during stage 2 than during stage 6 (Calvo *et al.,* 2001b).

In this respect, studies on pollen-rich marine cores provide an excellent opportunity for the development of land-sea correlations, allowing strong minimization of the age-scale uncertainties that are currently encountered in direct comparison of marine and continental paleoclimatic records (Sánchez Goñi *et al.,* 1999 and 2000a; Shackleton *et al.,* 2003). Recent studies involving direct correlations of pollen records with alkenone SST estimations, both measured in a marine core from the Alboran Sea, showed abrupt transformations of the whole vegetation cover over the South Iberian Peninsula paralleling abrupt SST changes in the Western Mediterranean (Sanchez-Goñi *et al.*, 2002). Another study combining biomarkers and pollen related lower *n*-alkane and pollen concentrations with higher dust flux during glacial times (Zhao *et al.,* 2003), evidencing a decrease of the density vegetation cover in the North West Africa, due to climate aridity.

This approach is undertaken here for sediments from the Alboran Sea (core ODP-977A), where terrestrial proxies (*n*-nonacosane, *n*-hexacosanol and pollen concentrations) and marine data ($U^{K'}_{37}$-SST and C_{37} alkenone concentration) are compared. This core was chosen because it recorded interesting abrupt $U^{K'}_{37}$-SST oscillations for the last 250 Kyr (Martrat *et al.,* 2004). In addition, higher *n*-alkane and *n*-alkan-1-ol concentrations were found in glacial than in interglacial periods (Fig. 2). These differences could reflect an increase of terrestrial material inputs during glacial times due to higher transport efficiency rather than increases in vegetation development. To test this hypothesis, selected sections of the core from the last two glacial/interglacial cycles, corresponding to significant $U^{K'}_{37}$-SST changes, have been analysed for sporo-pollen concentrations in the frame of the EAOG fellowship during a three months stage at the *Département de Géologie et Océanographie (Université Bordeaux I*). The preliminary sporo-pollen concentration records from this ODP Site had revealed that this marine core was suitable for pollen determinations and that development of these combined marine-terrestrial approaches was useful for paleoclimatic studies.

2. Environmental setting: present-day oceanographic conditions, climate and vegetation

The Alboran Sea is the westernmost basin of the Mediterranean Sea. North Atlantic Surface Water enters through the Strait of Gibraltar and after partial modification (Modified Atlantic Water), crosses the Alboran Sea, describing two anticyclonic gyres, the Western and the Eastern Alboran Gyres (Fig. 1) (Perkins *et al.,* 1990). The Mediterranean Outflowing Water (MOW) is formed mainly by the Levantine Intermediate Water (LIW) and, episodically, part of the Western Mediterranean Deep Water (Parrilla *et al.,* 1986; Millot, 1999). This water circulation system is determined by the climatic conditions over the Mediterranean region. Excess of evaporation over precipitation and river runoff generates the dense water masses that drive the anti-estuarine circulation in this semi-enclosed sea (Béthoux, 1979 and 1980; Lacombe *et al.,* 1981). The strong Azores anticyclone dominates the climate of the western Mediterranean in summer whereas southward migration of European depressions generates high instability and frequent incursions of north westerlies in winter (Sumner *et al.,* 2001).

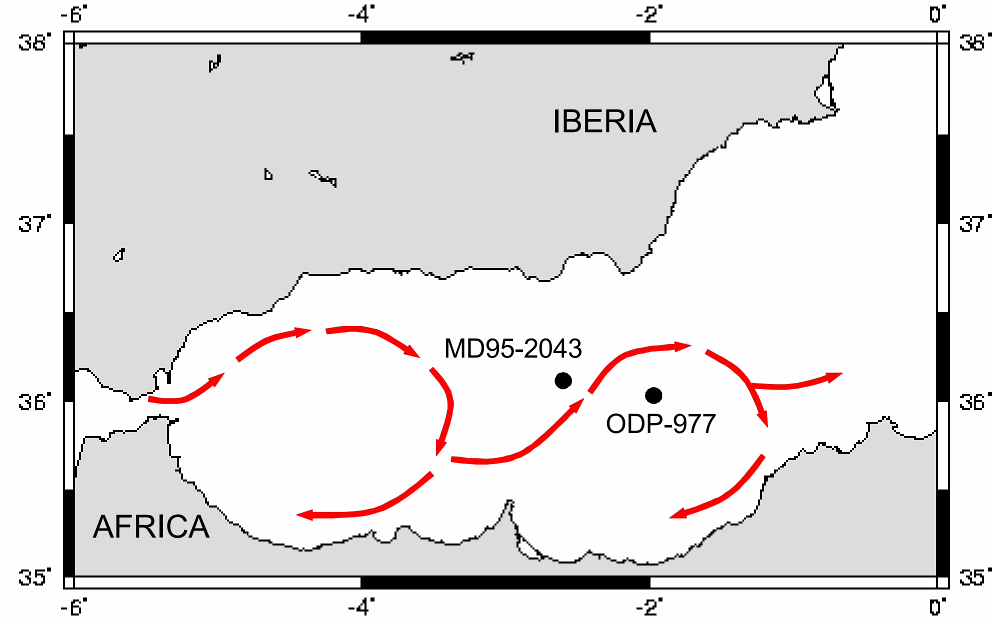

Figure 1 Location of the core studied (ODP Site 977A). The other Mediterranean core mentioned in the text is also shown (MD95-2043)

The Mediterranean climate that presently dominates the studied area is characterized by mild winters and hot and dry summers (Rivas-Martínez, 1987). These conditions promote the development of Sub-Mediterranean and Mediterranean vegetations, dominated by the evergreen oak forest (*Quercus rotundifolia*) that colonise the southern part of Iberia (Blanco Castro *et al.*, 1997). *Quercus rotundifolia* grows with thermo-Mediterranean species, such as

Olea europea ssp. *sylvestris*, *Chamaerops humilis, Pistacia lentiscus* and others, in the littoral zones (up to 1000 m a.s.l.), and with deciduous trees, such as *Quercus pyrenaica, Q. faginea, Taxus baccata* among others, in the Betic mountain chain (1000-2000 m a.s.l.). Altitudes between 2000 and 2900 m a.s.l. are occupied by *Pinus sylvestris* and some species of *Juniperus.* Pasturelands (*Festuca clementei, Artemisia granatensis*…) are found above 2900 m a.s.l., in the coldest hilltops of Sierra Nevada (Andalusia, South Spain) (Martínez Parras & Peinado Lorca, 1987).

3. Material and methods

Selected sediment samples from Site 977A were analysed for the present work. The core was recovered during leg 161 of the Ocean Drilling Program and it is located south of Cabo de Gata (Spain), in the eastern Alboran Basin, (36º1.9'N, 1º57.3'W; 1984 m water depth) (Fig. 1).

The analytical procedure for determining *n*-alkanes, *n*-alcan-1-ols and C_{37} alkenones is described in detail in Villanueva *et al.* (1997b). Briefly, sediment samples were freeze-dried and manually grounded. After addition of an internal standard containing *n*-nonadecan-1-ol, *n*-hexatriacontane and *n*-tetracontane, ca. 2 g of dry sediment were extracted with dichloromethane in an ultrasonic bath, followed by saponification with 6% potassium hydroxide in methanol. The lipidic compounds were recovered with hexane which was then evaporated to dryness under a N_2 stream. Finally, the extracts were redissolved with toluene, derivatized with bis(trimethylsilyl)trifluoroacetamide and analyzed by gas chromatography with a flame ionization detector.

Samples for pollen determination were prepared following the procedure described in de Vernal *et al.* (1996). A known number of exotic *Lycopodium* spores were added and the samples were chemically treated (cold 10%, 25% and 50% HCl, cold 48% and 70% HF). Afterwards, they were sieved through 10µm nylon mesh screens. The final residue for pollen analysis was mounted unstained in double distilled glycerine. Pollen grains were counted using a Zeiss Axioskop light microscope at x400 and x1000 (oil immersion) magnifications. A minimum of 100 pollen grains, excluding *Pinus* and spores, were counted in nearly all samples.

4. Results

Concentrations from conifer and non-conifer pollen grains, fern spores as well as total sporo-pollen concentration are shown in Figure 2, where they are compared with $U^{K'}_{37}$-SST, *n*-nonacosane and *n*-hexacosanol records. Sporo-pollen concentration values range between 115 and 23160 grains/cm^3 (conifer), 77 and 12875 (non-conifer), 0 and 60 (fern spores), resulting in total sporo-pollen concentration between 192 and 27249 grains/cm^3.

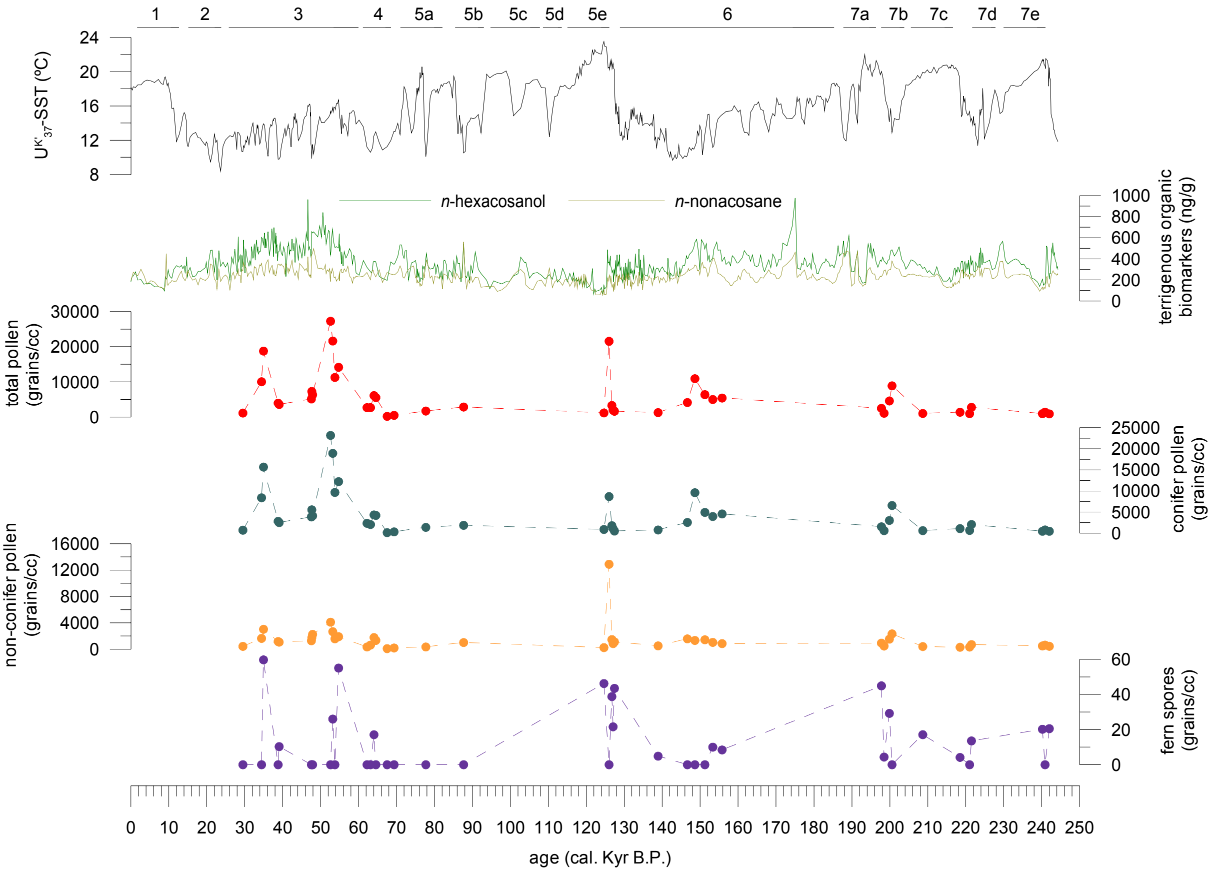

Figure 2 $U^{K'}_{37}$-SST, concentrations of *n*-nonacosane, *n*-hexacosanol, total sporo-pollen, conifer and non-conifer pollen grains and fern spores. Marine Isotopic Stages (1-7) are also indicated at the top of the graph (Martrat *et al.,* 2004)

In general, conifer and non-conifer pollen concentrations display a similar pattern during the last two glacial periods. Conifer pollen exhibits concentration values up to seven times higher than non-conifer, except in the transition to substage 5e, where non-conifer pollen double conifer concentration (Fig. 2). In contrast, the fern spore concentration exhibits a different profile (Fig. 2). In order to illustrate better the difference between glacial and interglacial periods, the concentrations of *n*-hexacosanol, *n*-nonacosane, C_{37} alkenones, total sporo-pollen and fern spores are shown in Figure 3, together with the $U^{K'}_{37}$-SST profile, during Marine Isotope Stages (MIS) 3-5 (Fig. 3a) and MIS 6-7 (Fig. 3b).

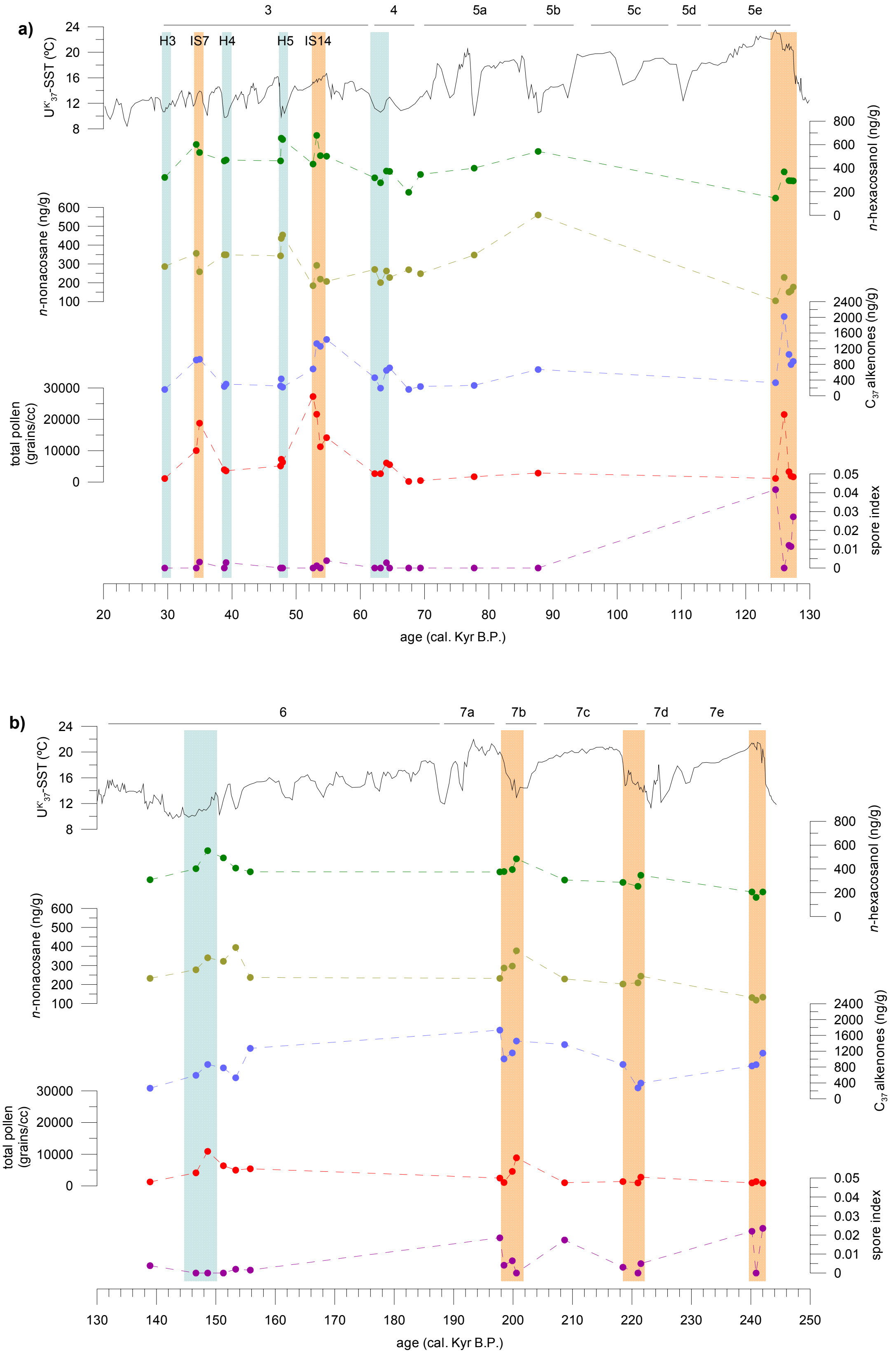

Figure 3 U$^{K'}_{37}$-SST and concentrations of *n*-hexacosanol, *n*-nonacosane, C_{37} alkenones, total sporo-pollen and spore index (fern spores / conifer + non-conifer), in the last glacial (MIS 3-5, Fig. 3a) and in the penultimate glacial cycles (MIS 6-7, Fig. 3b). Shaded bars indicate selected events in which sporo-pollen concentrations have been discussed.

Higher total sporo-pollen concentrations are observed in glacial than interglacial periods (Fig. 3). MIS 6 shows values around 5200 grains/cm^3. In the transition to the coldest event, at around 148 Kyr, the concentrations decrease, after reaching a maximum of 10900 grains/cm^3 (Fig. 3b). During the second part of MIS 4 (Fig. 3a), total sporo-pollen concentration also decreases from values of 5800 grains/cm^3 to around 2670 grains/cm^3. Higher concentration variability is found in stage 3 than in this previous period. Average values are around 10870 grains/cm^3. However, total sporo-pollen concentrations are in the order of 20450 grains/cm^3 during interstadial episodes, reaching relative minima at cold events (ca. 4560 grains/cm^3) (Fig. 3a).

Interglacial periods have the lowest total sporo-pollen concentration among the samples studied: around 2100 grains/cm^3 and 1600 grains/cm^3 in MIS 5 and 7, respectively (Fig. 3). Concentration maxima coincide with relative low $U^{K'}_{37}$-SST in the transition to substage 5e (21540 grains/cm^3, 20.5ºC, Fig. 3a) and in substage 7b (8870 grains/cm^3, 12.9ºC, Fig. 3b). They then diminish when $U^{K'}_{37}$-SST increase.

Comparison of total sporo-pollen concentration with *n*-nonacosane and *n*-hexacosanol records (Fig. 3) indicates better correlation with the latter. Anyway, both kind of terrigenous biomarkers exhibit a good parallelism during the last 250 ka. C_{37} alkenones and total sporo-pollen profiles describe a similar pattern, with major pollen peaks coinciding with higher alkenone values.

Interestingly, the records of fern spores exhibit higher concentrations during the warm periods (interglacials and D/O interstadials) and the lower values are found in the cold episodes, even during abrupt coolings of interglacial periods (Fig. 2). Comparison with terrigenous biomarkers displays no defined correlation pattern.

Ferns are preferentially stream-borne dispersion plants (Turon, 1984; Cour, 1974). Their spores had previously been measured together with other herbaceous hydrophilous taxa, like Cyperaceae, as markers of surface runoff and/or fluvial transport in cores off Portugal and off the Ivory Coast (Lézine and Denèfle, 1997). In the present work, in order to avoid sediment rate effects a spore index has been calculated as follows:

$$\text{Spore index} = \frac{\text{fern spore concentration}}{\text{conifer concentration + non-conifer concentration}}$$

The observed index values range between 0 and 0.042 (Fig. 3). They show lower variability than the spore concentration record (Fig. 2) during MIS 3. Higher values indicate significant fluvial transport, whereas a decrease in the fern spore concentration (lower index values) points to drier climatic conditions and enhanced aeolian transport (Lézine and Denèfle, 1997).

5. Discussion

Although the rivers that enter in the Alboran Sea have relative small, arid and abrupt catchment areas, the torrential rain regime that characterizes the region may provide large amounts of riverine material. Recently, the composition of particle fluxes from sediment traps in the Alboran Sea has been analysed (Fabres *et al,* 2002). The lithogenic component represented 68 to 76% of the collected particulate material. Comparing with maximum values of Saharan dust recorded in southern Spain (Diaz Hernandez & Miranda Hernandez, 1997), less than 12% of these lithogenic particles correspond to aeolian inputs. Moreover, recent sedimentological and geochemical studies performed in a nearby core (MD95-2043, Fig. 1) suggested that fluvial supply was the main source of detrital particles during the last glacial period (Moreno *et al,* 2002). Therefore, it can be hypothesized that most terrigenous organic matter (including pollen, *n*-alkanes and *n*-alcan-1-ols) sedimented in Site 977A during the studied period was fluvial transported.

On the other hand, north westerlies are the prevailing winds in the studied region whereas southern Saharan winds occur sporadically under specific climatic conditions (Rodríguez *et al,* 2001). Pollen and terrigenous biomarkers analysed in ODP-977A core essentially represent the vegetation from the south of the Iberian Peninsula.

Pinus pollen often is over-represented in marine sediments (e.g. Turon, 1984) and tends to mask the variation of other taxa. For this reason it is currently removed from pollen percentages and in the application of transfer functions to marine pollen records, e.g. Sánchez Goñi *et al.* (2002). However, in view of the observed good correlation between conifer (*Pinus* + *Cedrus*) and non-conifer pollen data (Fig. 2) in the present study total sporo-pollen concentration has been considered to be an indicator of the whole vegetation of the area. Additionally, the parallelisms in the variability of this proxy with *n*-nonacosane and *n*-hexacosanol records (Fig. 3) suggest that similar vegetation sources were responsible for both records. Variations in total sporo-pollen concentration and terrigenous biomarkers have therefore been interpreted in terms of changes in cover vegetation and/or transport efficiency.

5.1. Glacial periods

MD95-2043 core (Fig. 1) has provided good high resolution records for Marine Isotopic Stage 3 in Alboran Sea allowing the identification of the abrupt climatic oscillations (e.g. Cacho *et al.,* 1999; Moreno *et al.,* 2002; Pérez-Folgado *et al.,* 2003). Atmospheric circulation and surface and deep Mediterranean oceanography determined precipitation patterns, vegetation development and marine productivity during HE and D/O stadials and interstadials (Cacho *et al.,* 2000; Sánchez Goñi *et al.,* 2002; Moreno *et al.,* 2004).

In Site 977A (Fig 3a), the terrigenous markers are coherent with the MIS 3 climatic conditions described in marine pollen studies from the Mediterranean region and the Western Iberian margin (Combourieu-Nebout *et al.,* 2002; Roucoux *et al.,* 2001; Sánchez Goñi *et al.,* 2000b and 2002) and in geochemical analyses in Alboran Sea sediments (Moreno *et al.,* 2002; Moreno *et al.,* 2004). The higher sporo-pollen concentration values found in the interstadials could be associated to increasing cover vegetation due to more humid conditions. Conversely, the lower values registered in HE are related to colder and drier climate (e.g. Sánchez Goñi *et al.,* 2002). Both the relatively high fern spore index and the concentration of these spores suggest an increase in fluvial discharges during the onset of D/O interstadials linked to wetter conditions (Fig. 2 and 3a). These results are consistent with the changes in C_{37} alkenone concentrations that reflect higher marine productivity during the interstadials than during the HE (Fig. 3a) which can be attributed to surface water fertilization resulting from higher river runoff (Moreno *et al.,* 2004). Having in mind these observations the results of the last two climatic cycles may be interpreted in similar terms as MIS 3.

In the Marine Isotope Stage 6, the $U^{K'}_{37}$-SST record exhibit a prolonged cold episode that is maintained for nearly 10 Kyr, between 150 and 140 Kyr (Fig. 2 and 3b). During this episode, the concentrations of sporo-pollen, *n*-nonacosane and *n*-hexacosanol reach a maximum and then decrease suggesting a reduction of the vegetation cover and/or the material transported. Minimum values in the fern spore index suggest low fluvial transport (Fig. 3b). Low pollen and higher plant *n*-alkane concentrations have been documented in northwest Africa during dry glacial periods (Zhao *et al.,* 2003) reflecting sparse vegetation canopy due to continental aridity. In the Mediterranean region, terrestrial pollen records have also evidenced a reduced forest development during MIS 6 (Tzedakis *et al.,* 2003). However, in ODP-977 higher values of terrigenous proxies during the transition to the colder event, when fluvial transport is low, may reflect a higher eolian transport likely due to a strengthening of the wind system rather than increases of vegetation cover.

Thus, the decrease of terrestrial inputs during the second part of MIS 4 (Fig. 3a) could be related to the change to MIS 3 climatic conditions. The spore index also shows lowest values during this period indicating less fluvial transport.

Both in MIS 6 and 4, the increase in marine productivity evidenced by higher C_{37} alkenone concentration parallels higher amounts of the terrigenous biomarkers (Fig. 3) suggesting fertilization from terrestrial material inputs or increased water column mixing by higher winds.

5.2. Interglacial periods

During Termination II, the $U^{K'}_{37}$-SST record shows a decrease of 1ºC, at about 126 Kyr B.P. (Fig. 3a) that is paralleled by increases in the concentration of *n*-nonacosane and *n*-hexacosanol and maxima of sporo-pollen concentration (Fig. 3a). A similar climatic oscillation has been observed in mid-latitude marine cores located off Portugal (MD95-2042) and in the Alboran Sea (ODP-976) prior to the establishment of clear interglacial conditions (Sánchez Goñi *et al.,* 1999; Levi, 1999). In MD95-2042 this Younger Dryas-like event is characterized by cooler and drier conditions reflected in a slight interruption of the arboreal development and the persistence of semi-desert species as well as a peak in *Bitectatodinium tepikiense*, a dinocyst associated to cool waters (Sánchez Goñi *et al.,* 1999). Although Mediterranean forest was developed during Termination II, the vegetation cover was not much dense and the arboreal community was consequently more sensible to any environmental change. The drastic reduction of the spore index, indicating a decrease in fluvial transport along with the increase of terrestrial proxy concentrations, (Fig. 3a) would also imply an enhancement of the aeolian transport related to stronger atmospheric circulation in this short episode.

During the three temperate substages of Marine Isotopic Stage 7 (7e, 7c and 7a), similar forest development occurred in the Mediterranean region, as indicated by arboreal pollen values of Tenaghi Philippon record in northeast Greece (Tzedakis *et al.,* 2003). This pollen sequence also shows that the largest forest expansion of this interglacial period, with highest abundance of thermophilous taxa, corresponds to substage 7c. It seems that tree populations did not undergo a drastic reduction during the substage 7b, as it happened in 7d (Tzedakis *et al.,* 2003). This forest persistence is also observed in the pollen record from Valle di Castiglione in Italy (Follieri *et al.,* 1988), suggesting that the moisture conditions of existing during substage 7c continued during the following colder period.

In the Alboran Sea, low or decreasing concentrations of terrestrial proxies are found in the transitions to warmer substages (7e, 7c, 7a) (Fig. 3b), and they are partly associated to reduced terrestrial material transport. Maximum sporo-pollen and terrigenous *n*-alkane and *n*-alkan-1-ol concentrations were observed in substage 7b in association with low spore index values (Fig. 3b). This trend may reflect in part an enhancement of aeolian transport and in part higher biomass production during this period, in comparison to substage 7d, as documented in Tenaghi Philippon (Tzedakis *et al.,* 2003).

Finally, the increase in terrestrial material recorded by the higher amounts of sporo-pollen and higher plant *n*-alkanes and *n*-alkan-1-ols during the transition to MIS 5 and MIS 7b had likely provided nutrients to Alboran waters increasing marine productivity, as indicated by the maxima of C_{37} alkenone concentration following the terrigenous proxies (Fig. 3a and 3b).

6. Summary

In this pilot study, the combined palynological and biomarker analyses have provided substantial for the understanding of the climatic evolution of the Alboran Sea during the last two glacial and interglacial periods. Joint comparison of total sporo-pollen concentration, higher plant *n*-alkanes and *n*-alkan-1-ols and $U^{K'}_{37}$-SST has shown the occurrence of abrupt changes of the proxies recording terrigenous vegetation inputs to the marine environment during the transitions from colder or warmer periods.

The preliminary results of this work points to the eolian/fluvial transport efficiency rather than density vegetation cover as the main factor controlling terrestrial material delivering to Alboran Sea in the climatic transitions studied. Both spore concentration and spore index as complementary indicator of different transport patterns support this hypothesis.

Identification of the plant species contributing to the sporo-pollen concentration and higher plant *n*-alkanes and *n*-alkan-1-ols will allow more comprehensive interpretation of the observed abrupt changes in terrestrial proxies associated to vegetation and climatic changes. The identification of the species is expected to be carried out as part of a post-doctoral project in the D.G.O. in 2005.

Acknowledgements

I am very grateful to Maria Fernanda Sánchez Goñi for introducing me to “the palynology world”, for helpful discussions and improvement of this manuscript. I thank people from the D.G.O. who made my stage very pleasant. M.-H. Castera is thanked for preparing the pollen samples and B. Martrat for providing samples and biomarkers data. I also thank my Ph.D. supervisor, J.O. Grimalt, for helpful comments that have improved this report and for his support to this small research work which complements part of my Thesis. EAOG is greatly acknowledged for providing the fellowship that allowed the stage during summer 2004.

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Appendix

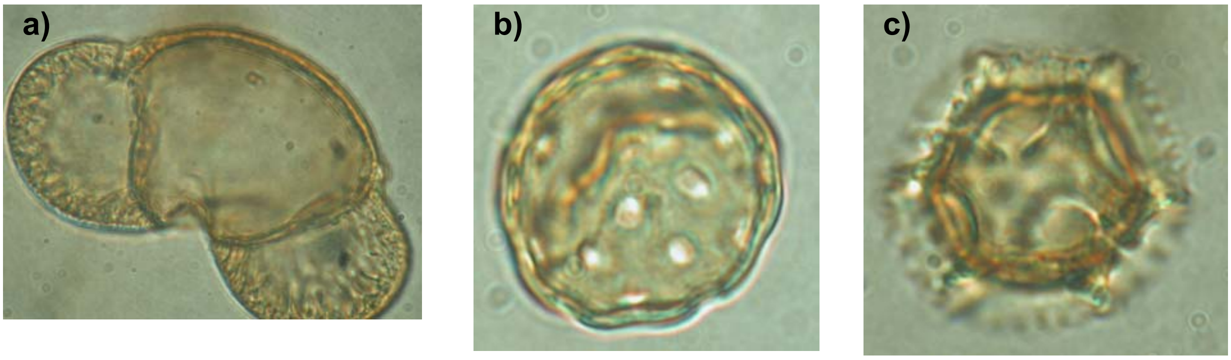

Plate I Photographs of different pollen taxa found in core ODP-977A, performed with a Nikon COOLPIX 995 digital camera: **a)** conifer (*Pinus*), **b)** non-conifer (*Chenopodiaceae*), **c)** non-conifer (*Taraxacum* type)