STABLE ISOTOPIC COMPOSITION OF THE KOMLÓ CALCAREOUS MARL FORMATION („SPOTTED MARL" S. STR.), MECSEK MOUNTAINS, S HUNGARY

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ABSTRACT

In the Mecsek Mountains (Tisza Unit, S Transdanubian, Hungary) an Aalenian-Bajocian rhythmic limestone-marlstone alternation crops out. Precise petrological analysis has not been accomplished on this formation and the origin of this characteristic bedding phenomenon has not been explained. Stable isotope values of carbonate samples show higher $\delta^{18}O$ and $\delta^{13}C$ values in limestone beds relative to marlstone beds. A correlation exists between the carbonate content and the stable isotope values. On the basis of this isotope data and other observations on the collected samples can be devised a depositional model of Komló Formation. The data suggest that production and/or dilution cycles may have formed the limestone-marlstone alternation; diagenetic overprint was only of minor significance.

INTRODUCTION

In the eastern part of Mecsek Mountains outcrops of Mesozonic sedimentary and volcanic formations of Tisza Unit can be studied over a relatively large area. A characteristic sedimentary unit is a limestone-marlstone series, the Komló Calcareous Marl Formation. This formation is pat of the so-called „spotted marl" series, that overlies a siliciclastic coal-bearing unit *(Fig. 1)*. the precise depositional age of the Komló Calcareous Marl is uncertain, although a rough range in Aalenian-Bajocian interval has been long ago given (VADÁSZ, 1935, FORGÓ et al., 1966). The most conspicuous character of this formation is its rhythmic limestone-marlstone alternation (FORGÓ et al., 1966). The origin of this bedding has not yet been studied in detail.

In this study carbon and oxygen isotopes are applied to elucidate the origin of the characteristic bedding. For ice-free periods during the Phanerozoic, $\delta^{18}O$ variations have been interpreted in terms of in salinity changes (PRATT, 1984) or changes in surface water temperatures (DE BOER, 1982), while $\delta^{13}C$ variations are thought to reflect organic productivity and water stratification (ARTHUR et al., 1984).

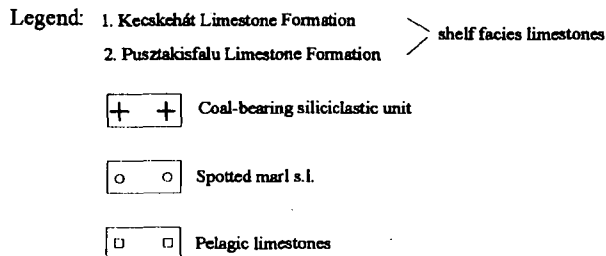---

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Tithonian	Márévár Limestone Formation
Kimmeridgian	Várkony Limestone Formation / Kisújbánya Limestone Formation
Oxfordian	Fonyászó Limestone Formation
Callovian	Dorogó Calcareous Marl Formation
Bathonian	Óbánya Limestone Formation
Bajocian	
Aalenian	Komló 2. Calcareous Marl Formation
Toarcian	Óbánya Aleurolite Formation
Pliensbachian	Mecseknádasd 1. Sandstone Formation
Sinemurian	Hosszúhetény Calcareous Marl Formation / Vasas Marl Formation
Hettangian	Mecsek Coal Formation

Legend: 1. Kecskehát Limestone Formation
2. Pusztakisfalu Limestone Formation

〉 shelf facies limestones

+ + Coal-bearing siliciclastic unit

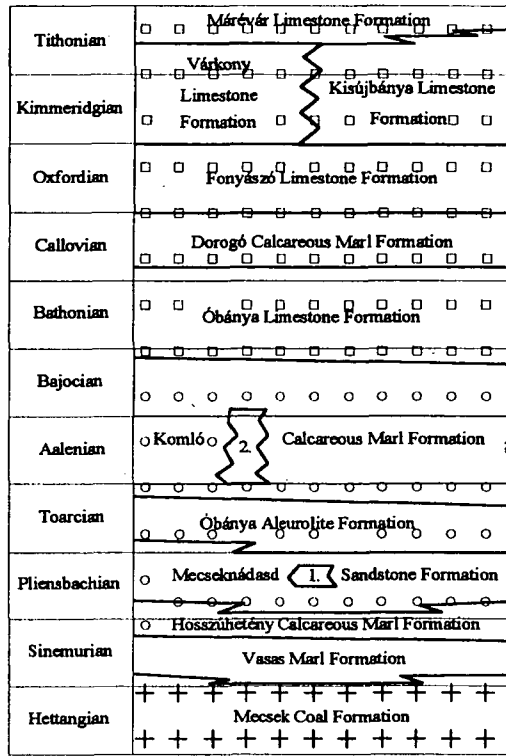o o Spotted marl s.l.

□ □ Pelagic limestones

Fig. 1. Stratigraphic units of Mecsek Mountains

GEOLOGICAL SETTING

Two sections north of Püspökszentlászló were studied in detail (Figs. 2, 3a, 3b). These sections consist an alternation of carbonate-rich and carbonate-poor layers (couplets) with centimeter-decimeter scale bed thickness. Petrographically, the carbonate-rich beds are argillaceous limestones, calcareous marls and marls, the carbonate-poor layers are marls, argillaceous marls. All of these have some silt grains, this silt content is mainly abundant in the carbonate-poor semicouplets. The beds are grey and greenish grey (on weathered surface

some yellowish shade) with abundant darker grey spots. The carbonate-poor beds are slightly darker than the limestones because of their higher clay content. Sharp and continuous bedding contact can also be observed. The carbonate-poor beds with highest clay content are thin bedded, nevertheless all of the carbonate-rich semicouplets are massives. All of beds are macroburrowed. Microburrowing or lamination are absent. Rock Eval pyrolysis did not detect significant organic matter (TOC values between 0.04 and 0.17 %, HETÉNYI, pers. comm.). These facts refer to oxic conditions during deposition of Komló Formation. Locally, sharp bedding contacts may be developed by turbidity current activity. Sedimentary structures, however, indicative of redeposition (including existence of fine-grained turbidities or contourites), e.g., cross lamination, normal or inverse graded bedding, fine lamination, lenticular bedding, fine, obscure silt lenses, tool marks on underlying bedding planes, complete or incomplete Bouma-sequence were not observed.

In thin section the limestones classify as bioclastic wackestones or packstones. The most abundant biogenes are radiolarians and siliceous sponges (commonly recrystallized), and filaments. A small amount of unrounded terrigenous quartz silt grain are present as well. Intensive bioturbation is conspicuous in all samples. Planktonic foraminifera are present in some thin sections. These forams can not be used for precise biostratigraphic dating, but they suggest normal marine salinity (RESCH, pers. comm.).
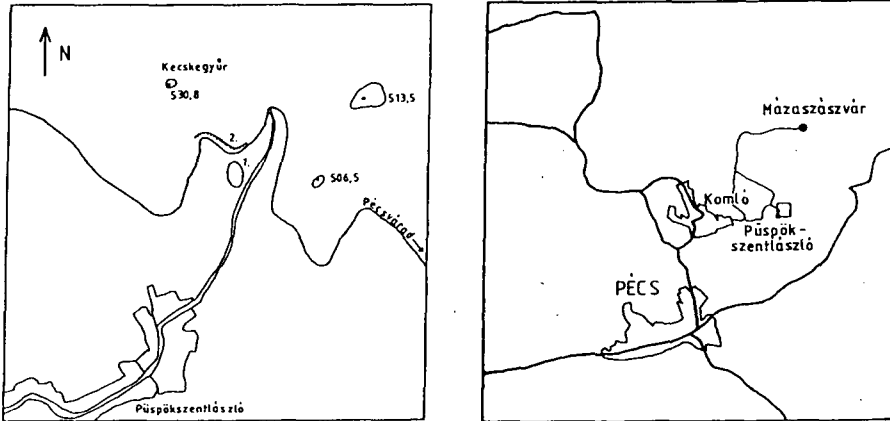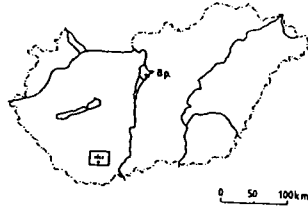

Fig. 2. Location map of the investigated profiles
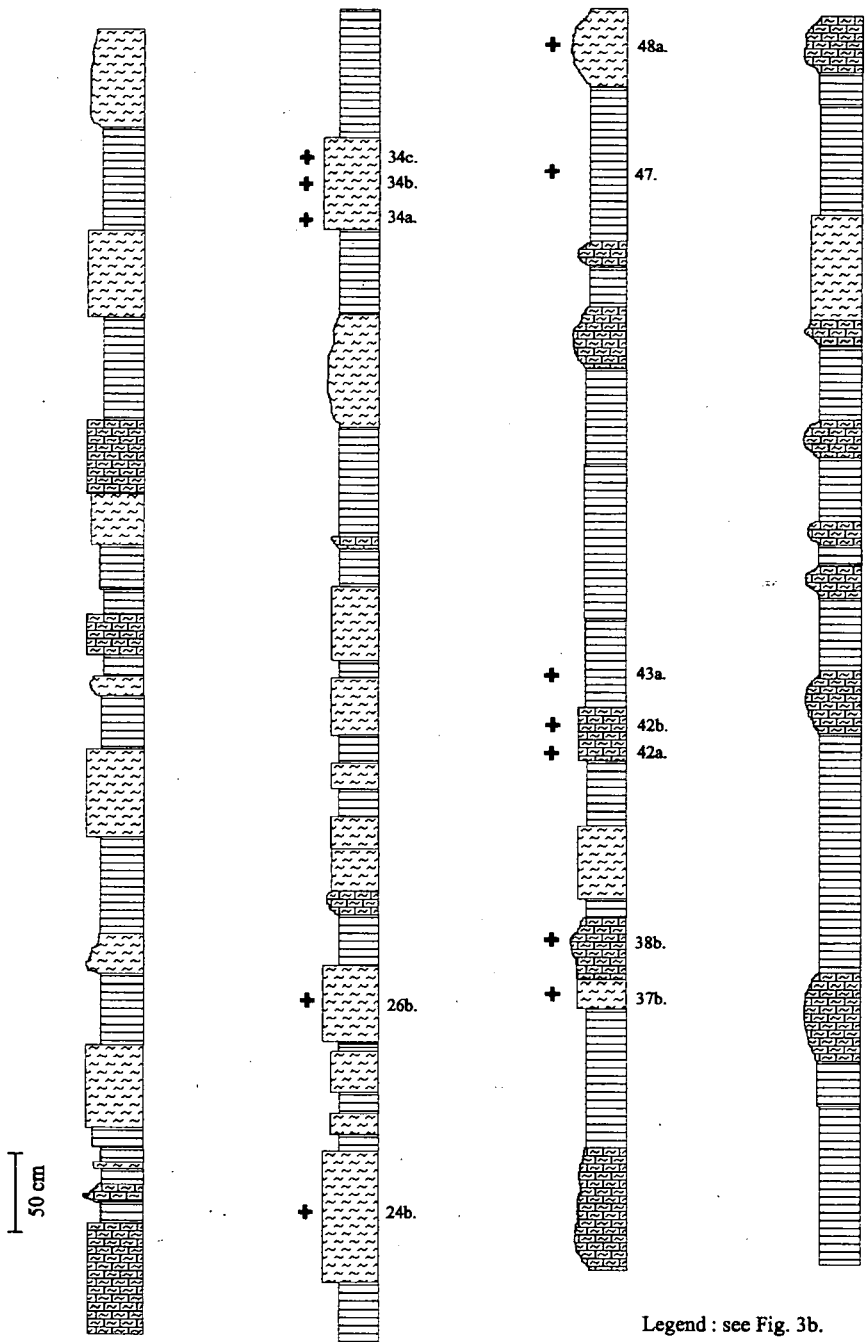Legend: 1. Section Püspökszentlászló II. 2. Section Kecskegyűr, road cut

Fig. 3a.: Lithologic column of section Püspökszentlászló II.

Column annotations (left to right):

Middle column:
- 56.
- 54c.
- 54b.
- 53.
- 52.
- 48.
- 32.
- 31.

Left column:
- 1a.

Right column:
- 59.
- 58b.
- 58a.
- 57.

Legend:

✚   sampled layer (with nummer)

argillaceous marl

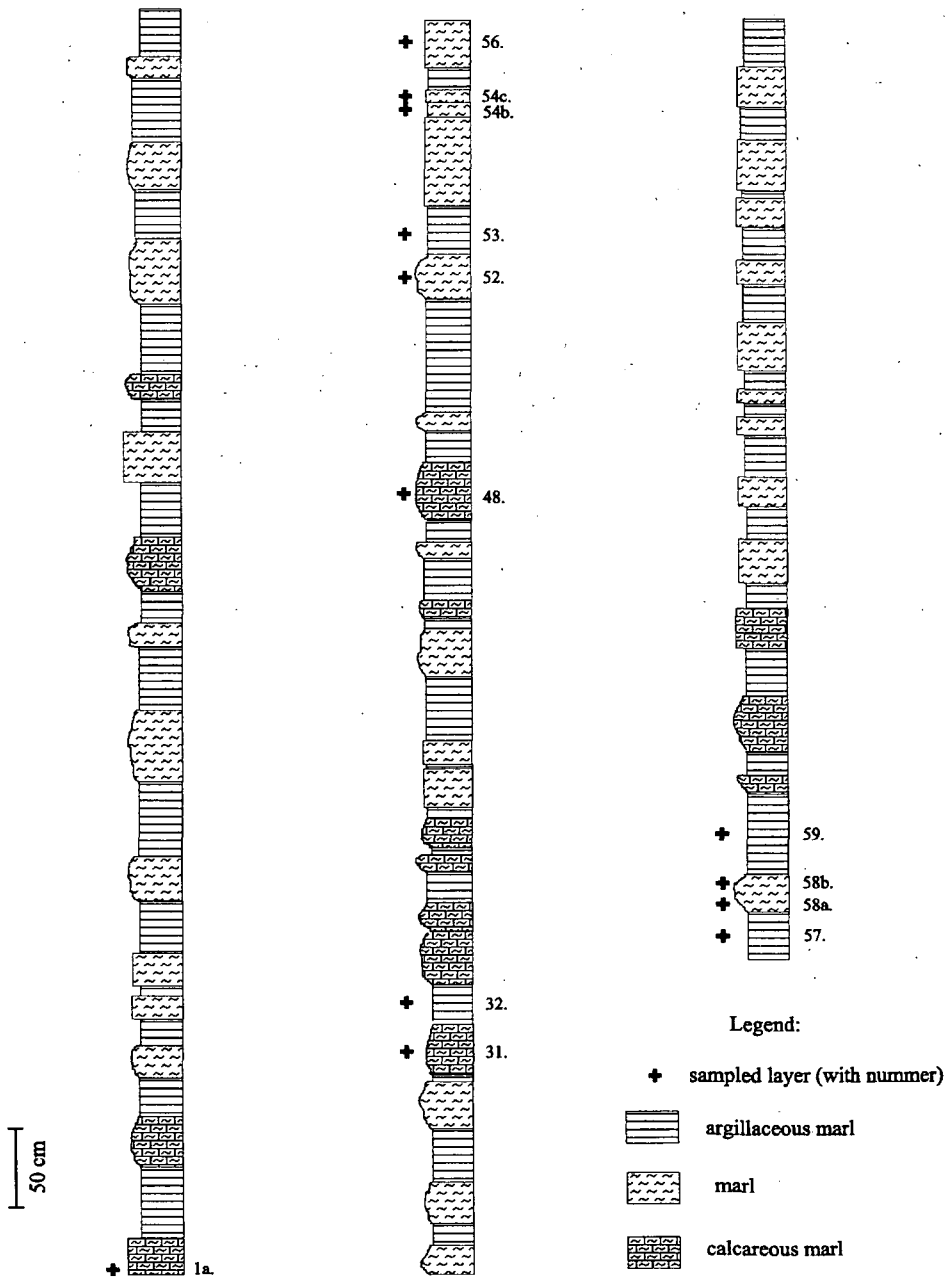marl

calcareous marl

50 cm

Fig. 3b.: Lithologic column of section Kecskegyűr, road cut

A few marlstone samples show fitted fabric structures. This fact and the partially dissolution of some of biogenic constituents indicate carbonate dissolution and reprecipitation during burial diagenesis. In outcrops weavy bedding surfaces are widespread; nevertheless carbonate concretions did not form, i.e. carbonate redistribution in the sense of HALLAM (1964) probably was not significant enough to cause a limestone-marl alternation.

In essence, this profile represents a basinal facies, dominated by hemipelagic processes. Sedimentation was pesumably continuous. Consequently, this succession is a good candidate for detailed analysis to examine the origin of rhythmic bedding.

METHODS

Bulk samples were milled in an agate mortar and the powder was analyzed by Attila Demény at the Geochemical Research Laboratory of the Hungarian Academy, Budapest. Carbon dioxide was produced following MCCREA (1950). The $^{13}C/^{12}C$ and $^{18}O/^{16}O$ ratios were determined using a Finnigan MAT delta S mass spectrometer. The isotope ratios are quoted in per mil relative to the PDB (Pee Dee Belemnite) and the V-SMOW (Vienna Standard Mean Ocean Water).

The reproducability of duplicate analyses is better than $\pm$ 0.1 ‰. The standard Harding Iceland Spar were also analysed, which yielded the following values: $\delta^{13}C$ = -4.88 $\pm$ 0.03 ‰; $\delta^{18}O$ = 11.85 $\pm$ 0.07 ‰; n = 4 (accepted values: $\delta^{13}C$ = -4.80 ‰; $\delta^{18}O$ = 11.78 ‰, LANDIS, 1983). PBD values were converted to V-SMOW values using the equations: $\delta^{18}O_{sample}/SMOW$ = 1.03091 x $\delta^{18}O_{sample}/PBD$ + 30.91 (COPLEN, 1988).

RESULTS

The results of stable isotope measurements are shown in the Table 1. In a given couplet $\delta^{18}O$, and $\delta^{13}C$ values of the marlstones are always lighter than those of limestones. The $\delta^{13}C$ and $\delta^{18}O$ values show a weak positive correlation (*Fig. 4*). A strong correlation exists between the percentage of calcite and the isotope ratios (*Figs. 5, 6*). In order to examine the potential effect of the noncarbonate fraction on the stable isotope values two samples with different carbonate content and different isotope excursion were treated with 10 % hydrochloric acid. The data show that the noncarbonat matrix has no effect on the stable isotopic composition of the bulk sample. The observed correlations belong to the samples, can be established (DEMÉNY, pers. comm.).

DISCUSSION

Changes ratio in deep-water production during glacial-interglacial transitions and related changes in carbonate dissolution probably can not be called upon to explain the cyclicity in Jurassic strata deposited during a period of globally warm climate. ARTHUR et al. (1984, 1986) proposed that periodic changes in insolation, evaporation, wind stress, and/or rainfall (these factors may be affected the climate under greenhouse conditions) in a wide variety of environments caused changes in input of terrigenous detritus, water mass stratification,
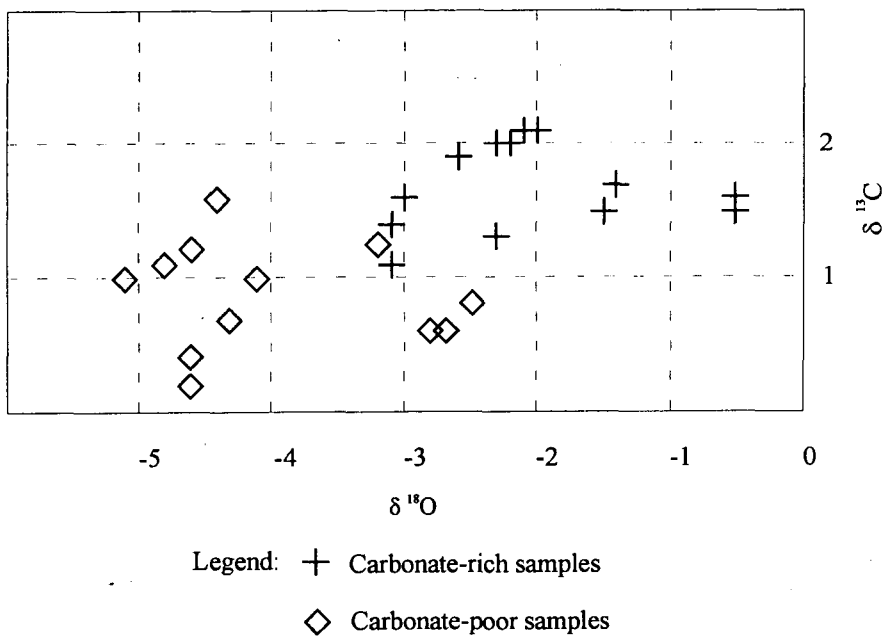
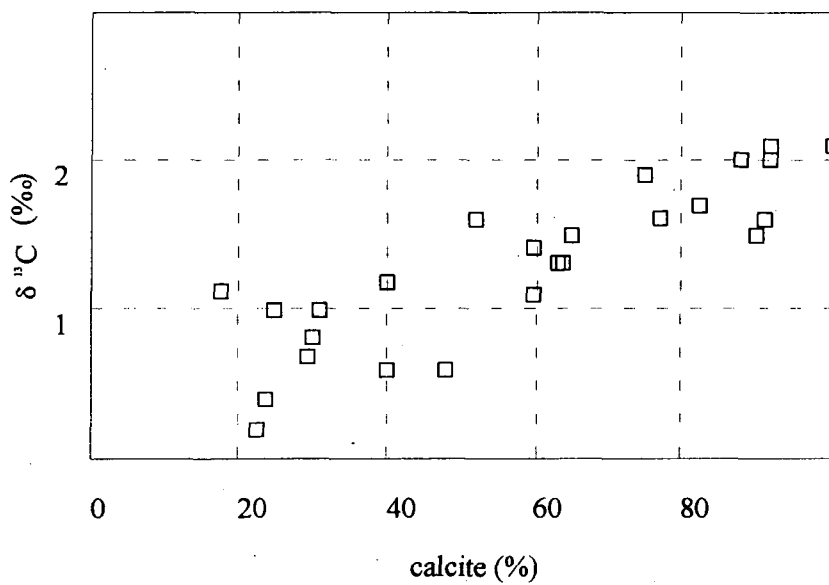*Fig. 4.:* Carbon isotope ratios in function of oxygene isotope ratios

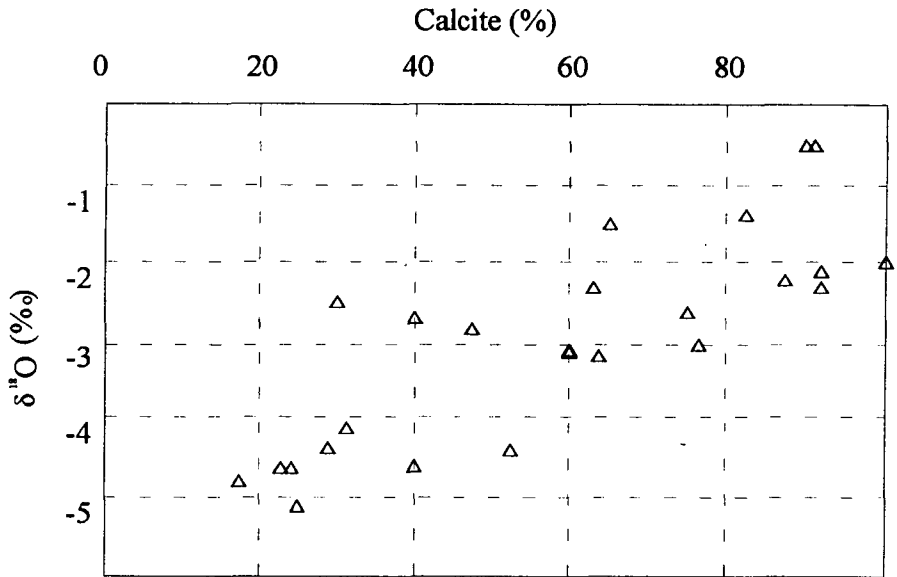*Fig. 5.:* Carbon isotope ratios in function of calcite content

Fig. 6.: Oxygene isotope ratios in function of calcite content

surface productivity, deep-water oxygen content, and rates of carbonate dissolution. So pelagic-hemipelagic limestone-marl alternations can generated under greenhouse conditions. Some of the above described factors can be detected by stable isotopic data.

Carbon isotope signal

The carbon isotopic composition of calcareous fossils reflects the composition of the dissolved carbon of the water from which the organisms precipitated their shells.

Several authors attempted to explain the excursions of $\delta^{13}C$ values:

Ocean surface water is enriched in ^{13}C relative to deep water (BERGER and VINCENT, 1986). THUNELL et al. (1991) proposed a model to explain the origin of Early Pliocene limestone-marl couplets in Calabria, Italy, which invokes ^{13}C enrichment of carbonate-secreting zooplankton as a result of an increase in surface waters phytoplankton productivity. This enrichment varies as a function of time, and this difference may become quite small in older sediments. Perhaps, this latter effect is due to diagenetic alteration, at least in part (KILLINGEY, 1983). Carbon isotope compositions of bulk rocks probably do not change a great deal during diagenesis because the volume of carbon within the pore-water reservoir is small and because the isotope fractionation between calcium carbonate and dissolved bicarbonate is small at low temperatures (EMRICH et al., 1970).

BARTOLINI et al. (1996) observed positive $\delta^{13}C$ anomalies in Early Bajocian and Callovian-Oxfordian carbonates in Central Italy that may record changes in global climate toward warmer, more humid periods characterised by increased nutrient mobilisation and increased carbon burial. High biosiliceous productivity and preservation appear to coincide with the observed anomalies, when the production of platform carbonates was subdued and ceased in many areas, with a drastic reduction of periplatform ooze inpot in many Tethyan basins. According to BARTOLINI et al. (1996) hydrothermal events related to rifting an/or

accelerated oceanic spreading may be the endogenic driving force that created a perturbation of the exogenic system.

BEAUCHAMP et al. (1987) proposed a basinal stagnation and thermohaline-stratification ocean model to explain the high $\delta^{13}C$ values for the evaporitic Permian Svedrup Basin, Canada. The model suggests that increased storage of ^{13}C-depleted sedimentary organic matter in the basin and high evaporation rates cause ^{13}C enrichment of the remaining seawater. MII et al. (1997), however, pointed out that the relatively constant $\delta^{18}O$ values across the $\delta^{13}C$ maximum suggest that the basinal-stagnation and thermohaline-stratification ocean model can not explain the $\delta^{13}C$ maximum in the isochron West Spitsbergen isotope stratigraphy. These globally recognized $\delta^{13}C$ shifts can be explained by changes in the size of the organic carbon reservoir (KUMP, 1991).

As pointed out above, diagenesis may alter the carbon isotopic composition. The light carbon isotope values, in particulary those of calcite in marlstones may be due to decomosition of organic matter.

According to a model proposed by RAISWELL (1987) during sulfate reduction, bacterial decomposition of organic matter leads to precipitation of ^{13}C-depleted calcite (up to -35 ‰). RICKEN and EDER (1991), however, emphasize that with increasing overburden, the sediments pass into the methane production zone, where isotopically light bicarbonate is removed by bacterial methane production. The remaining bicarbonate in the pore water and the precipitated câbonate therefore continuously shift from light $\delta^{13}C$ values to heavier ones.

In model proposed by JENKYNS and CLAYTON (1986), the lower $\delta^{13}C$ of the cement is compatible with the introduction of carbon dioxide derived from bacterial oxidation of organic matter. Low $\delta^{13}C$ values in carbonates associated with organic carbon contents support this model.

These observations are consistent with the isotopic record of Komló Formation inasmuch as the lowest $\delta^{13}C$ values are found in marlstones, which were probably deposited during periods of lower organic productivity.

Oxygen isotopic signal

The $\delta^{18}O$ value of bulk carbonates is determined by the $\delta^{18}O$ value of unaltered biogenic constituents (and primary micritic matrix) and the $\delta^{18}O$ value of the overgrowth cement on the biogenic particles. The latter will have a $\delta^{18}O$ value controlled by the temperature and the $\delta^{18}O$ value of the interstitial water. Thus, at greater burial depths, where cements are generally more abundant and most of the planktonic constituents have been dissolved, the $\delta^{18}O$ value of bulk samples reflects more closely the geologic conditions during burial diagenesis rather than the paleotemperature of the initial ocean water (COOK and EGBERT, 1983). MATTER (1974), MATTER et al. (1975), and SCHOLLE (1977) pointed out that as pelagic carbonates become more deeply buried, their porosity decreases and their $\delta^{18}O$ values become increasingly more negative. Highly negative oxygen isotope values are formed in carbonates deposited in fresh water environments or in carbonates precipitated at high temperatures. The oxygen isotopic composition of marls and limestone layers may be homogenized by diagenetic processes (EINSELE and RICKEN, 1991). During diagenesis, oxygen isotope ratios are likely to be far more readily altered than carbon isotope ratios. The ratio of oxygen in pore water to oxygen in the rock in initially extremely porous carbonate oozes is high, the inverse is true with respect to carbon. In addition, there is a large temperature fractionation of oxygen isotopes which can play an important role during burial diagenetic cementation. Such effects have been noted in many pelagic sequences (SCHOLLE, 1977).

Oxygen isotope ratios of pelagic carbonate-marl rhythms often show low values in the carbonate-poor intervals. Several well studied examples can be found in the literature of Cretaceous, and other ice-free periods of the Earth's history (WEISSERT et al., 1979, DE BOER, 1983, KAUFFMAN, 1988). Two mechanisms should be considered to explain the relatively low oxygen isotopic values of the carbonate in the marly beds:

1. Fluctuations of the surface water temperature: carbonates of carbonate-poor beds would have precipitated from relatively warm surface waters.

2. Fluctuations of the surface water salinity, proposed first by PRATT (1984); the salinity would have been relatively low at the times of formation of the marly semicouplets.

The first process, fluctuations of surface water temperature, fits in a model with variations of circulation and upwelling related to shifts of climatic zones due to orbital influences on the climate. Temperature differences of oceanic surface waters are not only due to changes in atmospheric temperature and insolation but also due to changes of ocean currents, upwelling, and the supply of waters derived from higher and hence cooler latitudes (DE BOER, 1991).

The second process, fluctuations of surface water salinity, was an more important process in the Jurassic ocean, while the reduced circulation of the ocean might well have led to a greater influence of evaporation/precipitation, resulting in a variable salinity (DE BOER, 1991). KAUFMANN (1988) suggested that to enhanced fresh water input could have caused slower circulation and lower organic production in the surface waters. BARRON (1986) stated that surface runoff patterns may have influenced stratification in marginal seas and could have had a global effect.

Processes such as dissolution and reprecipitation of carbonate are unlikely to cause the rhythmic layering in the sequence because of the stable oxygen isotope ratios of carbonate. These show lower values in the marly intervals whereas in the case of diagenetic redistribution of carbonate they would have shown lower values in the more cemented limestone beds. The difference between the oxygen isotope values measured in limestone and in marlstone in an given couplet are quite large. Diagenetic redistribution of carbonate would have lowered the mean $\delta^{18}O$ values of carbonate-rich beds and thus have diminished the original differences. The differences of the oxygen isotope values between the limestone and marl beds in a given couplet suggest a primary origin of the carbonate-marl rhythm. Thin section observations, however, show evidence of some dissolution and reprecipitation of calcite. Some foraminifera are partially dissolved, chambers of radiolarians and siliceous sponges are filled by calcite. Although burial diagenesis may have shifted the $\delta^{18}O$ signature of the limestones more negative values, I believe that the negative changes in amplitude of the $\delta^{18}O$ signal from bed to bed represent closely the original $\delta^{18}O$ pattern in the surface waters. According to ARTHUR and DEAN (1991), $\delta^{18}O$ values -2.5 to -3.5 ‰ are reasonable isotopic compositions for diagenetic unaltered marine carbonates formed in a warm, ice-free ocean. As show the Table 1. some marlstone samples have lower $\delta^{18}O$ values than -3.5 ‰. In case of these samples diagenetic modification of the original carbonate content can be not excluded. As stated above, the stable oxygen isotope values are higher in limestone beds than in over- and underlying marly intervals. This points to differences of temperature during time of formation, with the limestone beds having formed during periods of „cooler" and/or „more saline" surface waters (DE BOER, WONDERS, 1984).

TABLE I

Stable isotope ratios and calcite ontents of measured samples

sample	$\delta^{13}C$ (PDB) (‰)	$\delta^{18}O$ (PDB) (‰)	$\delta^{18}O$ (SMOW) (‰)	calcite (%)
PS 2-24 B	2.1	-2.0	28.9	100
PS 2-26 B	1.0	-5.1	25.7	25
PS 2-34 A	1.9	-2.6	28.3	75
PS 2-34 B	2.0	-2.2	28.6	88
PS 2-34 C	1.6	-4.4	26.3	52
PS 2-37 B	1.2	-4.6	26.1	40
PS 2-38 B	2.1	-2.1	28.8	92
PS 2-42 A	1.6	-3.0	27.8	76
PS 2-42 B	2.0	-2.3	28.6	92
PS 2-43 A	0.4	-4.6	26.2	24
PS 2-47	0.2	-4.6	26.1	23
PS 2-48 A	1.1	-4.8	26.0	18
KG-1 A	1.7	-1.4	29.4	82
KG-31	1.5	-0.5	30.4	90
KG-32	0.8	-2.5	28.3	30
KG-48	1.6	-0.5	30.4	91
KG-52	1.3	-2.3	28.5	63
KG-53	1.0	-4.1	26.7	31
KG-54 B	1.5	-1.5	29.4	65
KG-54 C	1.3	-3.2	27.7	64
KG-56	1.4	-3.1	27.7	60
KG-57	0.7	-4.3	26.5	29
KG-58 A	1.1	-3.1	27.8	60
KG-58 B	0.6	-2.8	28.0	48
KG-59	0.6	-2.7	28.1	40

TABLE 2

Compilation of stable isotopic data

Sample localities of isotopic data	$\delta^{13}C$‰ PDB values	$\delta^{18}O$‰ PDB values	References
DSDP Site 238, Indian Ocean, Upper Miocene	+2.0 – -0.5	no data	Foraminifera tests Vincent, Berger (1985)
DSDP Site 216, Indian Ocean, Miocene	+3.0 – -0.5	+3.5 – -0.5	Foraminifera tests Vincent, Berger (1985)
Moria, Apennines, Upper Albian	no data	-2.6 – -3.7	Pelagic limestones and marls de Boer (1991)
Hartland Shale Mbr., Rock Canyon, Colorado, Cenomanian	+1.0 – -1.0	-8.0 – -9.0	Brackish shales, marls Eicher and Diner (1989)
Bridge Creek Mbr., Pueblo, Colorado, Turonian	no data	-3.5 – -8.0	Pelagic limestone Eicher and Diner (1989)
Bridge Creek Mbr., Pueblo, Colorado, Turonian	no data	-6.0 – -8.0	Pelagic limestone Eicher and Diner (1989)
Niobrara Fm., Berthoud State #3 core, Denver Basin, Up. Cret.	+2.0 – +1.0	-6.0 – -10.0	Pelagic limestones, marls, black shales Arthur, Dean (1991)
DSDP Site 387, Atlantic Ocean, Neocomian	+1.5 – +1.0	-2.0 – -5.0	Pelagic limestones, marls, black shales Arthur, Dean (1991)
Breggia Gorge, Switzerland, Upper Liassic	+3.0 – +1.0	-1.0 – -3.0	Pelagic limestones, marls Jenkyns, Clayton (1986)
Val Cepelline, Lombardian Basin, Italy, Upper Liassic	+3.0 – +2.0	-1.0 – -3.0	Pelagic limestones, marls Jenkyns, Clayton (1986)
Valdorbia, Umbrian Basin, Italy, Toarcian	+5.0 – +2.0	0 – -2.5	Pelagic limestones, marls Jenkyns, Clayton (1986)
Bányahegy, Gerecse, Hungary, Upper Liassic	+4.0 – +2.0	-1.0 – -3.0	Pelagic limestones, marls Jenkyns, Clayton (1986)
Val Varea, Lombardian Basin, Italy, Upper Liassic	-1.0	-2.0	Mn-rich carbonates Jenkyns, Clayton (1986)
Trubi Marls, Calabria, Italy, Early Pliocene	-0.3 – -2.0	+0.2 – -1.0	Planctonic foraminifera tests in marls Thunell et al. (1991)
Trubi Marls, Calabria, Italy, Early Pliocene	0 – -1.5	+0.5 – -1.0	Planctonic foraminifera tests in limestones Thunell et al. (1991)

Depositional model

Summarizing the suggestions of data with a simple model can be explained the rhythmic layering of Komló Formation:

In the Jurassic ocean, thermohaline circulation was probably driven mostly by differences in salinity rather than by differences in temperature as it is the case in the modern ocean (BRASS et al., 1982). Bottom water was dense because it was more saline than the surface water, but it was less cold as in the present oceans. Dissolved bicarbonate in this bottom water was enriched in ^{18}O as a result of higher salinity. When vertical mixing in this ocean were slow, i. e. during deposition of marlstone beds of Komló Formation, the deeper, more saline water with its higher δ^{18}O value remained relatively separated from less saline surface water characterized by a lower δ^{18}O value. During periods of increased circulation, deeper waters mixed with surface water causing a rise in δ^{18}O of the upper waters. These upwelling bottom waters brought nutrients that fostered production of calcareous plankton in the photic zone. Deposition of the limestone beds probably occured during this period. The higher δ^{18}O values of limestones support this interpretation. Due to enhanced productivity more organic matter is deposited resulting in higher δ^{13}C values in limestones compared to marly intervals. The abundance of biogenic silica in the limestone beds is also consistent with this model of enhanced bioproductivity (HERBERT et al., 1986).

Under oxic conditions the organic matter is being oxidized in the water column, on the sea floor or in the sediments near the sediment-water interface, but in the limestones its original higher quantity can be traced by the δ^{13}C signal.

Proposing a dilution model to explain the marl-limestone rhythmicity, in which the „brackish" (or more diluted) surface seawater layer is inferred to have been in place during marlstone deposition, this δ^{18}O pattern would support it. The comparatively low δ^{18}O values in Komló Calcareous Marl marlstones, however, do not need to represent brackish water. Even the lowest of these values is heavier than the values from brackish Hartland Shale from the Western Interior Seaway, Cenomanian, USA (Table 2), and the Komló Formation contains a planktonic fauna indicating normal marine salinities (RESCH, pers. comm.).

Applying this simple model to the Komló Formation suggests that the rhythmic bedding is due to variations in bioproductivity (more calcareous plankton at the times of enhanced circulation, more upwelling, relatively „drier" and „cooler" periods resulting in limestone formation). This process could be superimposed variations in continental runoff (more terrigenous mud and silt carried from the continent at times of more humid and wet conditions resulting in marlstone deposition). The original isotope record was overprinted to some degree by diagenesis i. e., the isotope values of all marlstone and limestone beds may be shifted toward more negative values but burial diagenesis seems to have not been significant.

Of course, merely on the basis of stable isotopic data can not model any depositional system. The main problem is that precise dating of examined sequence have not been solved because of scarcity of index fossils. Thus, the sedimentation rate of the formation can not be stated. In order to establish a more accurate model, one has to examine tha clay mineral assemblage, the trace element record, the microfacies characteristic and the fossil assemblage.

CONCLUSIONS

The stable isotope values of limestone-marlstone couplets in the Komló Calcareous Marl Formation show consistently higher $\delta^{18}O$ and $\delta^{13}C$ values in limestones relative to marlstones. A significant correlation exists between the percentage of calcite and the isotopic composition. According to suggestions of data production and/or dilution cycle may be existed as depositional background of the origin of limestone-marlstone alternation. A depositional model is proposed for the rhythmic marl-limestone succession of the Komló Formation, whereby this bedding is the result of regular changes in ocean bioproduction and concomitant salinity changes. Postdepositonal alteration was minor and resulted in a general decrease in $\delta^{18}O$ and $\delta^{13}C$ values in both marls and limestones. In order to establish of the importance of the individual processes (whether the production or the dilution cycle was more important), additional sedimentological, geochemical and paleontological work is needed.

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