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STABLE ISOTOPES ($\delta^{18}O$, $\delta^{13}C$) OF MOLLUSK SHELLS IN ENVIRONMENTAL INTERPRETATIONS; AN EXAMPLE FROM SİNOP MIOCENE SUCCESSION (NORTHERN TURKEY)

Baki VAROL

ABSTRACT.-.Stable isotope values of mollusk shells representing centain levels in Sarmatian and Tshokrakian units <ithin Miocene succession in Sinop region have been respectively measured as δ^{13} C= between -0.32 and 0.78 ‰, and δ^{18} O = between -2.18 and -2.38 ‰, δ^{13} C= between-2.61 and 0.79 ‰, and ‰, δ^{18} O= between -6.46 and -0.10 ‰. When these values have been compared with values of mollusk fauna from recent Black Sea (δ^{13} C=between - 0.99 and 0.39 ‰, and δ^{18}O = between -2.32 and -0.41 ‰) and with values of surface waters of Black Sea (δ^{13} C = between 0 and 1 ‰, and δ^{18} O= -2.84 ‰ (in average)), it is shown from the stable isotope analysis point of view that Tshokrakion Sea resembles to recent Black Sea and the Sarmatian Sea displays a wide range from normal sea water to brackish waters.

INTRODUCTION

Application of stable isotope (δ^{18}O and δ^{13}C) analysis to shell-froming calcite and aragonite of organisms is a long-lasting practice in investigation of temperature and salinity changes in ancient seas (Epstein and Lowenstom, 1953). The main principle of this application is to have records of isotope values of the water in which organisms lived is reflected by the isotopic values within the shells. However, this is not a simple phenomenon and a series of complex events can take place. Therefore, following points must be taken in account in interpretation and usage of data obtained in these studies. The origin of calcite or aragonite, either inorganic or organic, constituting the mineralogical composition of a shell may differ in its isotope composition. For example, it has been recorded that aragonite inorganically crystallized at 25°C displays an increase of ‰ 0.6 in its isotope content with respect to inorganic calcite. However, stable isotope values in biogenic aragonite can have more positive or more negative values with respect to biogenic calcite (Arthur et al. 1983). Grosman and Ku (1986) similarly stated that rate of increase in δ^{18}O values of aragonite in foraminifera and mollusk shells with respect to calcite was not related to the temperature of the water. Moreover, these authors investigated that difference in &13C values of biogenic aragonite and dissolved inorganic carbon increased with decreasing temperature and recommended the application of this relationship in studying the paleo-temperatures of ancient oceans.

Isotope changes controlled by climatic and hydrological cycles are among the subjects that had been frequently studied. Isotopic changes in ocean water are mainly controlled by natural events such as glaciation, rain and flooding. For example, effects of Tertiary continental glaciation are reflected by important changes in δ^{18}O isotope values of deep marine sediments of the same time-span (Shackleton and Kennett, 1975). Periods of large floods enrich the waters in nutrients causing low salinity. Increasing organic production by this way causes the formation of sapropels as seen in Mediterranean basin (Calvert, 1983; Abrojen et al. 2002). Effects of this formation are recorded by enrichment of heavy isotopes (^{13}C) in surface waters and carbonates, although organic rich bottom sediments display enrichment in light isotopes (^{12}C). Within this process, it is observed that surface waters have lower $d^{18}O$ values but higher $d^{13}C$ values (Abrojen et al. 2002).

The escape of the light 16O isotope from the system causes enrichment of ^{18}O heavy isotope in global or seasonal warming periods, periods of increased evaporation and drought (Craig et al. 1963). Therefore evaporitic beach facies are characterized by sediments with higher $d^{18}O$ values with respect to temperate belts (Arthur et al. 1983).

Diagenesis is one of the most important factors effecting isotope values in the study of depositional environments by isotopes. Especially, in the isotope studies on shells of organisms, secondary calcite which can be formed depending on the chemistry of pore fluid in every stage of diagenesis, can effect the isotope character of the primary calcite (shell) and lead to misinterpretation of the results or mask the values obtained (Geary et al. 1989). Therefore, shells of planktonic organisms in deep marine sediments exhibiting properties of closed diagenetic environment have been frequently preferred due to balance between sediment and pore fluid (Resales et al. 2001). These authors emphasize to have a control on factors such as the disintegration of organic matters by bacterial effects or transformation of volcanic particles to clay minerals in the diagenetic environment that may result in unexpected low values of stable isotopes.

The subjects mentioned above are among the most important factors that affect the isotopic character of the carbonate association or of the shells which have gained their isotopic properties from their living environments. Moreover, many factors such as the vital effect and ontogenetic factor can also be affective on the isotopic character of the shells. For example, considareble increase in heavy isotope (^{13}C) values of shell structure in juvenile stage having high rate of growth and metabolic activations is frequently recorded (Romanek and Grossman, 1989). Similarly, metabolic carbonate which can enter to the shell structure can cause enrichment of shell with respect to light isotope (^{12}C) and deplete the shell with respect to heavy isotope (^{13}C). In this point of view, organisms having an effective respiratory system (athropods, mollusks, annelids) do not reflect the changes mentioned above as they do not including metabolic CO_2 in their skeletons (Weber, 1968).

In this study, isotope values obtained from shells representing the Sarmatian and Tshokrakian units have been correlated with those of the recent Black Sea fauna and with isotepe values of marine water in which they lived. So, by establishing the similarities or differences between recent Black Sea and the Sarmatian and Tshokrakian seas with respect to isotope values, approaches for depositional environments have been made. Application of these types of studies are very limited. Paleo-temperature records belonging to Romanian - Chauvda stages of Dardanelle Straight (Taner, 1996), isotopic characters of sediments with planktonic foraminifers of Campanian-Maastrichtian age around Hekimhan (Yıldız and Özdemir, 1999) are the studies published yet. That the Miocene succession of Sinop had been studied relatively well in regard to sedimentology and paleontology has facilitated this present study. The Miocene succession of Sinop region has quite different depositional environments within the scope of sedimentological and paleontological definitions. According to betterknown details of Eastern Tethyan chronostratigraphy the Sarmatian and Tszhokrakian sediments are found to be very suitable for the scope of this study. In the previous scientific reports, it is stated that most of the Tshokrakian fauna display very similar properties close to the salinity values of recent Black Sea (Özsayar, 1977). However, it has been recorded that Sarmatian was deposited in a depositional environment having a broad variety of salinity changes, ranging from hyper saline to brackish waters and include fauna associations reflecting this conditions (G6-rur et al. 2000; Varol et al. 2001).

MATERIAL AND METHOD

This study is carried out to test applicability of isotope values obtained from shells of organisms in interpretation of depositional environments. Number of samples for isotope analysis is compulsorily minimized due to financial restriction. Therefore, the more definite expression of the results obtained will be performed by another study including a larger number of samples in the furture. In sampling, great care was given to obtain samples from only a single group of organisms (Mollusks) in the levels which are rich in shells of sedimentologically well defined units. Whole shells that can be easily extracted from the base rock and cleaned were preferred. The extracted shells are washed with distilled water in an ultrasound tank to remove clays, oxidized matter, etc, and checked under the binocular microscope. The shells within the limestone in upper part of the Sarmatian succession are extracted only in fragments due to extensive diagenesis. Within these shells, samples which have no effects of diagenetic changes and are avoid of basement rock fragments were chosen. For comparison of the isotope values and referencing of the samples, recent samples from Black Sea fauna are obtained and especially certain different genus has been chosen within the beach sands from outer seaport region. All isotope analysis were carried out in the laboratories of the department of geochemistry in Tubingen University. SMOW and PDB standards were measured together with all samples and PDB is used for our anaysis to obtained more sensitive results for paleosalinity and paleotemperature interpretations.

SEDIMENTOLOGY

Sedimentology and paleogeography of Miocene succession of Sinop region is reported in detail by Görür et al (2000). From bottom to top, some local unconformities with low angle are observed in this succession represented by Paratethyan stages of Tarkhanian, Tshokrakian, Karaganian and Sarmatian. In this study, sedimentology of Tshokrakian and Sarmatian sediments in the studied region is described. Generalized geological map and stratigraphic section of Sinop Penninsula are illustrated in figure 1 and 2.

Tshokrakian: The rock units of this age are best observed around Kurtkuyusu in the west of Sinop Penninsula. The thickness of the unit ranges between 20-50 m. It is characterized by dark colored mud stone, rich in pyrite and including carbonatized plant fragments at the bottom, yellow colored weakly cemented massive sandstone with wave ripples and some large-scale cross-bedding in the middle and at the top (Fig. 3a). Properties of semi-closed depositional environment dominated by anoxic conditions rich in organic matters at the bottom turn in to high-energy environments with deposition of high amount of sand in middle-upper portions. Large -scale cross-bedden sandstones representing a large portion of the Tshokrakian succession display high energy sand accretion. Some preserved ripples within the succession reflect the same depositional development. The shell samples for isotope analysis are recovered from "Canbula sp.", which is widely found in thick sandstone levels forming an important portion of the Tushokrakian succession and from *"Acteocina* sp.", which is concentrated within silty-sandy mudstone forming intercalations in this unit.

Sarmatian: Thickness of this unit that outcrops in the seaward slopes and coastal margins around Kayıkbaşıburnu in Southwest of Sinop Peninsula ranges between 40-600 m. Moreover, this unit can be more than 1300 m thick in the reaion. It is divided into two formations, Kavıkbaşı Burnu and Yavkıl formations (Görür et al. 2000V The Sarmatian succession is Kavıkbaşı burnu and Yaykıl formations (Görür et al. 2000V The Sarmatian succession is described according to the facies types due to the purpose of this study and the sampling is described according to the facies types due to the purpose of this study and the sampling is realized within the same framework. The facies descriptions and sampling levels are described as followings.

Lensoidal Conglomerate: Conglomerates cropping out on the basement of Sarmatian occurrences in Sinop Penninsula are poorly sorted, clast-supported and block sized; Limestone pebbles and large pebbles derived from the basement mud stones are present in the constituent of the conglomerates. Internal structure of these conglomerates characterizes mass flow and lensoidal distribution of this conglomerate support basin magrin depositional system.

Shell bearing sandy limestone- This unit cropping out close to sea level in between Yaykıl and Kayıkbaşı headlands diplays planar and though cross-bedding and shell accretions (Fig. 3b). Shells are intensively cemented by carbonate and mixed embedded fragments or whole shells within the levels of sandy conglomerates and sandstones. Samples for isotope analysis are obtained from association characterized by Card/urn sp., and *Dreissena* sp.

This unit does not display long-distance lateral continuity and presents lensoidal out crops along the shore. In thes unit, low-angle unidirectional planar cross-beddings ana tnrough cross-oeddings support presence of wave case and nign-energy currents periodically activated in shore line belt respectively (Scholle and Spearing. 1988).

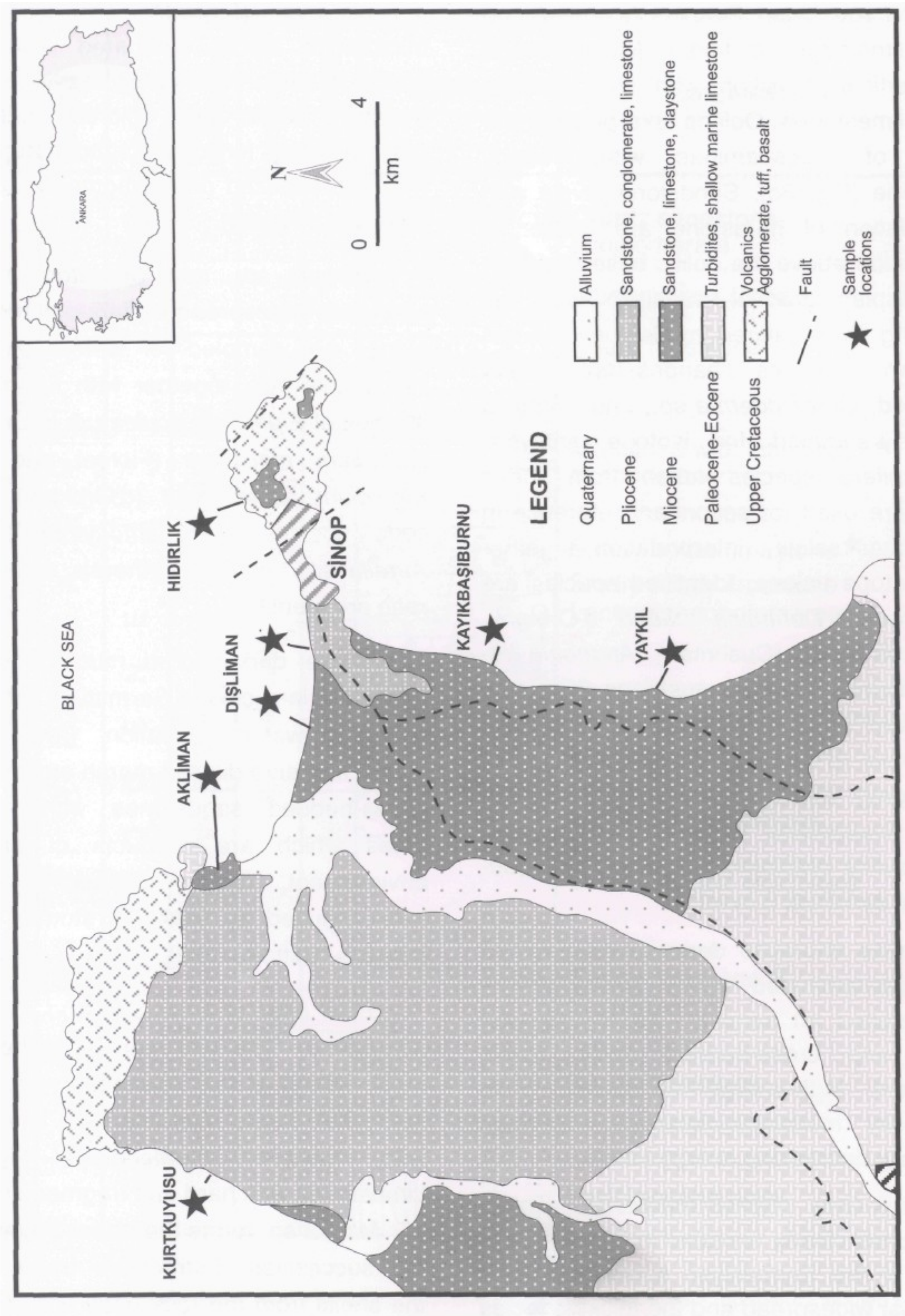

Fig. 1 - The generalized geological map of the study area and the sample locations

Sandy *oolitic limestone.-* This unit is a few meters thick and crops out in shore line between Kayıkbaşı Penninsula and Sinop with restricted distribution. It displays lateral and vertical transition with shell bearing sandy limestones. Oolites take place in the levels of cross-laminae within sandy limestone (Fig. 3c). Sandstone levels with intercalation of mudstone are thicker in succession above the oolitic base (> 15 m) and display gradual transition with the overlying dark colored mudstones. In this part where facies changes are clearly observed, *Cerestoderma* sp., and *Gibbula* sp., is sampled for isotope analysis. Foraminifera species taken from these levels are used for secondary reference in sea water salinity interpretation together with isotope values. Identified species are as follows: *Dentritina haueri* d'Orbigny, *Ammonia tepida* (Cushman), *Ammonia beccarii* (Linne), *Spirolina austriaca* d'Orbigny; *Elphidium reginum* (d'Orbigny), *Elphidium hauerinum* (d'Orbigny), *Elphidium rugosum* (d'Orbigny), *Elphidium macellum* (Fichtel et Moll), *Sinuloculina mayehana* (d'Orbigny), *Sinuloculina cyclostoma* (Reuss), *Ouinqueloculina seminula* (Linne).

These shoreline deposits represented by cross-laminated oolitic limestones is accumulated on sand shoals. Cross-laminated and cross-bedded oolitic levels represent high-energy conditions. Thin sandy-sity mudstone intervals represent gradually decreasing energy levels.

Dark-Gray colored mudstone- These mudstones clearly revealed in middle-upper levels of the Sarmasiyen succession form the most widespread and the thickest facies association. In the bedded portion between massive sections, many wave ripples, bioturbation structures, and score structures are observed (Fig. 3d). Syn-depositional microfaults within laminated levels are typical in this unit. In outer headland beach trenches, within dark colored mudstones cross-bedded lensoidal sandstone levels with carbonatized plant fragments are also observed (Fig. 3e).

Gibbula sp., and *Cerastoderma* sp., which are widespread in dark-colored mudstones are sampled for isotope analysis. Within this unit, together with foraminifera species of *Elphidium aculeatum* (d'Orbigny), *Elphidium macellum* (Fichtel and Moll), *Elphidium fichtelianum* (d'Orbigny), ostracoda species of *Cyamocytheridea* sp., *Auralia* sp. Leguminocythereis and *Cytheretta* are identified.

These dark-colored mudstones were deposited in a closed Sarmatian basin with restricted water circulation. Carbonatized plant fragments display marsh environment. Cross-bedded sandstones within mudstones which are products of stagnant environment, display fluctuating energy levels caused by seasonal storms or tide currents (Clifton, 1988).

Coal bearing limestone and mudstone alternation- This unit represents the upper most portion of the Sarmatian succession. Thin coal bands alternate with limestones with mudstone intercalations (Fig. 3f). Limestones with hard and fragmented shells of? *Mactra* sp. forms the upper boundary of the succession. Extractable fragments of the shells from the rock body are userd for isotope analysis.

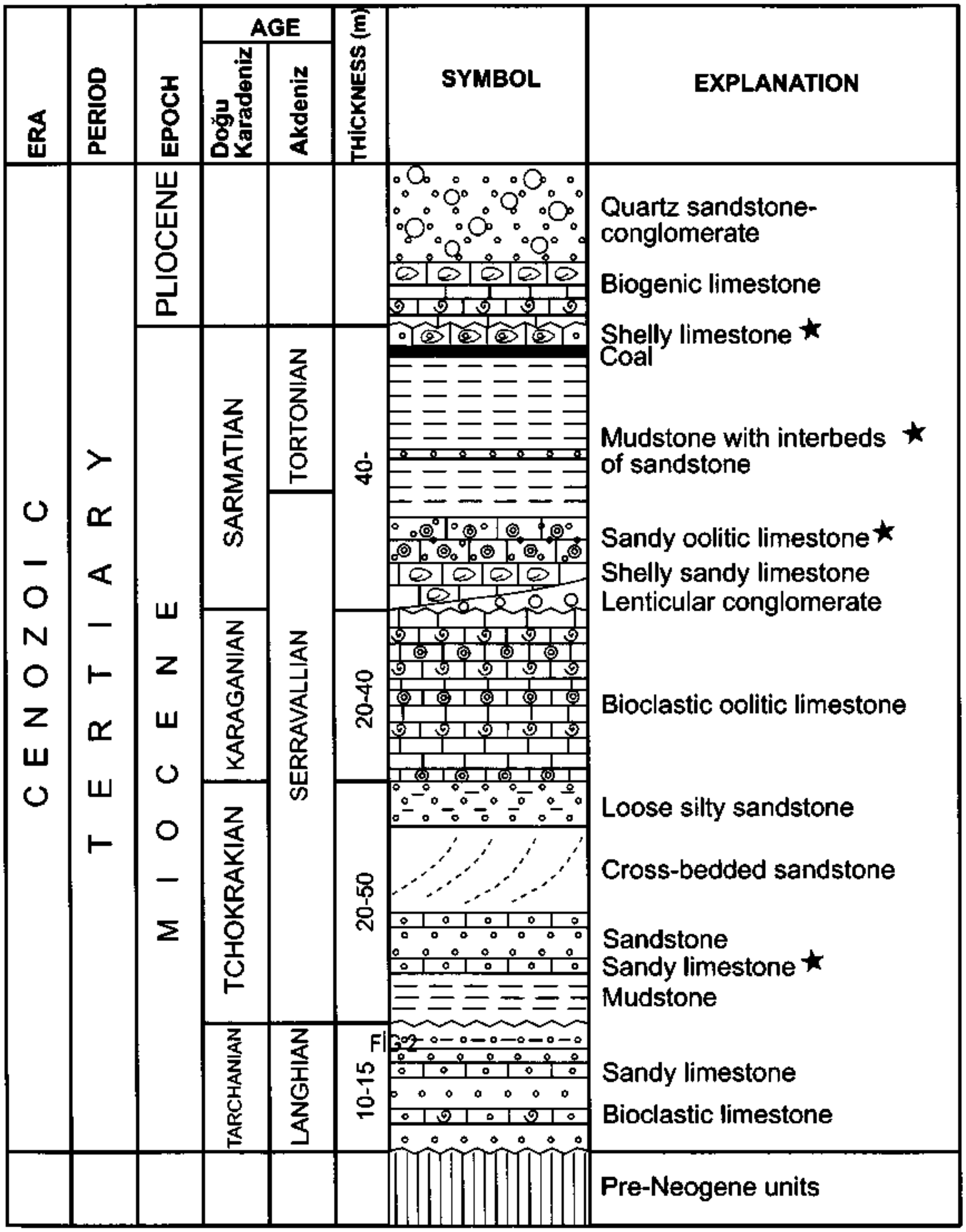

Fig. 2 - Generalized stratigraphic section of the Miocene succession; black stars represent sampled levels (modrified after Özsayar, 1977).

Except a few ostracoda, the limestone with mudstone intercalations forming the larger portion of the facies association does not include paleontological evidence. Coal bearing levels overlying these levels display coastal marsh development and increasing fresh water influences in Sarmatian marine environments.

STABLE ISOTOPES

The values belonging to shells are displayed in table 1. These values are combined in three groups as followings:

Recent Shells: $d^{13}C$= between - 0.99 and 0.48 ‰; $d^{18}O$= between -2.32 and -0.41

Sarmastian Shells: $d^{13}C$= between -2.61 and 0.79 ‰; $d^{18}O$= between -6.46 and -0.10‰;

Tshokrakian Shells: $d^{13}C$= between -0.32 and 0.84 %o; $d^{18}O$= between -2.18 and -2.38 ‰;

In addition to these, the values of recent Black Sea surfaces waters;$d^{13}C$= between 0 and 1 ‰; $d^{18}O$= -2.84 ‰ are obtained from Deuser (1972) and Rank et al. (1999).

When these isotope data are plotted in a diagram, the values for Sarmatian display distribution on a wider area with respect to values of reent Black Sea and values of Tshokrakian shells (Table 1; Fig. 4). However, the values of recent Black Sea and values of Tshokrakian shells display close aerial distribution to each other. The values representing Tshokrakian Sea are grouped in the same area and but the values representing Sarmatian Sea are spread in the diagram. Only one of the values of Sarmatian lies within the recent Black Sea zone, the others are distributed in very wide areas.

The isotope values of shells in the cross-bedded sandy-pebbly limestones with abundant shell accretions overlying the Sarmatian basal conglomerates ($d^{18}O$= between - 3.24 and -3.07 ‰) display considerable decrease in heavy oxygen isotope (^{18}O) with respect to the isotope values derived from oolitic limestones and mudstones overlying them. This kind of decrease in heavy oxygen isotope (^{18}O) in marine environments is due to mixing of fresh water or surface waters with marine water. Fresh waters enriched in light oxygen isotopes (^{16}O) causes dipletion of heavy oxygen isotope (^{18}O) in marine waters (Matyas et al. 1996). Being sedimentologically identified, gullied beach sediments associated with fresh water fluxes in the lower parts of the Sarmatian (Varol et al. 2001), are supporting evidences for isotopic variations in the shells of these units. The second succession representing the middle-upper part of the Sarmatian is composed of sandstone, oolitic limestone and mudstone and interpreted as deposited in shore and backshore environment (Görür et al. 2000; Varol et al. 2001). Heavy oxygen isotope (^{18}O) values of mollusk shells recovered from these levels display an increase with respect to previous levels. This indicates that fresh water influx to the system is stopped and probably that the marine conditions returned. Moreover, the isotope values concentrates around $d^{18}O$= between -0.1 and -0.47 ‰ includes heavier values than other species ($d^{18}O$= between -232 and -187) and the value measured in recent Black Sea waters (^{18}O= -2.84 ‰) except *Mytilus* sp. ($d^{18}O$= -0.41 ‰) living in recent

Table 1 - Table for the list of measured $d^{18}O$ and $d^{18}C$ isotope values of Tchokrakian and Sarmatian and recent Black Sea faunas.

Sample Number	Speices	$d^{13}C$	$d^{18}O$	Age
1	*Mytilus* sp.	-0,99	-0,41	Black Sea-Recent
2	*Cerastoderma* sp.	0,48	-1,87	Black Sea-Recent
3	*Dona* sp.	0,37	-2,26	Black Sea-Recent
4	*Chlayms* sp.	0,39	-2,32	Black Sea-Recent
5	*Mactra* sp.	-2,61	-6,46	Sarmatian
6	*Gibbula* sp.	-1,5	-0,16	Sarmatian
7	*Cerastoderma* sp.	-1,38	-0,2	Sarmatian
8	*Cerastoderma* sp.	-1,2	-0,1	Sarmatian
9	*Gibbula* sp.	-1,48	-0,16	Sarmatian
10	*Gibbula maeotica*	-1,78	-0,31	Sarmatian
11	*Cerastoderma* sp.	-1,69	-0,47	Sarmatian
12	*Dreissena* sp.	0,41	-3,07	Sarmatian
13	*Cardium* sp.	0,79	-3,24	Sarmatian
14	*Corbula* sp.	-0,78	-2,38	Tshokrakion
15	*Corbula* sp.	-0,78	-2,27	Tshokrakion
16	*Acteocina* sp.	-0,84	-2,26	Tshokrakion
17	*Acteocina* sp.	-0,32	-2,18	Tshokrakion

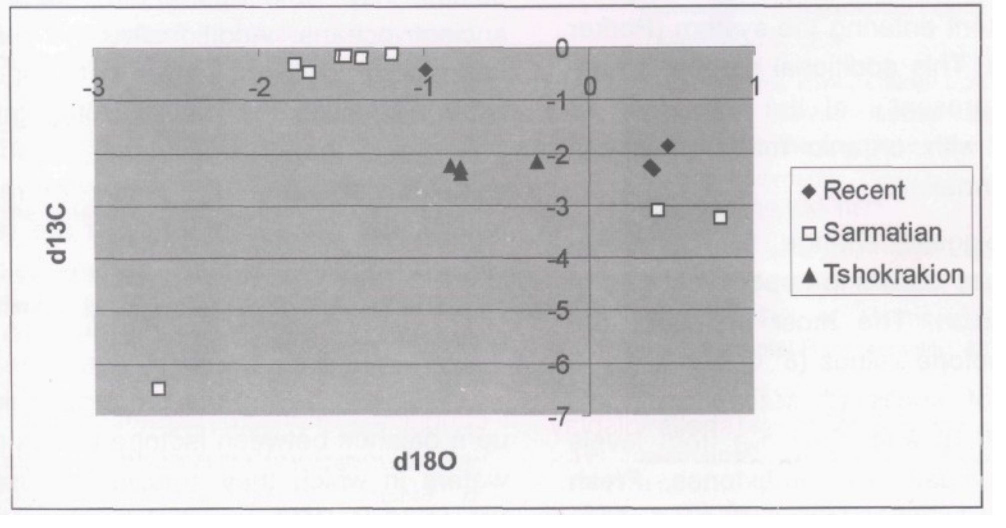

Fig. 3 - Correlation diagram for $d^{18}O$ and $d^{13}C$ isotope values between Tshokrakian and Sarmatian Tauna ang recent Black Sea surface waters

Black Sea. Additionally, *Dentritina* sp., and *Spirolina* -sp.., concentrated in sandy limestone intercalated with oolitic limestones which were sampled for isotope analysis are known as an association of organisms living in conditons, above the normal marine salinity (A. Poignant, 2000, personal communication). The relative enrichment of heavy oxygen isotope (^{18}O) in these levels in combination of paleontological and isotope data indicates that Sarmatian Sea is more saline and closer than recent Black Sea as indicated in sedimentological studies.

In addition to this, there are other ideas indicating that different organisms living in the same environments with habitats of $CaCO_3$ binding rate, different than marine waters, can cause differences in heavy oxygen isotope ^{18}O fractionation (Arthur, 1983). Depletion of heavy carbon isotopes ^{13}C and enrichment of ^{18}O in shells representing the lower-middle and partially upper portion of the Sarmatian succession can be interpreted as a increase in organic carbon content entering the system (Parker et al. 1972). This additional carbon is supported by presence of the thickening of mudstones with organic matter in these levels of Sarmatian.

The biggest change in Sarmatian isotope values occurs in upper most part of the succession. The most important decrease in isotope values ($\mathbf{d}^{18}O$ and $\mathbf{d}^{13}C$) of fragments of shells (? *Mactra* sp.), are recorded (-6.46 and -2.61‰) from levels with marls, coals and limestones. Fresh water with light isotopes derived from continental environments and transported to marine conditions decrease the concentration of heavy isotopes in marine environments (Arthur et al., 1983; Matyas et al. 1996). Enrichment in light isotope (^{12}C) in the analysed samples is the evidence of development of fresh water condition in the basin within time. Coal bands in these levels, which are indicators of marsh development, are sedimentological evidences supporting these environmental changes.

RESULTS

The isotope values of shells recovered from Tshokrakian and Sarmatian sediments of Miocene successions in Sinop region ($\mathbf{d}^{18}O$ and $\mathbf{d}^{18}C$) is in conformity with the paleogeographic and sedimentological data. When isotope values of shells from recent Black Sea and surface waters and shells from Tshokrakian and Sarmatian Sea are considered, Tshokrakian Sea displays similarities with the recent Black Sea, but the Sarmatian Sea data presents mixing of fresh water fluxes within time.

Stable isotope analysis of shells is a good application for the interpretation of paleosalinity and paleo-temperature of ancient oceans. Additionally, the number and type of the samples are very important factors affecting the results. Although it is considered that the restricted number of samples can be a risk factor for reliable results, the close conformity of the results with previously defined sedimentological properties of Tshokrakian and Sarmatian units decreases this risk factor.

Mollusks having the property of setting up a balance between isotope values of the waters in which they survive with isotope values of CaCO3 secreted for their shells, rae ideal organisms for this kind of studies. It must be considered that the usage of organisms with unknown isotopic fractionation of their shells in environments with indefinite sedimentological framework would be high risky for stable isotope analysis obtained from these organisms.

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PLATE

PLATE - 1

Fig. 1 - Yellow and thick bedded sandstone mudstone is a typical facies for the Tchokrakian succession (carta headland) in the Sinop region.

Fig. 2 - Gravelly and sandy limestone with shell accumulation is a characteric deposite for the basal part of the sarmatian succession (Kayıkbaşı heai dland).

Fig. 3 - Sandy oolitic beds within the sarmatian succession (Gelincik district, SW Sinop)

Fig. 4 - Black silty miudstone with wave ripples typify the middle and upper parts of the sarmatian succession (Yaykıl shore)

Fig. 5 - Cross - bedded sandstones is an indication of high energy deposition within the black mudstone unit with environmental quiescence (Disliman beach)

Fig. 6 - Yellow shelly sandstones and clayey limestones interbedded with coal-bearing layers indicate the upper most part of the sarmatian succession (Disliman beach)


Fig.1

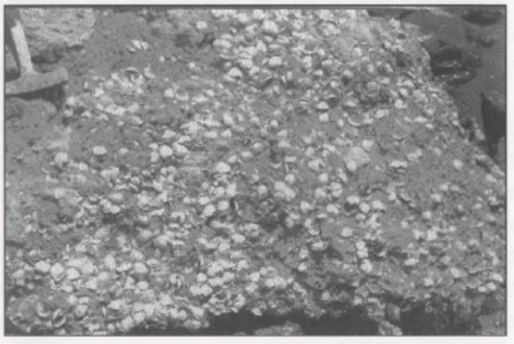
Fig.2


Fig.3

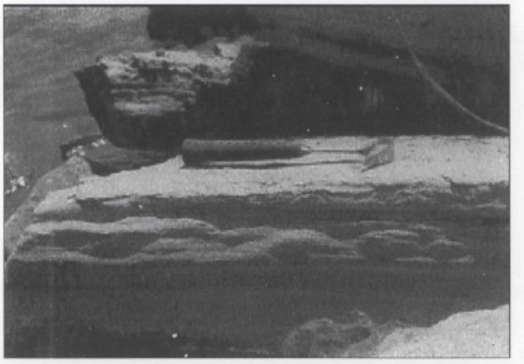
Fig.4

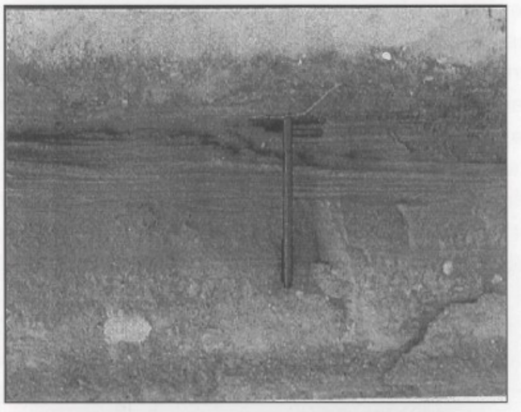
Fig.5

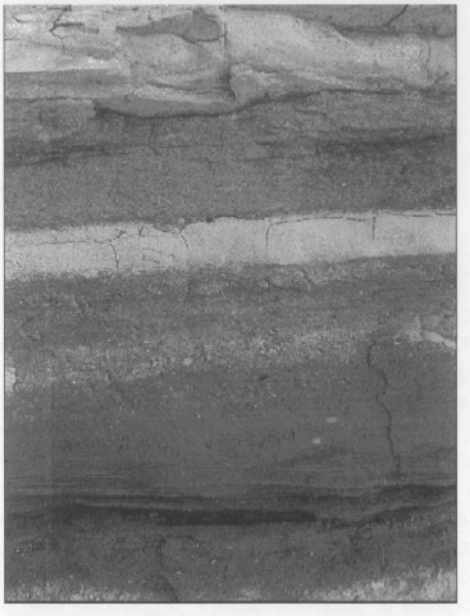
Fig.6