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Multi-proxy constraints on sapropel formation during the late Pliocene of central Mediterranean (southwest Sicily)



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ABSTRACT

The late Pliocene (Piacenzian) in the Mediterranean region was punctuated by short-lived episodes of widespread deposition of organic-rich sedimentary layers known as sapropels. The causes of their formation remain a long-standing debate in the science community, and require disentangling the roles of climatic/oceanographic processes that triggered higher primary productivity or enhanced organic matter preservation. The lack of data, especially of sea temperatures at sufficient temporal resolution, is one of the main challenges to solve this debate.

Here, we present new organic geochemistry and micropaleontological data from the late Pliocene at Punta Grande/Punta Piccola sections (southwest Sicily) that allow untangling the mechanisms that favored the formation of two sapropel series (noted S and A) in the central Mediterranean area during this period. Sea surface (SSTs) and subsurface temperatures were estimated using three distinct organic geochemical proxies namely the alkenone unsaturation index ($U^{K'}_{37}$), the long-chain diol index (LDI) and the tetraether index (TEX₈₆). Reconstructed SSTs are relatively stable throughout the late Pliocene and \sim 4 °C higher than modern Mediterranean SSTs, which is consistent with the climatic conditions inferred for this period from paleoclimate modeling. An increase in SST is, however, recorded by $U^{K'}_{37}$ and LDI proxies across each sapropel horizon, supporting that the two sapropel series S and A were formed during warmer climate conditions. The comparison of SST data with variations in accumulation rates of total organic carbon and lipid-biomarkers (alkenones, long-chain alkyl diols, archaeal and bacterial tetraethers), and with changes in calcareous nannofossil assemblages, indicates that the studied sapropels might have formed under different environmental conditions. The first series of sapropels (S), deposited between 3.1 and 2.8 Ma, is likely due to a better preservation of organic matter, induced by the development of a strong thermohaline stratification of the water column and to oxygen-depleted bottom waters. Higher terrestrial input that occurred between 3.1 and 2.8 Ma may interestingly explain the large discrepancies observed between TEX₈₆ and U^{K'}₃₇-LDI temperature values during this period. The second series of sapropels (A), deposited between 2.7 and 2.6 Ma, is more likely due to enhanced primary productivity in a weakly-stratified water column.

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1. Introduction

The late Pliocene (Piacenzian; 3.6–2.59 Ma) was characterized by the cyclic deposition of dark, organic matter-rich layers named sapropels in the Mediterranean Sea. Their formation is argued to be triggered by pulses of freshwater supply to the Mediterranean associated with the strengthening of the precessionally-controlled African monsoon (e.g., Béthoux, 1993; Foucault and Mélières, 2000; Combourieu-Nebout et al., 2004), which led to enhanced primary productivity and organic matter export to marine sediments

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(Pedersen and Calvert, 1990), and/or enhanced water column stratification and improved organic matter preservation (e.g., Béthoux, 1993). Yet, the relative importance of increased export productivity *versus* organic matter preservation for the formation of sapropels is still uncertain.

The sapropels from the Punta Grande/Punta Piccola sections (southwest Sicily: Fig. 1), considered as a reference for the cvclochronology of the middle Pliocene (Lourens et al., 1996), have been the subject of a number of chemical, mineralogical and palynological studies (e.g., Foucault and Mélières, 2000; Combourieu-Nebout et al., 2004; Beltran et al., 2007). However, studies have been focused on a relatively limited number of sapropel layers (S104-S107). In addition, only a few studies have attempted to reconstruct sea surface temperatures (SSTs) at high temporal resolution during the late Pliocene (Lourens et al., 1996; Herbert et al., 2015). Lourens et al. (1996) estimated SSTs at Punta Grande/Punta Piccola using oxygen isotopes and planktonic foraminifera assemblages (ratio warm versus cold taxa). In the Mediterranean, the interpretation of oxygen isotope data is, however, complicated by strong hydrological variability (e.g., Colleoni et al., 2012). More recently, Herbert et al. (2015) described a high-resolution reconstruction of Mediterranean SSTs based on the alkenone unsaturation index ($U_{37}^{K'}$; Prahl et al., 1988) to provide the first continuous record of Mediterranean for the late Pliocene-early Pleistocene (3.5-1.5 Ma).

Here, we have estimated marine temperatures from the Punta Grande/Punta Piccola section for the whole late Pliocene (3.6-2.6 Ma), using the indices $U^{K^\prime}{}_{37},\, \text{TEX}_{86},\, \text{and long-chain diols or}$ LDI (Prahl et al., 1988; Schouten et al., 2002; Rampen et al., 2012). The TEX₈₆ is based on archaeal lipids (isoprenoidal glycerol dialkyl glycerol tetraethers, also known as isoprenoidal GDGTs) and is generally considered to reflect the annual mean SSTs (e.g., Schouten et al., 2013). However, several studies have argued that GDGT-based temperature proxy may, in some regions, better reflect subsurface temperatures rather than annual mean SSTs (e.g., Huguet et al., 2007). TEX₈₆ is thus considered in the present study to integrate surface and subsurface water temperatures. We compared the reconstructed SSTs to changes in accumulation rates of calcareous nannofossils, total organic carbon (TOC) and lipid-biomarkers (alkenones, long-chain alkyl diols and isoprenoidal and branched GDGTs). Alkenones are produced by Haptophyte algae (Marlowe et al., 1990), long-chain alkyl diols by Eustigmatophyceae (1,13- and 1,15-diols) and/or Proboscia diatoms (1,14-diols) (Versteegh et al., 1997; Rampen et al., 2007), and GDGTs by aquatic Archaea (isoprenoidal GDGTs; Schouten et al., 2002) or terrestrial soil bacteria (branched GDGTs; Peterse et al., 2010). Changes in TOC, alkenone, long-chain diol, and isoprenoidal GDGT content, combined to calcareous nannofossil assemblages, provide reliable tracers of trophic conditions in the upper part of the water column (e.g., Rampen et al., 2007; Schouten et al., 2013), while branched GDGT contents reflect terrestrial input into marine sediments (e.g., Hopmans et al., 2004). These new data are used to provide a comprehensive understanding of the environmental context in which sapropels were formed and to constrain the main mechanisms that favored their formation.

2. Materials and methods

2.1. Geological setting and sampling

The Capo Rossello composite section is located in the Caltanissetta basin (southwest of Sicily) and presents an excellent stratigraphy based on magnetic-isotopic data and on calcareous nannofossil record (Rio et al., 1990; Hilgen, 1991; Lourens et al., 1996). The Punta Grande and Punta Piccola sections represent the upper part of the Capo Rossello composite section and outcrop some 10 km west of Agrigento along the road from Porto Empedocle to the Rossello beach (Fig. 1). The Punta Grande section and the lower part (first 14 m) of the Punta Piccola section are characterized by marls and marly limestones of the Trubi Formation (Hilgen, 1991). The uppermost part of the Punta Piccola section comprises regular alternations of grey marls and lightgrey marly limestones of the Monte Narbone Formation, with the cyclical occurrence of dark laminated (sapropel) marl layers noted S101 to S112 and A1 to A5 (Hilgen, 1991; Lourens et al., 1996; Sprovieri et al., 2006).

A total of 61 samples were collected with an average sampling spacing of 80 cm in the Punta Grande and Punta Piccola sections, including the sapropel layers S101 to S109, S111, S112, A4, A4/5 and A5 (Fig. 1). Both sections are beautifully exposed along the coast, with no tectonic disturbances or vegetation cover noticeable. Sampling was performed as deep as possible beneath the surface (i.e. 10 to 30 cm) to collect fresh sediment and avoid the weathered surface layer of the outcrops.

During the Pliocene, the studied sites were situated in an open marine slope-basin setting in the Sicily sill, at a water depth of about 600–800 m (Sgarrella et al., 2012). According to the age model of Lourens et al. (1996), the studied time interval spans the Piacenzian (late Pliocene), from the Zanclean/Piacenzian boundary (3.6 Ma) to the Piacenzian/Gelasian boundary coincident with the sapropel A5 mid-point (2.59 Ma), and comprises the Planktic Foraminifera Zones MPL4B and MPL5A (Cita, 1975) and the Calcareous Nannofossil Zones MNN 16a and MNN 16b-17 (Rio et al., 1990) (Fig. 1). This age model was used to determine the sedimentation rates in the studied sections.

2.2. Calcareous nannofossil analyses

Slides for quantitative counts of calcareous nannofossils were prepared following the Random Settling method of Geisen et al. (1999). A small amount of dried sediment powder (10 mg) was mixed with water (with basic pH, over-saturated with respect to calcium carbonate) and the homogenised suspension was allowed to settle for 24 h onto a cover slide. The slide was dried and mounted on a microscope slide with Rhodopass. Four hundred calcareous nannofossils (coccoliths and nannoliths) were counted in a variable number of fields of views (between 15 and 30 according to the richness of the sample) using a polarizing optical ZEISS microscope (magnification $1000 \times$).

Absolute abundance of calcareous nannofossils per gram of sediment was calculated using the formula:

$$X = (N \cdot V) / (M \cdot A \cdot H) \tag{1}$$

where X is the number of calcareous nannofossils per gram of sediment; N the number of nannofossils counted in each sample; V the volume of water used for the dilution in the settling device (475 cm³); M the weight of powder used for the suspension (g); A the surface considered for nannofossil counting (cm²); H the height of the water over the cover slide in the settling device (2.1 cm). Species-specific relative abundances (percentages) were also calculated from the total nannofossil content.

Taxonomic concepts adopted here followed the species classification according to Young and Bown (1997). The preservation of calcareous nannofossils (degree of etching and overgrowth) was estimated by optical and scanning electron microscopy observations, using the recommendations of Roth (1984).

2.3. Total Organic Carbon analyses

Sub-samples (ca. 50 mg of ground samples) were acidified with 2N HCl in pre-cleaned (combustion at $450 \,^{\circ}$ C) silver capsules un-

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