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Tracking atmospheric and riverine terrigenous supplies variability during the last glacial and the Holocene in central Mediterranean

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Abstract

The objectives were to retrace the eolian and fluvial terrigenous supplies in a sediment core from the Sicilian-Tunisian Strait by coupling mineralogical, grain-size and geochemical approaches, in order to get informations on the atmospheric versus riverine contributions to sedimentation on the southern side of central Mediterranean since the last glacial. The eolian supply is dominant over the whole interval, excepted during the sapropel S1 when riverine contribution apparently became significant, and particles provenance has been modified since Last Glacial. Saharan contribution increased during the Bølling-Allerød, evidencing the persistence of aridity over North Africa although the northern Mediterranean already experienced moister and warmer conditions. The Younger Dryas is marked by proximal dust inputs highlighting intense regional eolian activity. A southward migration of dust provenance toward Sahel occurred at the onset of the Holocene, likely resulting from a southward position of the Inter Tropical Convergence Zone, probably associated with a large-scale atmospheric reorganization. Finally, a peculiar high terrigenous flux associated with drastic modifications of the mineralogical and geochemical sediment signature occurred during the sapropel S1, suggesting the propagation of fine-particles derived from major floodings of the Nile River – resulting from enhanced rainfall on northeastern Africa – and their transportation across the Sicilian-Tunisian Strait by intermediate water-masses.

1 Introduction

The Mediterranean is a transitional area where northern and southern climatic influences tightly interact (e.g. Magny et al., 2009). Previous studies revealed that moist conditions developed in the Mediterranean during the early Holocene while a progressive orbitally-driven trend to aridification characterized the mid and late Holocene. This climatic evolution displays a contrasting regional pattern, with an abrupt transition at 5.7 kyr on the western Mediterranean and a more gradual transition in the eastern

Mediterranean (Cheddadi et al., 1991; Ariztegui et al., 2000; Magny et al., 2002, 2007a; Frigola et al., 2007; Tzedakis, 2007; Sadori et al., 2008; Roberts et al., 2008, 2011; Jalut et al., 2009; Peyron et al., 2011). Precipitation estimations based on lake levels, fire and pollen association, and on speleothem and isotope records, also provide evidences of contrasting seasonality across the Mediterranean during the Holocene (Magny et al., 2003, 2007, 2009; Zanchetta et al., 2007; Tzedakis, 2007; Roberts et al., 2008; Peyron et al., 2011; Vannière et al., 2011), but estimating the respective atmospheric and oceanic control on Mediterranean climatic evolution through their impact on eolian and fluviatile systems is still complex. A multiproxy study of the terrigenous supply would help retracing the variability of both eolian and fluviatile systems.

The nature and provenance of fine-grained terrigenous particles in the Mediterranean is mainly controlled by the balance between riverine supplies driven by precipitation regime on the surrounding continents and eolian supplies from the Sahara (Bergametti et al., 1989b; Matthewson et al., 1995; Guerzoni and Chester, 1996; Foucault and Mélières, 2000; Goudie and Middleton, 2001). Several studies used deep-sea clay mineral associations as tracers of source regions and as indicators of water mass fluctuations (Chamley, 1975; Petschick et al., 1996; Fagel et al., 1997, 2006; Gingele et al., 2001; Liu et al., 2003; Boulay et al., 2005; Colin et al., 2010). Indeed, the mineralogical nature of sediments, which depends on the petrographic characteristics of their source areas (e.g., Bout-Roumazeilles et al., 1999; Sionneau et al., 2008), has been used to retrace detrital particles provenance in the Mediterranean and thus to assess the respective eolian and riverine contributions to deep-sea sedimentation and provide valuable information on the sediment propagation pathways (Caquineau et al., 1998, 2002; Bout-Roumazeilles et al., 2007; Erhmann et al., 2007a; Hamann et al., 2009; Kandler et al., 2009; Formenti et al., 2011a, 2011b). The clay mineral fraction also provides information on climatic conditions, such as precipitation and runoff patterns over the adjacent continents (Chamley, 1989; Montero et al., 2009, 2010), as well as on the dynamics of river inputs (Pinsak and Murray, 1960). The geochemical signature of clays would complementary help characterizing the main sources and evidencing specific transportation patterns or transfer processes (Haug et al., 2001; Kandler et al., 2009; Formenti et al., 2011a, b). Coupling clay mineral and grain-size studies would help investigating the sedimentary transfer regime because sediment grain-size distribution, which is primarily driven by sedimentary processes, reflects transportation conditions (Ehrmann et al., 2007; Montero et al., 2009; Sionneau et al., 2010). In this frame, combining clay mineralogy with grain-size analyses and geochemical tracers would allow retracing significant variations of detrital supply in the Siculian-Tunisian strait and inferring any major modifications of both atmospheric and oceanic terrigenous transfers patterns since the last deglaciation.

2 Geographical settings

2.1 Oceanic circulation

The Mediterranean Sea is a concentration basin where evaporation exceeds precipitation plus freshwater discharge. The surface Atlantic water entering from Gibraltar Strait transforms into Modified Atlantic Water (MAW) as it flows eastward (Fig. 1). The eastern Mediterranean surface water displays a counterclock wise circulation (Pickard and Emery, 1982; Pinardi and Masetti, 2000). The Levantine Intermediate Water (LIW) forms in the northwest Levantine basin (Fig. 1) through convection cells at mid-depth during winter (Lascaratos et al., 1998). The LIW flows westward in sub-surface layers (∼ 150 to 600–800 m) towards the northernmost Adriatic Sea and western Mediterranean (Fig. 1) via the relatively shallow Siculo-Tunisian Strait. The dense Eastern Mediterranean Deep Water (EMDW) – formed in the Adriatic Sea (Fig. 1) and occasionally in the Aegean Sea – and Western Mediterranean Deep Water (WMDW) – formed in the Gulf of Lions – fill the deep basin (< 800 m depth) (Wüst, 1961; Pickard and Emery, 1982; Malanotte-Rizzoli and Hodt, 1988; Klein et al., 1999). The Mediterranean Intermediate Water represents up to 80 % of the Mediterranean Outflow (MOW) into the Atlantic Ocean (Bryden and Stommel, 1984). This general pattern is highly dependent on environmental conditions including eolian activity and precipitations distribution and recent alteration of the ocean/atmosphere coupling has resulted in enhanced deep-water formation in the Aegean Sea (Malanotte-Rizzoli et al., 1999).

2.2 Present-day river supplies

At present day, a major part of detrital clays is supplied to the Mediterranean via rivers (Fig. 1), the most important being the Nile River (from 120 to $230 \times 10^6\,\mathrm{t\,yr^{-1}}$) discharging in the eastern Mediterranean. The Po River, discharging into the Adriatic Sea ($17 \times 10^6\,\mathrm{t\,yr^{-1}}$) and southeastern European rivers associated with Turkish rivers, providing about respectively $30 \times 10^6\,\mathrm{t\,yr^{-1}}$ and $17 \times 10^6\,\mathrm{t\,yr^{-1}}$ to the Aegean Sea, are some major contributors to sedimentation into the central Mediterranean (Holeman, 1968; Milliman and Syvitski, 1992; Stanley et al., 1992; Erhmann et al., 2007b; Garzanti et al., 2006; Hamann et al., 2009). The detrital supply through the Dardanelles Strait is reduced ($0.9 \times 10^6\,\mathrm{t\,yr^{-1}}$) because most sediment is trapped within the Black Sea and Marmara Sea (Erhmann et al., 2007a). Central Mediterranean is not affected by detrital supply from the Rhône River nor from the Ebro River which discharge into the western Mediterranean. Finally riverine contribution to the central Mediterranean from areas bordering the southern shores is negligible due to the narrow drainage basin and sparse rainfall (Martin and Milliman, 1997).

2.3 Eolian supply

The estimation of the present-day eolian contribution to deep-sea sedimentation varies from $3.9 \times 10^6\,\mathrm{t\,yr^{-1}}$ to $120 \times 10^6\,\mathrm{t\,yr^{-1}}$ (Bergametti et al., 1989c; Matthewson et al., 1995; Guernozi and Chester, 1996; Goudie and Middleton, 2001). This wide range of estimation reflects intermittent and seasonal variations of dust outbreaks toward Europe (D'Almedia, 1986; Guerzoni et al., 1997). Satellite imagery, back trajectories and observations indicate that fine particles originating from North Africa (Fig. 1) are transported by southerly/southwesterly winds (Scirocco, Ghibbli) toward the central

Mediterranean (Ganor et al., 2000; Goudie and Middleton, 2001; Washington et al., 2005; Engelstaeder et al., 2006; Israelevich et al., 2012). Arid and semi-arid regions of North Africa (Fig. 1) are the main areas of production of dust all-year long (Coudé-Gaussen, 1982; Pye, 1987): Tunisia and northern Algeria, Morocco and western Sahara, the South Algeria-Mali region, the Bodélé depression and the southern Egypt-northern Sudan (Brooks and Legrand, 2000; Caquineau et al., 2002; Israelevich et al., 2002; Prospero et al., 2002; Goudie, 2003; Formenti et al., 2011a). Most of the northward aerosols transportation across the Mediterranean is linked to the seasonal displacement of cyclones over the Mediterranean (Folger, 1970; Guerzoni et al., 1997; Moulin et al., 1997; Rodriguez et al., 2001). Maximum aerosol is observed over central and eastern Mediterranean in spring (Luck and Ben Othman, 2002; O'Hara et al., 2006) and summer when anticyclonic conditions initiates drought over the area (Prospero et al., 2002; Koren et al., 2006; Roberts, 2008; Israelevich et al., 2012). Although Saharan fine particles are mainly produced during warm and hydrolysis period, it is transported from paleosols and little consolidated formations during dry intervals when vegetation is reduced and eolian erosion more efficient (Brooks and Legrand, 2000). During summer, the Saharan depression favors the transport of dust over the western basin, progressively moving toward central Mediterranean (toward Corsica and Italy) at the end of summer (Bergametti et al., 1989a; Moulin et al., 1997; Barry and Chorley, 1998).

3 Materials and methods

3.1 Materials

The core MD04-2797CQ ($36^{\circ}57'$ N-$11^{\circ}40'$ E) was taken during the PRIVILEGE-PRIMAROSA cruise in 2004 at 771 m water-depth in the Sicilian-Tunisian Strait (Fig. 1). According to its geographical setting, the sedimentation of the core is likely influenced by both marine and eolian supplies. The scarcity of rivers along the North African margin, and their non-permanent characters prevents the area from any major and perennial fluviatile contribution (O'Hara et al., 2006). By contrast, the area is submitted to intense wind-driven supply all year long, carrying along with the dominant south-southwestern low-altitudes dusty winds (Scirocco) (Barry and Chorley, 1998) which reach the Adriatic Sea by crossing the Mediterranean (Fig. 1). According to the main wind system, the proximal source of dust for the Sicilian-Tunisian strait is likely the Tunisia-northern Algeria area (Coudé-Gaussen, 1987). The Mali-Algeria frontier or the Moroccan Atlas (Brooks and Legrand, 2000; Caquineau et al., 2002; Prospero et al., 2002; Goudie, 2003) could also contribute to sedimentation either during intense dust episode or when specific and/or seasonal atmospheric configuration affect the main wind directions (Bergametti et al., 1989a, 1989b; Ganor et al., 1991; Pye, 1992; Avila et al., 1997; Goudie and Middleton, 2001; Prospero et al., 2002; Washington et al., 2005).

3.2 Chronostratigraphy

The age model is based on 13 AMS 14C datations (Rouis-Zargouni et al., 2010, 2012). The ages were corrected using reservoir ages of 400 yr during the Holocene, the Younger Dryas and the late glacial, 560 yr during the Bølling-Allerød and 800 yr during the Heinrich event 1 and Older Dryas (H1-OD), following recommendations of Siani et al. (2001) (Table 1). The top of the core is dated at 668 ka cal. BP, suggesting that the sea-sediment interface was not preserved during coring. The Terrigenous Mass Accumulation rates (MART) were calculated as follow: MART (g kyr^{-1} cm^{-2} = Linear Sedimentation Rates (cm kyr^{-1}) ·dry density (g cm^{-3}) · (100-(%opal + %$CaCO_3$)).

3.3 Clay minerals

The analyses were performed according to the protocol described in Bout-Roumazeilles et al. (1999). All samples were first decalcified with 0.2 *N* hydrochloric acid. The excess acid was removed by repeated centrifugations. The clay-sized fraction

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(< 2 µm) was isolated by settling, and oriented on glass slides (oriented mounts). Three XRD (X-ray diffraction) determinations were performed using a Bruker D4 Endeavor coupled with Lynxeye detector: (a) untreated sample; (b) glycolated sample (after saturation for 12 h in ethylene glycol); (c) sample heated at 490 °C for two hours. Each clay mineral is then characterized by its layer plus interlayer interval as revealed by XRD analysis. Smectite is characterized by a peak at 14 Å on the untreated sample test, which expands to 17 Å after saturation in ethylene glycol and retracts to 10 A after heating. Illite presents a basal peak at 10 Å on the three tests (natural, glycolated, and heated). The illite crystallinity, or Kübler Index is based on the expression of the width of the illite peak at 10 Å and allows identifying the anchizone, the limit of diagenesis and the onset of the epizone. Chlorite is characterized by peaks at 14 Å, 7 Å, 4.72 Å and 3.53 Å on the three tests. Kaolinite is characterized by peaks at 7 Å and 3.57 Å on the untreated sample and after saturation in ethylene glycol. Both peaks disappear or are strongly reduced after heating. Palygorskite presents a basal peak at 10.34 Å, accompanied by a weaker peak at 6.44 Å, on both untreated and glycolated tests. The 10.34 Å peak collapses at 10 Å after heating (Brindley and Brown, 1980). The presence of palygorskite has been confirmed by MET observations of the palygorskite-rich samples. Semi-quantitative estimation of clay mineral abundances, based on the pseudo-voigt deconvolution for the doublets illite-palygorskite (10 Å–10.34 Å) and kaolinite-chlorite (3.57 Å–3.53 Å), was performed using the software MacDiff developed by Petschick (2001).

3.4 Grain-size

Grain-size analyses were performed on the carbonate- and opal-free fraction of the sediment using a Malvern Mastersizer 2000 laser (0.02–2000 µm) following protocols described in details in Montero et al. (2009). After defloculation, an aliquot of the sample was measured using 10 % ultrasonication in the Hydro S dispersion cell, once beam obscuration ranges between 12 and 15 %. The mode (i.e. most frequent grain-size in µm), the percentage of clay (< 2µm), cohesive silt (2–10 µm), sortable-silt (10–63 µm) and sand (> 63 µm) are reported.

3.5 XRF-scanning

Elemental abundances were measured on U-channels using the Avaatech core scanner from the EPOC laboratory at the University of Bordeaux. The sediment was protected with Ultralene@ X-ray transmission foil in order to avoid contamination. The data were acquired at a 30 s count time, using 10 kV voltage and 400 mA intensity. The results are expressed in counts per second, with a 2 % precision according to standard samples. Trace elements are normalized to Al (Tribovillard et al., 2008), in order to avoid dilution effect by carbonates.

4 Results

4.1 Clay mineralogy

The clay mineral fraction is composed of kaolinite (45 %), smectite (25 %) and illite (15 %). Chlorite and palygorskite are secondary components with 10 and 6 % of the clay content respectively (Table S1). The composition of the clay mineral assemblage displays some variations over the studied interval (Fig. 2). The kaolinite content remains slightly below 45 % between 18.5 and 11.7 ka. After what it increases and represents 50 % between 10.5 and 8.5 ka. The percentage of kaolinite decreases at 8.5 ka and remains low between 8 and 5.8 ka. It then increases progressively between 5.8 and 2 ka and reached its maximum (> 50 %) around 2 ka. The smectite remains stable between 18.5 and 8.5 ka excepted two maxima around 12 ka and 11 ka (Fig. 2). The content in smectite increases up to 38 % of the clay mineral fraction between 8.5 and 5.8 ka as the kaolinite content is minimum. This smectite-rich interval is interrupted by a slightly depleted level around 7.2 ka (Fig. 2). The smectite content decreases progressively since 5.8 ka down to 15 % at the top of the core. The illite content is maximum

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between 18.5 and 11.7 ka, then it drops down and remains low (< 15 %) between 11.7 and 5 ka. Then illite increases slightly and remains stable around 15 % over the more recent part of the record (Fig. 2). The Kübler Index is low (< 0.4°2θ) indicating high structural order when the content in illite is maximum. By contrast, the Kübler Index is higher (< 0.5°2θ) – low ordering- between 11.7 and 5 ka when the percentage of illite is reduced. The illite to kaolinite ratio (I/K) is characterized by a drastic change at 11.7 ka from values around 0.35 to values as low as 0.25. The chlorite content does not correlate with other clay species and shows slight variations, being higher than average between 18.5 and 15.5 ka. The palygorskite is less abundant between 8.5 and 5.8 ka, while smectite is maximum (Fig. 2).

4.2 Grain-size

The grain-size varies between 4 µm and 10 µm, indicating the detrital fraction is mainly composed of cohesive silt-size particles. Cohesive particles indeed represent 50 to 70 % of the total terrigenous fraction, being slightly less abundant between 18.5 and 11.7 ka and between 7.5 and 5.5 ka (Fig. 3). By contrast, the clay-size fraction is generally low (around 11 %), excepted between 8.5 and 5.5 ka when it represents up to 25 % of the detrital material. The sortable silt (10 µm to 63 µm) composes 25 % of the detrital fraction, being more abundant between 13 and 12 ka where it represents 45 %. The sand-size fraction is rare, but shows two maxima between 14 and 12 ka and between 8.5 and 7.5 ka (Fig. 3).

4.3 Elemental geochemistry

The Al content displays a decreasing trend from 18.5 to 14 ka (Fig. 4). The Ca to Fe ratio is used to distinguish areas dominated by carbonates (Sahara) from areas where hydrolysis is intense (alterites). The Ca/Fe ratio is characterized by large variations, with maximum values between 15 and 13 ka, and between 5 and 3 ka, while the ratio is minimum before 15 ka and between 2 and 1 ka. Some intervals are characterized by higher than average K/Al ratio, between 14 and 11.7, between 5.8 and 5.5 ka and between 2.5 and 1 ka. The two latter intervals are also characterized by the highest values of the Ti/Al ration (Fig. 4). The Zr/Al ratio displays a peculiar behavior, peaking at 13.4 ka, 7 ka, 6.4 ka, 1.3 ka and being high between 12.8 and 12.3 ka.

5 Discussion

5.1 Mineralogical and geochemical characterization of particle provenance and transport patterns

Illite and chlorite are abundant in sediments from the Ionian Sea (Fig. 1), where they mainly derive from the southern European rivers flowing into the Adriatic (Po River) and Aegean Seas (southeastern European Rivers) (Venkatarathnam and Ryan, 1971; Dominik and Stoffers, 1978; Chamley, 1989; Alonso and Maldonado, 1990; Tomadin, 2000). Smectite predominates in the eastern basin, being abundant in the Marmara Sea (Ergin et al., 2012; Armynot et al., 2012), in the Levantine basin (Hamann et al., 2009) and in the southeastern Aegean Sea (Erhmann et al., 2007a). Smectite characterizes the Nile River suspended loads, deriving from Cenozoic volcanic provinces of the Ethiopian Highlands (Ukstins et al., 2002; Hamann et al., 2009), and is further redistributed as suspended clay particles within the Mediterranean surface water (Venkatarathnam and Ryan, 1971; Foucault and Mélières, 2000; Hamann et al., 2009). Palygorskite has been shown to be abundant in tunisian dunes and is used as a main eolian tracer, because it may be transported over long-range distance, crossing the Mediterranean through meridian transfers (Coudé-Gaussen et al., 1982; Robert et al., 1984; Molinaroli, 1996). Illite is also a dominant component of eolian dust originating from Sahara when associated with palygorskite (Coudé-Gaussen and Blanc, 1985; Guerzoni et al., 1999; Foucault and Mélières, 2000; Goudie and Middleton, 2001), whereas illite associated with abundant kaolinite originate from southeastern areas of North Africa (Chester et al., 1977; Stanley and Wingerath, 1996; Foucault and Mélières, 2000; Goudie and Middleton, 2001; O'Hara et al., 2006; Hamann et al., 2009). The illite to kaolinite ratio (I/K) is a useful tracer of Saharan versus Sahelian dust provenance (Schütz and Sebert, 1987; Caquineau et al., 1998, 2002; Formenti et al., 2011a), which is thought to be conservative after long-range transport (Chester et al., 1972; Caquineau et al., 1998, 2002; Kandler et al., 2009). Highest ratio characterizes the northwestern areas (Fig. 1), whereas low ratio retraces the Sahelian belt: the lowest ratios are observed in dust collected in Sahel (0.1 to 0.3), intermediate values characterize south and central Sahara (0.3 to 0.7), moderate to high values are observed in Tunisia and northern Algeria (1 to 2) whereas highest ratios (1.3 to 2.6 up to 6) occur in northwestern Sahara (Fig. 1) and in Morocco (Chester et al., 1972; Glaccum and Prospero, 1980; Paquet et al., 1984; Coudé-Gaussen, 1991; Kiefert et al., 1996; Avila et al., 1997; Caquineau et al., 1998, 2002; Kandler et al., 2009). The I/K ratio also displays a longitudinal gradient (Fig. 1) with ratio decreasing from northwestern Africa (I/K = 2 to 1.1) toward northeastern Africa (I/K = 0.2 to 0.7) (Gomes et al., 1990; Ganor and Forer, 1996; Caquineau et al., 1998).

Geochemical data are used to pinpoint the origin of the terrigenous fraction and to retrace transportation processes. North and central Sahara are characterized by carbonate-rich soils, whereas the Sahelian area submitted to intense hydrolysis is depleted in Ca and enriched in Fe (Kandler et al., 2009; Formenti et al., 2011a, b). As a result, the Ca/Fe ratio is used to distinguish the Saharan and Sahelian provenance, complementary with the I/K ratio: Tunisia-northern Algeria source is characterized by high carbonate (up to 50 %) and low Fe contents (Chester et al., 1984; Bergametti et al., 1989), Moroccan Atlas has a high carbonate content (up to 70 %) and a rather high Fe content (Bergametti et al., 1989; Avila et al., 1997; Kandler et al., 2009), Libya displays intermediate Fe and carbonate contents (Guieu et al., 2002; O'Hara et al., 2006) whereas southern Algeria shows intermediate Fe content associated with low carbonate content (< 25 %) (Paquet et al., 1984; Bergametti et al., 1989; Alastuey et al., 2005).

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The grain-size of dust displays strong seasonal variations (Guerzoni et al., 1997), with a fine population (2 to 10 µm, mode around 2.7 µm) characterizing long-distance and high latitude transport and a coarser population (5 to 50 µm, mode around 20 µm) typical of dust storms carried over few km, being restricted to adjacent continental and marine areas (Prospero, 1981; Torres-Padron et al., 2002). Comparison between Ti/Al and Zr/Al ratios and grain-size distribution are thus used to identify specific eolian vs. riverine transport processes, associated with the development of sand dunes during the Holocene, or with remote fine-grained eolian outbreaks or coarse-grained fluvial supply (Haug et al., 2001; Martinez-Ruiz et al., 2003; Erhmann et al., 2007b; Hamann et al., 2009; Montero et al., 2009; Skonieczny et al., 2011).

5.2 Bølling-Allerød

The Bølling-Allerød (B-A) is marked by enhanced terrigenous supply starting at 14.6 ka, associated with increased contributions of both illite and palygorskite while kaolinite and chlorite decrease (Fig. 2). The relationship between illite content and sortable silt-size fraction (14 µm) indicate that illite is mostly transported through eolian processes. The concomitant increase of the Ca/Fe and I/K ratio suggests that the increased eolian supply is associated with a modification of the main detrital source (Fig. 5a). Indeed, increased proportion of Ca vs. Fe during the B-A may basically be interpreted as reflecting higher productivity but this hypothesis is not supported by the dinoflagellete record from core MD04-2797 (Rouis-Zargouni et al., 2010). The increased proportion of illite may indicate a northward migration of the main clay provenance over Libya, as illite has been shown to be more abundant in dust collected over northern Libya (O'Hara et al., 2006). Nevertheless the I/K ratio range – varying between 0.35 and 0.4 – during the B-A is rather low compared with observed values for Libya and does not further support this hypothesis (Fig. 5a). The observed sedimentological characteristics tend to indicate Tunisia-Algeria as the main dust provenance (Formenti et al., 2011a). According to previously published data, the mineralogical composition is indeed consistent with a source from northern or central Algeria (Fig. 7a), but the low content in palygorskite ruled out any major contribution from tunisian loess (Bout-Roumazeilles et al., 2007). The development of humid conditions during the B-A is evidenced by an increased contribution of river vs. eolian supply in the Aegean Sea (core SL128, Fig. 1) (Hamann et al., 2008), by pollen association from the Alboran (ODP976, Fig. 1) and Adriatic Sea (Combourieu-Nebout et al., 1998, 2002; Fletcher and Sanchez-Goñi, 2008) and by a speleothem record from the eastern Mediterranean (Bar-Matthew et al., 2003), contrasting with the clay mineral record from the Sicilian-Tunisian Strait. The Sea Surface Temperatures (SST, Fig. 3) – as reconstructed from planktonic *foraminifera* at site MD04-2797 – are high during the Bølling-Allerød (Ellassami et al., 2007), consistently with general warm oceanic conditions, relatively high humidity and temperatures compared with present-day in Sicily and central Mediterranean (Ramrath et al., 2000; Allen et al., 2002; Zielhofer et al., 2008; Incarbona et al., 2010). The dominance of semi-desert plants at site MD04-2797 (Desprat et al., 2012) confirms the persistence of dry continental conditions over Africa during the B-A. These results tend to confirm contrasting climatic evolution (e.g. Roberts et al., 2008) with humid conditions on the northeastern and eastern Mediterranean whereas aridity still persisted on the southern Mediterranean.

5.3 Younger Dryas

A major peak in the Zr/Al ratio is observed between around 12.8 ka, associated with an increase in the abundance of both sortable-silt and sand-size particles (Fig. 5b). The clay association does not exhibit major modification but smectite is more abundant at the expense of kaolinite, displaying a peak at 12 ka (Fig. 2). These characteristics indicate the supply of coarser particles that may either be associated to a volcanic supply, or to river supply or to an intense dust episode (Martinez-Ruiz et al., 2003; Erhmann et al., 2007b; Hamann et al., 2009; Skonieczny et al., 2011). Smectite peaking at 12 ka supports the hypothesis of a tephra (Fig. 5d). Moreover, when compared with the chronology of tephras (e.g. Zanchetta et al., 2011), the increase of Zr may be associated with the Agrano Pomici Principali (12.8–12.2 ka) tephra (DiVito et al., 1999; Siani et al., 2004). Nevertheless, optical observations of the sediment do not give any support to this hypothesis. The hypothesis of enhanced river supply is also unlikely considering the general arid climatic conditions that prevailed during the Younger Dryas (YD) (Lézine et al., 1995; Gasse et al., 2000). This specific level could thus alternatively reflect intense Sahara eolian activity that has previously been reported in sediment cores from the northeastern Tropical Atlantic (De Menocal et al., 2000; Cole et al., 2009; Skonieczny et al., 2012).

Cold intervals are generally associated with intense dust deposition/eolian activity resulting from reduced vegetation promoting the erosion of soils, low moisture availability and enhanced atmospheric circulation. The North Atlantic Oscillation (NAO), which is responsible for some part of the Mediterranean climate variability since its impact on the wind activity and precipitation balance (Pittalwala and Hameed, 1991; Moulin et al., 1997; Sanchez-Goñi et al., 2002), has been shown to be positive during the YD (Kim et al., 2007). A positive phase of the NAO is thought to be responsible for the development of dry conditions over southern Europe and North Africa and for promoting northward intrusions of Saharan air masses toward the western Mediterranean (ODP976, Fig. 1) during cold climatic events of the last climatic cycle (Combourieu-Nebout et al., 2002; Bout-Roumazeilles et al., 2007). These cold events were characterized by increased palygorskite supply associated with the dominance of arid steppe (*Artemisia*) and the occurrence of endemic pollen (*Argania*) pinpointing a dust source from western Morocco being transported along the North African coast (Bout-Roumazeilles et al., 2007). The absence of specific enrichment in palygorskite during YD in the Sicilian-Tunisian Strait indicates that the main long-range transportation pattern of fine Saharan dust is not responsible for the peculiar characteristic of sediments deposited in the Sicilian-Tunisian Strait. This is in agreement with grain-size and geochemical data, which rather point to medium to short-range transportation of coarse particles from proximal continental areas (Pye and Tsoar, 1987; McTainsh et al., 1997; O'Hara et al., 2006). The occurrence of such a coarse-grain proximal supply during the YD, consistent with the presence of loess deposits and coastal dunes in Tunisia (Coudé-Gaussen et al., 1987;

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Grousset et al., 1992; Crouvi et al., 2010) and further supported by model suggesting that Libyan desert and coastal areas are major coarse-grain dust sources for central and eastern Mediterranean (Callot et al., 2000), may thus reflect local eolian activity forced by ocean-atmosphere linkage at global scale (Fig. 7b).

5.4 Onset of the Holocene

The end of the Younger Dryas (11.7 ka) is marked by a drastic change in the clay assemblage (Figs. 5a, 6a and b) and a decrease of the sortable silt proportion. The kaolinite increases while illite diminishes, resulting in a progressive decrease of the I/K ratio. This shift, associated to a modification of the illite structural composition, evidences a major change of provenance of the clay fraction, interpreted as an increase contribution of the Sahelian area (Fig. 6a). This observation is supported by the synchronous changes in geochemical signature (Fig. 6c) confirming enhanced contribution of strongly hydrolyzed domains. Low I/K indicates enhanced contribution from the southern Algeria-Mali border region (Paquet et al., 1984; Bergametti et al., 1989b; Alastuey et al., 2005; Bout-Roumazeilles et al., 2007), but the clay association suggests a provenance from southern Algeria rather than from northern Mali, because kaolinite is more abundant southward (e.g. Bout-Roumazeilles et al., 2007), becoming dominant in southern Algeria whereas the Mali border is characterized by smectite-rich assemblages (Fig. 7c). The abundance of semi-desert pollen taxa exhibits a sharp decrease at 12.25 ka (Desprat et al., 2012), before the mineralogical transition, suggesting that the modification of vegetation, i.e. a major change in steppe composition and a development of scarce deciduous oak woodlands, might prevent soils from weathering, displacing southward the areas of eolian erosion. The end of the YD is marked by a similar modification of the clay mineral fraction in a core from the northeastern Tropical Atlantic off Senegal (Skonieczny et al., 2012) and interpreted as recording a southward shift of the Inter Tropical Convergence Zone (ITCZ), which strongly affect the transport of Saharan dust toward the Atlantic Ocean. Moreover, the onset of the Holocene is also marked by a decrease of the I/K ratio off Portugal (Stumpf et al., 2011). The synchronicity of the clay modification at these sites, submitted to distinct wind regimes, signify that the end of the YD is marked by a large-scale atmospheric reorganization over the North African continent, with the development of dry conditions during the early Holocene, affecting both the high-latitude long-range transportation and the regional low-latitude wind regime over the Mediterranean.

5.5 Sapropel S1

The time interval between 8.6 and 5.5 ka is marked by the highest terrigenous flux and smectite content, with a significant grain-size fining (Figs. 2 and 5d). The associated increase in particulate organic carbon (POC) during the whole interval suggests presapropelic conditions (Essallami et al., 2007). Both timing and peculiar characteristics suggest this interval to be related to the most recent organic-rich sapropel S1 (Kidd et al., 1978) deposited during the mid-Holocene in the eastern Mediterranean (Emeis et al., 1996; Emeis et al., 2000; De Lange et al., 2008). During the sapropel, increased precipitation on the surrounding continents provided freshwater discharge and promoted strong stratification of the water column and enhanced nutrient supply, resulting in increased productivity and improved preservation due to oxygen depletion of the water column (Rossignol-Strick et al., 1982; Rohling, 1994; Abu-Zied et al., 2008). Increased precipitations are thought to be linked to either orbitally-forced enhanced monsoon activity (Rossignol-Strick et al., 1982; Hilgen, 1991; Lourens et al., 1996; Emeis et al., 2000) or to increased rainfall over the Mediterranean region (Kallel et al., 1997; Magny et al., 2002). Indeed, although some data appear to be consistent over the Mediterranean basin (Sadori et al., 2011), rainfall records display seasonal modulation (Sadori et al., 2004, 2008; Frisia et al., 2006; Magny et al., 2007, 2012; Kotthoff et al., 2008; Jalut et al., 2009; Colonese et al., 2010; Giraudi et al., 2011; Peyron et al., 2011). Speleothems indicate increased moisture availability in both western (Corchia Cave, Zanchetta et al., 2007), and eastern Mediterranean (Soreq Cave, Bar-Matthews et al., 2000) consistent with lakes records from East Anatolia (Stevens et al., 2001; e.g. Roberts et al., 2008) but also evidence contrasting moisture originating either from

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the Atlantic (Bard et al., 2002) or from the Mediterranean (Arz et al., 2003). Moreover rainfall patterns also display slightly different chronology in Italian lakes (Roberts et al., 2008) and stalagmite records (Frisia et al., 2006; Zanchetta et al., 2007). Consequently, most studies focused on the precipitation distribution and moisture availability that prevailed on the continent at the time of S1.

But investigations of the marine propagation of the sapropelic layer from eastern toward western Mediterranean evidenced the absence of true sapropels and/or synchronous events (Mercone et al., 2000) in the western basin, suggesting lower export of production fluxes into the basin (Martinez-Ruiz et al., 2003) or different hydrographic conditions (Weldeab et al., 2003). Smectite provenance during the sapropel in the Sicilian-Tunisian Strait would provide additional information on the modifications of oceanic environmental conditions. Indeed, central Mediterranean is depleted in smectite (Bout-Roumazeilles et al., 2007), which is rare in southern Algeria and absent from northern and central Algeria. By contrast, the southern Algeria-Mali border (Fig. 1) is locally characterized by a high content in smectite (Bout-Roumazeilles et al., 2007). But, at present-day, this area is submitted to trade-winds, promoting a dominant low-altitude westward transportation and ruling out any significant contribution of this area to sedimentation in the central Mediterranean. A volcanic origin of smectite from Sicilia is not supported by optical observations or by the chronology of the main tephra (Siani et al., 2004). The enhanced supply of smectite may originate from tunisian loess (Fig. 1) (e.g. Bout-Roumazeilles et al., 2007), but the low content of palygorskite during the smectite-rich interval does not further support this interpretation. Moreover, the enhanced moisture availability over that period, associated with pollen association – i.e. open oak forest with heath underbrush or maquis and Asteraceæ-Poaceæ-Cyperaceæ steppe (Desprat et al., 2012) preventing soils from erosion – and the geochemical signal give little support to significant eolian activity. These observations highlight that smectite may alternatively be supplied toward the Mediterranean by the Nile River and transported as suspended particles within the Mediterranean surface water (Ventakatarathnam and Ryan, 1971; Stanley and Wingerath, 1996; Foucault and Mélières, 2000; Hamann et al., 2009). This smectite-rich interval is indeed characterized by an increase abundance of palladium content, suggesting the presence of Pd-bearing smectite, which are generally associated with mafic and ultramafic rocks (Sawlowicz, 1993) and may indicate a specific contribution from the volcanic and ultramafic materials outcropping in Ethiopia (Fig. 7d). This interpretation is consistent with an increased contribution of the Blue Nile draining the Cenozoic volcanic provinces of Ethiopian Highlands (Ukstins et al., 2002), during the African Humid period (Revel et al., 2010). In the Levantine basin (SL112, Fig. 1), the terrigenous fraction, deriving from the Nile River (Krom et al., 2002; Schilman et al., 2001; Hamann et al., 2008), reveals an increase in cohesive fine-grained particles during the Sapropel S1 (Hamann et al., 2010), interpreted as enhanced pluvial period in Egypt (Revel et al., 2010) during the African Humid Period, AHP (Cole et al., 2009; De Menocal et al., 2000).

Considering uncertainties on the age models, the enhanced contribution of fine particles is synchronous in the Levantine Sea and in the Sicilian-Tunisian Strait, supporting a provenance of the fine smectite-rich particles from the Eastern Mediterranean basin. The abundance of fine particles may thus result from slightly slower oceanic circulation during the Sapropel S1 promoting the deposition of the finest fraction through settling processes (Cram and O'Sullivan, 1999). The circulation pattern is consistent with this hypothesis since the Levantine Intermediate Water (200–600 m water-depth) overflows toward the western Mediterranean basin across the Siculian-Tunisian Strait. Moreover, cold-water corals and benthic *foraminifera* assemblages evidenced the impact of sapropelic conditions in shallow water (600 m water-depth) from the Ionian Sea (Fink et al., 2012), giving additional support to a westward propagation of sapropelic conditions through intermediate water-masses (Fig. 7d). The occurrence of a smectite-rich interval in the Alboran Sea (Bout-Roumazeilles et al., 2007), and on the Atlantic side of the Gibraltar Strait during the AHP, suggest a potential transport of smectite through the Mediterranean Overflow Water (Stumpf et al., 2010).

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5.6 Late Holocene

The upper part of the record is marked by an increase of the Ti/Al and Zr/Al ratios (Fig. 5c), associated with a high supply in quartz, both suggesting enhanced wind activity. The dominance of silt-size particles and the stable MART (Fig. 5c) suggest long-range transportation rather than enhanced eolian supply. The increases in kaolinite and palygorskite abundances (Fig. 5c) as well as the low Ca/Fe and I/K ratios indicate a provenance from the Sahelian area and/or the southwest Morocco (Fig. 7e). This event is synchronous with the occurrence of cool/arid events in the Mediterranean (Rapid Climate Coolings-RCC; e.g. ~ 3.5–2.5 kyr BP), suggesting relationships with high-latitude climatic phenomena and altered runoff regime (Mayewski et al., 2004). Previous studies evidenced a climate change around 4.5 ka cal. BP, with distinct regional late Holocene climatic evolution in the Mediterranean (Roberts et al., 2008; Sadori et al., 2008; Peyron et al., 2011; Sadori et al., 2011; Vannière et al., 2011; Magny et al., 2012). Southern sites experienced a drastic decrease in summer precipitations (Magny et al., 2012) while northern sites were submitted to increased precipitation. According to present-day atmospheric pattern, the late Holocene mineralogical and geochemical records are consistent with a mean southward position of the ITCZ which may also explain the low-latitude aridity associated to the RCC (Haug et al., 2001; Hodell et al., 2001). A southward position of the ITCZ would indeed explain the geographically contrasted Mediterranean climatic evolution by promoting the aridification of North Africa while increased summer precipitation occurred in northern Mediterranean (Vannière et al., 2011) as well as enhanced high-altitude export of dust (Skonieczny et al., 2011).

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6 Conclusions

The clay mineral, grain-size and geochemical studies of sediment deposited on the Sicilian-Tunisian Strait help retracing atmospheric vs. riverine terrigenous supplies variability since the last glacial in central Mediterranean. Although the eolian supply is dominant at the studied site – excepted during the sapropel S1 – both flux and the main provenance of particles display strong variations, related to aridity/moisture balance and vegetation cover, driven by large-scale atmospheric reorganization.

The Bølling-Allerød is marked by increased terrigenous flux while both illite and palygorskite became main components of the clay mineral fraction. The concomitant dominance of silt-size particles indicates an eolian origin for this enhanced detrital supply. The geochemical and mineralogical data indicate a dominant Saharan contribution, pinpointing central and northern Algeria as a main provenance. This increased eolian contribution from Sahara highlights contrasting regional climatic evolution, with the development of moist conditions over the North and eastern Mediterranean during the Bølling-Allerød, while glacial aridity persisted on the southern Mediterranean.

A short-term coarse-grained and Zr-rich interval characterized the Younger Dryas, as arid and cold climatic conditions promoted soil erosion. Similar events characterize the western Mediterranean and the northeastern Tropical Atlantic Ocean, although clay mineralogy clearly indicates different provenances. These results suggest local increase of wind activity driven by wide ocean-atmosphere interactions.

The onset of the Holocene is marked by a major change of clay mineralogy and cristallinity corresponding to a southward migration of the main clay provenance toward the Sahelian belt. This change is associated with a progressive development of the Mediterranean vegetation and the southward shift of the ITCZ. Similar signals are recorded in the northeastern tropical Atlantic Ocean and off Portugal, suggesting a large-scale atmospheric reorganization.

High terrigenous flux associated with the dominance of very fine-grained Pd-rich smectite characterized the early to mid-Holocene in central Mediterranean, while the

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organic content indicates pre-sapropelic conditions. The sedimentological characteristics rule out any eolian supply but rather suggest enhanced riverine contribution from remote area during the sapropel S1. The peculiar signature of this event in the central Mediterranean and in the Levantine basin suggests the propagation through intermediate water-masses of fine-grained smectite originating from the Nile River during the sapropel S1, resulting from enhanced precipitations on northeast Africa, coincident with the late phase of the AHP.

Finally, an increase supply of dust originating from northern Sahel or southern Morocco characterizes the last 3 kyr, indicating intense eolian activity linked to modification of the rainfall regime.

Supplementary material related to this article is available online at: http://www.clim-past-discuss.net/8/2921/2012/cpd-8-2921-2012-supplement.pdf.

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Table 1. Radiocarbon ages performed on core MD04-2797. The ages were corrected using reservoir ages of 400 yr during the Holocene, the Younger Dryas and late glacial, 560 yr during the Bølling-Allerød and 800 yr during the Heinrich event H1 and the Older Dryas, following recommendations of Siani et al. (2001).

Depth (cm)	Species	^{14}C age (yr BP)	Error $\pm 1\sigma$	Corrected age (yr BP)	Error $\pm 1\sigma$	Calibrated age (cal yr BP)	Error $\pm 1\sigma$	Laboratory
0	*G. inflata*	1105*	20	705	20	668	9	ARTEMIS
80	*foraminifera*	5496	95	5093	95	5827	144	ARTEMIS
160	*foraminifera*	6700	85	6300	85	7241	120	ARTEMIS
199	*foraminifera*	7523	81	7123	81	7967	71	ARTEMIS
240	*foraminifera*	8113	81	7713	81	8488	98	ARTEMIS
330	*G. ruber*	8888	110	8488	110	9477	112	ARTEMIS
410	*foraminifera*	10 863	32	10 463	32	12 493	49	ARTEMIS
470	*G. inflata*	12 728	173	12 168	172	14 016	35	ARTEMIS
510	*G. ruber*	13 900	141	13 100	141	15 934	621	ARTEMIS
610	*G. ruber*	15 590	50	15 190	50	18 725	370	ARTEMIS
700	*foraminifera*	17 660	70	17 260	70	20 414	180	ARTEMIS
940	*foraminifera*	23 415	163	23 015	163	27 879	331	ARTEMIS
1030	*foraminifera*	26 095	7	25 695	7	30 490	156	ARTEMIS

* Data from Rouis-Zargouni et al. (2010).

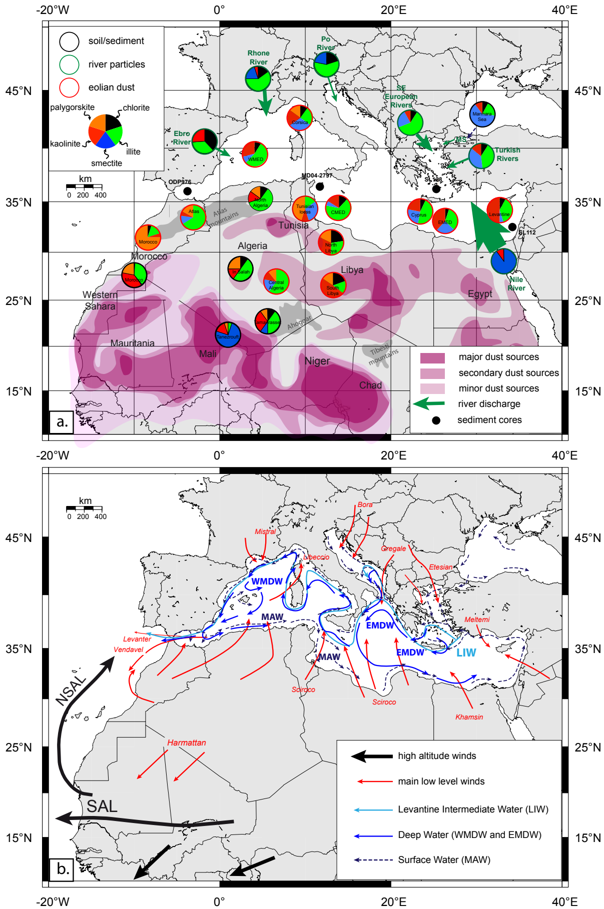

Fig. 1. Caption on next page.

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Fig. 1. Geographical settings: **(a)** clay mineralogy of peri-Mediterranean river particles (green circles), sediments/soils (black circles) and dust particles (red circles), modified from Bout-Roumazeilles et al. (2007), additional data from Cyprus and Levantine Sea: e.g. Hamann et al. (2009); data from northwest Aegean province and West Turkey province: e.g. Ehrmann et al. (2006); data from Marmara Sea: Armynot du Châtelet et al. (2012); North and South Libya dust from O'Hara et al. (2006). Position of the sediment cores mentioned in this study. Major, secondary and minor dust sources modified from D'Almeida (1998); Brooks and Legrand (2000); Caquineau et al. (2002); Prospero et al. (2002); Israelevich et al. (2002); Goudie (2003); Formenti et al. (2011a, b). Main low level (red arrows) and altitude winds (black arrows), rivers supply (green arrows, thickness proportional to annual suspended supply) in the Mediterranean Sea. SAL: Saharan Air Layer, NSAL: northern branch of the Saharan Air Layer. The limit between Sahara and Sahel is reported. Main surface (MAW), intermediate (Levantine Intermediate Water, LIW) and deep water (WMDW and EMDW) masses (Saliot, 2005).

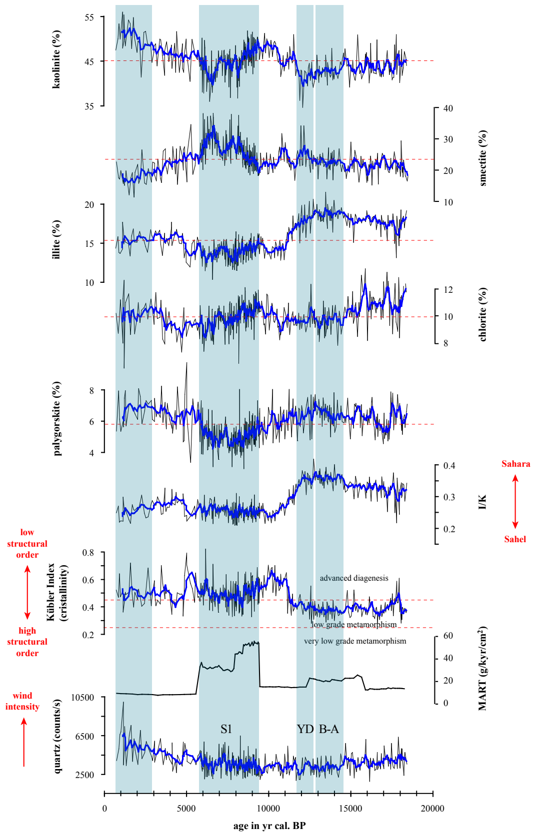

Fig. 2. Clay mineralogy of core MD04-2797: kaolinite (%), smectite (%), illite (%), chlorite (%), palygorskite (%), I/K : illite to kaolinite ratio, Kübler index (crystallinity), (MART) Terrigenous Mass Accumulation Rates (g kyr^{-1} cm^{-2}) and quartz content (counts/s) for the last 20 000 yr. B-A = Bølling-Allerød, YD = Younger Dryas, S1 = sapropel S1 (Mercone et al., 2000).

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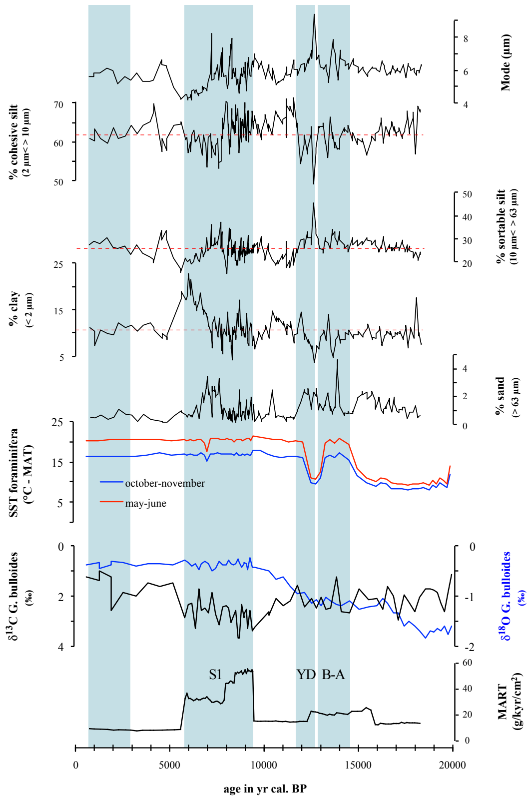

Fig. 3. Grain size analysis of core MD04-2797: mode (µm), % of cohesive silt (2 <> 10 µm), % of sortable silt (10 <> 63 µm), % of clay (< 2 µm) and % of sand (> 63 µm). MART from Fig. 1. Sea Surface Temperatures (SST) for May–June and October–November based on *foraminifera* association (Essallami et al., 2007), δ^{18}O (‰) and δ^{13}C from planktonic *foraminifera G. bulloides* (Essallami et al., 2007).

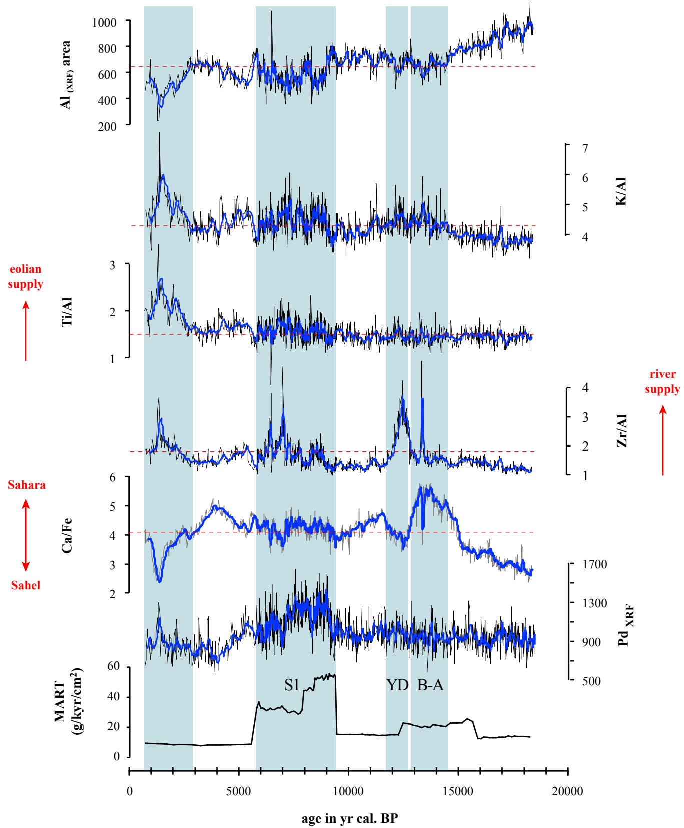

Fig. 4. Geochemical data for core MD04-2797, Al content (counts/s), Ti/Al ratio, Zr/Al ratio, Ca/Fe ratio and Pd content (counts/s). MART from Fig. 1.

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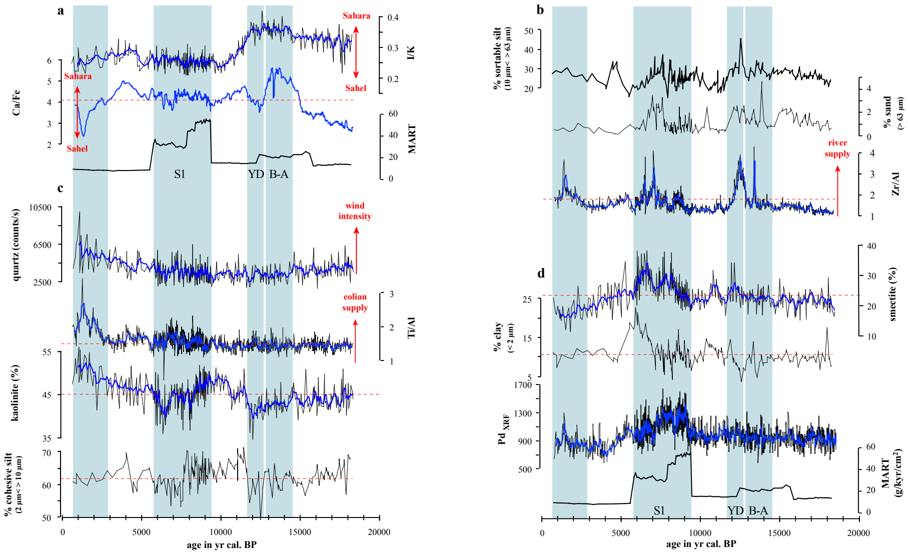

Fig. 5. Comparison of multiproxies data, **(a)**: I/K, Ca/Fe and their interpretation on particles provenance for the Bølling-Allerød, **(b)**: grain-size compared with Zr/Al during the Younger Dryas, **(c)**: quartz content (counts/s), Ti/Al ratio and kaolinite % over the last 3000 yr, **(d)**: Smectite (%), characterizing the sapropel S1, % of clay, content in Pd (counts/s) and MART.

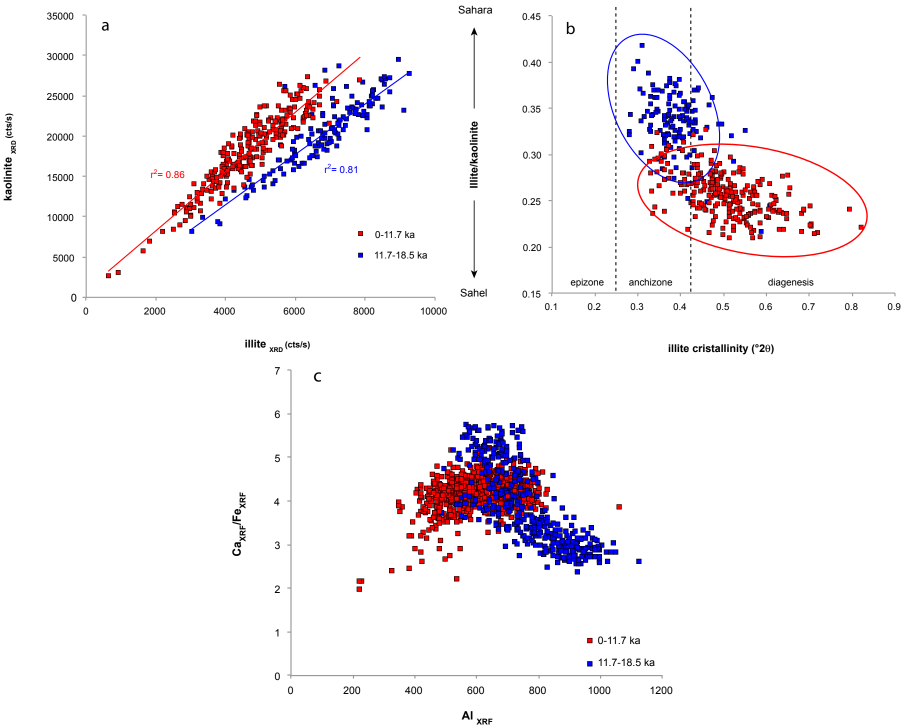

Fig. 6. Comparison of the mineralogical and geochemical signatures before and after 11.7 kyr, **(a)**: kaolinite versus illite (XRD counts/s) evidencing contrasting provenance between 18.5 and 11.7 ka (red squares) and between 11.7 ka and top of the core (blue squares); **(b)**: illite to kaolinite ratio as a function of illite crystallinity (Kübler Index), diagenesis, epizone and anchizone limits, **(c)**: Ca content versus Fe content (counts XRF).

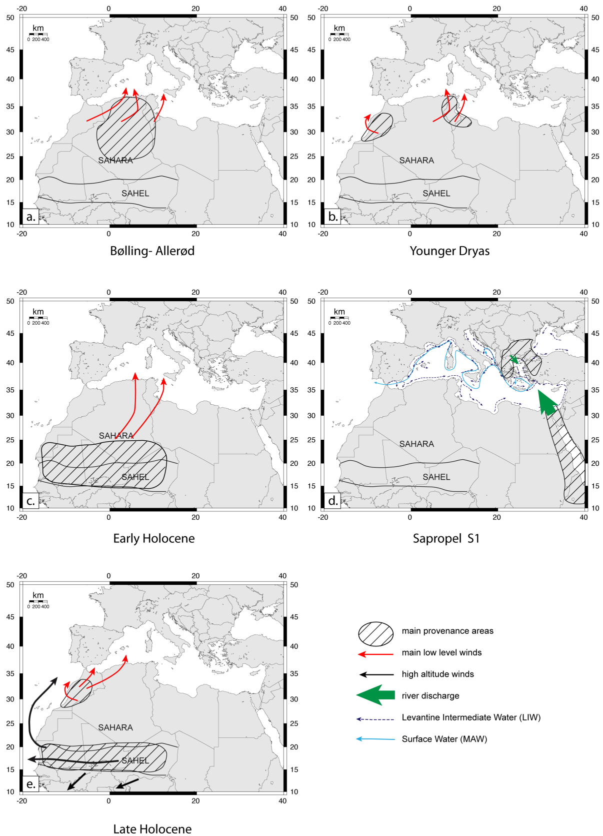

Fig. 7. Main provenance (dashed areas) and potential eolian – low level (red arrows) or altitude winds (black arrows) – and riverine (green arrows) transportation patterns of clay-mineral particles for 5 timeslices, **(a)** Bølling-Allerød; **(b)** Younger Dryas; **(c)** early Holocene; **(d)** Sapropel S1; **(e)** late Holocene.