The Dark Side of the Mediterranean Geological Record: the sapropel layers and a case study from the Ionian Sea

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Abstract

A peculiar feature of the Neogene Mediterranean marine and land sequences is the quasi-cyclic occurrence of organic carbon-rich layers named sapropels. Their occurrences in the sedimentary record usually correspond to periods of enhanced monsoon rainfall during precession minima and summer insolation maxima. Nevertheless, the causal factors that led to their formation are still highly debated. Integrated multi-proxy investigations document that during sapropel deposition important changes occurred in the entire water column: freshwater lenses in the surface waters led to stratification of the water column and to hypoxic or totally anoxic bottom waters. Sapropels offer the unique opportunity to perform studies on climatic, oceanographic and environmental changes at an extraordinary resolution allowing detailed insights into short-scale climatic fluctuations.

Micropaleontological and magnetic signatures demonstrate that oceanographic conditions conducive to sapropel formation were not confined to the eastern Mediterranean sea but occurred also and possibly simultaneously in the entire Mediterranean. The differences appear a consequence of different preservation, changes in water column depth and local hydrographic conditions. Here we report the main features characterizing the youngest Mediterranean sapropel (S1) deposited during the Holocene in the Ionian basin

1 Introduction

Neogene sediments of the Mediterranean Sea are characterized by the occurrence of organic carbon-rich (with TOC usually $>$ 2%) layers named sapropels [1]. Their formation seems to be mainly controlled by astronomical forcing, usually corresponding to phases of precession-induced insolation maxima [2] leading to periods of wetter climate in the Mediterranean region. The word “sapropel” was introduced in literature by Potoniè [3] to indicate dark sediments with decomposing organism deposited under stagnant water. Sapropel is a contraction of the literal translation of the German words Fäulniss and Schlamm into ancient Greek (sapros and pelos, meaning putrefaction and mud respectively). At first discovered in marine sediments in the ’50 [4, 5] the sapropels have been subject of a plethora of studies during the last decades and several models have been proposed to explain the mechanism leading to their deposition (e.g., [6]).

In spite of the large numbers of studies performed in the last 40 years, and two Ocean Drilling Program expeditions (ODP legs 160 and 161 in 1995), the causes of sapropel formation are still debated.

At present, Mediterranean sediments are characterized by low organic carbon content ($<0.5\%$ organic carbon) due to low surface water nutrient levels (hence generally low productivity) and oxic bottom waters due to a vigorous deep-water formation and circulation (details in [7]). Sapropels were instead likely deposited under hypoxic or anoxic deep water conditions strictly related to deep water stagnation due to a heavily reduced or halted deep (or even intermediate) water circulation. The deposition of organic-rich layers such as the sapropels must have hence required major changes to the present water circulation patterns. At first, their occurrence was linked to improved preservation of organic matter under anoxic bottom water conditions. The explanation for the anoxia was related to density stratification of the water column, limiting water circulation and supply of oxygen to deep water. At least for the youngest sapropel the water stratification takes into account increasing freshwater input originated from the Nile River during period of enhanced monsoon regime in the equatorial region [8, 9, 10, 11, 12, 13, 2]. Other proposed triggering mechanisms consider also the increasing organic matter accumulation related to the enhancement of primary productivity [14, 15, 16, 17, 18, 19, 2]. Certainly an increase in productivity in the Mediterranean cannot be achieved with the present-day oceanographical features and for this reason some authors invoked a reversal of the water circulation [16] or a shoaling of the density gradient (pycnocline) into the photic zone [17]. The debate concerning the roles and the importance of productivity and preservation in sapropels formation is still on-going.

Even if Total Organic Carbon (TOC) content was originally chosen as key parameter to identify sapropels, other proxies have been successively used. Among those, geochemical elements (e.g., Fe, Mn, Al, S, Ba, V, Mo, As, I) [16, 20, 21] magnetic parameters such as susceptibility, anysteretic (ARM) and isothermal remanence (IRM) [22, 23, 24, 25], microfaunal taxa [8, 26, 27] are sensitive to the anoxic conditions and can be considered indicators of the sediments deposited under anoxic conditions.

Sapropels offer the unique opportunity to perform climatic, oceanographic and environmental reconstructions at an extraordinary resolution allowing detailed insights into short-scale climatic and environmental oscillations.

Here, after a general discussion on the main features of sapropels, we present, as a case study, the sapropel S1 in a sediment core (ET 99-M11) collected in the Ionian Sea.

2 Sapropels across the Mediterranean

Most of the literature on sapropels considers the eastern Mediterranean, where these layers have been at first discovered and defined. However organic rich layers occur also in the western part of the basin, although they appear more scattered and less developed [28]. This implies that the Sicily channel sill may act as a barrier against processes that favour sapropel deposition. The present-day anti-estuarine Mediterranean circulation, clearly influenced by the Gibraltar strait sill must have still played a role even at the time of the sapropel formation.

TOC contents in the western Mediterranean show maximum values of up to 6% (in the Tyrrhenian Sea) and appear to decrease toward the western areas were TOC hardly reaches the 2-3% [28]. For this reason Murat [28] suggested to redefine a sapropel as an organic-rich lithologic layer (ORL) deposited in open sea, with at least 0.8% of TOC. The sapropels or organic rich layers observed in the Western Mediterranean are more scattered throughout the time even if the timing (at least for the late Quaternary <400 Ka) is synchronous with that of sapropels deposition in the eastern part of the basin [29]. The only exception is the Alboran Sea, where background TOC values are already above 0.4-0.5% [28, 26] and the timing of occurrence of ORL does not appear to match the timing observed for sapropels in the other parts of the basin. In particular the youngest ORL observed at ODP Sites 976 and 979 is strictly coincident with the Younger Dryas ($\sim$12.5-11.5 Ka BP) which means it is older than Sapropel S1 deposited during the early Holocene (9.5-6.Ka BP). However the micropaleontological signal indicates that the planktonic foraminifer assemblage typical of a sapropel layer occurs above the ORL and is coincident with the timing of sapropel S1. It is also noteworthy that TOC peaks were observed in cores from the Alboran Sea at around 55 Ka BP corresponding to the rarely found sapropel S2 [26] and also in correspondence of the I-cycle 4-6 (31-38 Ka BP) that never expresses sapropels in other areas. The presence of sapropels in both the western and eastern Mediterranean indicates that in some cases the entire basin responded in unison at precession minima. This seems true especially in correspondence of interglacial stages as indicated by the finding of warm sapropels such as the S5 ($\sim$125-119 Ka BP) throughout the basin.

3 Magnetic signature of sapropels

In sub-oxic/anoxic conditions as those typical of sapropel depositions, magnetic Fe oxides dissolve, resulting in a decrease of magnetic concentration coupled with an increase in magnetic grain-size and in coercivity [31, 32, 33, 24, 34]. The bacterial degradation of organic matter is a diagenetic process leading to sulphate reduction and methanogenesis that clearly occurs in sapropels. As evidenced during analysis of core from the ODP leg 160, the process can be so severe to be responsible of a magnetic enhancement observed in several sapropels recovered in the Eastern Mediterranean Sea [23]. A ferrimagnetic iron-sulphate phase is responsible of a high magnetization that is directly proportional to the organic carbon content found within the sapropels [23]. On the base of the magnetic properties Larrasoana [25] grouped the sapropels in the 3 different types corresponding to different anoxic conditions. The dissolution or enhancement of the magnetic signal within sapropel layers represents a distinctive feature that can be easily identified already in whole-core measurements (e.g. magnetic susceptibility, K) and represents a marker that can be used for tuning a sedimentary sequence to orbital scale (Figure 1). Concentration-related magnetic parameters such as K, ARM, IRM can be indicative of the presence of sapropel layers even in sediments where the lithologic expression is not clearly visible (“missing sapropels”, [22]). Possibly, the best indi-

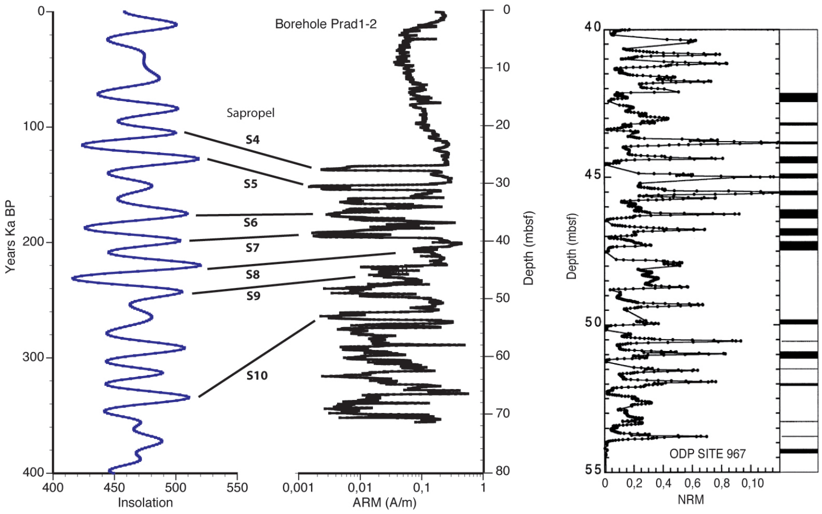

Figure 1: Magnetic parameters (ARM and NRM) from Borehole Prad1-2 (Adriatic Sea) and ODP Site 967 (eastern Mediterranean). Minimum ARM values related to magnetic dissolution occur in the sapropelitic layers recognized in the Adriatic Sea [30] and correlated with insolation maxima. Peaks in the NRM observed at ODP Site 967 reflect precipitation of iron sulphides in correspondence of sapropel layers.

cator of a sapropel layer is the ARM because it is more sensitive to the presence of fine grained ferrimagnetic materials. On the contrary, magnetic susceptibility could also represent an unclear indicator as it is also influenced by the presence of the paramagnetic contribution of the clay minerals. Another distinctive feature of the sapropel layers is the precipitation of Fe oxides at the oxygenation front that causes higher magnetic intensities [35] due to the formation of iron oxides and also Fe sulphides [23].

4 Foraminifera signature of sapropels

Several studies have shown that an unusual planktonic foraminiferal fauna characterizes the sapropel layers consisting of an increase in the occurrence of warm subtropical species *Globigerinoides ruber* (var. *rosea* and *alba*) and the SPRUDTS group (see [36]), or, in many cases, in the exclusive presence of high productivity water indicators such as *Neogloboquadrina dutertrei* and *Globigerina bulloides* [8, 10]. Some authors (see for details [26] and [37]) noted that it is possible to recognize the late Quaternary sapropels on the basis of the quantitative and qualitative variations in planktonic foraminifera assemblage, either when sapropels are deposited in warm or in cold intervals. Generally, sapropel layers deposited during warm intervals are characterized by peaks abundance of *G. ruber* and occurrence of *G. ruber* var. *rosea* (e.g. S1, S3, S5, S7, S9, and S10), while abundance of *N. dutertrei* characterizes the sapropel layers deposited in cold intervals (e.g. S4, S6 and S8).

Both these species inhabit surface waters and are well documented in literature to be related to low salinity [36, 38, 8, 39, 40].

Qualitative foraminifer analyses show that many taxa are characterized by an increase in size (e.g. *Orbulina universa*, *G. ruber* $>$ 150 micron) and that the tests are thinner with large pores, often covered by diffuse pyrite crystals and sometimes entirely or partially filled by pyrite.

During times of sapropels formation benthic foraminiferal abundances and diversity strongly decrease till the near exclusive presence of deep infaunal taxa. Microfauna even disappear in some levels suggesting extremely low oxygen values on the sea bottom [41, 27]. The diversity reported in the pre- and post sapropel benthic assemblages from different sites suggest that the evolution of the dysoxic-anoxic conditions, as well as the re-oxygenation pattern at the end of the stagnant period, were characterized by spatial and temporal variability, possibly controlled by basin physiography and, in particular, by the water column depth [27, 42, 43, 44]. In addition, high resolution benthic foraminiferal distributional trend during times of sapropel S5 and S6 deposition indicated that water column stratification and deep-water formation was unstable and reflected the climate fluctuations at millennial time scale.

Based on these biological features the sapropel deposition appears the result of different oceanographic phases related to stratification of the water column, reduced ventilation in intermediate/deep water and changes in nutrient regimes.

5 Geochemical signature of sapropel

Sapropels are generally reported to contain higher concentrations of trace metals relative to the surrounding sediments (in particular Fe, Mn, Al, S, Ba, V, Mo, As, I) [16, 20, 45, 21]. Enrichment of redox-sensitive elements such as barium is considered the best indicator of sapropels as confirmed by the good correlation existing between this element and the organic carbon. This led to assume that Ba, present as biogenic barite, is a good paleoproductivity proxy and the best indicator of the sediments deposited under anoxic conditions.

Another useful geochemical proxy in sapropel studies is a peak in Mn/Al, which usually delineates the post-depositional oxidation front and marks the thickness of the original sapropel. Post-depositional oxidative alteration in the sediment is documented at the top of many S1 intervals. These geochemical alterations modify the TOC profile but can be detected by studying redox-sensitive elements like Fe and Mn. During and after sapropel deposition, oxygen is consumed in situ by oxidation of organic matter. The oxygen-depleted conditions lead to remobilization and upward diffusion of Mn. When the organic carbon is consumed, oxygen diffuses into the sediments from above and oxidizes the reduced species, Mn and Fe oxyhydroxides which then precipitate at the oxidation front, enriching it in these elements.

6 Sapropel S1: case study from the Ionian Sea

Most of the information available concerning the sapropels is derived by high reso-

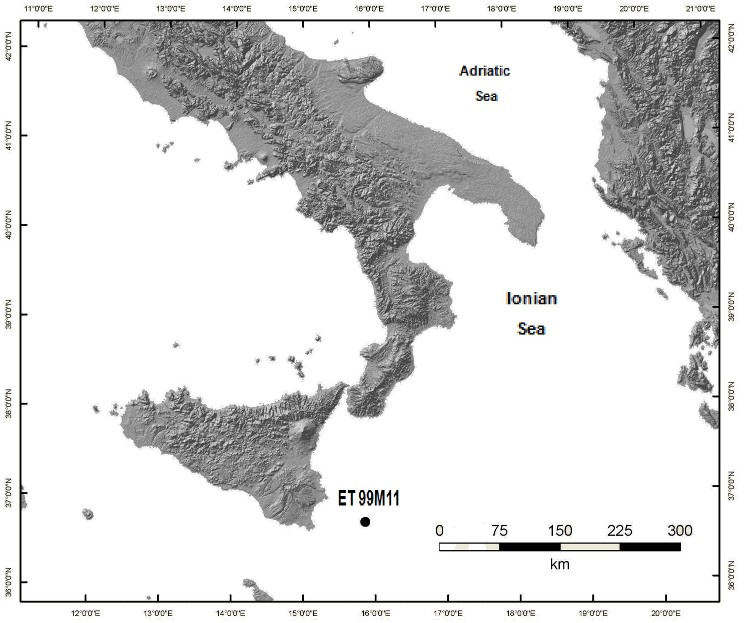

Figure 2: Location map of the core ET 99 M11.

lution studies performed on the most recent one (S1) deposited during the early Holocene as it can be easily recovered with simple coring equipment used at sea and its exact age determined by accurate radiocarbon dating methods.

Multiproxy investigations have been used to identify the precise boundary of sapropel S1 in several cores covering the entire eastern Mediterranean basin demonstrating that the timing of its deposition occurred between 9.8 - 5.7 14C Ka BP (or 10.8- 6.1 Ka cal. BP) [46]. In this paper we discuss, as case study, the results of a sapropel S1 recovered in Core ET99-M11 (Ionian Sea, Lat 36°44'04"N, Long 15°50'94"E, 2600 m water depth) (Figure 2).

This site was chosen because it is close to one of the present day sources of deep-water formation for the Eastern Mediterranean Sea, the Adriatic Sea, and it is close to the ODP Hole 964 where more than 50 sapropel layers were recovered in an excellent and complete sediment section spanning the interval from lower Pliocene to the Holocene [47].

Sapropel S1 is identified by about 38 cm of brown muddy sediment visually different from the surrounding sediments (Figure 3). The TOC content reaches a maximum value of 1.6% well distinct from the 0.2-0.3% background values. The chronological framework is constrained by three 14C AMS datings obtained from planktonic foraminifera. A multiproxy investigation carried out at high resolution (1-cm sampling corresponding to about 100 years in time resolution) and including geochemical, planktonic foraminifer assemblage and rock-magnetic analyses indicates that three different environmental scenarios related to changes in anoxia, productivity and seasonal stratifications existed during the sapropel deposition as already proposed by Rolhing et al. [48] for the Adriatic Sea and observed in other cores from the eastern Mediterranean.

1. *S1a sub-unit (from 9.8 to 8.2 Ka BP calibrated age).* A clear and drastic change of several parameters marks the beginning of the sapropel deposition at about 9.8 ka. The interval is characterized by the strongest anoxic conditions as indicated by peak values in the Barium, Ba/Al ratio and TOC content, minima in the magnetic concentration (low K and ARM values) coupled with increasing magnetic grain size (low Karm/K) and coercivity (Figure 3). This is related to magnetite dissolution and reductive diagenesis of magnetic minerals as consequence of suboxic/anoxic conditions occurring during the sapropel deposition. The planktonic foraminifer assemblage is characterized by elevated percentages of the low salinity water indicator *Globigerinoides ruber* var. *rosa* and by the increase in frequency of spinose species typical of tropical and subtropical areas such as *Globigerinella siphonifera*, *Globigerinella digitata*, *Globoturborotalita rubescens*, *Globoturborotalita tenella*, and *Globigerinoides trilobus* suggesting warmest surface water conditions with low salinity lenses. The peak abundance of *Globigerina bulloides* opportunistic species, thriving in any eutrophic setting [49], documents that sapropel formation coincided with a marked increase in nutrient availability in the surface waters.
2. *Sapropel interruption (8.2-7.9 Ka BP calibrated age).* Both TOC and Ba content decrease for an interval of few centuries, at around 8 Ka BP, marking the interruption of Sapropel S1. This interval is characterized by the peak in frequency of *Globorotalia inflata* and *Neogloboquadrina pachyderma* species living in cool and well mixed layer with intermediate to high nutrient levels [49]. Their occurrences represent a short period of improved deep water oxygenation probably triggered by cold conditions synchronous to the 8.2 Ka event [48]. Magnetic parameters also indicate a decreasing dissolution (increasing Karm and Karm/K values) related to lower anoxic conditions (Figure 3). This interruption is characterized by colder water conditions coupled to fairly high productivity.
3. *S1b sub-unit (7.9- 5.9 Ka BP calibrated age).* The reestablishment of anoxic conditions is well defined by increasing TOC and Ba values (Figure 3). The gradual decrease of *G. ruber* together with the increase of *N. pachyderma* dextral and *G. inflata* (Figure 3) marks a significant change in the upper water column. In particular the distributional trend of the last species records the development of frontal systems in the surface/sub-surface water, leading to the demise of the stratification before the end of sapropel deposition. Magnetic parameters indicate low magnetic content with increasing grain size in the first part of this interval whereas an opposite trend related to the precipitation of Fe oxides is observed at the end of the sapropel layer.

The timing of the sapropel boundaries is well defined by several proxies (TOC, Ba, magnetic parameters, planktonic

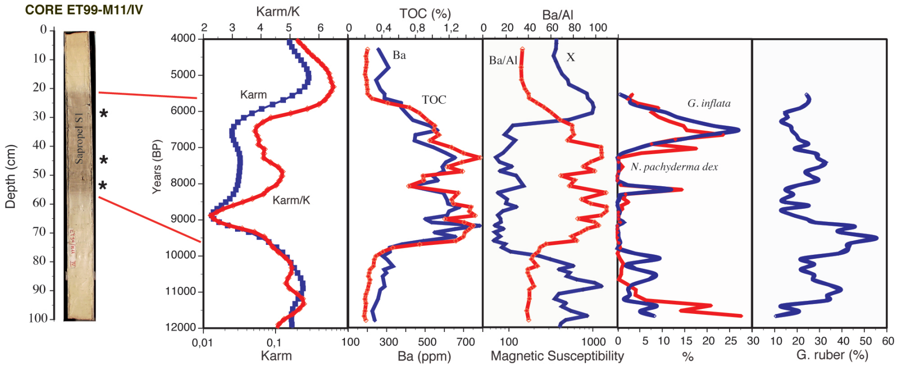

Figure 3: Magnetic, Geochemical and Micropaleonttological record of sapropel S1 from core ET99-M11. ⋆ symbol refers to ^{14}C dating points. Karm and Karm/K give indication of magnetic concentration and grain size respectively. Bariun and Ba/Al ratio are among the best indicator of Sapropels reflecting precipitation associated with primary productivity. Mass-specific magnetic susceptibility (x) was measured on discrete samples whereas Karm and Karm/K represent whole-core (U-channel) measurements. In the planktonic assemblages, the relative abundances of the deep-dwelling taxa *N. pachyderma* and *G. inflata* and the percentages of the warm subtropical species *G. ruber* allow to identify the paleoenvironmental sub-units of S1 layer (for details see text).

foraminifera) indicating that the anoxic conditions started about 9.8 calibrated Ka BP. Minimum content of magnetic particles (low susceptibility and ARM) with large grain-size (lower Karm/K) indicate that reductive dissolution (i.e. anoxic conditions) reached a peak around 9.1 Ka BP. For an interval of about 3-4 centuries centered around 8.2 Ka BP the anoxic deposition was interrupted by a re-oxygenation phase. This is evident by a decrease in TOC and Ba content, an increase in magnetic content and in planktic foraminifer microfauna indicating deep seasonal mixed layer. After the interruption, the environmental conditions show a second interval characterized by anoxic conditions that appear less developed than those in sub unit S1a. At about 5.8 Ka BP, TOC and Ba return to values close to the background indicating the end of the sapropel. Biological proxies point out to a water mass which began to mix earlier than showed by end of the lithological sapropel. Magnetic parameters show an increase in fine grained minerals as an effect of precipitation of Fe-oxides at the top of the oxidation front.

7 Conclusions

In conclusion, our high resolution multiproxy analysis of sapropel S1 from the Ionian Sea shows similar features to synchronous sapropels recovered from the eastern Mediterranean sea. A first phase of bottom water anoxia and sea surface high productivity (due to increase freshwater discharge from the continents) is followed by a short interruption in the anoxic conditions provoked by mixing of upper and intermediate water, which still sustain fairly high productivity. The third phase of sapropel S1 deposition shows that upper water stratification was generally less intense than in the first phase and that the complete demise of stratification occurred before the lithological evidence for the end of the sapropel S1.

This study demonstrates that a combination of micropaleontological, magnetic and geochemical data is a good strategy to reconstruct the relative role of the factors (productivity, bottom waters ventilation, preservation) leading to sapropels formation.

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