Proxy based reconstructions of late Miocene climatic and tectonic driven changes in the Eastern Mediterranean

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Cornelia Maria Köhler
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Gutachter:
Prof. Dr. Tilo von Dobeneck
Dr. Wout Krijgsman

Prüfer:
Prof. Dr. Thomas Pichler
Prof. Dr. Rebecca Rendle-Bühring

The courage of the water drop is to dare falling into the desert

- Touareg Saying -

It's just gone noon
Half past monsoon
On the banks of the River Nile

- Night boat to Cairo, Madness, 1979 -

The research for the thesis was carried out at:

Department of Marine Geophysics, University of Bremen, Klagenfurterstr. 2, 28359 Bremen, Germany

Palaeomagnetic laboratory ‘Fort Hoofddijk’, Faculty of Earth Sciences, University of Utrecht, Budapestlaan 17, 3584 CD Utrecht, The Netherlands

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Abstract

Numerous Plio-Pleistocene palaeoclimatic changes have been identified and quantified using proxy records archived in Mediterranean sediments. For example, the Mediterranean region experienced palaeogeographic reorganisations and an intensification of the North African monsoon during the late Miocene. In this study, the Eastern Mediterranean Metochia section (Gavdos, south of Crete) and Monte Gibliscemi section (Sicily) are studied. The sections consist of cyclic sapropel-marl sediments, and the cycles have been used to precisely tune the pattern to the appropriate orbital solutions. Therefore, the ages of palaeoenvironmental changes obtained in this study are very accurate and make it possible to compare the timing of tectonic and climatic changes affecting the region.

The marl beds were analysed using environmental magnetic and geochemical proxies to precisely date the palaeoenvironmental changes. Decreases in the dust abundance in the sediments, which can be traced by their reduced hematite content, are indicative of increases in continental humidity. In addition, because Ti is associated with windblown dust, changes in the transport mechanism of material from North Africa, i.e. aeolian vs. fluvial pathways, are identified using the ratio Ti / Al.

Eastern Mediterranean sediments consist to a large extent of terrigenous material originating from North Africa and Europe. Changes in provenance are traced in the sedimentary archive of the Metochia section, using a combination of geochemical, environmagnetic and diffuse reflectance spectrometric proxies. The different source areas and the complicated interplay of the proxy parameters are unravelled using fuzzy *c*-means cluster analysis (FCM) combined with non–linear mapping (NLM). After performing the FCM and NLM analysis, a four cluster solution has the greatest stability and describes the paleoenvironmental changes in both the African and European source areas. The data is partitioned according to high and low Aegean input cluster centres and arid vs. humid climate cluster centres. The cluster solution clearly reveals the change from predominately terrigenous input in the lower parts of the Metochia section to mainly marine input at the top of the section. In addition a shift in the paleoenvironmental conditions in North Africa from arid to more humid conditions at ~8.2 Ma is also identified.

The break-up of the Aegean landmass, traced by the starvation of the Aegean sediment source, is timed at ~8.2 Ma. Furthermore, a climatic change in North Africa towards more humid conditions is also documented in the Eastern Mediterranean sediments at ~8.2 Ma. The tectonic and palaeoclimatic changes, recorded in the Metochia sediments, are therefore coeval. In order to argue for a common cause for these changes, precise ages are required. Within a narrow time interval between approximately 8.2-8 Ma, major palaeogeographic reorganisations occurred in the Mediterranean, e.g. the opening of the Tyrrhenian Sea and changes in the configuration of the Mediterranean-Atlantic gateways. The North African monsoon is coupled with the Asian monsoon system, which also strengthened during the late Miocene as a result of a tectonic uplift phase in the Himalaya-Tibetan plateau. It is therefore possible that the palaeogeographic reorganisations in the Mediterranean and the synchronous Himalayas tectonic uplift are coupled by large scale tectonic interactions.

The late Miocene palaeoenvironmental conditions in North Africa are traced in the sedimentary archive of the astronomically tuned marls of the Monte Gibliscemi section using geochemical proxies. The terrigenous input in the section is dominated by material from North Africa, which was transported by the river systems draining into the Eastern Mediterranean: Gabes, Libyan, Eosahabi/Chad and Nile. The proxy parameters indicate that the palaeoenvironmental conditions in North Africa were humid from 9.5 Ma with a comparatively high fluvial input to the Eastern Mediterranean. Furthermore, tectonic reorganisations of the Mediterranean-Atlantic gateways pertaining to the Betic and Rifian corridors occurred. These changes were initiated at ~8.4 Ma and produced sediments in the Eastern Mediterranean which are geochemically similar to those of Messinian age in the same region. This indicates similar palaeoenvironmental conditions for the times between ~9.4-7.8 Ma and the Messinian, when the Mediterranean gradually became isolated. The geochemical proxy parameters indicate conditions of enhanced biosiliceous productivity and the clay mineralogy indicates the presence of authigenic clay formation. The increases in the geochemical ratios indicate sluggish circulation in the Mediterranean, which is thought to be related to late Miocene gateway dynamics of the Betic (southern Spain) and Rifian (Morocco) Mediterranean-Atlantic connections. The sudden termination of restrictive at ~7.8 Ma is associated with a transgression linked to the opening of the Rifian corridor.

This study shows that the late Miocene sedimentary sections provide suitable archives with which to reconstruct and precisely date palaeoenvironmental changes in the Mediterranean region. Changes in the North African palaeomonsoon are recorded by changes in dust abundance, but also by tracking changes in aeolian vs. fluvial input from the continent towards the Mediterranean. The fragmentation of the Aegean is dated precisely and the accuracy of the astronomical age enables a comparison with other tectonic reorganisations and the activity of the North African monsoon.

Zusammenfassung

An Sedimentarchiven des Mittelmeerraums wurden zahlreiche Paläoklimaänderungen im Plio-Pleistozän anhand von Proxyparametern identifiziert und rekonstruiert. Während des späten Miozäns erlebte die Mittelmeerregion paläogeographische Reorganisationen und eine Intensivierung der nordafrikanischen Monsunsystems. In dieser Studie, wurden die Metochia-Sektion (Gavdos, südlich von Kreta) und der Monte Gibliscemi-Sektion (Sizilien) des östlichen Mittelmeeres untersucht. Diese Sektionen bestehen aus rhythmischen Mergel-Sapropel Abfolgen, deren sedimentäre Zyklen entsprechenden orbitalen Zyklen zugeordnet und auf diese Weise datiert wurden. Daher ist das Alter der in dieser Studie untersuchten Sedimente sehr genau bestimmt, was wiederum ermöglicht tektonische und klimatische Veränderungen in dieser Region zu vergleichen.

Die Mergelschichten wurden mittels umweltmagnetischer und geochemischer Proxyverfahren analysiert um das genaue Alter der Paläoumweltveränderungen zu bestimmen. Eine Verringerung des Staubeintrags in die Sedimente lässt sich aus einem reduzierten Hämatitanteil bestimmen und ist bezeichnend für humide klimatische Bedingungen. Darüber hinaus wird Ti im Zusammenhang mit äolischem Staub gebracht, weshalb Veränderungen des Ti / Al Verhältnis den Transportmechanismus von nordafrikanischen Sedimentkomponenten, d.h. äolische vs. fluviale Transportwege, ermittelt.

Die Zusammensetzung der Sedimente des östlichen Mittelmeer wird beeinflusst von terrigenem Eintrag aus Nordafrika und Europa. Um Veränderungen der Herkunft der Sedimente zurückzuverfolgen, wurde die Sedimente der Metochia-Sektion mittels einer Kombination aus geochemischen, umweltmagnetischen und spektrophotometrischen Proxyparameter untersucht. Die verschiedenen Herkunftsgebiete und das komplizierte Zusammenspiel der Proxyparameter wurden mit fuzzy *c*-means cluster analysis (FCM), in Verbindung mit non-linear mapping (NLM) entschlüsselt. Nach der Durchführung der FCM und NLM Analysen, zeigte eine Vier-Cluster Lösung die größte Stabilität und beschreibt die Paläoumwelt-

veränderungen in den afrikanischen und europäischen Herkunftsgebieten am besten. Die Clusterzentren wurden unterteilt in Cluster mit hohen und niedrigen Ägäiseintrag und in arides vs. humides Klima in Nordafrika. Die Clusterlösung zeigt deutlich die Umstellung von hohem terrigenem Eintrag in den unteren Teilen des Metochia Abschnittes zu erhöhten marinen Eintrag an dem oberen Ende der Sektion. Darüber hinaus wurde eine Veränderung in den Paläoumweltbedingungen in Nordafrika von ariden zu humideren Bedingungen auf 8.2 Ma datiert

Der Zerfall der ägäischen Landmasse wurde durch die Verringerung der ägäischen Sedimentquelle aufgespürt. Der Zerfall fand um 8.2 Ma statt, als es in Nordafrika ein Wechsel zu humideren Umweltbedingungen stattfand. Die tektonischen und paläoklimatische Veränderungen in den Sedimenten von Metochia fanden daher zeitgleich statt. Falls diese Veränderungen durch eine gemeinsame Ursache stattfanden braucht man eine präzise Altersangabe. Zwischen etwa 8.2-8 Ma fanden im Mittelmeerraum große paläogeographische Veränderungen statt, z.B. die Öffnung des Tyrrhenischen Meeres und Änderungen der Mittelmeer-Atlantik Gateways. Der nordafrikanische Monsun ist gekoppelt mit dem asiatischen Monsunsystem, welches im späten Miozän intensivierte als Folge einer tektonischen Hebungsphase des Himalaya-Tibetischen Plateaus. Es ist daher möglich, dass die paläogeographischen Umstrukturierungen im Mittelmeerraum und die zeitgleiche tektonischen Hebung des Himalayas das Ergebnis von groß angelegten tektonischen Wechselwirkungen ist.

Die Paläoumweltbedingungen des späten Miozäns in Nordafrika wurden in dem Sedimentarchiv der astronomisch datierten Mergel des Monte Gibliscemi Abschnitt mit Hilfe von geochemischer Proxies rückverfolgt. Der terrigene Eintrag ist dominiert von Material aus Nordafrika, welches in den östlichen Mittelmeerraum durch die Flusssysteme Gabes, Libyen, Eosahabi / Tschad und Nil transportiert wurden. Die Proxyparameter deuten darauf hin, dass die Paläoumweltbedingungen in Nordafrika seit 9.5 Ma humid waren, mit vergleichweisen erhöhten fluvial Eingang in das östliche Mittelmeer. Darüber hinaus gab es tektonischen Reorganisationen der Mittelmeer-Atlantik Gateways (Betische und Rif Strasse). Diese Änderungen wurden um 8.4 Ma eingeleitet; es fällt auf, dass diese Sedimente des östlichen Mittelmeers geochemisch ähnlich mit messinischem Sedimenten aus der gleichen Region sind. Dies deutet auf ähnliche Umweltbedingungen zwischen 8.4-7.8 Ma und des späteren Messinium hin, als das Mittelmeer zunehmend isoliert wurde. Die geochemischen Proxyparameter zeigen Bedingungen mit erhöhter biogener Opal-akkumulation und Anwesenheit von authigenen Tonmineralen. Die Erhöhung der geochemischen Proxyparameter deuten auf eine stagnierende Zirkulation des Mittelmeers, welches im Zusammenhang mit den späten Miozän Veränderungen der Betischen (Südspanien) und Rif (Marokko) Strassen steht. Um 7.8 Ma endet die Isolation des Mittelmeers vermutlich mit der Öffnung der Rif Strasse.

Diese Studie zeigt, dass die Sedimentarchive des späten Miozän dazu geeignet sind, Paläoumweltveränderungen in der Mittelmeerregion präzise zu datieren. Veränderungen des nordafrikanischen Paläomonsuns werden durch Wechsel der Staubmenge, aber auch durch Veränderungen im äolischen vs. fluvialen Eintragsverhältnis, aufgespürt. Der Zerfall der Ägäis wurde präzise datiert und die Genauigkeit des astronomischen Alters ermöglicht einen Vergleich mit anderen tektonischen Veränderungen und der Tätigkeit des nordafrikanischen Monsuns.

Contents

1. Introduction

Northern Africa of today is an arid to hyperarid region, covered to a great extent by the vast Sahara desert. But during the late Miocene, from about 11 to 5.9 Million years before present, the palaeoenvironmental conditions of this region were remarkably different: Central North Africa was drained by river networks feeding into the Mediterranean Sea (Burke and Wells, 1989; Griffin, 2002; Griffin, 2006; Gladstone et al., 2007) and 'Mega Lake Chad' was a large water body (Fig. 1) where (semi-)aqueous fauna existed (Vignaud et al., 2002; Lihoreau et al., 2006) and early hominids lived (Brunet et al., 2005). These and many more pieces of evidence indicate humid conditions in the late Miocene times, implying that the North African monsoon belt must have expanded north of the central Saharan watershed.

1.1 Aim of the Study

This study focuses on the late Miocene, when climatic changes and tectonic reorganisations were ongoing in the Mediterranean realm. The approach and objective is to combine environmental magnetic and geochemical proxies in order to reconstruct and precisely date climate variations over northern Africa during the late Miocene with a particular emphasis on the quantification of aeolian dust abundance and river runoff. Palaeogeographic reorganisations in the Mediterranean Sea are traced by changes in sediment composition and provenance, which should occur as a result of the disappearance of a sediment source when a landmass breaks up or the appearance when a landmass emerges. Furthermore, the thesis focuses on changes in productivity caused by varying circulation patterns and intensities of the Mediterranean, themselves controlled by tectonic modifications of the gateways between the Atlantic and Mediterranean.

In the last decade, the astronomical polarity time scale was extended into the late Miocene using the palaeoclimatic and palaeomagnetic information recorded in uplifted Eastern Mediterranean marine sediments (Hilgen et al., 1995; Krijgsman et al., 1995; Krijgsman et al., 1997; Hilgen et al., 2001). These sections consist of cyclic sapropel-marl sequences and contain records of

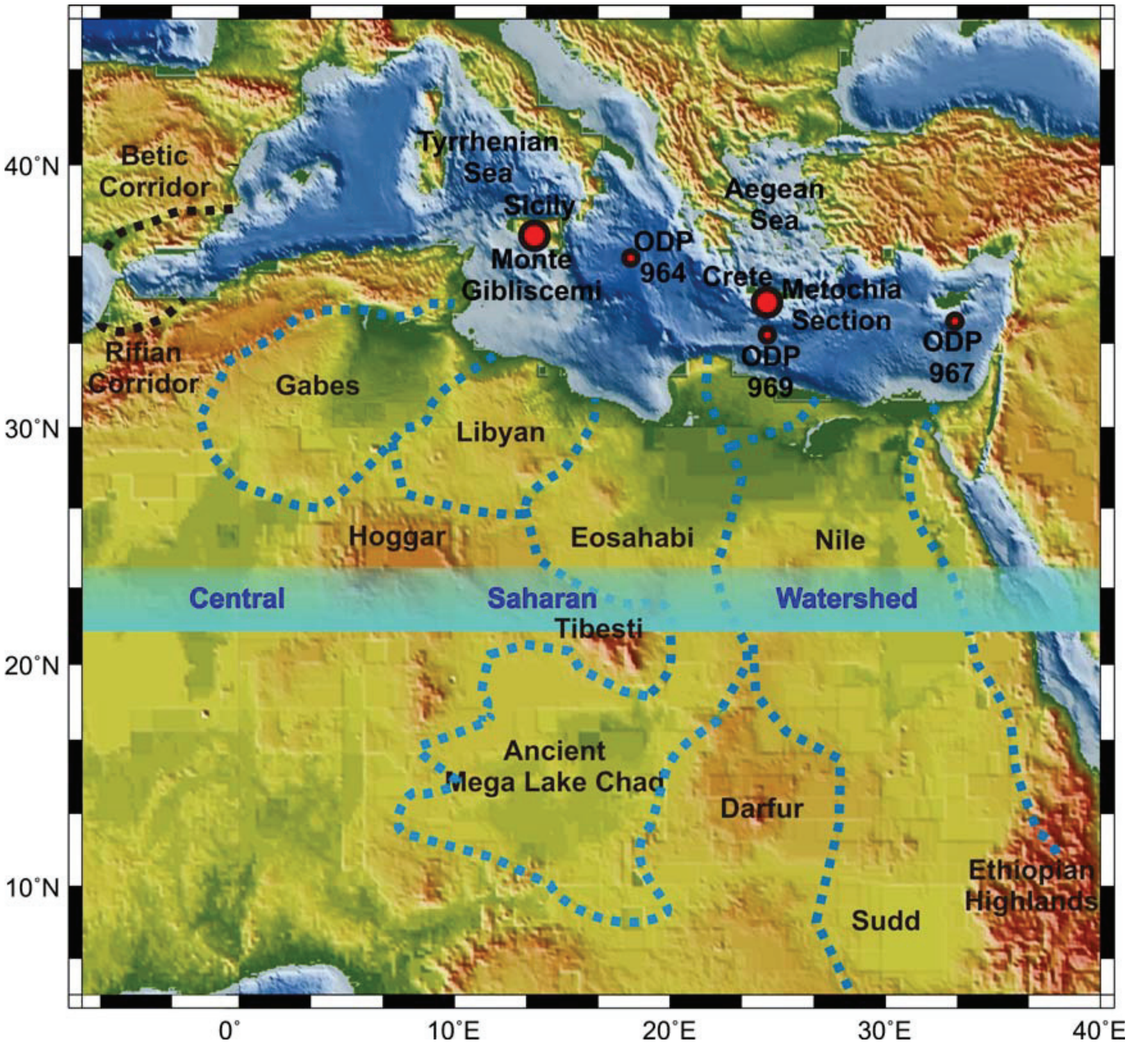


Fig 1: Map of the Mediterranean region and North Africa. The location of the sections used in this study is shown, as well as the ODP Sites. The Miocene gateways are indicated in black (Krijgsman et al., 1999; Krijgsman et al., 2000). The location of possible drainage basins in North Africa and the location of Ancient Lake Chad during the late Miocene are based on Griffin (2000), Griffin (2006) and Gladstone et al. (2007) and shown in blue. The approximate location of the central Saharan watershed is around approximately 21-22 °N (e.g. Larrasoaña et al., 2003a).

palaeoenvironmental changes in the Mediterranean region. The archives used in this study are uplifted marine sediments exposed as parts of the Metochia section, located on Gavdos Island south of Crete, and the Monte Gibliscemi section of southern Sicily (Fig. 1). The sediments in these sections also consist of intercalated hemipelagic marls and sapropels, which can be placed within an orbitally tuned chronology (Hilgen et al., 1995; Krijgsman et al., 1995). Although the organic-rich sapropel layers are too intensely modified by diagenetic processes to yield reliable information on aeolian dust content, the more oxic marl layers should carry a reliable pristine signal and were thus in the focus of this study.

1.2 North African Climate

The North African monsoon is influenced by the position of the tropical rainbelt, which is connected with the intertropical convergence zone (ITCZ) (Griffin, 2002; Jury and Mpeta, 2005). The ITCZ is a low pressure belt of rising air, which moves within the tropics in the course of a year. The northward penetration of the tropical rainbelt reaches a maximum during the summer months and is ultimately controlled by the northward extent of the ITCZ (Chao and Chen, 2001).

The northern boreal summer ITCZ limit determines the distribution of rainfall and the prevalence of more arid or more humid environmental conditions in North Africa. Its position is influenced by several factors and on various timescales. Long term aridification in North Africa results from the northward movement of Africa due to continental drift (Axelrod and Raven, 1978), changing the areas affected by the ITCZ. Short-term and even interannual changes in rainfall in North Africa can result from changes in the surface temperatures of the world oceans (Fontaine and Janicot, 1996; Janicot et al., 1997; Hunt, 2000). North African rainfall decreases with warmer oceans surrounding the continent, by decreasing the land-water temperature contrast (Giannini et al., 2003). On an orbital timescale, the North African monsoon also shows variation, especially influenced by the precession cycle; more humid conditions occur during precession minima (Lourens et al., 2001; Larrasoaña et al., 2003a; Tuenter et al., 2003).

Tectonic uplift also shifts the position of the ITCZ, an example being the late Miocene uplift phase of the Himalaya-Tibetan plateau (Clemens and Prell, 1991; Kroon et al., 1991; Filippelli, 1997; An et al., 2001). During northern hemisphere summer, the Himalaya-Tibetan plateau is heated and air rises, shifting the tropical rainbelt northward (Ramstein et al., 1997; Fluteau et al., 1999). Increased wind strength across the Arabian Sea from ~8.5 Ma (Kroon et al., 1991) carries moisture towards the Indian continent. In Africa, there are also ‘barriers’: the Hoggar massif in the West and the Ethiopian highlands in the East (Fig. 1) (Axelrod and Raven, 1978; van Zinderen Bakker and Mercer, 1986). The Hoggar massif influences the heating of North Africa and hence the position of the ITCZ (Drobinski et al., 2005). The Ethiopian highlands act as a rainshadow, forming a barrier that blocks the moist winds originating in the Indian Ocean (Bonnefille, 1985). During the late Miocene, the Ethiopian highlands experienced an uplift phase (Gani et al., 2007), which is an important aspect as it is the catchment area of the Blue Nile. .

1.3 Mediterranean Tectonics

The Mediterranean basin is an active tectonic region, where Africa and Europe converge and the African plate is subducted under the European margin (e.g. Krijgsman, 2002; Meulenkamp and Sissingh, 2003). During convergence in the early-middle Miocene, the eastern Tethyan Gateway, connecting the Mediterranean with the Indian Ocean, was gradually closed (Harzhauser et al., 2002; Krijgsman, 2002). Other late Miocene palaeogeographic changes included the opening of the Sea of Crete in the Aegean region and the Tyrrhenian Sea in the central Mediterranean.

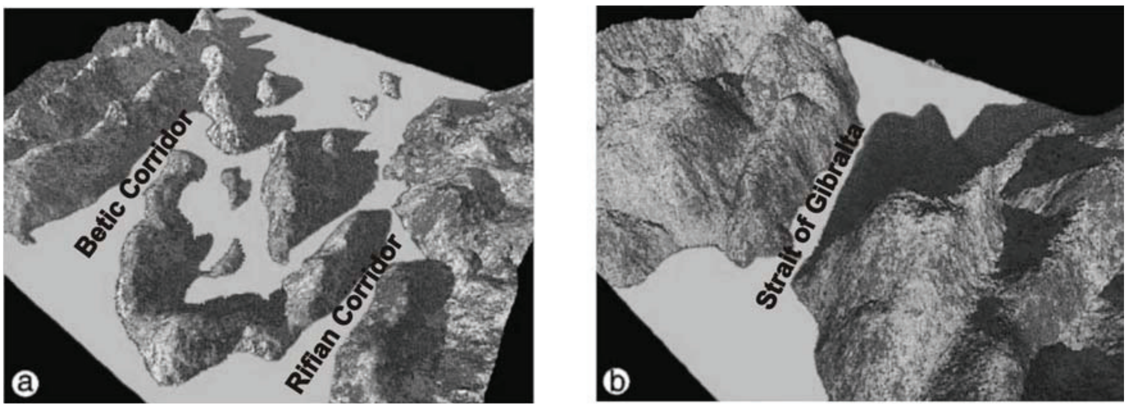


Fig. 2: The late Miocene Mediterranean-Atlantic gateways and the modern gateway are shown. The figure is taken from Krijgsman (2002).

During the late Miocene, two western gateways connected the Mediterranean with the Atlantic: the Betics in southern Spain (Soria et al., 1999) and the Rifian corridor in northern Morocco (Fig. 2) (Krijgsman et al., 1999). The western Atlantic gateways were also affected by the large scale late Miocene tectonic reorganisation (Krijgsman et al., 1999; Soria et al., 1999). Disruptions in the Atlantic-Mediterranean connections led to the gradual isolation of the Mediterranean basin, eventually causing the Messinian Salinity

Crisis, which resulted in the desiccation of the Mediterranean (Hsü et al., 1973).

1.4 Proxy Parameters

Proxy parameters are measurable manifestations and hence approximations of past or present system conditions in the absence of a physical law or formula. Proxies for past climates and environments have been deduced from chemical, biological or physical properties of the studied sediments, providing a detailed insight into past conditions and changes. Since palaeoclimatic proxy parameters are indirect measures of climate they often reflect multiple aspects of the climate system leaving space for uncertainty and interpretation. The choice of suitable and selective proxies is crucial and interpretations must be treated with caution as these parameters provide only approximations of past environments and their validation, calibration and interpretation often relies only on modern analogues.

The exact position of the ITCZ cannot be easily determined in palaeoclimatic studies, but the palaeoenvironmental changes resulting from its influence can be traced using proxy parameters. Conditions are arid when the ITCZ does not cross the central Saharan watershed, enhancing aeolian dust production and accumulation, which can be detected in Mediterranean sedimentary sections. As an example, a suitable proxy to identify such conditions is the abundance of detrital hematite in Mediterranean sediments, which depends on both arid conditions to drive its formation in African soils and the strength of the monsoonal winds that transport it northwards out the sea.

1.5 Thesis outline

The first proxy study presented in chapter 2 identifies changes in sediment provenance recorded in the Metochia section south of Crete. The archive spans a time interval during the late Miocene when the Aegean landmass fragmented and an intensification of the North African monsoon occurred. The multiproxy parameter set consists of environmagnetic, geochemical and spectrometric parameters capable of distinguishing between Aegean and North African terrigenous source areas. The resulting multivariate data set was analysed using fuzzy *c*-means cluster analysis (FCM) and non-linear mapping (NLM). The statistically and palaeoenvironmentally most coherent model consists of four clusters, divided on the basis of decreasing terrigenous sediment supply from the Aegean landmass and changes in aridity and humidity on North Africa. The decrease in terrigenous input from the Aegean was associated with the fragmentation of the Aegean landmass. This major tectonic change is found to be nearly synchronous with changes from arid to more humid conditions in North Africa, which are related with an enhancement of the North African monsoon system. The analysis of the Metochia archive revealed that the Aegean landmass fragmented coevally with a climatic change in North Africa at ~8.2 Ma.

The tectonic and climatic information preserved in the Metochia marls are discussed in chapter 3. This manuscript focuses on the coeval timing of the fragmentation of the Aegean landmass and the intensification of the North African monsoon. It is hypothesised that these events may have been driven via a large scale link with the late Miocene tectonic uplift of the Himalayas. During the Tortonian, the entire Mediterranean was undergoing a tectonic reorganisation, for example the fragmentation of the Aegean landmass, the opening of the Tyrrhenian Sea and changes in the Mediterranean-Atlantic gateways. These are likely linked to a common cause (e.g. Jolivet and Faccenna, 2000; Meulenkamp and Sissingh, 2003). The North African monsoon is coupled to the Asian monsoon system (Griffin, 2002), which intensified at ~8 Ma as a result from a tectonic uplift pulse of the Himalaya-Tibetan plateau. It is therefore possible that the Alpine-Himalayan wide tectonic changes during the late Tortonian affected the entire African-Eurasian plate boundary. The mechanistic link would suggest a causal relationship between Alpine-Himalayan wide tectonic events and global climate.

The manuscript presented in chapter 4 focuses on the sedimentary archive of the Monte Gibliscemi section, Sicily. The section is located close the African continent and in the proximity of to the drainage basins of ancient rivers that would have drained the continent towards the central Mediterranean. The hemipelagic marls were analysed geochemically and show that the palaeoclimatic conditions in central North Africa were humid and that the river systems were active. Monte Gibliscemi’s location close to the Sicilian Sill, which forms a barrier between the Western and Eastern Mediterranean basins, makes it sensitive to changes in the Mediterranean-Atlantic connection. Tectonic changes in the gateways, first the near closure of the Betic corridor and then the opening of the Rifian gateway reduced the circulation of the Mediterranean Sea. The geochemical proxy record and the clay mineralogy indicate that these changes led to the formation authigenic clays in the sediment from ~8.4-7.8 Ma.

2. Tracking provenance change during the late Miocene in the Eastern Mediterranean using geochemical and environmental magnetic proxies[1]

Abstract

The astronomically tuned marls of the Metochia section (Gavdos, Greece) are analyzed using geochemistry, environmental magnetics and diffuse reflectance spectrometry to study late Miocene paleoenvironmental changes in the Eastern Mediterranean region. Fuzzy *c*-means cluster analysis (FCM), combined with non–linear mapping (NLM), is performed on a multiproxy data set to identify and characterize terrigenous source areas from North Africa and the Aegean margin. The proxy parameters included in the FCM are selected on the basis of their univariate characteristics and ability to trace input changes from regional source areas. The magnetic hard isothermal remanent magnetization (HIRM), calculated on a carbonate-free basis and the elemental ratio Ti / Al are used to distinguish aeolian dust and transport mechanisms of material originating from North Africa. Mass accumulation rates of Al and Ni are employed to represent the input of terrigenous material originating from the Aegean region. Finally, $CaCO_3$, calculated from Ca, provides information concerning marine productivity and Mn / Al is used to trace diagenetic reprecipitation in the marls. After performing the FCM and NLM analysis, a four cluster solution shows the greatest stability and describes the paleoenvironmental changes in both the African and European source areas. The cluster solution clearly reveals the change from input dominated by terrigenous material in the lower parts of the Metochia section to marine input at the top of the section, but also changes in the paleoenvironmental conditions in North Africa from arid to more humid conditions at ~8.2 Ma.

[1] This chapter has been submitted for publication as: C.M. Köhler, D. Heslop, M. Dekkers, W. Krijgsman, D.J.J. van Hinsbergen, T. von Dobeneck: Tracking provenance change during the late Miocene in the Eastern Mediterranean using geochemical and environmental proxies. *Geochemistry, Geophysics, Geosystems*

1. Introduction

The relationships between different proxies, especially when combining geochemical and environmental magnetic parameters, are often complex. However, subtle, but important changes can be identified and better interpreted when applying fuzzy c-means cluster analysis (FCM) (Bedzek, 1981), commonly combined with the non–linear mapping (NLM) dimension reduction technique (Sammon, 1969). Both techniques show groupings of the samples and when these are similar, the cluster solution becomes more interpretable (Vriend et al., 1988; Dekkers et al., 1994). FCM is a powerful tool to unravel the changes in paleoenvironmental information recorded in sediments (Dekkers et al., 1994; Kruiver et al., 1999; Schmidt et al., 1999; Urbat et al., 2000; Hanesch et al., 2001; Kruiver et al., 2002; Vlag et al., 2004). It is important to have well-defined input variables with precise interpretations assigned to them, in order to be able to understand and interpret the clustering solution. The aim of this study was to investigate uplifted marine marls from the Metochia section on Gavdos Island (located to the South of Crete (Krijgsman et al., 1995)) using FCM applied to a carefully chosen geochemical and environmental magnetic proxy data set, to discriminate between the different European and African input sources and visualize changes in the sediment provenance.

The astronomically dated marl-sapropel sequences of the Metochia B and C sections on Gavdos Island are an environmental archive spanning the time interval 9.4 – 6.9 Ma, from which sediment supply and the development of source areas such as the former southern Aegean land mass and northern Africa can be reconstructed (Fig. 1). Because sulfate reducing diagenesis in sapropel layers can modify strongly the primary terrigenous input signal (e.g. Passier et al., 1998), only samples from the marls were selected for analysis.

A variety of different approaches have been adopted previously in an attempt to identify the various sources that contribute material to the Eastern Mediterranean Sea. For example, Venkatarathnam and Ryan (1971) used seafloor surface sediments and Foucault and Mélières (2000) employed Pliocene sediments to associate different clays and clay mineral assemblages with European and African sources. North African dust can be traced by identifying the antiferromagnetic minerals hematite and goethite, which provide information pertaining to the abundances of transported dust (Larrasoaña et al., 2003a). Geochemical data concerning the provenance of sediments in the Eastern Mediterranean region provide a means with which to discriminate between materials originating from the Aegean realm and North Africa (Bergametti et al., 1989; Wehausen and Brumsack, 1998; Wehausen and Brumsack, 2000; Lourens et al., 2001).

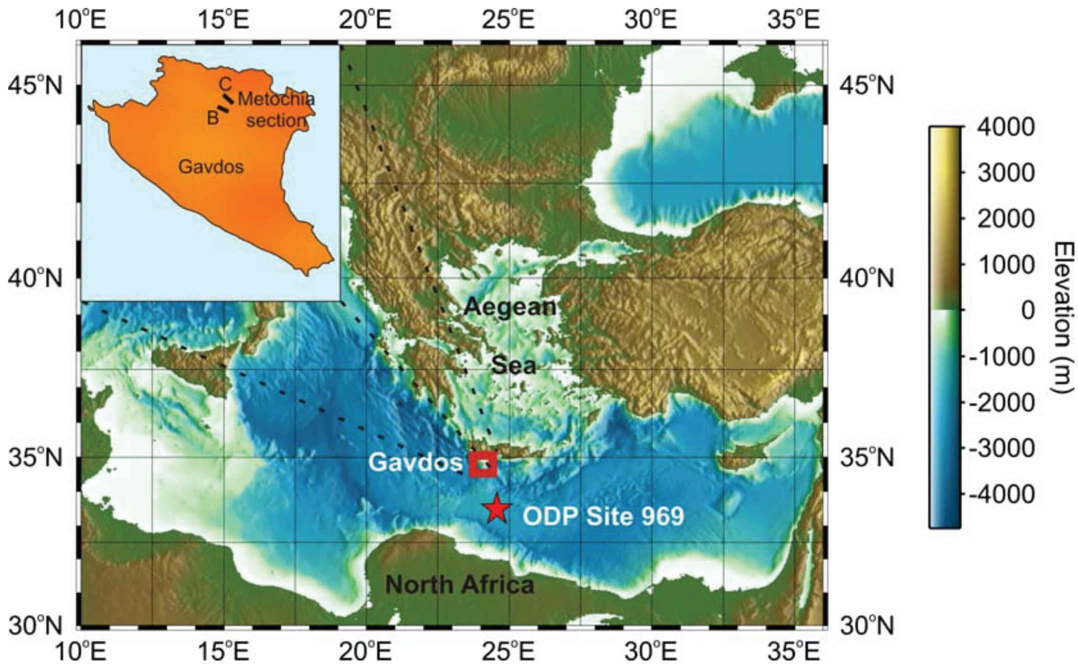


Fig. 1: Present day map of the Eastern Mediterranean showing the location of the Metochia section on Gavdos (South of Crete) between the Aegean rim and North Africa and the location of the ODP Site 969. The inset panel shows Gavdos Island and the location of the Metochia section.

Previous investigations have demonstrated that the elements Al and Ca, and the ratios Ni / Al, Si / Al, Fe / Al and Ti / Al are indicative of different sediment origins and can be linked to the environmental conditions in certain source areas (e.g. Bergametti et al., 1989; Wehausen and Brumsack, 1998; Lourens et al., 2001). Ni is associated with the ultramafic rocks, which in this region are exclusively found in the Aegean region, whereas Si / Al and Ti / Al represent the African source area (Wehausen and Brumsack, 2000). Based on these classifications, Lourens *et al.* (2001) and Larrasoaña *et al.* (2003a) employed specific elemental ratios in order to reconstruct changes in the Pliocene-Pleistocene climate of North Africa. The geochemical proxies of the

Miocene sediments are of interest because they can be compared to the Pliocene records of Wehausen and Brumsack (1998; Wehausen and Brumsack, 2000), who worked on ODP Site 969 located to the South of Crete, which contains a mixture of sediments of Aegean and North African sources.

By making use of the precise astronomical age model of the Metochia sediments and selecting a meaningful proxy data set, we will show that FCM can identify changes in the origin of the sediments and paleoenvironmental conditions in the source areas.

2. The Metochia section and sampling

The island of Gavdos is situated 30 km to the South of Crete and represents the southernmost exposed part of the Hellenic arc system. The Metochia section is located on the northeastern part of the island and is composed of open marine sediments (Fig. 1). The sedimentation history of the Gavdos basin and the position of the Metochia section on the Aegean rim make it an ideal archive for recording regional paleoenvironmental events. Paleobathymetry information from the Metochia section based on the contribution of planktonic species to the total foraminifera assemblage indicates that Gavdos was situated at a water depth of more than 1000 - 1200 m during the late Miocene (van Hinsbergen and Meulenkamp, 2006).

The section itself is composed of two subsections, Metochia B and C, which display a characteristic alternating pattern of intercalated hemipelagic marls and sapropels (Krijgsman et al., 1995). The base of Metochia B consists of shallow marine sands (Postma et al., 1993; Krijgsman et al., 1995), which were not considered as part of this study as they mainly consist of locally derived clastic sediments. The Metochia sediments were dated precisely using a combination of cyclo-, magneto- and biostratigraphic methods (Hilgen et al., 1995; Krijgsman et al., 1995). Krijgsman *et al.* (1995) identified 17 magnetic polarity reversals within the Metochia section that could be correlated to the geomagnetic polarity time scale (GPTS) (Cande and Kent, 1995) and spanned the age interval 9.6 – 6.9 Ma.

The Metochia sapropel pattern was originally tuned by Hilgen *et al.* (1995) to the 65ºN summer insolation curve of Laskar (1990), with modern-day values for dynamical ellipticity of the earth and tidal dissipation by the moon. Individual sapropels were correlated to precession minima, whilst small and large-scale sapropel clusters were matched to maxima in the 100 and 400 kyr eccentricity cycles, respectively (Hilgen et al., 1995). The coherent cyclicity of the Metochia sediments resulted in the section becoming a fundamental component of the stratigraphic framework for the Mediterranean Upper Miocene and played a key role in the extension of the astronomical polarity time scale from ~5 – 10 Ma (Hilgen et al., 1995). Revised sapropel ages in accordance with the more recent orbital solution of Laskar *et al.* (2004) are employed in this study. These updates to the age model are in the range of 0.15 – 15.2 kyr, resulting on average in a 2.5 kyr shift to younger ages.

The Metochia sample set collected during the investigation of Krijgsman *et al.* (1995) was used in this study. In general, 2 – 4 virgin samples were selected from each marl bed (average spacing = 20cm), with the number of specimens depending on the thickness of each bed and the availability of material. A total of 208 samples were investigated, however, no sample material was available for three marl intervals (Metochia B: 8.839 – 8.821 Ma, Metochia C: 7.826 – 7.817 Ma and 7.798 – 7.786 Ma).

3 Methods

In order to reconstruct a sedimentary history from the Metochia section it is necessary to define proxies that have the ability to discriminate between the terrigenous contributions originating from the various source areas around the Eastern Mediterranean. Source area indicative geochemical ratios will be combined with environmental magnetic parameters and diffuse reflectance data in order to characterize the Metochia sediments. As the information provided by the various measured proxies is too complex to be unravelled by uni- or bivariate approaches, multivariate fuzzy *c*-means cluster analysis (FCM) and non–linear mapping (NLM) methods will be employed to determine how the different proxies are environmentally associated and controlled.

3.1 Geochemistry

Terrigenous Al is a constituent of clays and silts, which are transported to the Eastern Mediterranean from the Aegean region and North Africa via both aeolian and fluvial pathways (Foucault and Mélières, 2000; Lourens et al., 2001). Al abundance also plays an important role in the geochemical interpretation because the abundances of other elements can be normalized against it to compensate for dilution effects (Wehausen and Brumsack, 1998). $CaCO_3$ was calculated from Ca, assuming that 98% of the Ca resides within $CaCO_3$, providing a proxy representing marine biogenic input. In contrast to the sapropel layers that form under humid conditions, with high fluvial runoff and having a low $CaCO_3$ content (Schenau et al., 1999), the Eastern Mediterranean marls are rich in $CaCO_3$. The Pleistocene marls from this region, for example, contain between ~52 and ~62 % $CaCO_3$ (Wehausen and Brumsack, 1998). More arid conditions during periods of marl formation result in reduced fluvial runoff and increased transport of northern African aeolian material to the Eastern Mediterranean.

Dust originating from North Africa is rich in both Si and Fe, hence the ratios Si / Al and Fe / Al have previously been used to identify and trace North African aeolian dust (Bergametti et al., 1989; Chiapello et al., 1997). Fluctuations in Ti / Al reflect variations in the relative importance of fluvial and aeolian transport mechanisms, as Ti is associated with aeolian dust, whereas Al is found in both aeolian and smectite-rich fluvial materials (Lourens et al., 2001), but can also originate from the Aegean. When both Ti and Al are associated with a North African origin, higher values of Ti / Al therefore indicate a predominantly aeolian flux, whilst lower ratios signify fluvial transport (Lourens et al., 2001; Larrasoaña et al., 2003a).

The characteristic Jurassic and Cretaceous ultramafic rocks which are found in mountain ranges in the Aegean landmass (Jacobshagen, 1986), including modern day Crete, provide a signature which can be distinguished from North African terrigenous material. Ni and Mg are associated with these rocks (Wedepohl, 1969; Wehausen and Brumsack, 2000). Ni is only associated with Aegean sources, whereas Mg can also be present in clays derived from North Africa, e.g. palygorskite, thus the information of the

Mg-content is therefore ambiguous. For this reason, Ni, instead of Mg, is used as the proxy to trace the Aegean source area.

To test for the possible influence of diagenetic processes on the marl samples and the presence of unidentified oxidized sapropels in the section, the Mn / Al ratio is compared to the Fe / Al ratio for coincidence of peaks. Both Mn and Fe have been described as being mobilized and reprecipitated as a result of the downward movement of the oxidation zone during early diagenesis (van Santvoort et al., 1997; Kouwenhoven et al., 1999; Kouwenhoven et al., 2003). The detection of oxidized sapropels is of importance as Ni is enriched in sapropels (van Santvoort et al., 1997; Wehausen and Brumsack, 2000) and the diagenetic enrichment has to be distinguished from the detrital Aegean signal.

To determine the geochemical composition of the Metochia marls, X-ray fluorescence analysis (XRF) was performed on the planar surfaces of solid cylindrical samples (~4 g) using a *Spectro Xepos* energy dispersive polarization X–ray fluorescence analyzer. The measuring time was 300 seconds per sample. Accuracy and precision were checked using the internal marine standard Mag-1 (certified USGS marine sediment standard reference material) and duplicate measurements. The measurement errors are <5 % for the elements that will be employed in this study. For Al and Ni, mass accumulation rates (MAR) were calculated by forming the product of the XRF results (mg / kg), bulk density (kg / m^3) and sedimentation rate (m / kyr) (Peterson et al., 2000).

3.2 Environmental Magnetism

To complement the geochemical dataset a number of indicative environmental magnetic parameters were measured. Magnetic susceptibility and ‘hard’ isothermal magnetization (HIRM) are important tracers of terrigenous material. Values of volume dependent magnetic susceptibility (κ) when calculated on a carbonate free basis (cfb) provide a measure of the total magnetic mineral concentration, which in the case of the marine sediments will be predominantly linked to the terrigenous magnetite content (deMenocal et al., 1991). Paramagnetic clay minerals can also contribute to κ, becoming important when the concentration of magnetite is low. κ was measured on individual samples using a *KLY2 AGICO Kappabridge* instrument (noise level ~4×10^{-8} SI).

The iron oxides hematite and goethite are important constituents of North African dust (e.g. Bloemendal et al., 1992). These minerals are antiferromagnetic and their high coercivity means that they are magnetically "harder" than ferrimagnetic materials such as magnetite. The carbonate free 'hard' isothermal remanent magnetization ($HIRM_{cfb}$) provides a measure of the antiferromagnetic component. Each sample was pulsed in a 2.5 T field in order to at least partially magnetize the hematite and goethite component, yielding the isothermal remanent magnetization ($IRM_{2.5T}$), which was subsequently overprinted with a backfield of –0.3 T to allow the calculation of HIRM (Stoner et al., 1996). After each treatment in the pulse magnetizer the individual samples were measured on a 2G *DC*-SQUID magnetometer (noise level ≤ 1×10^{-12} Am^2). Based on the measurements of HIRM the combined abundance of the hematite and goethite components can be assessed.

3.3 Diffuse Reflectance Spectrometry

In addition to their magnetic properties, hematite and goethite are also strong pigmenting minerals and even in trace amounts can exert a significant influence on the reflectance characteristics of a sediment (Deaton and Balsam, 1991). Diffuse reflectance spectrometry (DRS) measures the ratio of incident to reflected light intensity as a function of wavelength. DRS spectra spanning the wavelengths of visible light (400-700 nm, in 10 nm increments) were determined for all the samples using a Minolta CM - 2002 Spectrophotometer. The dominant wavelength of a measured spectrum (which is linked to the hue of the sediment color) can provide important information concerning hematite and goethite content (Cornell and Schwertmann, 1996). Specifically, the dominant wavelength of mixtures of hematite and goethite are in the range of 580-595 nm (Madeira et al., 1997). The dominant wavelength of each of the Metochia samples was determined from the measured reflectance spectra in order to identify possible compositional changes within the terrigenous component.

3.4 Fuzzy c-means clustering and non-linear mapping

FCM is a multivariate partitioning technique (Bedzek, 1981), which was applied to the Metochia data to help quantify the subtle relationships among the different measured proxies. FCM has been successfully applied in previous environmental studies, which have combined geochemical and environmental magnetic proxies (Dekkers et al., 1994; Hanesch et al., 2001; Kruiver et al., 2002; Vlag et al., 2004). The aim of FCM is to define the similarities and dissimilarities that exist between a collection of points within a multivariate data space. A given number of so-called cluster centers are represented as points within the data space and the similarity of each measured sample can be compared to the individual cluster centers based upon a predefined distance metric. Memberships define the similarity of the points to the cluster centers, with values varying between zero (no similarity) and one (identical) for each cluster center, under the constraint that for any given sample the memberships across all the cluster centers must sum to unity. The identification of the optimal cluster centers is an iterative procedure based upon an objective function which aims to find a balance between producing the maximum separation between the cluster centers and minimizing the combined distance between the samples and the cluster centers. It is possible that a given sample may not show a strong similarity to any of the given cluster centers, in this study any sample for which $u_2 > 0.75u_1$ (with u_1 indicating the highest membership and u_2 the second highest membership) was assigned as a "transitional" case. The fuzzy exponent, q, which determines the fuzziness of the model, was chosen to be $q = 1.5$ (Bedzek et al., 1984). Before performing the FCM the distribution of each parameter was normalized with a mean of zero and standard deviation of one in order to ensure that all parameters have equal weight in the analysis. A number of FCM solutions were calculated with between two and nine cluster centers using the software of Vriend *et al.* (1988). The stability of the cluster solution was tested by repeating the analysis from different starting configurations, but also by including and excluding different input parameters. The final solution of the model is therefore considered to be robust.

Non–linear mapping (NLM) is a dimension reduction technique, which attempts to form an undistorted low-dimensional (typically two-dimensional) representation of a multi-dimensional data set (Sammon, 1969). This is achieved by iteratively searching for a configuration of points in a low dimensional space that accurately preserves the distance matrix which describes the relative positions of the data points in the high dimensional measurement space. When FCM and NLM show similar sample groupings, it supports and adds meaningfulness to the cluster solution (Vriend et al., 1988; Dekkers et al., 1994). NLM has therefore been used in combination with FCM to confirm the validity of a given cluster solution.

Input parameters for the FCM and NLM were chosen carefully under the criterion that in order to be included in the analysis a proxy must have a clear interpretation and not represent the same process as another proxy already selected for analysis. In this way the input parameters were not biased due to over-representation of any given process.

4. Results and Interpretation

4.1 Univariate Statistics

Source identification and variation of terrigenous input from different Eastern Mediterranean sources will be discussed based on the measured geochemical and magnetic proxies. Given that the marl – sapropel formation mechanism has not changed from the late Miocene through to the Pleistocene (Schenau et al., 1999), the results of this study are compared to data of Pliocene sediments from ODP Site 969 located to the South of Crete (Fig. 1). Due to different land-water distributions resulting from tectonic reorganizations during the late Miocene in the Aegean region (Meulenkamp and Hilgen, 1986; Meulenkamp et al., 1994; van Hinsbergen and Meulenkamp, 2006), comparisons may not be straightforward. However, both archives, ODP Site 969 and the Metochia section, should contain a mixed terrigenous component from the Aegean region and Northern Africa.

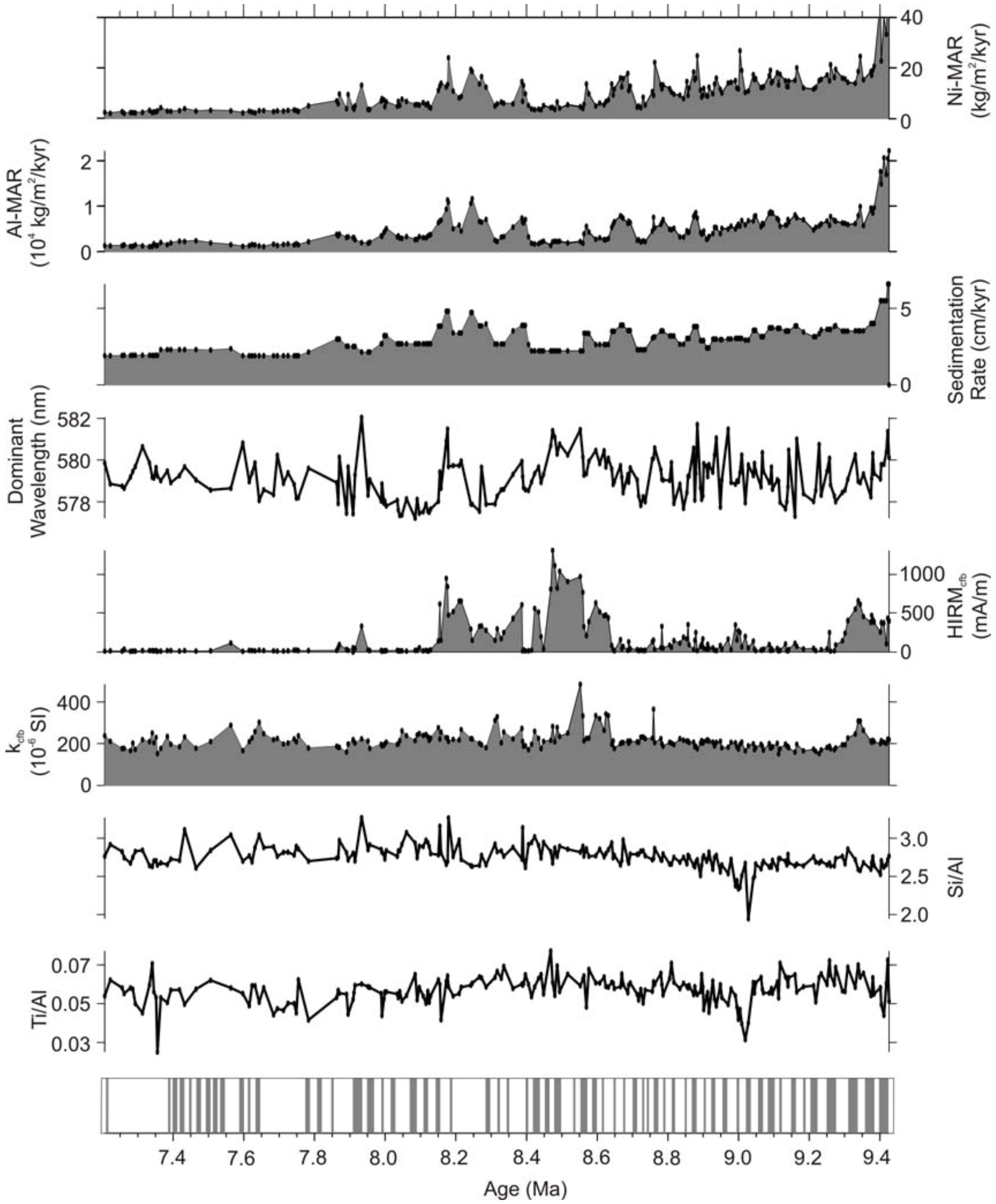


Fig. 2: Age profiles of geochemical, environmental magnetic and DRS proxies. The elemental abundances are normalized to Al abundance or shown as MAR, $HIRM_{cfb}$ is the "hard" isothermal remanent magnetization on a carbonate free basis. The stratigraphy of the Metochia section is shown in the lower most panel. Sapropels are shown by the grey intervals, the homogenous marls, which are used in this study, by the white intervals.

The Ti / Al ratios of the Metochia marls are on average 0.057 (Fig. 2), which is lower than the values of Pleistocene marls of ODP Site 969, which are in the range of ~0.07 – 0.08 (sapropels values are ~0.06) (Wehausen and Brumsack, 2000). Higher Ti / Al ratios observed in Site 969 marls are associated with Ti-rich North Africa aeolian dust (Lourens et al., 2001;

Larrasoaña et al., 2003a). The marls of the Metochia section have low Ti / Al ratios and are in the range of the Pliocene sapropels which could result from reduced aeolian dust input, lowering the Ti content of the marls. Alternatively, increased Al input could also lower the ratio. Especially when the Aegean input is high and dominates the terrigenous input, Al influences and lowers the Ti / Al ratio, it is therefore no longer a proxy for aeolian dust input. However, when the Aegean terrigenous contribution decreases, the Ti / Al ratio is influenced by the Ti associated with aeolian dust derived from North Africa and hence represents the aeolian fraction associated with marls.

The North African source proxy Si / Al shows no major variations and the mean value is ~2.75 and therefore slightly below the values reported for Pliocene sapropel–marl sediments (Wehausen and Brumsack, 2000), but within the range of modern Saharan dust values (Bergametti et al., 1989) (Fig. 2), indicating that there was a continuous input of North African material to the Metochia section during the late Miocene. The lower values than in the Pleistocene records can be linked to higher Al-input during the Miocene. Prior to ~8.5 Ma, the Si / Al ratio is slightly below the average, but then increases to a value of 2.8. The Fe / Al ratio is generally lower than the values reported for ODP Site 969 (Wehausen and Brumsack, 2000), but the values are still within the range of 0.35 – 0.7 reported for African dust by Bergametti *et al.* (1989) (Fig. 2). The low Fe / Al ratios and the presence of a minimal number of elevated values in the data illustrate that remobilization of Fe seems unlikely to have occurred in the vast majority of the marls. However, in isolated cases, the peaks in Fe / Al correspond with peaks in Mn / Al, indicating the possibility of remobilized Fe and Mn as the result of early diagenesis (Fig. 3) (van Santvoort et al., 1997). When these intervals coincide with elevated Ni values, being above the background values, it suggests diagenetic overprint in the marls, e.g. at ~9.0, ~7.9 and ~7.3 Ma.

Apart from the isolated diagenetically altered samples, Ni is a proxy unambiguously associated with ultramafic rocks, exclusive to the region to the Aegean landmass. The Ni / Al ratio (Fig. 3) shows important variations through the Metochia section, with the highest values occurring from the base of the record until ~8.4 Ma. These higher values are within a similar range as the ODP Site 969 material, containing a mixture of sapropels and marls, sapropels commonly being enriched in Ni (Wehausen and Brumsack, 2000). The Ni / Al values decrease after 8.4 Ma. As most samples of the Metochia section are unaffected by diagenetic alteration of the Ni / Al ratio, the values imply a change in the source area of the material reaching the Metochia section.

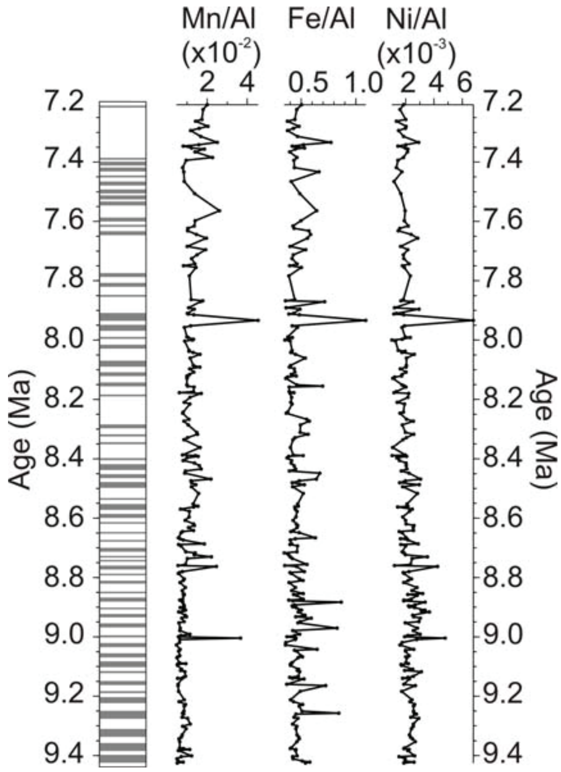


Fig. 3: Age profiles of elemental abundances normalized to Al abundance. Where high values of Mn / Al, Fe / Al and Ni / Al correspond, e.g. at ~7.9 Ma, there is the presence of a hidden sapropel (a sapropel layer has also been reported in this time interval in the Monte Gibliscemi section on Sicily (Hilgen et al., 1995)). The stratigraphy of the Metochia section is included, grey intervals are sapropels, white intervals are homogenous marls.

κ_{cfb} has low values at the base of the Metochia section, which increase steadily to a maximum at ~8.5 Ma and lie in the range ~170 - 350 x 10^{-6} SI (Fig. 2). After 8.5 Ma the κ_{cfb} decreases again to values between 180 – 270 x 10^{-6} SI. The low κ_{cfb} values suggest that the signal of the marls is dominated by the presence of paramagnetic clay minerals and that magnetite is only a minor component. However, κ_{cfb} remains fairly constant throughout the section. There is a minor gradual decrease in the upper part of the section which should imply a drop in detrital magnetite, as the overall sediment input decreases in this time interval. At the top of the section, the κ_{cfb} does not follow the trend of the sedimentation rate, which steadily decreases throughout the section and shows a drop after 8.1 Ma (Fig. 2).

HIRM indicates that the antiferromagnetic content of the marls varies substantially (Fig. 2). Prior to 8.1 Ma, the $HIRM_{cfb}$ shows the highest variability and this is the time interval when elevated hematite and / or goethite abundances occur in the marls. Also the magnetite content varies slightly in this interval as is indicated by the minute changes in κ_{cfb}. Within two intervals of the section (9.4 – 9.3 Ma and 8.65 – 8.15 Ma), the $HIRM_{cfb}$ shows maximum values (Fig. 2), therefore the marls contain high amounts of antiferromagnets. These peaks correspond to the intervals described by Krijgsman *et al.* (1995) to contain a mixture of hematite and magnetite. Intermediate $HIRM_{cfb}$ values are present between 9.3 – 8.65 Ma. The marls should therefore still contain antiferromagnets, but only a minor magnetite component as the κ_{cfb} has its lowest values at this time interval. After 8.15 Ma, the $HIRM_{cfb}$ drops to its lowest values and remains low, indicating that antiferromagnetic minerals are only a minor component in the magnetic mineral fraction. The κ_{cfb} increases in this time interval, and indeed, Krijgsman *et al.* (1995) also found that magnetite is present, but they could not identify an antiferromagnetic phase in this time interval.

The dominant wavelength of the reflectance fluctuates around 577 - 582 nm (Fig. 2) which is within the lower range of the 580-595 nm envelope expected for hematite and goethite mixtures (Madeira et al., 1997). Prior to 8.7 Ma, values fluctuate around 580 nm and show highest values between 8.7 and 8.45 Ma. These correspond with the maximum $HIRM_{cfb}$ values and follow the observed pattern of hematite and goethite content. After 8.45 Ma, the values of the dominant wavelength become lower and fluctuate around ~579 nm, indicating the presence of at least traces of hematite and goethite in the marls, despite the low values of the magnetic parameters (Fig. 2).

4.2 Multivariate Statistics

The selected input parameters for the FCM and NLM (Table 1) represent the various environmental processes which may be recorded within the Metochia sediments. Al is associated with alumno–silicate minerals which are of terrigenous origin and can originate potentially from either the Aegean region or North Africa. The Al – MAR is therefore used as proxy to reflect the total terrigenous supply to the sediments in the Gavdos Basin. The marine component, $CaCO_3$, is expected to behave in an inverse manner to the Al – MAR.

Parameters typifying the input of North African material are $HIRM_{cfb}$ and Ti / Al. From the age profiles (Fig. 2) it can be expected that the $HIRM_{cfb}$ will show the greatest variation of these two parameters. The Ti / Al ratio describes transport mechanisms from the North African source area to the basins of the Eastern Mediterranean and can provide information on how the fluvial – aeolian system varied during the studied time interval. As the relationship of the two North African input parameters is not straightforward from the univariate plots, the FCM could provide insights into how the parameters co-vary.

Table 1: Input parameters for the cluster analysis and what they represent in the FCM solution.

Input Parameter	**'Simplified' FCM Interpretation**
Al – MAR (kg / m^2 / kyr)	Total alumno – silicate input, overall terrestrial input
$CaCO_3$ (%)	Biogenic production, marine component
Mn / Al	Diagenetic changes in the marls
Ti / Al	Fluvial vs. aeolian North African transport (when input from the Aegean is low)
Ni – MAR (kg / m^2 / kyr)	Aegean input
$HIRM_{cfb}$ (mA / m)	Antiferromagnetic (hematite and goethite) North African dust component

The proxy representing the Aegean region is the Ni – MAR; unlike in the univariate analysis the Ni – MAR is preferred over the Ni / Al ratio in the FCM and NLM analysis because the mass accumulation rate better describes the total input variation of Ni, a trace component in the sediment. In contrast, the Ni / Al ratio was used in the age profiles to track diagenetic alterations and provide a direct comparison to the studies of Wehausen and Brumsack (2000), comparing the values against their scenario of an uplifted Crete during the Pliocene.

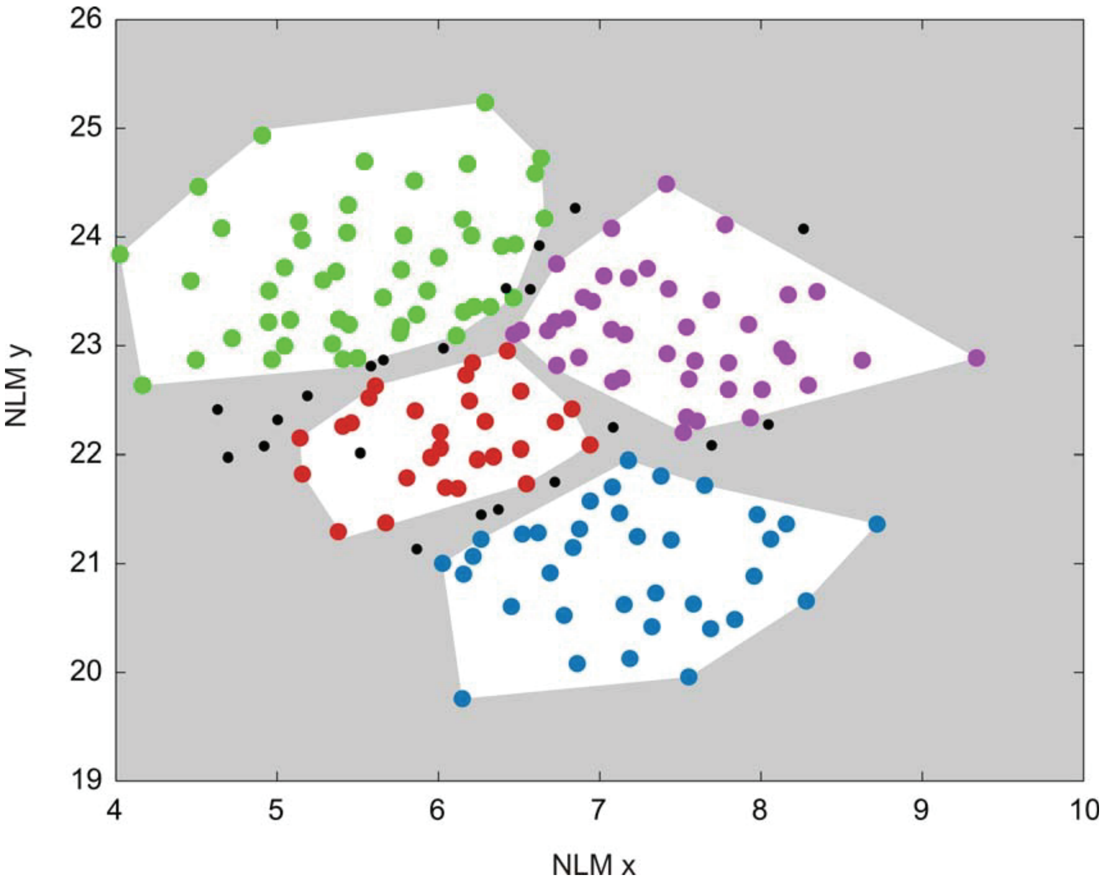


Fig. 4: Non–linear map showing the four cluster separation and the transitional cases. Transitional cases are shown as small black dots. The axes on the NLM are arbitrary. The Aegean cluster is colored green, Humid North Africa cluster purple, Arid North Africa cluster red and the Low Aegean / Humid North Africa cluster blue.

Mn / Al is used to trace diagenetic processes in the marls, as Mn is a redox sensitive element, and remobilized during diagenesis (van Santvoort et al., 1997). To balance the input parameters in order to avoid overrepresentation, some proxies presented in the univariate approach are not used for the FCM. These include the North African source area proxies Si / Al and Fe / Al, the dominant wavelength, representing hematite and goethite variations and κ_{cfb} indicating total magnetic mineral input, a proxy for terrigenous sediment input. The source areas or processes described by these proxies are already represented by the chosen input parameters, which allow for better interpretations.

Whereas repeated runs of the algorithm testing for the stability of the solution showed that two, three, four and five clusters solutions were viable from a statistical viewpoint, the four cluster model seems to be the optimal cluster solution. The partitioning factor F' was maximized and the entropy H' minimized (Bedzek et al., 1984; Vriend et al., 1988) in the two cluster solution, but the four cluster solution is a refined version of the lower cluster number solutions, as individual clusters split into coherent parts. The two cluster solution separates the data into a “terrigenous” and “marine” cluster; the three cluster solution further divides the “terrigenous” cluster into two separate clusters, which is continued in the four cluster solution. The five cluster solution has very small cluster sizes, further dividing the terrigenous clusters into smaller clusters, making the solution not interpretable. The four cluster solution has 22 transitional vs. 186 cluster-associated samples which is acceptable when studying the major and long term changes rather than sample to sample variation. The locations of all the samples, including the transitional cases, are shown in the NLM plot (Fig. 4). When the samples are grouped according to their FCM memberships, the NLM provides clear support for the four cluster solution.

5. Discussion

5.1 Paleoenvironmental Interpretation

The results of the FCM analysis make it possible to highlight the different paleoenvironmental settings present during the late Miocene on the basis of the measured proxy data set. The first task is to study the positions of the cluster centers within the measurement space in order to define the paleoenvironmental conditions they represent. The locations of the cluster centers are given in Table 2. The description and interpretation of the clusters in terms of sediment input is provided in Fig. 5, which shows that two cluster centers are contrasted by changes in the terrigenous component (cluster centers 1 and 4) and that cluster centers 2 and 3 are characterized by changes concerning North Africa. The results demonstrate clearly that the

Cluster Center 1 **Aegean Cluster** **High:** Al – MAR, Ni – MAR **Low:** $CaCO_3$ **Intermediate:** Mn / Al, Ti / Al, $HIRM_{cfb}$	**Cluster Center 2** **Humid North Africa Cluster** **Low:** Ti / Al, $HIRM_{cfb}$ **Intermediate:** $CaCO_3$, Mn / Al, Ni – MAR, Ali – MAR
Cluster Center 3 **Arid North Africa Cluster** **High:** Ti / Al, $HIRM_{cfb}$ **Intermediate:** Mn / Al, $CaCO_3$, Al – MAR, Ni - MAR	**Cluster Center 4** **Low Aegean/Humid North Africa Cluster** **High:** $CaCO_3$ **Low:** Al – MAR, Ti / Al, Ni – MAR, $HIRM_{cfb}$ **Intermediate:** Mn / Al

Fig. 5: A schematic representation of the cluster center characteristics and their assigned source areas for the four cluster solution.

cluster separation depends on changes in the overall terrigenous input, especially concerning the North African dust proxy $HIRM_{cfb}$ and the Ni – MAR. As expected, the marine component $CaCO_3$ increases as the terrigenous supply decreases.

Examining Table 2 reveals that the ratio Mn / Al is consistently low across all the cluster centers, demonstrating that the effects of diagenesis do not strongly influence the cluster solution. The samples associated with high peaks present in the Mn / Al age profile (Fig. 3) are transitional cases. Therefore the environmental signals recorded by the samples represented in the FCM can be considered to be of a primary origin. Cluster center 1 is located at a position of the measurement space with high terrigenous input and low $CaCO_3$ values. The Al – MAR and the Ni – MAR in particular, have the highest values for any cluster center indicating the importance and predominance of terrigenous input, especially from the Aegean. Proxies corresponding to North African input ($HIRM_{cfb}$, and Ti / Al) are at intermediate levels. The Ti / Al ratio in this cluster is influenced by the high Al-input from the Aegean, which should lower the ratio. It therefore cannot be interpreted in representing North African input pathways, but rather reflects Aegean vs. North African input.

The largest contrast to the terrigenous input represented by cluster center 1 is observed in cluster center 4, which is dominated by high $CaCO_3$ contents, suggesting that this cluster represents marine input. This hypothesis is supported by the low values of the terrigenous proxies Al – MAR and Ni – MAR. The input parameters representing North African sources, $HIRM_{cfb}$ and Ti / Al, are also low in this cluster center. Based on its position within the measurement space it is apparent that cluster center 4 represents predominantly marine input.

Cluster centers 2 and 3 are distinguished on the basis of contrasts in the North African source indicators. The terrigenous input parameters, especially concerned with the Aegean source and the marine component, remain at intermediate values in both cluster centers. The Al-input is no longer dominated by the Aegean source area, but a mixture from the Aegean and North Africa. Therefore the cluster solutions can be interpreted as representing an African signal. Cluster center 2 has low $HIRM_{cfb}$ and Ti / Al values suggesting that the North African dust input is not important. Cluster center 3, on the other hand, is distinguished by very high $HIRM_{cfb}$ values suggesting high aeolian dust supply. The Ti / Al values are also highest in this cluster center, supporting the importance of aeolian dust transport from North Africa.

To summarize, cluster center 1 ("Aegean cluster") has a high terrigenous component, the most important source area being the Aegean landmass. It becomes gradually replaced by cluster center 4 ("low Aegean / humid North Africa cluster") via cluster centers 2 and 3. These cluster centers are influenced by changes concerning the dust input from North Africa. Cluster center 2 ("humid North African cluster) has low aeolian dust input, whereas cluster center 3 ("arid North African cluster") is characterized by high aeolian dust. The increased dust input described by

Table 2: Locations of the cluster centers within the measurement space for the four cluster model.

	Cluster Center 1 Aegean Cluster	Cluster Center 2 Humid North African Cluster	Cluster Center 3 Arid North African Cluster	Cluster Center 4 Low Aegean /Humid North African Cluster
Al – MAR ($kg / m^2 / kyr$)	7343	4488	4160	1489
$CaCO_3$ (%)	40	49	51	60
Mn / Al	0.0065	0.0092	0.011	0.014
Ti / Al	0.0581	0.0554	0.0622	0.0552
Ni – MAR ($kg / m^2 / kyr$)	15.84	8.65	9.92	2.84
$HIRM_{cfb}$ (mA / m)	97	31	341	15

cluster center 3 suggests more arid paleoenvironmental conditions in North Africa, allowing for enhanced dust production. Other intriguing relationships also appear, for example the Al – MAR, associated with terrigenous input is highest in the Aegean cluster, therefore showing that a large amount of sediment supply comes directly from the Aegean landmass and is dominant over the North African source. This is not surprising considering the closeness of Gavdos to the former Aegean landmass, which was drained by a fluvio-lacustrine system towards the Gavdos Basin (Fortuin, 1978; Meulenkamp et al., 1994; van Hinsbergen and Meulenkamp, 2006). As Al seems to be associated with the Aegean source, it can explain the ratios indicating North African sources being somewhat lower than the Pliocene values of previous studies by Wehausen and Brumsack (1998; 2000) (e.g. Fe / Al, Ti / Al). Especially in the Aegean cluster the low Ti / Al can be explained by the high Al amount from the Aegean, the ratio therefore reflecting an Aegean vs. North African signal. However, in the three other cluster centers, the Al – MAR is intermediate or low, indicating less influence from the Aegean region. The variations in the Ti / Al ratio in these cluster centers should therefore be related to change in the African transport system from more aeolian (arid North Africa) to more fluvial / less aeolian transport (humid North Africa). The relation to changes in North Africa is supported by the fact that the Ti / Al ratio and the $HIRM_{cfb}$ describe similar paleoenvironmental conditions in these cluster centers (Fig. 5).

By considering the membership values of the individual Metochia samples it is possible to employ the FCM solution to reconstruct the sedimentary inputs as a function of time (Fig. 6). The dominant feature of the membership plot of the individual cluster centers shows the Aegean cluster being replaced over time by the low Aegean / humid North Africa via the North African clusters. On the basis that the depositional environment of Gavdos did not change during this time interval (van Hinsbergen and Meulenkamp, 2006), it is apparent that the changes observed in the cluster memberships represent actual modifications to the composition of the terrigenous component. The Aegean cluster is dominant prior to ~8.8 Ma and gets replaced by the low Aegean / humid North Africa cluster after ~8.1 Ma. Isolated peaks in low Aegean / humid North Africa cluster prior to ~8.1 Ma can be related to high $CaCO_3$ contents. The starvation of Aegean sediment supply after ~8.1 Ma can be linked to the break up of the Aegean landmass during the late Miocene (Meulenkamp et al., 1994; van Hinsbergen and Meulenkamp, 2006). The break up would have caused the submergence of the landmass, resulting in the drowning of the sediment source area. The North African clusters show that the arid North African cluster is dominant prior to ~8.15 Ma, when it ‘competes’ with the Aegean cluster for dominance. The humid North African cluster becomes dominant after ~8.15 Ma, also when the low Aegean / humid North Africa cluster becomes more dominant. As the low Aegean / humid North Africa cluster also has low Ti / Al values, there appears to be change towards less aeolian input from North Africa, which is consistent with a previously proposed shift towards more humid conditions in North Africa during the late Miocene (e.g. Ruddiman et al., 1989; Griffin, 1999).

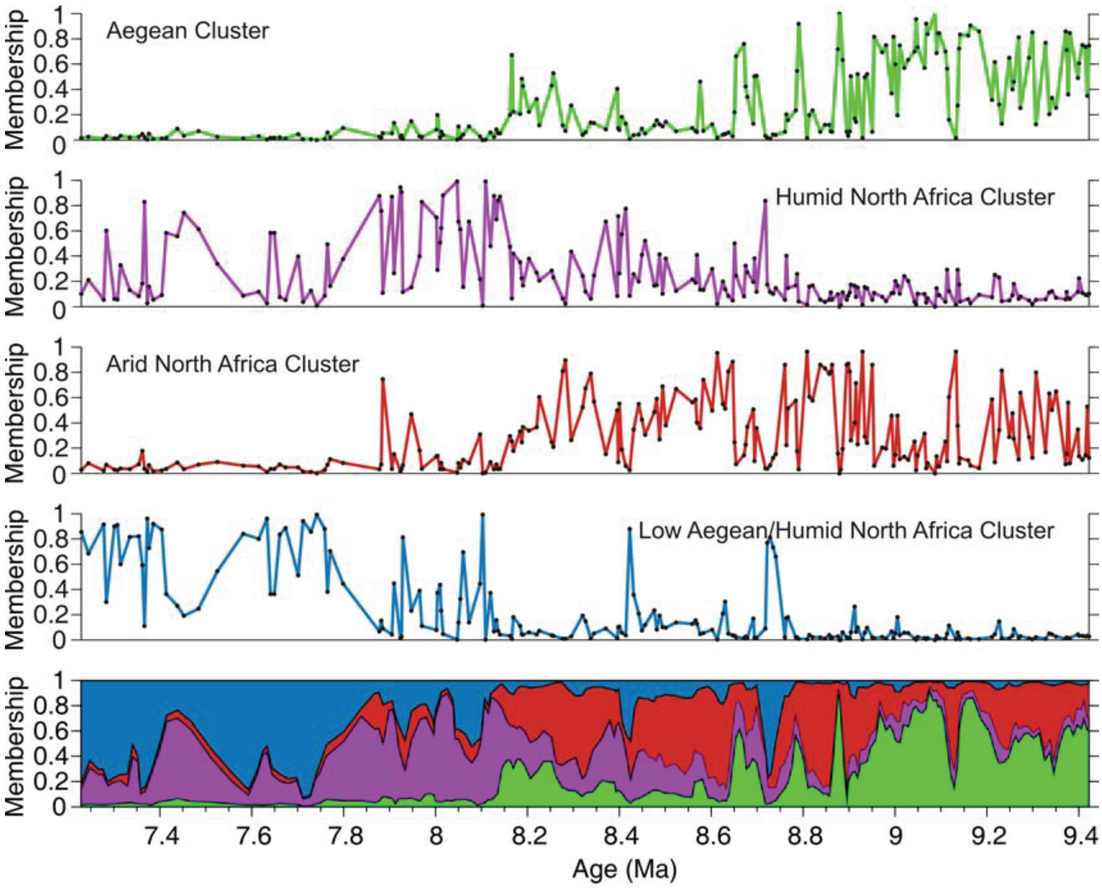


Fig. 6: FCM membership variation of the different cluster centers of the Metochia section as a function of time. The membership values define the similarity of the sample to the clusters. The cumulative membership plot is smoothed using a 3-point average. For cluster names and color coding see Fig. 4.

6. Conclusions

The sedimentary history of the Metochia section was investigated in detail using geochemical, environmental magnetic and DRS proxies. The multiproxy data set was constructed in such a way that it was possible to discriminate between terrigenous material originating from North Africa and the Aegean region. A number of indicative proxies suggested that changes in sediment source area may have occurred between 8.4 – 8.2 Ma, but these changes were not clear throughout the univariate data set.

The multivariate approach using FCM and NLM was employed to characterize a carefully chosen geochemical and magnetic data set in order to unravel the relationships that existed between the different proxies. A four cluster solution was selected because it best described the complicated interplay of paleoenvironmental and tectonic changes during the late Miocene.

The cluster solution shows a distinct decrease of terrigenous input and in the Ni – MAR over time. The Aegean cluster, with high terrigenous input, dominates the section from its base and becomes replaced by the low Aegean / humid North Africa cluster at ~8.1 Ma, with a minimal contribution from the Aegean region. This change can be linked to the late Miocene break-up of the Aegean landmass.

Cluster center variations concerned with the North African dust input show that during the late Miocene both arid and humid phases were present. More arid periods are characterized by high $HIRM_{cfb}$ prior to ~8.15 Ma, representing the presence of hematite and goethite in the sediment, and indicating enhanced aeolian dust input. A switch towards less arid environmental conditions with low $HIRM_{cfb}$ and Ti / Al values suggest less aeolian dust input after ~8.15 Ma, indicating more humid conditions in North Africa.

7. Acknowledgments

The samples were collected and provided by Fort Hoofddijk, Utrecht University. This study was financed by the DFG International Graduate College EUROPROX. We are thankful for the laboratory assistance of T. Frederichs, L. Brück, C. Hilgenfeldt and K. Enneking.

3. Concurrent tectonic and climatic changes recorded in upper Tortonian sediments from the Eastern Mediterranean: Is there a causal relationship?[2]

Abstract

During the late Miocene, the Mediterranean experienced palaeogeographic reorganisations and the intensity of the North African monsoon increased. In order to argue for a common cause for these changes, exact ages are needed. Here we present a precise age for the late Miocene fragmentation of the Aegean landmass using astronomically-tuned marine sediments from the Metochia section, Gavdos (South of Crete) by a multiproxy approach. The break-up of the Aegean landmass, traced by the starvation of the Cretan sediment, is timed at 8.2 Ma. Furthermore, a climatic change in North Africa towards more humid conditions is also documented in the Eastern Mediterranean sediments at 8.2 Ma. The North African monsoon is coupled with the Asian monsoon system, which also strengthened during the late Miocene as a result of an uplift phase in the Himalayas. Therefore we hypothesise that the palaeogeographic reorganisations in the Mediterranean and the synchronous Himalayas tectonic uplift result from large scale tectonic interactions.

Keywords: late Miocene, Eastern Mediterranean, palaeoclimate

[2] This chapter has been submitted for publication as: C.M. Köhler, W. Krijgsman, D.J.J. van Hinsbergen, D. Heslop: Concurrent tectonic and climatic changes recorded in upper Tortonian sediments from the Eastern Mediterranean: Is there a causal relationship? *Terra Nova*

Introduction

The semi-enclosed land-locked configuration of the Mediterranean region at the European-African collision zone makes it very suitable to register sedimentary provenance changes resulting from both plate tectonic processes and changes in palaeoclimate (e.g. Krijgsman, 2002). The Miocene geodynamic evolution of the Mediterranean region was controlled largely by the subduction of the African plate under the European margin. These plate tectonic processes were ongoing throughout the entire Miocene, but were also marked by a strong, Mediterranean-wide, increase of tectonic activity in the late Tortonian (Carminati et al., 1998a; Jolivet and Faccenna, 2000; Wortel and Spakman, 2000; Garcés et al., 2001; Meulenkamp and Sissingh, 2003). Late Tortonian climatic reconstructions also reveal substantial changes, especially on the North African continent which is thought to have been much more humid than today (e.g. Griffin, 2002; Gladstone et al., 2007). Previous studies of Mediterranean sedimentary archives have shown that certain geochemical proxies can be indicative of different sediment origins and that they can be linked to changes in the environmental conditions of Mediterranean and African source areas (e.g. Bergametti et al., 1989; Wehausen and Brumsack, 1998; Lourens et al., 2001; Larrasoaña et al., 2003a; Köhler et al., submitted-a).

In this study, we attempt to reassess the timing of climatically and tectonically induced changes in sediment provenance and their temporal evolution in the eastern Mediterranean region by studying the provenance changes in the astronomically dated marls of the Metochia section on the island of Gavdos, located south of Crete (Fig. 1). This section provides an ideal archive spanning the period between 9.7 – 6.6 Ma (Krijgsman et al., 1995), covering the proposed time frame of palaeogeographic fragmentation of the eastern Mediterranean (van Hinsbergen and Meulenkamp, 2006) and palaeoclimatological changes on the North African continent (Griffin, 2002). The late Tortonian location of the section within the fluvio-lacustrine drainage system of the southern Aegean landmass (e.g. Fortuin, 1978) and its relative proximity to the North African margin, make it ideal to trace variations in

Aegean sediment supply as well as changes in the North African monsoon system.

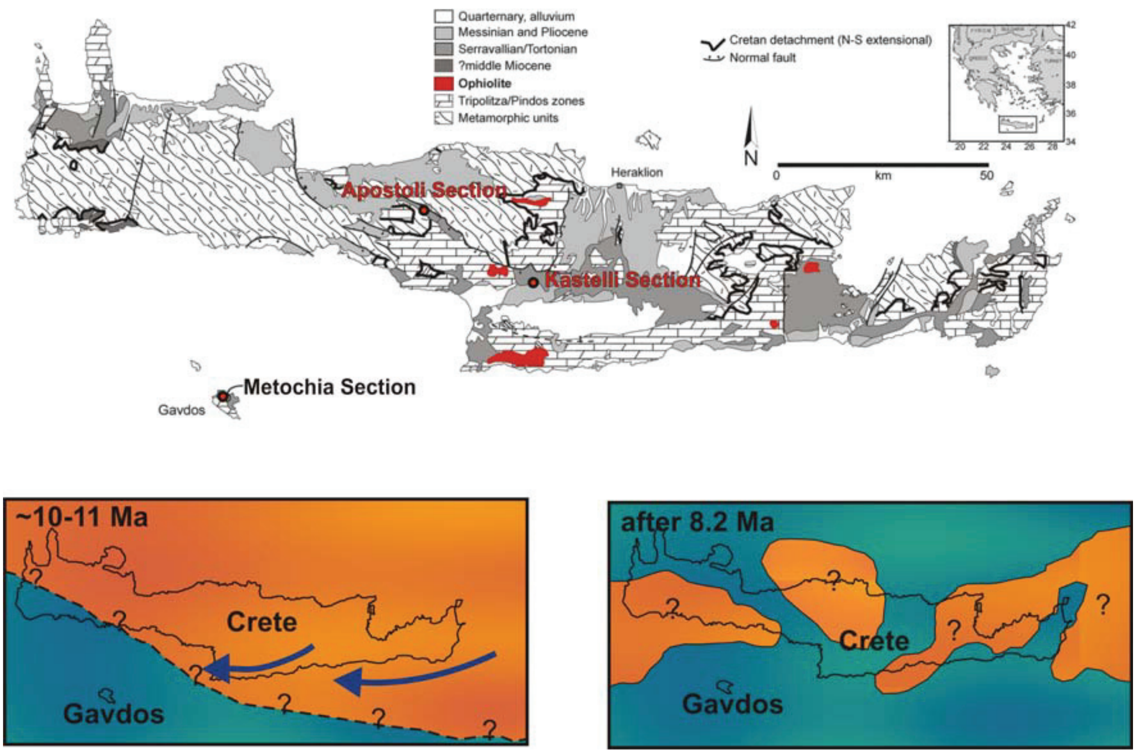


Fig. 1: A geological map of Crete and Gavdos (Meulenkamp et al., 1988; van Hinsbergen and Meulenkamp, 2006). The Ni-rich source areas (ophiolites) are marked in red; they are located within the drainage area of the fluvio-lacustrine system. Also shown are the two settings described in the text: Crete and the Aegean region forming a landmass and being drained to the West; the Aegean land mass being fragmented (modified from: Meulenkamp and Hilgen, 1986; Meulenkamp et al., 1988; van Hinsbergen and Meulenkamp, 2006).

Multiproxy analysis of the Metochia marls

A precise age model of the Metochia section was previously constructed using astronomical tuning, magnetostratigraphy and biostratigraphy (Hilgen et al., 1995; Krijgsman et al., 1995), but revised sapropel ages in accordance with the more recent orbital solution of Laskar *et al.* (2004) are employed in this study (Fig. 2). A detailed study using geochemistry and environmental magnetics showed that the marls of the Metochia section contain a mixture of sediments from the Aegean and North African regions (Köhler et al., submitted-a). The sedimentation rate, which is presumed to be dominated by Aegean terrigenous supply being drained by fluvial systems into the Gavdos basin

(Fortuin, 1978; van Hinsbergen and Meulenkamp, 2006), shows a steady decline and remains at low values from 8.15 Ma onwards (Fig. 2).

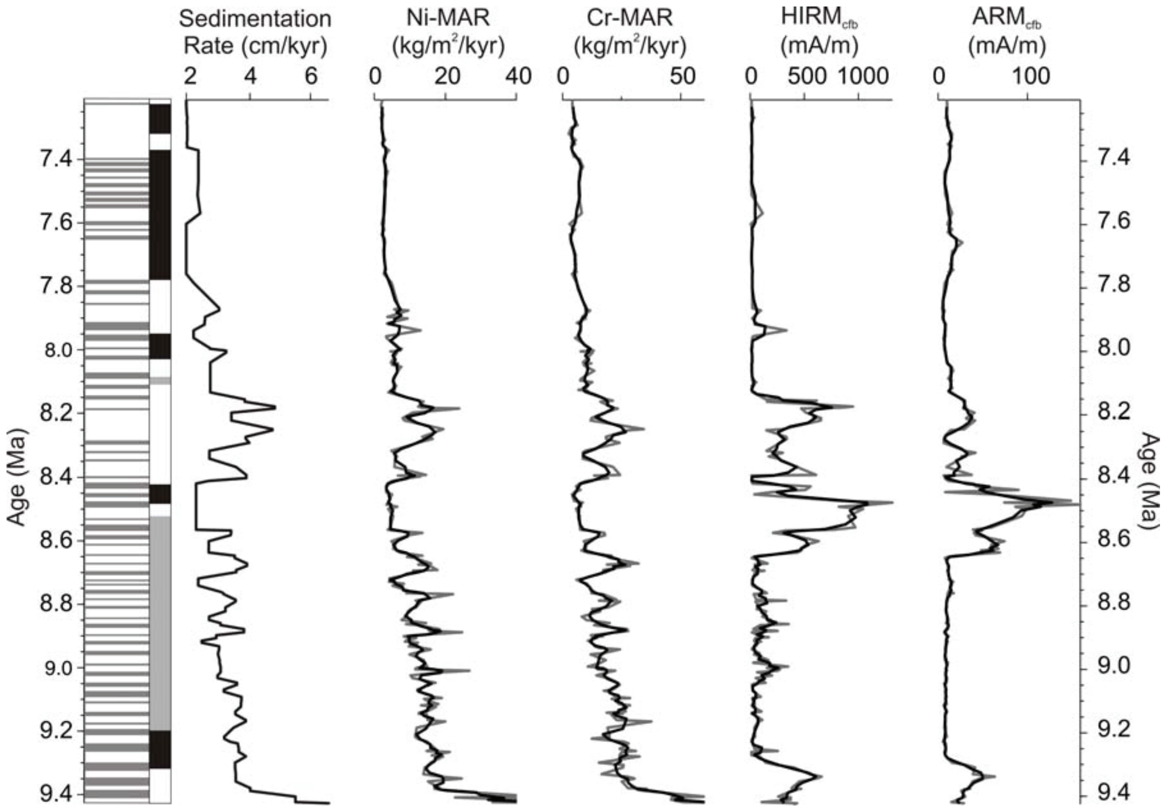


Fig. 2: Lithology and magnetostratigraphy (modified from Krijgsman, et al. (1995)) and age profiles of proxies of the Metochia section described in the text. The lithology: white intervals represent homogeneous marls, the dark grey sapropels. The magnetostratigraphy: black = normal and white = reversed polarity; grey = unreliable directions. The Metochia section was correlated to the GPTS of CK95 (Cande and Kent, 1995) by Krijgsman, et al. (1995). The HIRM was calculated using a saturation field of 2500 mT and a backfield of 300 mT (Köhler et al., submitted-a), the ARM was imparted under a DC bias field of 50 μT and a peak alternating field of 100 mT. The light grey lines indicate the data, the thick, black lines represent a three point running mean plot to highlight the important changes.

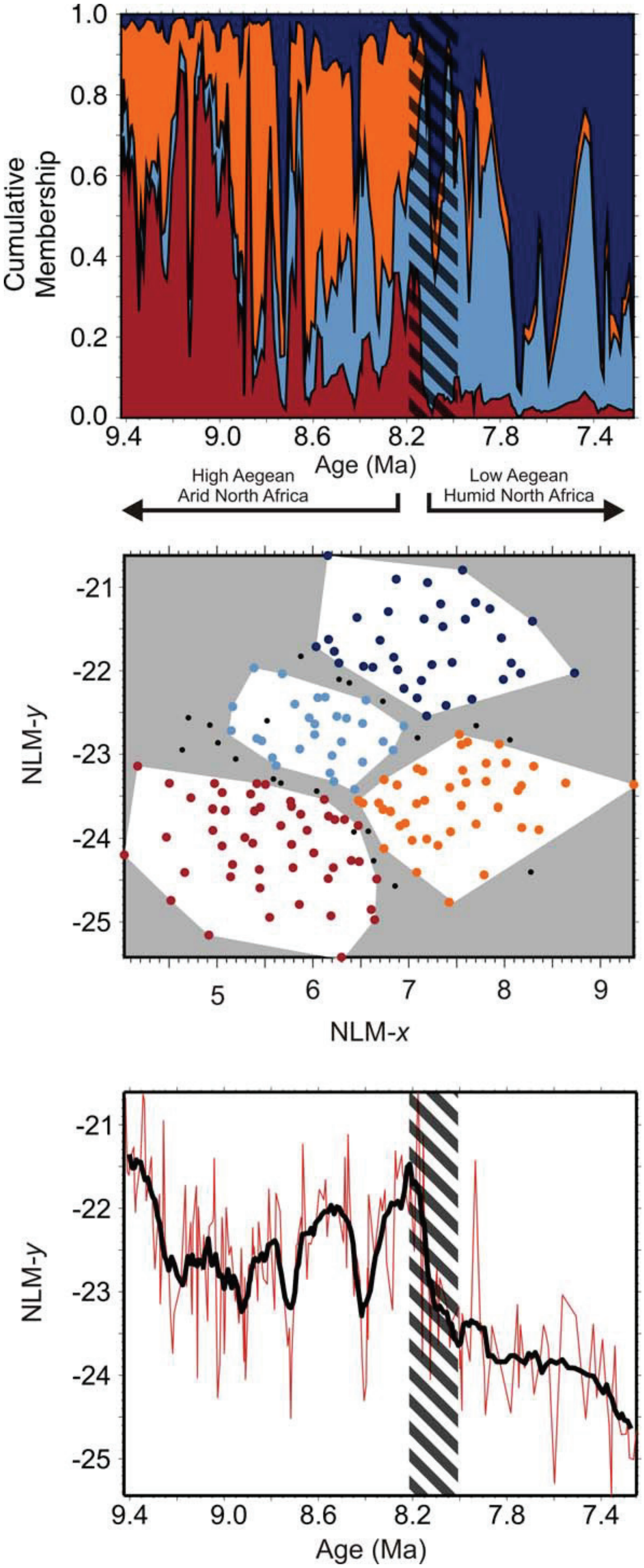
The elements Nickel (Ni) and Chrome (Cr) provide suitable proxies to identify changes in the Aegean region, as their presence can be linked to the erosion of ultramafic rocks, having high Ni and Cr concentrations (Wedepohl, 1969; Wehausen and Brumsack, 2000). The Ni- and Cr-MARs (mass accumulation rate) both show a strong decline at 8.15 Ma and subsequently remain at low values (Fig. 2), confirming that significant changes in the Aegean source area and/or its drainage system must have occurred at that time.

North African aeolian dust can be traced by magnetic minerals using the hard isothermal remanent magnetisation (HIRM) and anhysteretic remanent magnetisation (ARM), respectively (e.g. Larrasoaña et al., 2003a). Both African dust proxies show a similar temporal variability in the Metochia marls, with highest peaks and fluctuations prior to 8.2 Ma (Fig. 2), although these peaks do not necessarily correspond in magnitude. The dust input drops to minimal values at 8.15 Ma (Fig. 2), which reflects a change to more humid conditions, probably by an intensification of the monsoon circulation in North Africa (Rohling et al., 2002).

Through the combination of univariate results, fuzzy *c*-means clustering analysis (FCM) (Bedzek et al., 1984) and non-linear mapping (NLM) (Sammon, 1969) it is possible to partition the samples into a four cluster model (Fig. 3a and b) (Köhler et al., submitted-a). Two contrasting 'pairs of clusters' were identified: high vs. low Aegean input clusters and arid vs. humid North African climate clusters (Fig. 3a and b). Memberships of the samples, where 0 indicates no similarity and 1 shows that the sample is identical with the cluster centre, show that the high terrigenous cluster centres dominate from 9.4 – 8.2 Ma, and low Aegean and humid North Africa cluster centres become important from 8.2 - 8.15 Ma. The cumulative membership plot of the cluster model furthermore indicates that the drop in Aegean sediment supply and the switch to more humid conditions in North Africa occur coevally at 8.2 Ma (Fig. 3a). Here, we present an alternative approach with which to more clearly visualise the timing of the changes in Aegean and African sediment supply by direct combination of both the FCM and NLM solutions (Fig. 3c). Given that vertical shifts on the NLM appear to represent the movement between terrigenous and marine sediment sources, the *y*-coordinates are of particular interest. The NLM *y*-axis values of the samples plotted as a function of age reveal a change from high Aegean to low Aegean input. Starvation of Cretan sediment supply thus occurs between 8.2 – 7.9 Ma, when the *y*-coordinate values of the samples in the NLM decrease towards the values associated with the cluster centre representing low input of Aegean material (Fig. 3b).

Fragmentation of the Aegean region

The sudden disappearance of the Aegean sediment source in the Metochia marls suggests the E-W drainage system along southern Crete,



1.0
0.8
0.6
0.4
0.2
0.0
Cumulative Membership
9.4
9.0
8.6
8.2
7.8
7.4
Age (Ma)
High Aegean
Arid North Africa
Low Aegean
Humid North Africa
-21
-22
-23
-24
-25
NLM-y
5
6
7
8
9
NLM-x
-21
-22
-23
-24
-25
NLM-y
9.4
9.0
8.6
8.2
7.8
7.4
Age (Ma)

Fig. 3 (previous page): The results from the FCM and NLM (Köhler et al., submitted). The cumulative membership plot obtained from the FCM shows how the memberships of the individual samples vary over time; dark red: high Aegean input and arid North Africa, orange: arid North Africa, light blue: low Aegean and humid North Africa, dark blue humid North Africa. The NLM provides a low-dimensional representation of the proxy data set, individual samples are marked with points which are colour-coded according to the fuzzy cluster-centre to which they are assigned (transitional samples which have no clear assignment are shown as smaller black circles) (taken from Köhler et al., submitted-a). The NLM *y*-axis values of the samples plotted as a function of age reveal a change from high Aegean to low Aegean input (the black line is a 3-point running mean). The grey bar indicates the time interval when the high Aegean cluster becomes replaced by the low Aegean cluster, describing a change in terrigenous input from the Aegean region.

which was responsible for transporting material eroded from the southern Aegean landmass towards the Gavdos basin, underwent a tectonic fragmentation at 8.15 Ma. During the early Tortonian, the Heraklion region of Central Crete was part of this fluvio-lacustrine system marked by the fossil mammal-bearing continental deposits of the Kastellios Hill section (Sen et al., 1986). The lagoonal to non-marine sediments of Kastellios Hill are overlain by 25 m of shallow-marine (pebbly) sandstones and detrital limestones containing calcareous red algae (*Lithothamnium*) and *Heterostegina*. This transgressive surface is recognised Cretan-wide and has an age of 8.2 Ma in the Apostoli section (Meulenkamp, 1969). This date is based on using the duration of 194 kyrs for the *Sphaeroidinellopsis seminulina* zone (Hilgen et al., 1995) to calculate the sedimentation rate for this section.

The upper Tortonian marine sediments of the Heraklion region are best exposed in the Kastelli section (Langereis, 1984; Krijgsman et al., 1994) and have been analysed as part of palaeobathymetric reconstructions (van Hinsbergen and Meulenkamp, 2006). The results show that a progressive deepening from ~400 - 900 m took place between 7.9 - 7.5 Ma, after which the basin can be considered as deep-marine until at least 7.1 Ma (Fig. 4). Linear extrapolation of the palaeodepth curve for the Kastelli section back to 0 m, places the age range for the drowning phase between 8.4 – 8.1 Ma (Fig. 4). We conclude that the starvation of Aegean sediment supply on Gavdos is therefore likely to be related to the onset of basin formation on Crete.

Late Tortonian basin formation on Crete has been associated with the break-up of the southern Aegean landmass and the formation of the Sea of Crete during curvature of the Aegean arc (e.g. van Hinsbergen and Meulenkamp, 2006). The reason for this fragmentation is generally believed to be associated with south(west)ward roll-back by the subducted African slab (Le Pichon and Angelier, 1981; Meulenkamp et al., 1988; Jolivet, 2001; van Hinsbergen et al., 2005). During the early to middle Miocene, N – S extension was active in the Aegean region, resulting in the large-scale exhumation of metamorphic rocks (Fassoulas et al., 1994; Jolivet et al., 1996; Thomson et al., 1998; Thomson et al., 1999; Rahl et al., 2005). In the late Tortonian, a new tectonic phase caused E – W extension and a rapid high-angle break-up of the Aegean landmass (Meulenkamp et al., 1988; Fassoulas et al., 1994; van Hinsbergen and Meulenkamp, 2006).

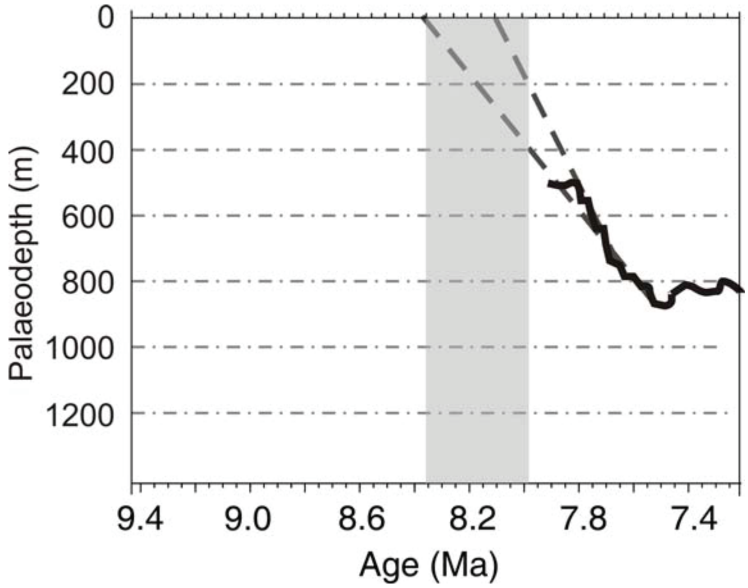


Fig. 4: The Kastelli section with the extrapolated age is modified from (van Hinsbergen and Meulenkamp, 2006). The grey bar indicates the transitional period identified in the FCM, which covers the time interval when the high Aegean cluster becomes replaced by the low Aegean cluster, describing a change in terrigenous input from the Aegean region.

The island of Crete formed and was subsequently transformed into a mosaic of small-sized submerging subbasins (Meulenkamp et al., 1994). The late Tortonian sedimentary sequences on Crete, that are supposed to document this tectonic phase, mainly consist of course clastics which appeared to be unsuitable to provide more precise ages for the break-up phase. Our multi-proxy analyses on the Metochia marls, combined with interpolated accumulation rates of the Cretan basins, now shows that the main fragmentation of the Aegean landmass can be precisely dated at 8.2 Ma.

Interaction of tectonics and climate

The late Tortonian of the Mediterranean region is characterised by major palaeogeographic changes that significantly affected paleoenvironmental conditions in various marine settings (e.g. Kouwenhoven and van der Zwaan, 2006) in addition to palaeoceanographic circulation patterns and water exchange with the Atlantic (e.g. Benson et al., 1991). In the Central Mediterranean, the Tyrrhenian Sea opened at 8 Ma (Kastens et al., 1987; Carminati et al., 1998b) and a sedimentological change in the Northern Apennines, possibly linked to an uplift phase, was astronomically dated to have taken place at 8.2 Ma (van der Meulen et al., 1999). The cyclical late Tortonian deposits of this region are furthermore characterised by the presence of numerous tephra layers indicative of increased volcanic activity (Vai et al., 1993; Laurenzi et al., 1997; Montanari et al., 1997). Late Tortonian tectonic processes in the Gibraltar region caused a reorganisation of the Mediterranean-Atlantic gateways through the Rifian foredeep of Morocco (Krijgsman et al., 1999) and via the Betics of southern Spain (Soria et al., 1999; Garcés et al., 2001), a first step to the progressive restriction of the Mediterranean during the Messinian salinity crisis (Hsü et al., 1973).

It is tempting to link all these late Tortonian palaeogeographic reorganisations, that seem to appear within a narrow time frame of 8.2 – 8 Ma to changes in the Europe-Africa plate motion velocities (Jolivet and Faccenna, 2000) or to large-scale changes in geodynamic processes affecting the entire collision zone (e.g. Meulenkamp and Sissingh, 2003). In addition, it is very intriguing to speculate if and how these tectonic processes affected the paleoclimatic system.

In this context, our multiproxy analysis on the Metochia marls indicates that the North African monsoon system intensified synchronously with the tectonic events in the Aegean and Mediterranean. The African monsoon system is largely influenced by the location of intertropical convergence zone (ITCZ), which is a global phenomenon (Griffin, 2002). To create humid conditions in North Africa, the northern limit of the ITCZ needs to shifts as far as ~22° N (Fluteau et al., 1999), passing the central Saharan watershed (Rohling et al., 2002; Larrasoaña et al., 2003a). In modelling studies, changes in the Mediterranean Sea during the late Tortonian, mainly had an influence on Mediterranean circulation patterns and the depth of the circulation cells (Meijer et al., 2004). An influence of the changing Mediterranean on the position of the ITCZ can not be disregarded, although the opening of new basins (e.g. Aegean or Tyrrhenian) seems unlikely to have had a dramatic effect, as they did not modify the water exchange with the Atlantic. Tectonic changes in the Aegean region should therefore not have been able to affect the position of the ITCZ, as even changes in the Mediterranean circulation do not seem to have a significant impression on North African climate.

The African monsoon system has also been linked to changes in the Asian monsoon (e.g. Fluteau et al., 1999; Griffin, 2002), implying that the North African monsoon intensification may result from strengthening of the Asian monsoon system. In addition, the enhancement of the Asian monsoon system has been linked to an uplift phase of the Himalayan-Tibetan Plateau (e.g. Ramstein et al., 1997; An et al., 2001). If these hypotheses are correct, our Metochia results thus indicate that the timing of enhancement of the North African monsoon system at 8.2 Ma coincides with an uplift phase in the Himalayas. This age is in large agreement with observed paleoenvironmental changes in the continental Siwalik sediments of Pakistan and in the marine sediments of the Arabian Sea (e.g. Quade et al., 1989; Kroon et al., 1991; Filippelli, 1997; An et al., 2001). The Mediterranean tectonic events could thus even be related with Himalayan uplift, suggesting that large-scale, Alpine-Himalayan wide tectonic changes during the late Tortonian around 8.2 Ma affected the entire African-Eurasian plate boundary, which may provide the context for the reconstructed coinciding tectonic and climatic events.

Conclusions

The late Miocene tectonic reorganisation of the Aegean region is traced by changes in the proxy record of the Eastern Mediterranean Metochia section. By quantification of these proxies the reconstructed break-up of the Aegean landmass has been dated to 8.2 Ma, placing it within the time frame of Tortonian Mediterranean-wide tectonic reorganisations. Interestingly, the Mediterranean tectonic reorganisations are coeval with a North African climate change towards more humid conditions, which could be potentially linked to an uplift phase in the Himalayan region at 8.2 Ma via an intensification of the Asian monsoon. Such a mechanistic link would suggest a causal relationship between Alpine-Himalayan wide tectonic events and global climate. The potential of multiproxy studies to date tectonic events shows that it is possible to obtain more accurate ages with which to test potential linkages between tectonic and climatological events.

Acknowledgments

The samples were collected and provided by Fort Hoofddijk, Utrecht University. This study was financed by the DFG International Graduate College EUROPROX. We are thankful for critical and helpful discussions with G. Dupont-Nivet and S. Hüsing, and for the laboratory assistance of T. Frederichs, L. Brück, C. Hilgenfeldt and K. Enneking.

4. Late Miocene palaeoenvironmental changes in North Africa and the Mediterranean recorded by geochemical proxies (Monte Gibliscemi section, Sicily)[3]

Abstract

The astronomically tuned marls of the Monte Gibliscemi section, Sicily, constitute an archive to trace the late Miocene palaeoenvironmental conditions (~9.7 - 7.0 Ma) in North Africa. Here we have utilised carbonate content and the Al-normalised geochemical proxies Ti / Al, Si / Al, Mg / Al, Fe / Al, Mn / Al and V / Al. The terrigenous input in the section is dominated by North African river systems draining into the Eastern Mediterranean: Gabes, Libyan, Eosahabi/Chad and Nile. When placed within an astronomically tuned chronology the proxy parameters indicate that the palaeoenvironmental conditions in North Africa were humid from 9.5 Ma with high fluvial input to the Eastern Mediterranean. Increases in the Si/Al and Mg / Al ratios occurred from 8.4 - 8.2 Ma and from 8.05 - 7.75 Ma, with maximum values similar to those of the Messinian diatomite sediments at the Monte Gibliscemi and Metochia (Gavdos, South of Crete) sections. These peaks indicate conditions of enhanced biosiliceous productivity and the presence of authigenic clay formation. Sluggish water circulation in the Mediterranean during those times is also inferred from the Mn / Al and V / Al behaviour. Late Miocene changes of the Betic (southern Spain) and Rifian (Morocco) Mediterranean-Atlantic gateways are interpreted as the driving force for the changes in water circulation. The transgression associated with the opening of the Rifian corridor can be accurately dated at 7.8 Ma.

Keywords: Geochemical proxy parameters, palaeoclimate, Eastern Mediterranean, late Miocene, biosiliceous production.

[3]This chapter has been submitted for publication as: C.M. Köhler, D. Heslop, W. Krijgsman, M.J. Dekkers, T. von Dobeneck: Late Miocene palaeoenvironmental changes in North Africa and the Mediterranean recorded by geochemical proxies (Monte Gibliscemi section, Sicily). *Palaeogeography, Palaeoclimatology, Palaeoecology*

1. *Introduction*

The late Miocene phase of North African climate is generally considered as much more humid than the more recent periods (Ruddiman et al., 1989; Griffin, 2002; Lihoreau et al., 2006; Gladstone et al., 2007; Köhler et al., submitted-a). More humid palaeoenvironmental conditions should support a denser vegetation cover binding the soil and reducing dust production (Middleton, 1985; Larrasoaña et al., 2003a). Higher precipitation, especially in North Central Africa, could also feed several rivers that drained into the central and eastern Mediterranean Sea (e.g. Burke and Wells, 1989; Griffin, 2002; Gladstone et al., 2007), leaving records of North African climate change in the Mediterranean sediments. African terrigenous supply can be traced by specific elemental ratios, e.g. Si / Al, Ti / Al and Mg / Al (e.g. Bergametti et al., 1989; Wehausen and Brumsack, 2000; Lourens et al., 2001). The elements Si, Ti, Mg and Al are associated with certain mineral phases and weathering conditions, indicating changes in North African palaeoenvironment, and dust production (Lourens et al., 2001; Larrasoaña et al., 2003a).

In a recent sediment provenance study, African source area changes during the late Miocene were reconstructed using geochemical and environmental magnetic proxies obtained from the astronomically dated sedimentary Metochia sequences of Gavdos Island in the Eastern Mediterranean (Köhler et al., submitted). A number of indicative proxies showed that such changes occurred between 8.4 – 8.2 Ma, suggesting less aeolian dust input after 8.2 Ma and thus more humid conditions in North Africa. However, the Gavdos record is also modulated by changes in the Aegean source area due to concurrent regional tectonics (Köhler et al., submitted-b).

Here, we aim to track and distinguish late Miocene climatic changes over North Africa in the sedimentary record of the Monte Gibliscemi section in southern Sicily using geochemical data. Monte Gibliscemi provides a suitable archive of astronomically dated marine sediments spanning the time period from 9.7 – 7 Ma, thus covering the proposed time interval of inferred climate change. The section is located in the central Mediterranean basin close to the North African continent and far away from other continental source areas, including Italy, which was located further to the west (Orszag-Sperber et al., 1993). Consequently, it is reasonable to anticipate that the African signal dominates the terrigenous input.

2 Materials and Methods

2.1 Section, age model and samples

The Monte Gibliscemi section is located in southern Sicily (Fig. 1) and consists of subsections A and B (Krijgsman et al., 1995). The section has a characteristic cyclicity of alternating sapropels and homogeneous marls. A precise age model by (Krijgsman et al., 1995) was based on cyclostratigraphy and biostratigraphy. Thirteen planktonic foraminifera and nine dinoflagellate bioevents were identified confirming the cyclostratigraphic ages. The characteristic sapropel patterns were tuned to the cyclic record to the 65°N summer insolation curve of Laskar (1990) with modern values for dynamical ellipticity of the Earth and tidal dissipation by the moon. Individual sapropels were correlated with precession minima, small sapropel clusters to 100 kyr eccentricity maxima and large scale sapropel clusters to 400 kyr eccentricity maxima (Hilgen et al., 1995). The Monte Gibliscemi sediments were deposited under deep marine conditions of ~1200 m (Krijgsman et al., 1995; Kouwenhoven et al., 2003) and span the time interval from 9.77 – 6.97, with a hiatus between 7.52 – 7.24 Ma (Hilgen et al., 1995; Krijgsman et al., 1995).

In the present study, only the homogeneous marls of the late Tortonian were investigated because they are less affected by diagenetic alterations than sapropels and are therefore more likely to reflect a primary terrigenous signal. The marls represent the arid phase of the sapropel-marl cycle (e.g. van Os et al., 1994; Rossignol-Strick et al., 1998), making them a suitable archive of changes in North African palaeoenvironmental conditions. In total, 268 pristine marl samples from the original stratigraphic sample set of Hilgen et al. (1995) and Krijgsman et al. (1995) were used, with three samples per marl bed giving an average temporal spacing of 0.009 Myr.

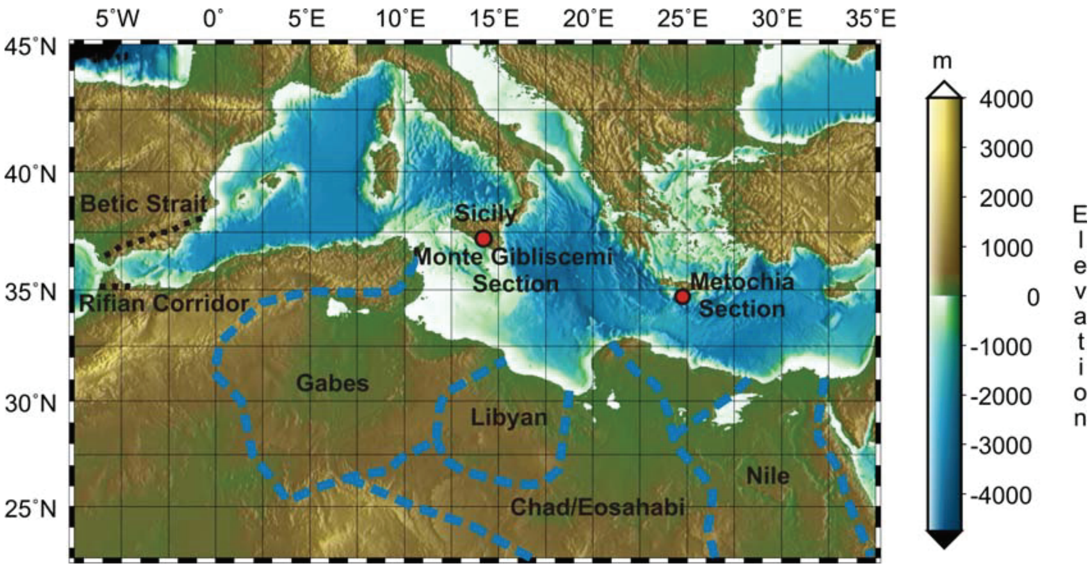


Fig.1: Map of the Mediterranean Sea and North Africa. The locations of the studied sections are marked with circles. The boundaries of the late Miocene river systems of North Africa are indicated with dashed lines (Griffin, 2002; Gladstone et al., 2007). The late Tortonian gateways connecting the Mediterranean Sea with the Atlantic Ocean are marked with black dotted lines (Krijgsman et al., 1999).

2.2 Geochemistry

To determine the geochemical composition of the Monte Gibliscemi marls, X-ray fluorescence analysis (XRF) was performed on the planar surfaces of solid cylindrical samples (~4 g) using a *Spectro Xepos* energy dispersive polarization X–ray fluorescence analyzer. The measuring time was 300 seconds per sample. Accuracy and precision were checked using the internal marine standard Mag-1 (certified USGS marine sediment standard reference material) and duplicate measurements. Measurement errors are <5 % for the elements Al, Ca, Si, Mg, Ti, Fe and Mn in this study; V has a higher error of ~20%.

The stable terrigenous element aluminium plays an important role in the geochemical interpretation because the abundances of other elements can be normalised against it to compensate for dilution effects (Wehausen and Brumsack, 1998; Schenau et al., 1999; Wehausen and Brumsack, 2000; Lourens et al., 2001; Larrasoaña et al., 2003a). The normalisation further enables comparison with other late Miocene geochemical studies (e.g. Köhler et al., submitted-a) and Pleistocene studies of the Eastern Mediterranean (e.g. Wehausen and Brumsack, 1998; Wehausen and Brumsack, 2000).

$CaCO_3$ was calculated from Ca, assuming that 98% of the Ca resides within $CaCO_3$, providing a proxy representing marine biogenic input. Marl sediments are associated with higher $CaCO_3$ content than sapropels; Eastern Mediterranean marls have typical $CaCO_3$ concentrations around 50 % (van Os et al., 1994; Wehausen and Brumsack, 1998). During marl deposition, arid North African climatic conditions are characterised by lower riverine run-off and enhanced aeolian dust production (Larrasoaña et al., 2003a). Sapropels are formed under more humid conditions with higher fluvial input and generally contain 20 – 30 % $CaCO_3$ (van Os et al., 1994). Fluctuations in Ti / Al reflect variations in the relative importance of fluvial and aeolian transport mechanisms as Ti is associated with aeolian dust while Al is found in both aeolian and fluvial materials (Lourens et al., 2001). In Eastern Mediterranean marine sediments higher values of Ti / Al (~0.065) therefore indicate a terrigenous input dominated by aeolian dust, whilst lower ratios (~0.055) signify fluvial transport (Wehausen and Brumsack, 2000; Lourens et al., 2001; Larrasoaña et al., 2003a).

The Si / Al ratio reflects both terrigenous North African and biosiliceous material. When originating from North Africa, Si is associated with quartz and Al with detrital clays, both terrigenous components of the sediments (John et al., 2003). When this relationship changes, and Si is associated with a biosiliceous origin, the proportion of Si to Al increases. Mg is incorporated into clay minerals, secondary carbonates and sea salts (Wehausen and Brumsack, 1998). Palygorskite, Mg-rich smectites and sepiolite have all been associated with a North African origin (Foucault and Mélières, 2000). Increasing Si / Al and Mg / Al ratios therefore either result from enhanced terrigenous supply of Si- and Mg-rich material or from changes related to restriction phases when biosiliceous productivity can be enhanced (Rouchy et al., 1995; Suc et al., 1995; Bellanca et al., 2001).

Changes in bottom water ventilation and productivity are traced by the Mn / Al, V / Al and Fe / Al ratios. These ratios help identify disruptions in the Atlantic-Mediterranean connection, which can result in stagnant bottom-water conditions (van Santvoort et al., 1997; Larrasoaña et al., 2003b). Furthermore, the ratios can identify diagenetic imprints of sapropels on the marl beds. Both, Mn and Fe, are remobilised and reprecipitated as a result of the downward movement of the oxidation zone during early diagenesis (van Santvoort et al., 1997). Higher V concentrations indicate reduced bottom-water ventilation (van Santvoort et al., 1997; Wehausen and Brumsack, 2000; Larrasoaña et al., 2003b). Enhanced Mn / Al, V / Al and Fe / Al ratios are therefore also indicative of increased diatomite productivity (Nijenhuis, 1999).

Mineral identification using X-ray diffraction (XRD) was performed on a few selected samples using a *Philips X'Pert Pro* multipurpose diffractometer with a Cu tube and an automated divergence slit of 110° 2θ. The measurements were performed as continuous scans from 3-85° 2θ with a calculated step size of 0.016° 2θ. Data were analysed using the *QUAX* software package, which provides a semi-quantitative abundance for each identified mineral. For a detailed description of the method see Vogt et al. (2002).

3. Results

Variations in sediment supply are traced in the marls below the hiatus by sedimentation rate and Al mass accumulation rate (MAR) (Peterson et al., 2000). The sedimentation rate at Monte Gibliscemi has its lowest values of ~2 cm / kyr in the older parts of the section. From 9.6 Ma values increase and remain high throughout the section, with maximum values of ~7.5 cm / kyr (Fig. 2). There are two drops at 8.5 Ma and 7.75 Ma, after which the sedimentation rate recovers again to its highest values (Fig. 2). The Al-MAR has its lowest values at the base of the section and increases from 9.4 Ma, reaching maximum values prior to 8.5 Ma (Fig. 2). The values then decrease to an intermediate level at 8.5 Ma, before recovering to close to the maximum towards the top of the section (Fig. 2). The Al-MAR is spikier than the sedimentation rate curve; distinct peaks at e.g. 9.1 Ma, 8.9 Ma and 8.75 Ma correspond to minimum $CaCO_3$ values (Fig. 2). However, the highest Al spike at 8.65 Ma is not accompanied by a $CaCO_3$ low.

The $CaCO_3$ content in Monte Gibliscemi nearly mirrors the Al content. High values at the base of 40 – 50 % decrease to minimum values of ~30 % between 9.1 – 8.75 Ma (Fig. 2). Then the values increase again to ~50%,

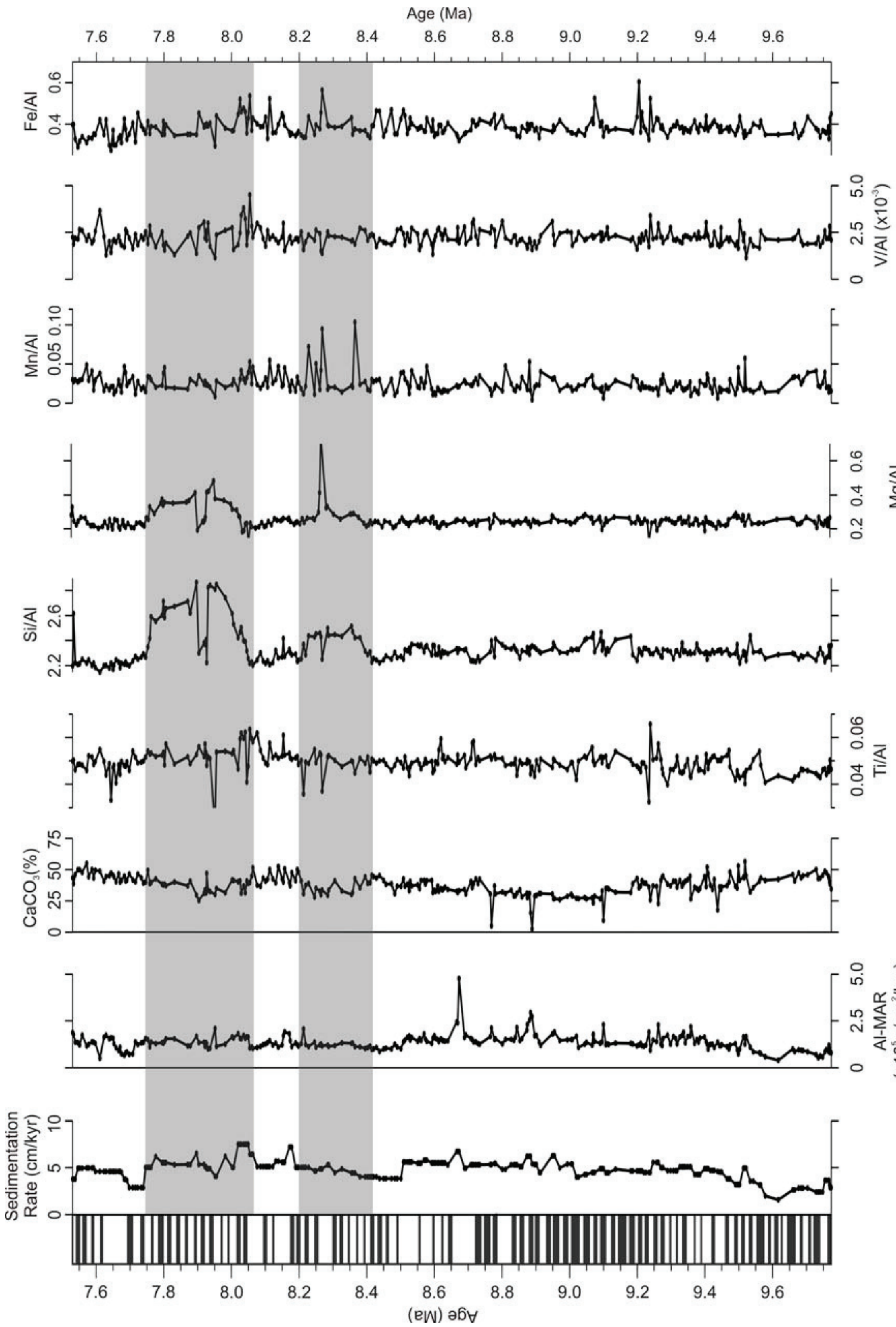


Fig. 2: Age profiles of the proxies discussed in the text, below the hiatus, and a stratigraphy column to show the marl-sapropel pattern of the Monte Gibliscemi section. The stratigraphy is indicated, where white = homogenous marls, black = sapropels; (taken from: Krijgsman et al., 1995). The grey bars indicate two intervals of enhanced Si / Al and Mg / Al ratios, when also ratios indicative of reduction in the bottom-water ventilation are elevated.

however between 8 – 7.9 Ma, values drop to ~30%. The $CaCO_3$ concentrations are fairly low for marl intervals, but still fall within the ranges of Pleistocene carbonate cycles described by van Os et al. (1994) and are above the reported values of sapropels (25 – 30%) in their study. Low $CaCO_3$ values can be explained by dilution effects from high terrigenous sediment supply or by changes in palaeoproductivity (van Os et al., 1994) as carbonate dissolution is unlikely to have occurred at the depositional depth of ~1200 m.

Except for isolated peaks, the Ti / Al ratio at Monte Gibliscemi does not show any major variations or trends. From the base to 9.5 Ma, the values of the ratio are distinctly lower than 0.05 and then they vary around 0.05 to the top of the section Fig. 2). Values below 0.05 are the lower end member of Ti / Al ratios for modern rivers (Nijenhuis, 1999). In some intervals (9.5 - 9 Ma, 8.8 - 8.6 Ma, 8.2 - 8 Ma) (Fig. 2), the ratio is approximately 0.06 or higher, which is closer to the values of aeolian dust ratios in the central Mediterranean (Nijenhuis, 1999; Wehausen and Brumsack, 2000). Hence it is possible that these intervals represent a relative increase in aeolian dust input.

Si / Al values at Monte Gibliscemi fluctuate around 2.3 prior to 8.4 Ma, without major variations (Fig. 2). A first peak with values up to 2.45 is observed in the time interval 8.4 – 8.2 Ma, followed by a second peak with values of 2.6 – 2.8 from 8.05 – 7.75 Ma (Fig. 2). Then the values drop again to 2.2 which is the lowest in the section and increase to 2.6 just prior to the hiatus at 7.52 Ma (Fig. 2). In the second peak, there is a drop in values at 7.9 Ma, which coincides with the diatomite layer in the section (Sprovieri et al., 1999). This indicates that the Si / Al ratio does not necessarily reflect the presence of diatomites but rather traces high biosiliceous productivity. The Mg / Al ratio in Monte Gibliscemi shows a similar pattern to the Si / Al ratio. Relatively constant values, fluctuating around 0.25, are present prior to 8.4 Ma (Fig. 2). The Mg / Al ratio has elevated values between 8.4 – 8.25 Ma and the double peak described in the Si / Al ratio between 8.05 – 7.75 Ma is also present, with values of ~0.4. The ratio drops at 7.9 Ma (Fig. 2) where the diatomite layer is present. After 7.75 Ma, low Si / Al values of ~0.2 return. Constant values of Si / Al and Mg / Al suggest a consistent North African source area.

The increase in Si / Al and Mg / Al ratios indicates a higher Si and Mg accumulation on condition that Al flux remained constant. A decrease in terrigenous contribution from North Africa could enhance the signal, but this is in disagreement with the intermediate to high values of the Al-MAR record (Fig. 2). The increase in Si could also be related to enhanced biogenic silica production. Preferred conditions for biogenic silica production are commonly related to riverine input (Van der Zwaan, 1979; van der Zwaan, 1982) or upwelling conditions (McKenzie et al., 1979-1980).

Prior to 8.4 Ma, Mn / Al and V / Al ratios show variations around their respective means of ~0.26 and ~0.002; no coeval peaks are present (Fig. 3). Only Fe / Al shows isolated peaks at 9.25 and 9.1 Ma (Fig. 3) that can be associated to the presence of Fe-rich minerals. Diagenetic enrichment of Fe resulting from burnt down sapropels is unlikely as there are no related Mn enrichments at these peaks (Fig. 2). However, volcanic ash layers are known from the lower Tortonian of Sicily and can result in Fe enrichment (Kuiper et al., 2005). Between 8.4 – 8.2 Ma peaks are present in the Mn / Al and Fe / Al profiles which are nearly synchronous (Fig. 2). The coincidence of the peaks suggests that both elements have been remobilised and precipitated as part of the oxidation front related to diagenesis (van Santvoort et al., 1997). The proxy parameters show that isolated intervals have to be treated with care as remobilisation and diagenetic overprinting may be present in the marls. Coincident peaks in V / Al and Fe / Al are also present between 8.1 – 8 Ma, when Mn / Al does not show much variation (Fig. 3). V / Al is a proxy for bottom water ventilation; enhanced V indicates stagnation of the deep water (Larrasoaña et al., 2003b). Under the assumption of stable Al fluxes this suggests restricted bottom water circulation during this time period. Similar conditions were present at 7.6 Ma prior to the hiatus, when both the V / Al and Fe / Al ratios show a coeval peak. The first appearance of peaks in the redox sensitive ratios is synchronous with changes in Si / Al and Mg/Al (Fig. 3), suggesting that there may be a possible link between them.

4. Discussion

4.1 Late Miocene climate of North Africa

The geochemical proxy records of the lower part (9.5 - 8.4 Ma) of the Monte Gibliscemi section show little variation, suggesting stable climatic conditions (Fig. 2). The sedimentation rate is high and the relatively low $CaCO_3$ content indicates high terrigenous input originating from the North African margin. The low Ti / Al ratios, around ~0.05 (Fig. 2) are in the range of what is expected of fluvial sediment supply (Nijenhuis, 1999). The geochemical records from Monte Gibliscemi thus indicate that North African rivers were draining into the Central Mediterranean during the Tortonian, likely the Gabes (Fig. 1), which would imply more humid conditions from at least ~9.5 Ma in northwest Africa.

The humid conditions recorded at Monte Gibliscemi during the late Tortonian are not in agreement with the previous results from the Metochia section of the Eastern Mediterranean (Köhler et al., submitted-a). On Gavdos, aeolian dust input was identified prior to ~8.2 Ma and more humid conditions thereafter (Köhler et al., submitted-a). Other studies have also recorded a late Miocene reduction in the dust flux off the West and East coasts of Africa (Ruddiman et al., 1989; deMenocal and Bloemendal, 1995), in particular a long record from the West coast suggests a climatic change towards more humid conditions in Central and North Africa prior to ~8 Ma (Ruddiman et al., 1989). Stable warm and humid conditions in northwest Africa, following a short interval of pronounced aridity, have been related to changes in global climate during the early late Miocene (van Zinderen Bakker and Mercer, 1986). It is therefore possible that the Monte Gibliscemi area was influenced by more humid palaeoenvironmental conditions in northwest Africa, which may have occurred earlier than the changes reported in northeast Africa (Griffin, 1999).

During the latest Tortonian, North Africa was probably drained by several river systems (Burke and Wells, 1989; Griffin, 2002; Griffin, 2006; Gladstone et al., 2007). In order for these rivers to flow, at least seasonally, the North African climate must be more humid and the intertropical convergence zone (ITCZ) must have been located northwards of its modern day position, (Fluteau et al., 1999; Gladstone et al., 2007). The ITCZ must cross the central

Saharan watershed of ~21° N (Rohling et al., 2002; Larrasoaña et al., 2003a) to enable rivers other than the Nile to drain into the Mediterranean (Rohling et al., 2002; Larrasoaña et al., 2003a; Griffin, 2006). The River Nile, commonly considered as an indicator of African monsoon variability, is not necessarily representative of North African climate. Its catchment area extends into (sub)-tropical regions (e.g. Rohling et al., 2002), where the Ethiopian highland, experienced a late Miocene uplift phase (Gani et al., 2007) and rainfalls fed the Sudd region in Sudan (Said, 1993; Griffin, 2002).

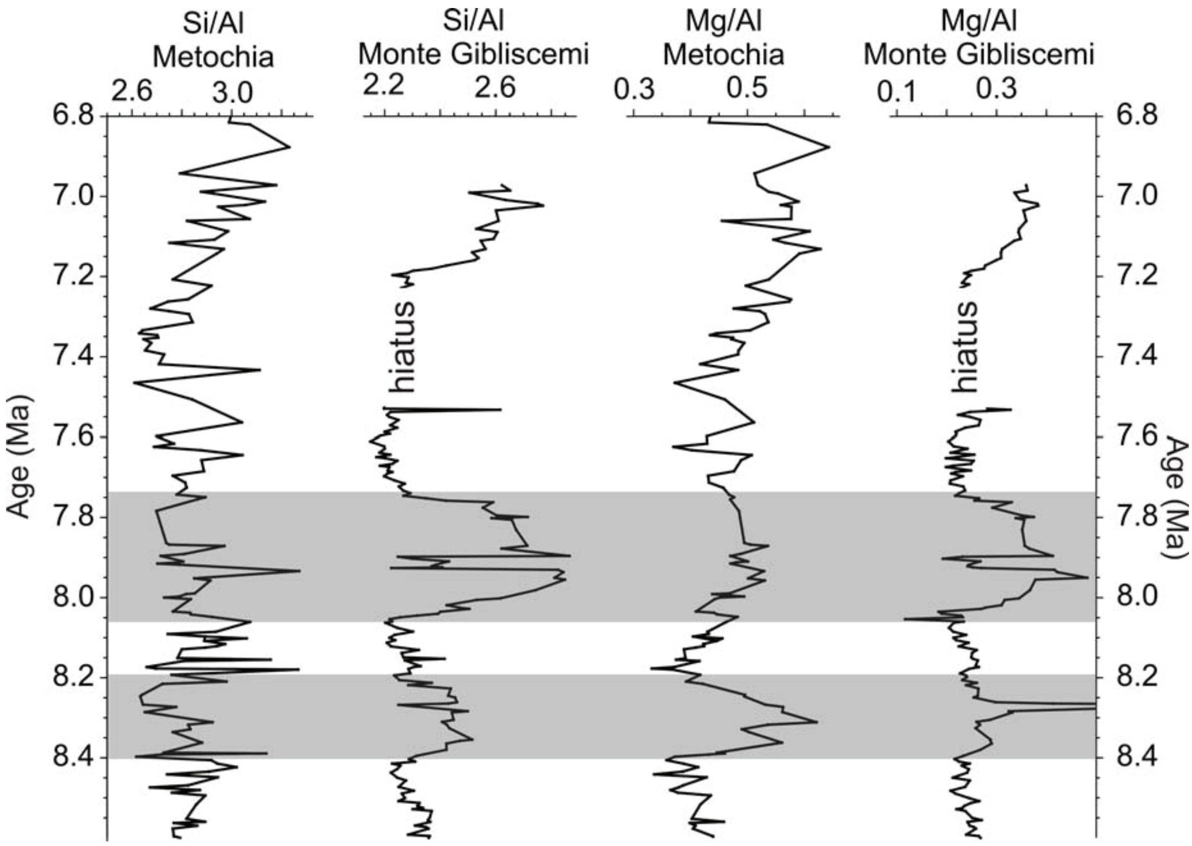


Fig. 3: Si / Al and Mg / Al ratios from the Metochia section, Gavdos, compared to those from Monte Gibliscemi, extended to the time period after 7.16 Ma (early Messinian) when restrictive conditions commenced in the Mediterranean as a result of changes in the Mediterranean-Atlantic connection. The base of the Tripoli Formation on Sicily is at 7.005 Ma (Hilgen and Krijgsman, 1999).

Modelling studies show that the freshwater input from the North African rivers into the Mediterranean was threefold higher than today (Gladstone et al., 2007), influencing the Eastern Mediterranean hydrological budget (Griffin, 2006). Knowledge of the hydrological budget of the late Miocene is important for model calculations of salt precipitation during the Messinian Salinity Crisis, which currently rely on modern hydrological data (Benson et al., 1991; Flecker et al., 2002; Meijer and Krijgsman, 2005). It was shown by Meijer and Krijgsman (2005) that the degree of saturation required for salt precipitation depends on the freshwater input into the Eastern Mediterranean; therefore knowledge of the river runoffs becomes very important. The Monte Gibliscemi proxy data indicate fluvial input from at least 9.5 Ma, and the locations of the North African rivers indicate late Miocene drainage of at least one of the systems into the westernmost tip of the Eastern Mediterranean (Fig. 1).

4.2 Late Tortonian restriction of the Mediterranean

The geochemical data from Monte Gibliscemi show two large peaks in Si / Al and Mg / Al between ~8.4 and 7.8 Ma;. both ratios also increase prior to the hiatus at 7.52 – 7.24 Ma (Fig. 2). As this hiatus is a post-depositional shear plane, these changes are not associated with it. The Si / Al and Mg / Al ratios also increase above the hiatus during the Messinian (Fig. 3). The Metochia section of Gavdos Island, situated south of Crete, is also a late Miocene section and covers the time interval corresponding to the hiatus of the Monte Gibliscemi section (Fig. 3). Interestingly, when comparing the Si / Al and Mg / Al ratios of both sections (Fig. 3), only the Mg / Al ratio shows a similar variation from 8.4 Ma in the Metochia marls. The Si / Al ratio only increases from 7.25 Ma onward (Fig. 3). If high Si / Al is associated with biosiliceous productivity resulting from enhanced riverine input, it would indicate that the Gavdos Basin (Metochia Section) was less influenced by riverine input. This scenario is possible as most North African rivers drained into the Eastern Mediterranean further west, i.e. closer to Monte Gibliscemi (Fig. 1). Increased fluvial input from North Africa can provide the nutrients needed for the enhanced biosiliceous productivity. However, the Ti / Al ratio does not show major changes (Fig. 2) over the time interval of interest. Therefore, it is unlikely that riverine input changed considerably from the conditions prior to the increase of biosiliceous productivity. Hence conditions not related to African climatic change appear to be the likely cause of the increase in Si and Mg.

The two phases of enhanced Si and Mg coincide with peaks in V / Al, Mn / Al and Fe / Al at the beginning of each Si / Al and Mg / Al ‘pulse’ (Fig. 2). The peaks of V / Al and Mn / Al are indicative of less oxygenated bottom-water conditions (Kouwenhoven et al., 1999; Larrasoaña et al., 2003b), whereas the higher Fe abundances under such conditions can be ascribed to the presence of pyrite (van Os et al., 1994). In present-day hypersaline anoxic basins in the Mediterranean, decreased oxygenated bottom water and increased salinity have been reported, along with an increased silica concentration of bottom waters (de Lange et al., 1990; Nijenhuis, 1999). Similar conditions have been described during oceanic convergence stages in the Neo-Tethys (Shoval, 2004), when high silica concentration conditions, combined with hypersaline bottom waters, caused authigenic formation of Mg-rich clays like palygorskite and sepiolite (Shoval, 2004).

We will now address the mineralogical composition of our samples derived from X-ray diffraction analysis. The sediments consist of a mixture of calcite, quartz, clay minerals, feldspars and traces of other minerals like pyrite and opal (cf. Table 1). The $CaCO_3$ content is between 30-44%, with just one sample containing only 20%, and is therefore within the range of calculated $CaCO_3$ from the XRF data (Fig. 2). Quartz fluctuates between 11-18 %, and the sum of the clay minerals between 37-50%, where the sample with the lowest $CaCO_3$ has a clay content of 61%. The clay mineralogy is mainly kaolinite, illites, smectites, palygorskite and chlorites. Moreover in some samples sepiolite is determined. Traces of clinoptilolite are identified in isolated samples (Table 1).

Of particular interest to this study are the amounts of the clay minerals kaolinite, palygorskite and sepiolite. Kaolinite is present in all samples and varies around ~20 % in the clay mineral fraction. Palygorskite is not present in all samples however it is a component of the samples from 8.3 Ma onwards and then ranges between 12-19%. The clay mineralogical fraction of the sample at 6.95 Ma has a palygorskite content of ~35 %. Sepiolite is present in a sample at 7.1 Ma.

For comparison, selected samples of the Metochia section are presented during the same time interval. $CaCO_3$, quartz and the total amount of clay minerals are within similar ranges as in the Monte Gibliscemi marls. Kaolinite is also present in most samples, only at ~8.5 Ma no kaolinite is present in the clay fraction, but sepiolite is found instead. From ~8.5 Ma onward, palygorskite is found in the clay minerals.

Kaolinite is a detrital mineral commonly associated with humid conditions in North African (John et al., 2003). Palygorskite, when of detrital origin, reflects aridity (Chamley, 1989), however authigenic formation in marine sediments has been reported (Shoval, 2004). Kaolinite is prominently present in the marls, indicating humid palaeoenvironmental conditions in North Africa (Table 1), as is also indicated by the XRF elemental chemistry data. From 8.5 Ma onward in the Metochia section (and 8.3 Ma at Monte Gibliscemi – different samples had to be analysed) palygorskite is found along with kaolinite. There are traces of opal and clinoptilolite present in the bulk sediment and one sample of each of either section has a notable sepiolite content (Table 1). Clinoptilolite can form in high productivity environments, needing the presence of biosiliceous material (e.g. John et al., 2003), like authigenic palygorskite. It is therefore possible that the conditions needed for authigenic clay formation were present from 8.4 Ma onward in the high Si / Al zones.

Early Messinian sediments of the Mediterranean (Tripoli formation of Sicily) are also characterised by increased biosiliceous sedimentation and hypersaline conditions (Suc et al., 1995; Bellanca et al., 2001; Rouchy, 2001; Blanc-Valleron et al., 2002). During the early Messinian interval, the Si / Al and Mg / Al ratios of the Monte Gibliscemi section (Fig. 3) show similar values as in the earlier two intervals of elevated ratios between 8.4 and 7.8 Ma (Figs 2 and 3), suggesting similar palaeoconditions. Increased biosiliceous production in Messinian times has commonly been associated with restriction phases of the Mediterranean (e.g. Bellanca et al., 2001), because the Mediterranean-Atlantic connection through southern Morocco rapidly deteriorated at 7.1 Ma (Krijgsman et al., 1999). Therefore, increases in the Si / Al ratio are taken as a first approximation for restrictive phases present in the record. Elements associated with changes in the bottom water oxygenation, e.g. V and Mn (van Santvoort et al., 1997; Kouwenhoven et al., 1999; Larrasoaña et al., 2003b) can be expected to be elevated during enhanced biosiliceous productivity (Nijenhuis, 1999). Therefore the peaks in the ratios of the redox sensitive elements can be indicative for restrictive phases.

Restrictive phases have previously been identified in the foraminifera record of Monte Gibliscemi at 8.5 Ma and 8 Ma, with benthic foraminifera diversities changing towards more stress-tolerant species (Kouwenhoven et al., 1999; Seidenkrantz et al., 2000). These changes in the late Miocene Mediterranean sediments were linked to Mediterranean-Atlantic gateway dynamics.

Table 1: XRD derived abundances of minerals of the bulk sediment of the Monte Gibliscemi and Metochia sections. The clay mineralogy is calculated from the sum of the phyllosilicate fraction of the bulk Metochia sediment

Section	Monte Gibliscemi							Metochia				
Sub-Section	GIA	GIA	GIB	GIB	GIB	GIB	GIB	GP	GP	MC	MC	MC
Sample	25	103●◊	218*◊	62*●	58	26*●◊	1	55	258◊	49	82	191*
Age (Ma)	9.68	9.26	8.32	7.53	7.23	7.08	6.96	9.40	8.51	8.18	7.99	7.29
$CaCO_3$ (%)	**44**	**20**	**38**	**31**	**37**	**33**	**30**	**19**	**35**	**35**	**48**	**50**
Quartz (%)	**12**	**18**	**16**	**15**	**13**	**13**	**18**	**17**	**11**	**20**	**15**	**8**
Sum of clay minerals (%)	**35**	**61**	**42**	**50**	**37**	**49**	**37**	**54**	**42**	**33**	**32**	**31**
Other mineral components (%) **	**9**	**1**	**4**	**4**	**13**	**5**	**15**	**10**	**12**	**12**	**5**	**11**
Kaolinite (%) ***	**31**	**22**	**21**	**22**	**19**	**14**	**27**	**11**	**0**	**24**	**13**	**10**
Palygorskite (%) ***	**0**	**0**	**12**	**18**	**19**	**18**	**35**	**0**	**10**	**18**	**16**	**6**
Sepiolite (%) ***	**0**	**0**	**0**	**0**	**0**	**22**	**0**	**0**	**20**	**0**	**0**	**0**

* traces of opal, ● traces of clinoptilolite, ◊ traces of pyrite, ** includes feldspars and minor components, *** calculated from total phyllosilicate amount

During the late Miocene, the Mediterranean was connected to the Atlantic ocean via two gateways: the Rifian corridor (Morocco) (Krijgsman et al., 1999) and the Betics (southern Spain) (Soria et al., 1999). These connections became gradually restricted, causing the Mediterranean to become more isolated, finally resulting in the Messinian Salinity Crisis (Hsü et al., 1973). The first connection that closed was the gateway through southern Spain (Soria et al., 1999; Betzler et al., 2006). Biostratigraphic results from the Guadix basin, which has a central position in the gateway, indicates that the progressive closure took place during the late Tortonian. Magnetostratigraphic dating of the sedimentary sequences of the Fortuna basin in the east part of the gateway, indicated that open marine sedimentation entirely ceased at ~7.8 Ma, culminating in the deposition of diatomites and evaporites of the Tortonian salinity crisis of the eastern Betics (Krijgsman et al., 2000). As a result of these conditions the circulation in the Mediterranean may have become more sluggish causing water body stratification, increasing oxygen stress and bottom water salinity (Kouwenhoven et al., 1999; Seidenkrantz et al., 2000; Kouwenhoven et al., 2003) and the formation of anoxic basins (Kouwenhoven and van der Zwaan, 2006). To drive such weak circulation supporting the formation of biosiliceous sediment and authigenic clays, the connections of Mediterranean-Atlantic must be deep enough to allow water exchange (Meijer et al., 2004). However, the depth of the Sicilian sill should not be neglected here, which forms the boundary between the Western and Eastern Mediterranean basins. Its depth is important in controlling circulation in the Eastern basin (Meijer et al., 2004). The circulation in the Eastern Mediterranean is influenced by the salinity in the Adriatic Basin, where lower salinities, caused by higher influx of Atlantic water (deeper sill), reduce the dense deep water (Meijer et al., 2004). As a result, the deep circulation of the Eastern Mediterranean decreases (Meijer et al., 2004).

The sudden termination of the Si / Al and Mg / Al peaks at ~7.8 Ma suggests that restricted conditions in the Mediterranean also terminated rapidly, with open connections being re-established quickly. Since the Betic gateway is supposed to be closed at that time, and Gibraltar probably did not yet exist, it must have been the Rifian Corridor that caused this change. Indeed, the late Miocene sedimentary records of the Rifian foredeep show a significant marine transgression during the late Tortonian which has been interpreted as the opening of the Rifian Corridor (Krijgsman et al., 1999). Magneto-biostratigraphic age constraints on this transgression event (between 8 and 7.6 Ma) were extrapolated from the base of the section indicating deepening of the Rifian corridor and are not very accurate, but our results from Monte Gibliscemi suggest that it occurred at 7.8 Ma.

5. Conclusions

The sedimentary archive of Monte Gibliscemi is a late Miocene record of North African palaeoenvironmental conditions and disruptions in the Mediterranean-Atlantic connection. The North Africa were humid with rivers draining into the central Mediterranean, and fluvial input dominating over aeolian contributions from 9.5 Ma throughout the section. There are two phases between 8.4 – 7.8 Ma, when the geochemistry of the marls is similar to early Messinian sediments with enhanced biosiliceous sedimentation, resulting from weaker circulation in the Eastern Mediterranean. These conditions enabled the formation of authigenic palygorskite in the basin and are linked to disruptions of the Mediterranean-Atlantic connections, which changed gradually from the late Tortonian. The data shows that the transgression associated with the opening of the Rifian corridor occurred at 7.8 Ma.

6. Acknowledgements

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5. Conclusions

In this study, late Miocene changes in the North African monsoon system and Mediterranean palaeogeographic reorganisations were reconstructed. Eastern Mediterranean uplifted marine sections, which were astronomically dated during previous studies, provided suitable archives to study and date these changes.

A key aspect of this research was the unravelling of the different terrigenous source areas which contributed sedimentary material to the Mediterranean and then identifying changes in the palaeoenvironmental conditions. This was especially important in the more eastern Metochia section, which was dominated by input from the former Aegean landmass, but also had aeolian dust input from North Africa. The identification and differentiation of these two source areas was performed successfully on the basis of the different geochemical signatures of the terrigenous sediments and the key factor that hematite was only associated with aeolian dust from North Africa. The terrigenous component reaching the Monte Gibliscemi section during the late Miocene was derived mainly from North Africa. This situation fits well with the proximity of the section to the North African continent and it is important to note that during the late Miocene Italy was located further west and was therefore unlikely to be a major source of sedimentary material.

Variations in the strength of the North African monsoon were traced by changes in the relative importance of fluvial and aeolian transport mechanisms and the abundance of aeolian dust in the Mediterranean sections. In the more eastern Metochia section, the contribution of aeolian dust, identified by the hematite abundance, was reduced to a minimum after ~8.2 Ma. The signal in the Monte Gibliscemi section is different with fluvial input present from ~9.5 Ma. Late Miocene river systems were present across much of North Africa and drained towards the more central region of the Mediterranean, where the sediments of Monte Gibliscemi were deposited. The identification of fluvial sediments in the Monte Gibliscemi section demonstrates that these ancient river systems were active from at least 9.5 Ma.

Tectonic reorganisations in the Mediterranean were also traced in the sediments of the studied sections. The Metochia section covers the time interval when the Aegean landmass fragmented, which is recorded in the sediments as a dramatic cessation of the supply of terrigenous material. and the Sea of Crete opened. The identification and dating of this episode means that an accurate age of ~8.2 Ma can be assigned to the opening of the Sea of Crete.

In Monte Gibliscemi, the tectonic changes in association with the dynamics of the Mediterranean-Atlantic gateways are recorded. First, the near closure of the Betic corridor and then the opening of the Rifian gateway reduced the circulation of the Mediterranean Sea. The geochemical proxy record and the clay mineralogy indicate that these changes led to the formation authigenic clays in the sediment from ~8.4-7.8 Ma. The transgression associated with the opening of the Rifian corridor can be accurately dated at 7.8 Ma.

It has been shown in this study that the late Miocene sediments of Mediterranean sections are a high fidelity archive of information with which North African climate change and palaeogeographic reorganisations can be reconstructed. It would therefore be profitable to extend the analysis to additional sections across the Mediterranean. This will provide a better understanding of the changing palaeoenvironmental conditions in North Africa and help palaeoclimatic modelling studies, which need more proxy data to provide realistic boundary conditions. Also, the tectonic reorganisations affecting the Mediterranean regions can be put into a better context when precise ages are known. The data and ages can be used to argue for a common cause, such as the northward movement of the African plate, for apparently disparate events.

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Chapter 2
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