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Unraveling North-African riverine and eolian contributions to central Mediterranean sediments during Holocene sapropel S1 formation

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Hydroclimate variability has exerted a fundamental control on the alternating deposition of organic-lean marl and organic-rich sapropel sediments in the eastern Mediterranean Sea (EMS). However, the exact mechanisms regarding the freshwater sources and related changes are still debated. Here, Sr and Nd isotopes and high-resolution elemental data are used to constrain different riverine and eolian supplies to the central Mediterranean over the past 9.8 ka. The detrital sediments in core CP10BC, taken at the margin of the Libyan shelf in the southwestern Ionian Sea, can be described by a three-endmember mixing system based on Sr and Nd isotopic compositions. The same systematics can also be deduced from Ti and K compositional variability. The endmembers comprise: Saharan Dust, Aegean/Nile, and Libyan Soil, representing the eolian supply from North Africa, the riverine inputs from the Aegean/Nile areas, as well as the riverine and shelf-derived fluxes from the Libyan-Tunisian margin, respectively. For the sapropel S1 period in particular, we find important detrital supplies from fossil river/wadi systems along the Libyan-Tunisian margin, activated by intensified African monsoon precipitation. Combining the temporal profiles with the consistent variability observed in the $^{87}Sr/^{86}Sr$–1000/Sr diagram, such Libyan contribution has been most prominent during the uppermost period of sapropel S1 in core CP10BC. This observation is in agreement with hydroclimate reconstructions of northwestern Libya. Comparison of the Sr-Nd isotope data between core CP10BC and four cores taken along a west–east transect throughout the EMS shows that this detrital supply originated mainly from western Libya/Tunisia, and was transported as far eastward as ~25°E while being diluted by an increasing Nile contribution.

1. Introduction

The Mediterranean lies at the interface between the European temperate and African tropical zones. In addition, its semi-enclosed basin setting makes it particularly sensitive to changes in the hydrological cycle (Tzedakis, 2007; Magny et al., 2013; Rohling et al., 2015). This sensitivity is best witnessed by the rhythmic occurrence of sapropels, organic-rich sedimentary units deposited in the eastern Mediterranean Sea (EMS) at an astronomically determined cyclicity (Rossignol-Strick et al., 1982; Hilgen, 1991; Tuenter et al., 2003). The knowledge of sapropel formation mechanisms mostly relies on the last, Holocene, sapropel S1 (Rohling, 1994; Rohling et al., 2015; and references therein), because it is more easily accessible and also is the only sapropel within the range of precise radiocarbon dating. Warm and humid conditions between ~10.8 and 6.1 ka cal. BP (hereafter referred to as "ka") have caused a pronounced density stratification of the water column and/or high primary productivity in the surface waters, ultimately resulting in deepwater stagnation and the basin-wide S1-formation in the EMS (e.g. Emeis et al., 2000; Mercone et al., 2000; Slomp et al., 2004; De Lange et al., 2008; Grimm et al., 2015; Tachikawa et al., 2015; Van Helmond et al., 2015). Enhanced freshwater influx at sapropel times has been demonstrated through $\delta^{18}O$ of planktonic foraminifera (e.g. Fontugne et al., 1994; Kallel et al., 1997; Emeis et al., 2000, 2003; Rohling et al., 2002, 2004; Hennekam et al., 2014; Toucanne et al., 2015) and of cave speleothems (e.g. Bar-

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Matthews et al., 2000, 2003; Bard et al., 2002; Zanchetta et al., 2007; Spötl et al., 2010; Zhornyak et al., 2011). Although the general principles are clear, the exact mechanisms with regard to freshwater sources and associated hydroclimate changes in the circum-Mediterranean remain highly debated.

Pioneering studies have proposed increased precipitation around the Northern Borderlands of the Eastern Mediterranean (NBEM) concurrent with sapropel depositions (Rossignol-Strick, 1987; Rohling and Hilgen, 1991). This increase is corroborated by speleothem $\delta^{18}O$ data from the NBEM (e.g. Zanchetta et al., 2007; Spötl et al., 2010; Zhornyak et al., 2011) and by their correspondence with the records of Soreq Cave (Bar-Matthews et al., 2000, 2003). Marine records from the eastern and central Mediterranean also confirm that increased rainfall was widespread over the NBEM during sapropel formation (e.g. Kallel et al., 1997; Emeis et al., 2000; Toucanne et al., 2015; Filippidi et al., 2016). Being seasonally specified, palynological studies demonstrate that enhanced winter precipitation was responsible for the NBEM wet conditions at times of sapropel deposition (e.g. S1: Kotthoff et al., 2008; Peyron et al., 2011; S5: Milner et al., 2012). Summer rainfall was thought to have been increased too, but this is questioned by Tzedakis (2007). Recent work has revealed north–south contrasts of precipitation seasonality in the Holocene central Mediterranean, which can reconcile the discrepancies from various proxies (e.g. Magny et al., 2011; Peyron et al., 2011, 2013; Giraudi et al., 2013; Goudeau et al., 2014, 2015). The evidence suggests that, there were also increases in 1) winter precipitation in the NBEM and 2) summer and winter precipitation in the southern EMS borderlands during S1 period, in addition to the enhanced Nile discharge (see review by Magny et al., 2013). Possible impacts of the Black Sea overflow and meltwater runoff of European ice-sheets during sapropel deposition have been excluded (Sperling et al., 2003).

Sapropels occurred at times of Northern Hemisphere insolation maxima related to the orbital cycle of precession, when the African monsoon was intensified (Rossignol-Strick et al., 1982; Rossignol-Strick, 1987; Hilgen, 1991; Tuenter et al., 2003). Even though the African monsoon precipitation did not directly extend to the EMS, the monsoon-fuelled Nile discharge is recognized as a key freshwater source for sapropel formation. This has been explicitly illustrated by the temporal coincidence between the S1 deposition and Nile discharge (e.g. Rossignol-Strick et al., 1982; Mercone et al., 2000; De Lange et al., 2008; Revel et al., 2010; Box et al., 2011; Hennekam et al., 2014). Apart from the Nile flooding, intensification of African monsoon precipitation had a profound and more widespread influence on the EMS during sapropel times.

Archaeological surveys suggest human occupation and dispersal over the Libyan-Saharan desert along the interlinked river-lake-wetland systems during the African Humid Period (AHP; ~11–5 ka) (e.g. Mandel and Simmons, 2001; Drake et al., 2011; Lézine et al., 2011; Manning and Timpson, 2014). Based on planktonic foraminiferal $\delta^{18}O$ data, Rohling et al. (2002, 2004) suggested that the summer monsoon penetrated northward beyond the central Saharan drainage basin at ~21°N during sapropel S5 deposition. As a consequence, activated fossil river/wadi systems along the wider North-African margin that are now buried beneath sand dunes could have debouched into the EMS. Major paleo-river systems have been revealed via satellite imagery (Paillou et al., 2009, 2012) and paleohydrological modeling (Coulthard et al., 2013). Influence of runoff from the central Saharan mountains towards the Gulf of Sirte during sapropel S5 period is also shown in Nd isotopes of planktonic foraminifera (Osborne et al., 2008, 2010). This freshwater route into the EMS may have rivaled the Nile runoff in magnitude (Scrivner et al., 2004), favored human migrations out of Africa (Osborne et al., 2008; Coulthard et al., 2013), and possibly operated for a majority of Quaternary sapropels (e.g. S1: Fontugne et al., 1994; S6: Emeis et al., 2003). Although the origin of Nd recorded in planktonic foraminifera is still controversial (see review by Tachikawa et al., 2014), this scenario is supported by dust variations over the past 3 Ma in combination with other Saharan data (Larrasoaña et al., 2003, 2013). However, so far there is no marine evidence whether the fossil river/wadi route was active during sapropel S1 time. In this paper we focus on such riverine contribution by analyzing the detrital component of EMS sediments.

It has been suggested that clay minerals of surface sediments over most of the EMS can be described by a simple two-endmember mixing model, with Saharan dust and Nile sediment as the dominant sources (e.g. Venkatarathnam and Ryan, 1971; Ehrmann et al., 2007a; Hamann et al., 2009). From a long-term perspective, various records of clay minerals and major elements reflect consistent sapropel-related alternations between riverine and eolian endmembers (e.g. Wehausen and Brumsack, 1999, 2000; Foucault and Mélière, 2000; Lourens et al., 2001; Zhao et al., 2012, 2016). However, only a few high-resolution studies on the detrital composition of marine sediments exist, and most of these are focused on the Nile-dominated region (e.g. Hamann et al., 2009; Revel et al., 2010, 2015; Box et al., 2011; Hennekam et al., 2014, 2015). Very little is known about the supplies of numerous small rivers from other sources through time. In particular, potential riverine supplies from the wider North-African margin may have been underestimated, or even neglected.

Paired Sr and Nd isotopes have shown to be a powerful tool for provenance studies. Their isotopic ratios in lithogenic sediments have characteristic fingerprints of source rocks, with little effect of transport processes and diagenesis (e.g. Freydier et al., 2001; Weldeab et al., 2002a, 2002b; Revel et al., 2010, 2015; Rodrigo-Gámiz et al., 2015; Castañeda et al., 2016). In addition to radiogenic isotopic tracers, elemental geochemistry of marine sediments can be used to track changes in detrital inputs (e.g. Wehausen and Brumsack, 1999, 2000; Bout-Roumazeilles et al., 2013; Wu et al., 2013; Klaver et al., 2015). Lower Ti/Al and Zr/Al values and higher K/Al ratios during humid sapropel periods are generally interpreted as a sign of reduced eolian and enhanced riverine fluxes, and vice versa for more arid conditions (see review by Martinez-Ruiz et al., 2015). Here, combining Sr and Nd isotopes with major elements, we present a high-resolution record of the origin of detrital sediments over the past 9.8 ka using a well-dated boxcore. Core CP10BC was strategically retrieved from the southwestern Ionian Sea, at the margin of the Libyan shelf (Fig. 1). The sediment provenance for this area is thought to be influenced by Saharan dust and riverine sources from northern and southern EMS borderlands, with a presumably limited Nile contribution. Therefore, this core represents an excellent location to understand the nature and importance of rivers flowing north out of the Sahara during sapropel S1 time. Furthermore, by comparing the Sr-Nd isotope data of core CP10BC with four cores taken along a west–east transect throughout the EMS, a detailed evaluation can be made of the potential distribution of detrital material.

2. Recent regional settings

2.1. Hydroclimate

The Mediterranean climate is characterized by marked seasonality in rainfall (Magny et al., 2013; Rohling et al., 2015 and references therein; Castañeda et al., 2016). General features are summarized as follows (Fig. 1a). During summer, the Mediterranean is dominated by the subtropical high-pressure belt resulting in dry and hot conditions, especially in the southeastern sector (Lolis et al., 2002). In winter, the high-pressure belt migrates southward and the Mediterranean comes under the control of the