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Identifying Late Miocene episodes of connection and isolation in the Mediterranean–Paratethyan realm using Sr isotopes

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Abstract

After decades of research, the timing and nature of Late Miocene connections between the Mediterranean, Paratethys and the global ocean are still speculative. The hydrologic flux implications of exchange or isolation are central to all hypotheses for generating the major lithological changes that represent the Messinian Salinity Crisis. Moreover, differences in the hydrologic fluxes envisaged are the primary distinction between models. Despite this, these fluxes remain largely unconstrained. This paper describes the basis for using Sr isotope data innovatively combined with salinity data through hydrologic budget modelling to determine the timing and nature of Mediterranean hydrologic connectivity. We examine the hypotheses for three Late Miocene events to illustrate how this approach allows us to test implied hydrologic scenarios and exclude incompatible models. 1) Pre-evaporite restriction of the Mediterranean; 2) the initiation of salt precipitation; 3) connection between the Sea of Marmara and both Paratethys and the Mediterranean during the Messinian. This process suggests that the Atlantic–Mediterranean exchange was significantly reduced up to three million years before evaporite precipitation. It also indicates that end-member hypotheses for initiating salt precipitation in the Mediterranean (desiccation and connected basin models) are inconsistent with Sr isotope data. A contrasting model where evaporite formation was triggered by Atlantic transgression into a strongly evaporation-dominated Mediterranean is shown to be more compatible with available datasets. The application to Sea of Marmara samples indicates that salinity changes in the basin were not caused by changes to the amount of inflow from either Paratethys or the Mediterranean. Other possible as yet untested applications important for constraining different aspects of the Messinian Salinity Crisis are highlighted.

Keywords: Sr isotopes; Salinity; Oceanic-exchange; Fauna; Lithology; Messinian Salinity Crisis

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1. Introduction

Salinity fluctuations in marginal marine basins are dependent on changes to the system's hydrologic budget. When these are sufficiently large, such fluctuations cause a lithological or faunal response which maybe preserved in the geological record. Successions from the Mediterranean Messinian Salinity Crisis (MSC) for example, include faunal and/or lithological data which indicate periods of hypersalinity, brackish water and normal marine conditions. The foundations to all hypotheses explaining the transition from one salinity state to another are changes to the basin's hydrologic fluxes (Atlantic inflow, Mediterranean outflow,

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evaporation and precipitation) although these may not be explicitly described. However, until recently, it has been difficult to investigate the nature of past flux changes directly, so that rejection of hypotheses through rigorous testing has proved impossible.

Good illustrations of this problem are the hypotheses for triggering large-scale evaporite deposition in the Mediterranean (Benson and Rakic-El Bied, 1991). Most of the principle models, whether they envisage desiccation in a Mediterranean isolated from the global ocean (Hsü et al., 1972, 1973, 1977; Cita, 1982) or evaporite precipitation under conditions of continued Mediterranean–Atlantic connection (Fabricius and Hieke, 1977) require reduced Mediterranean–Atlantic exchange relative to the present day. However, the nature of that restriction is different for each model. Desiccation models for the onset of salt precipitation propose that the salinity crisis was triggered by sea level fall (drawdown) in the Mediterranean as a result of very strong restriction or termination of Atlantic–Mediterranean exchange (i.e. both inflow and outflow). By contrast, models which envisage continued connection with the Atlantic state that inflow of Atlantic water to the Mediterranean was maintained throughout the MSC and imply a change in the balance between Mediterranean water loss by evaporation and outflow. This is because achieving salt concentrations high enough to result in evaporite precipitation when inflow of ocean water is maintained requires a significant reduction in outflow and/or an increase in evaporation. When Mediterranean water loss is dominated by the evaporation flux, salt concentration in the remaining water rises. Discrimination between these two end-member models requires the contrasting hydrologic flux scenarios to be tested. Thirty years of unresolved debate testifies that this has yet to be satisfactorily achieved.

In the Mediterranean, direct study of past hydrologic flux changes is complicated further because of its mid-latitude setting (Fig. 1). Traditionally, geologists have used faunal assemblages and lithological information to identify when periods of restricted exchange with the global ocean and complete isolation occur. However, neither fauna nor lithology responds to restriction directly. Instead, they respond to a variety of environmental conditions such as salinity, nutrient supply, oxygen level and stratification, which are in part controlled by restriction. The other controls on these environmental conditions complicate the use of faunal and lithological change in the interpretation of restriction histories. For example, since salinity is a function of the evaporation flux as well as inflow and outflow (restriction), where evaporation exceeds precipitation, as it does in the mid-latitudes (Fig. 1), net evaporation rather than restriction is the dominant control on salinity. An increase in salinity deduced from changes in the faunal assemblage and/or lithological signal may therefore represent either changes to global ocean exchange or an increase in the evaporation flux (Flecker et al., 2002).

For mid-latitude basins then, the faunal and lithological record alone is an insufficient dataset for studying a basin's hydrologic budget and cannot be used to discriminate between different hypotheses for the causes of salinity change. An additional proxy that responds directly and exclusively to exchange is required. This work builds on the generic Sr–salinity modelling of marginal marine systems whose chemical and mathematical basis are described in Flecker et al. (2002). Using examples from different periods in the history of MSC, this paper describes how this approach can be used to interpret key periods of the Mediterranean's restriction history and clarify which models are

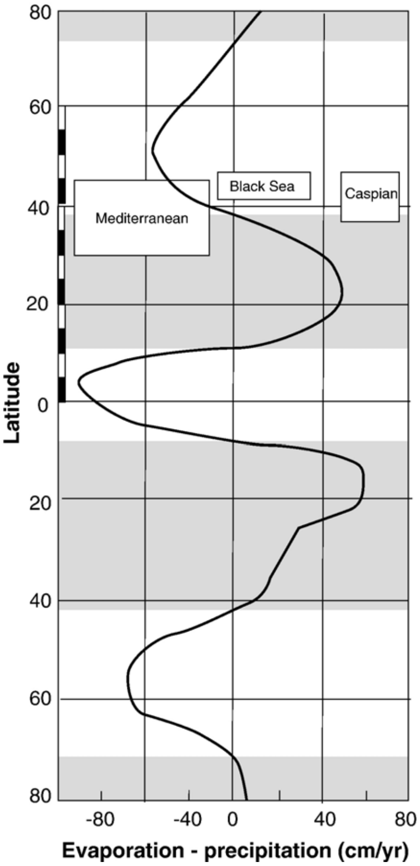

Fig. 1. Present day mean global evaporation minus precipitation showing the latitude of the Mediterranean, Black and Caspian seas (after Peixoto and Kettani, 1973).

least likely to be erroneous. It also demonstrates how subtle aspects of marginal marine connectivity can be investigated.

2. Methods

2.1. Sr isotope ratio of marginal marine water

The Sr isotope ratio of ocean water varies through time (Fig. 2). Because the residence time of Sr is significantly longer than the mixing time of the ocean, at any one time the ratio is the same wherever in the ocean you measure it.

The Sr isotope ratio of water in a semi-enclosed basin connected to the global ocean is controlled by simple mixing between ocean water and other Sr-bearing water sources feeding the basin e.g. river run-off and rain (Ingram and Sloan, 1992; Reinhardt et al., 1998). If available, hydrothermal fluids may also contribute to the Sr isotope signature of the marginal marine basin (Brass, 1976). However, the composition and magnitude of the hydrothermal flux values are difficult to constrain in the past (Teagle et al., 2003) and, for the purposes of this paper, are ignored.

Sr concentration differs from one water source to another. Ocean water has a Sr concentration ~ 40 times higher than average global river water (Palmer and Edmond, 1992). Nile and Rhone water have slightly higher Sr concentrations than the global average for rivers (Brass, 1976; Albarède and Michard, 1987), but even so the ocean water Sr isotope ratio dominates the Mediterranean water ratio. The degree of domination depends on the Sr concentration of river water and the isotopic contrast (i.e. $^{87}Sr/^{86}Sr$ difference) between oceanic and river $^{87}Sr/^{86}Sr$. Taking into account the analytical uncertainties on both individual Sr isotope measurements and the ocean water Sr isotope curve, the proportion of river water needs to exceed ~50% of the total inflow in order to generate a distinct non-oceanic Sr isotope signal in the Mediterranean (Flecker et al., 2002). Only when ocean water inflow drops below this threshold can the marginal marine basin $^{87}Sr/^{86}Sr$ evolve towards non-oceanic ratios. Progressive increase in the proportion of fresh water feeding the system results in a progressive increase in the deflection towards fresh water Sr isotope ratios. Where fresh water inflow has a strongly contrasting $^{87}Sr/^{86}Sr$, the resulting deflection will be greater than the precision of the measurement.

River water may have a strongly contrasting Sr isotope ratio as a result of dissolution of the rocks in the drainage basin (Palmer and Edmond, 1992). In most areas of the world, the continental crust is predominantly composed of old crystalline lithologies with high Rb/Sr that have evolved high $^{87}Sr/^{86}Sr$. The Sr isotope ratio of river water draining such crust is generally higher than that of ocean water at any time, largely because ocean water carries a component of low $^{87}Sr/^{86}Sr$ derived from hydrothermal alteration of basalts which were themselves derived recently from the mantle (Palmer and Edmond, 1992). The Mediterranean is unusual in being surrounded by atypical continental crust including large amounts of carbonates and mantle-derived volcanics. Different rivers flowing into the Mediterranean have different Sr isotope ratios (Fig. 2). Almost all of them have Sr isotope ratios lower than present-day sea water (Brass, 1976; Albarède and Michard, 1987; Palmer and Edmond, 1992). Some rivers such as the Nile have lower values than oceanic Sr isotopic records at any time in the Proterozoic (Fig. 2, McArthur et al., 2001) In contrast with most marginal marine systems, the mixing of substantial quantities of river water with oceanic inflow entering the Mediterranean, commonly results in a depression of the $^{87}Sr/^{86}Sr$ with respect to the coeval ocean water ratio.

2.2. Interpreting ancient Sr isotope ratios

Evaporite, some biogenic carbonate and phosphate mineralization incorporate the $^{87}Sr/^{86}Sr$ of the water in which they precipitated without fractionation. Unless post-secretion diagenesis has altered the primary Sr

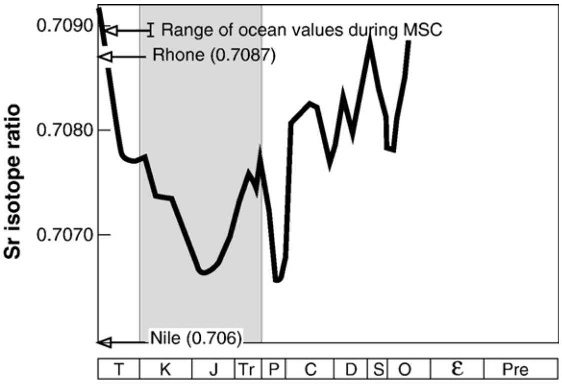

Fig. 2. Sr isotope record of ocean water through time (after Koepnick et al. 1988). Sr isotope ratios of Nile and Rhone river water today (Brass, 1976; Albarède and Michard, 1987) and the range of ratios for ocean water during the Messinian Salinity Crisis (MSC; McArthur et al., 2001) are indicated. The shaded area shows the range of Sr isotope ratios of Mesozoic carbonate from which much of the northern hinterland of the Mediterranean is made. River water draining areas dominated by such rocks might be expected to support a lower Sr isotope ratio than Late Miocene sea water.

isotope ratio, non-oceanic $^{87}Sr/^{86}Sr$ measured in fossil fauna that dwelt in a marginal marine setting therefore indicate that the proportion of ocean water entering the basin at the time had dropped below a level controlled by the Sr concentration of river water and isotopic contrast between oceanic and river water.

Early interpretations of Miocene Mediterranean Sr isotope records from foraminifera, were hampered by a partial understanding of the relationship between positive net evaporation and salinity in mid-latitude marginal marine settings. Typically these records comprise samples yielding both oceanic and non-oceanic Sr isotope ratios (Müller et al., 1990; Montanari et al., 1997). Despite the absence of visual evidence for preferential secondary alteration of foraminifera with non-oceanic $^{87}Sr/^{86}Sr$, these samples were interpreted as having a diagenetic origin, while samples with oceanic $^{87}Sr/^{86}Sr$ were assumed to be primary. This interpretation was based on inferring salinity from the Sr isotope ratios measured (Müller et al., 1990; Ingram and Sloan, 1992; Reinhardt et al., 1998). Non-oceanic Sr isotope ratios imply substantial dilution of the ocean water component by river water. If the evaporation flux is ignored, the calculated salinities are well below the salinity tolerance of the foraminifera analysed (Flecker et al., 2002). For example, some of the Tyrrhenian Sea (Fig. 3) foraminiferal data (Fig. 4B) have Sr isotope ratios 150 ppm lower than coeval ocean water (Müller et al., 1990; Müller and Mueller, 1991). This Sr isotope anomaly implies a salinity of around 10 g/l if the salinity of the water is assumed to be the product of mixing ocean water with fresh water without any evaporation effects (e.g. curve marked 0 on Fig. 5). This apparent inconsistency led Müller et al. (1990) to argue that all foraminiferal samples with Sr isotope ratios lower than coeval oceanic ratios were diagenetically altered.

However, it is only possible to deduce salinity levels from Sr isotope data where dilution of ocean water with fresh water is the sole control on salinity (Flecker et al., 2002). In mid-latitude semi-enclosed basins (Fig. 1), positive net evaporation can raise salinity to levels significantly higher than those resulting from simple mixing between ocean and fresh water (Flecker et al., 2002). The Mediterranean today is a good example. The largest water flux of the hydrologic budget is Atlantic inflow (Bryden et al., 1994). The evaporation flux is only ~12% of this, but because net evaporation slightly exceeds the net Mediterranean–Atlantic exchange (inflow–outflow), this is sufficient to increase Mediterranean salinity by 2 g/l above Atlantic inflow levels. Evidence of hypersaline conditions during the salinity crisis indicates that the evaporation flux was a more dominant component of the hydrologic budget in Messinian times than today. This means that dilution

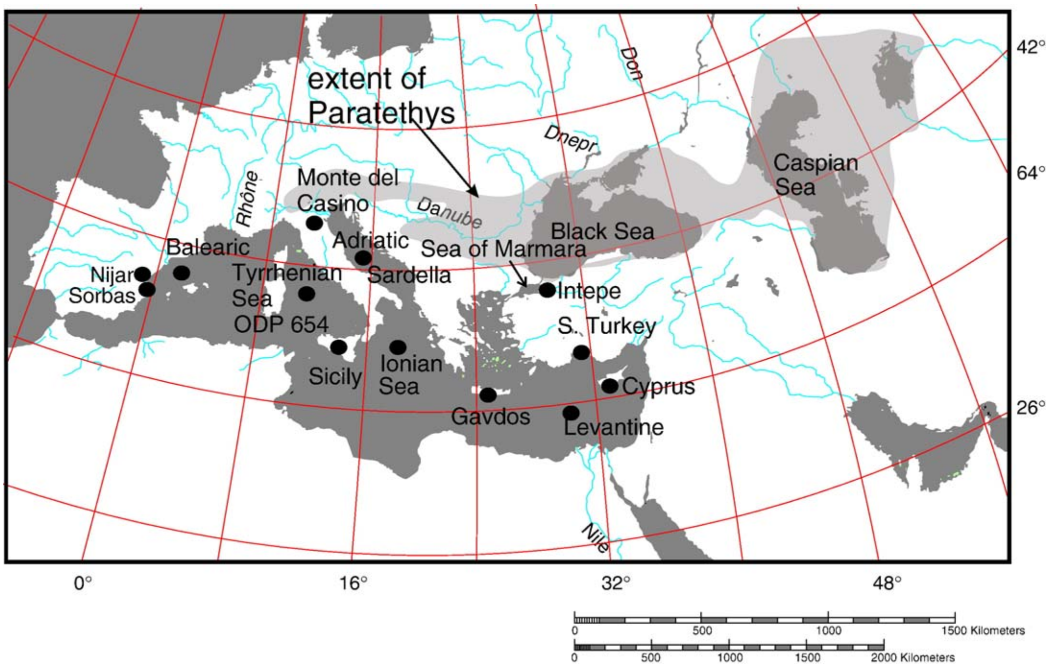

Fig. 3. Map of the Mediterranean and Paratethys showing the location of samples for which published Miocene Sr isotope data exist. Other sites mentioned in the text are also shown.

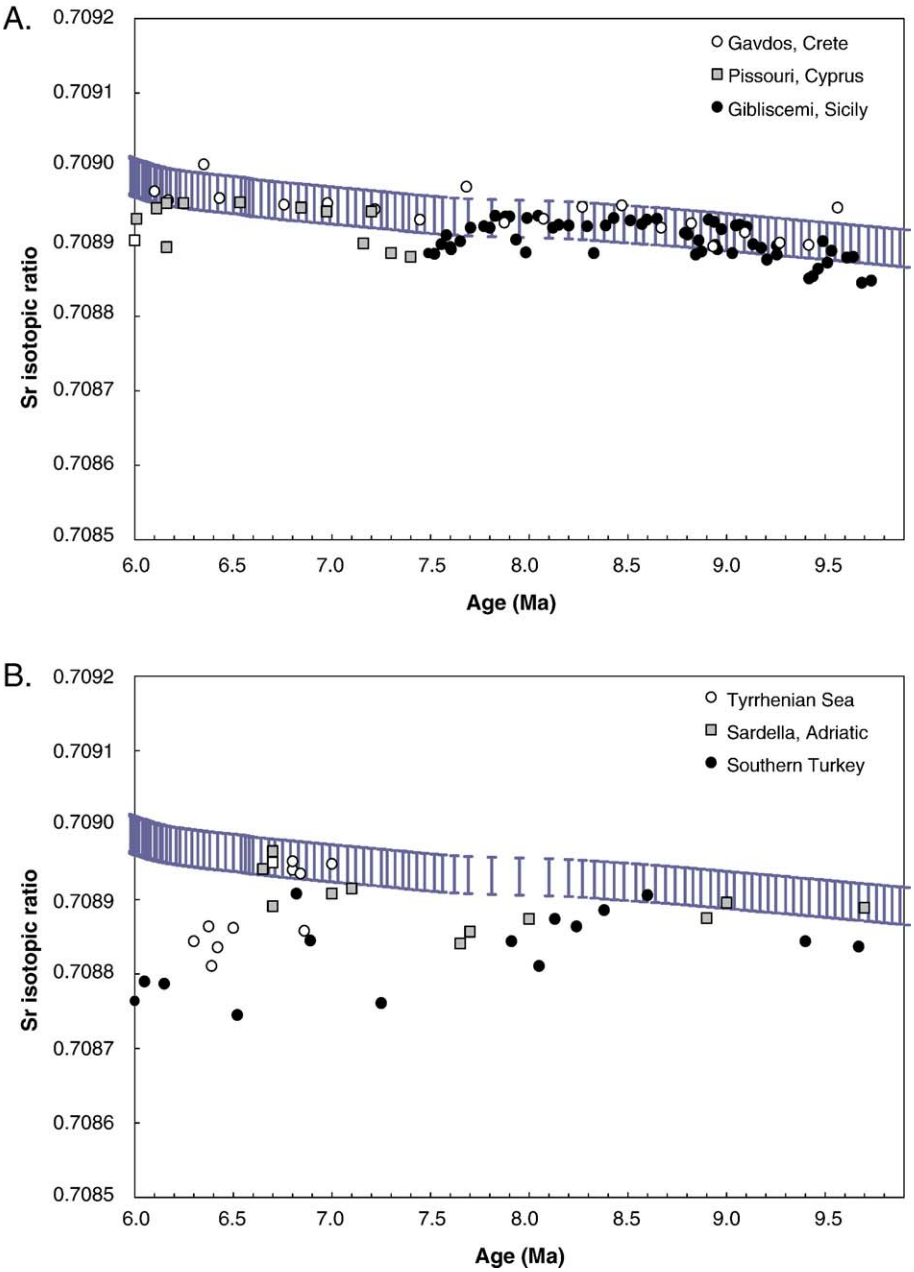

Fig. 4. Late Miocene pre-evaporite Sr isotope data from foraminifera plotted against the ocean Sr isotope record (McArthur et al., 2001). A) Geographically central Mediterranean sites. Pissouri data is listed in Table 1. Gavdos and Gibliscemi datasets can be found in Flecker et al. (2002) and Sprovieri et al. (2003) respectively; B) Northern sub-basins of the Mediterranean: Tyrrhenian Sea ODP654 (Müller et al., 1990; Müller and Mueller, 1991); Sardella (Montanari et al., 1997); Southern Turkey (Flecker and Ellam, 1999).

calculations based on non-oceanic Sr isotope ratios will significantly underestimate salinity if they ignore the evaporation flux (Flecker et al., 2002). There is therefore no inconsistency between low Sr isotope ratios and foraminiferal salinity tolerances.

In order to avoid confusion we use the term "oceanic" to describe water sourced from the Atlantic/global ocean with typical marine salinity and oceanic Sr isotope ratio. The term "sea-" or "marine-water" is not used to imply anything other than typical marine salinity (e.g. 35–38 g/l).

We acknowledge that it is difficult to demonstrate negligible diagenetic alteration (Hodell and Woodruff, 1994). However, where care has been taken to exclude adhering detrital (Flecker and Ellam, 1999) or biogenic

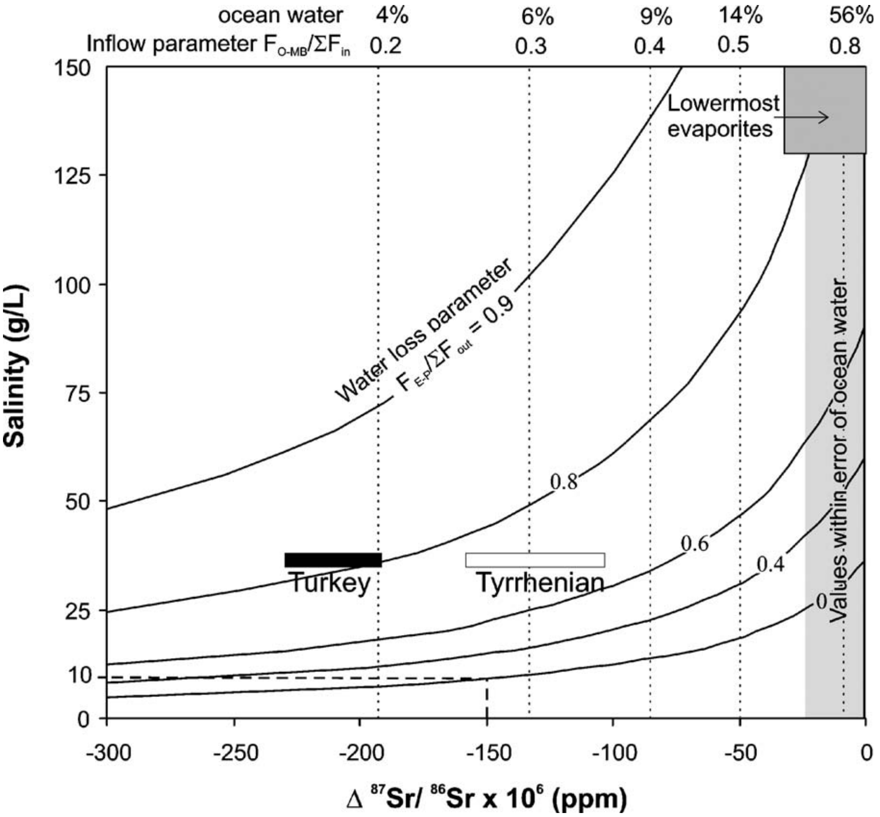

Fig. 5. Salinity derived from faunal tolerance and gypsum saturation constraints plotted against –Sr isotope data expressed as the difference between measured $^{87}Sr/^{86}Sr$ (see caption to Fig. 4) and coeval ocean water values (McArthur et al., 2001). The proportion of river and ocean water these Sr isotope ratios equate to, has been calculated assuming modern river water fluxes into the Mediterranean (Flecker et al., 2002). Contours of the water loss parameter (F_{E-P}:$\sum F_{out}$) map the relationship between river and ocean water input (the inflow parameter) and salinity (Flecker et al., 2002).

material (Reinhardt et al., 2000) and perform visual and chemical tests for alteration, a blanket rejection of non-oceanic Sr isotope ratios is simplistic and may exclude important information. Some papers listing Sr isotope ratios also tabulate other chemical indices such as stable isotope and Sr/Ca data measured on the same sample. Where these additional chemical data lie outside the normal range of values, it is possible to exclude Sr isotope results on the basis of chemical diagenetic alteration. Both Sr/Ca and stable isotope data exist for some Late Miocene Mediterranean datasets containing non-oceanic Sr isotope ratios (Table 1; Müller et al., 1990). These data indicate that there is no evidence for preferential alteration of samples with non-oceanic or oceanic Sr isotope ratios. This does not provide conclusive proof of the absence of non-visual chemical alteration, since the range of values for both stable isotope data and Sr/Ca ratios is large enough to encompass significant natural variability, particularly in marginal marine settings.

In theory these additional chemical data should provide some test of the balance between fresh and oceanic inflow calculated from the Sr isotope ratios. However, this too is problematic. Many of these chemical species do not respond simply to the fresh water flux, but to a combination of controls. $\delta^{18}O$ is a good example; its signature is controlled in the global oceans by ice-volume, temperature and salinity (evaporation). In marginal marine settings the controls over $\delta^{18}O$ are much less well constrained. Freshwater input may contribute to the signal, but under conditions of net evaporation it is likely to be evaporation, rather than the ocean–river water balance that dominates.

The combination of multiple controls and large ranges of natural variability means that stable isotope and elemental ratio data cannot be used to quantify past hydrological fluxes. In many circumstances these data are also unable to establish the primary or secondary nature of Sr isotope data. While excluding diagentically altered Sr isotope data remains a priority, in the absence of compelling visual or chemical evidence for diagenesis, we assume non-oceanic $^{87}Sr/^{86}Sr$ to represent the Sr isotope composition of marginal marine water at the time.

Table 1
Sr isotope data from the Pissouri Section, Cyprus

Sample	Stratigraphic height below base of gypsum (m)	Age (Ma)	Sr ratio	2σ	Sr/Ca	2σ	δ^{18}O	δ^{13}C
7005	−0.72	<5.96	0.708929	2.41E−05				
7005d	0.87	5.967	0.708929	1.99E−05	0.585	7.02E−05	1.298	0.379
7008	3.33	5.986	0.708902	2.41E−05	0.667	8.00E−06	1.016	0.476
7010	4.17	5.992	0.708931	1.56E−05			1.008	0.854
7014	11.09	6.083	0.708945	1.7E−05	0.7757	1.09E−05	0.790	0.501
7018	14.26	6.166	0.708952	2.41E−05	0.8303	4.98E−06		
			0.708893	0.000112				
7026	19.61	6.254	0.708952	1.99E−05	0.997	3.99E−05		
7038d	29.64	6.482	0.708953	1.7E−05	1.04	4.16E−05	−0.558	0.150
7046	34.6	6.768	0.708946	1.7E−05	1.0501	1.68E−05	0.838	0.375
7053	36.82	6.938	0.708941	1.7E−05	1.129	3.39E−04	1.155	1.467
7063	40.19	7.147	0.708898	2.41E−05	1.095	6.57E−05	0.344	1.856
7066	41.21	7.210	0.708941	3.4E−05	1.326	3.18E−04	0.135	1.237
7096	48.5	>7.532	0.708885	2.98E−05	1.2373	1.98E−05	0.245	1.136
7098	49	>7.532	0.70888	2.84E−05	1.201	4.80E−05	2.045	0.274
							2.026	0.216

The age of the samples was calculated using magnetostratigraphic and biostratigraphic data presented in Krijgsman et al. (2002). Planktic foraminifera were picked from washed and sieved whole rock samples and repeatedly ultrasonicated in 18.2 MΩ Milli-Q water to remove clay particles. Samples were leached with 1 N ammonium acetate (Gorokhov et al., 1995) to remove readily exchangeable Sr, and then dissolved in 2.5 M HCl (Flecker and Ellam, 1999). Sr was separated using conventional cation exchange procedures. Total procedure blanks were below 1 ng, which is negligible compared to the >1 μg Sr sample sizes used. Samples were loaded on Re filaments with a Ta activator similar to that described by Birck (1986), but prepared from high purity Ta metal. Sr isotope ratios were measured on a VG Sector 54–30 mass spectrometer in dynamic multi-collection mode. Instrumental mass fractionation was corrected using an exponential law and ^{86}Sr/^{88}Sr=0.1194. NIST SRM987 values over the period the measurements were carried out were: ^{87}Sr/^{86}Sr=0.710244 ± 17 (2 SD) n=31.

2.3. Modelling the hydrologic budget of marginal marine systems

Primary Sr isotope records derived from biogenic or authigenic mineralization monitor changes in the proportion of ocean and river water entering a marginal marine system (Ingram and Sloan, 1992; Reinhardt et al., 1998). By combining this information with paired faunal and/or lithological estimates of palaeosalinity, the proportion of water lost by evaporation and outflow can also be constrained (Flecker et al., 2002). This approach to hydrologic budget modelling provides a mechanism for evaluating changes in past hydrologic fluxes and for testing hypotheses which advocate particular palaeohydrologic flux scenarios. The model is described in detail in Flecker et al. (2002) and the Sr concentration and isotope ratio data used to calculate the palaeohydrologic fluxes for the Mediterranean are listed there in Table 1.

3. Discussion of results

Sr isotope, faunal and lithological records from three events during the Messinian Salinity Crisis are described below. These illustrate how changes to the hydrologic budget of marginal marine systems can be investigated. The examples enhance our understanding of the timing and nature of exchange between marginal and oceanic water bodies and enable us to assess the utility of different models.

3.1. Pre-evaporite successions in the Mediterranean

The pre-evaporite Late Miocene sequences in the Mediterranean have been extensively described. They typically comprise well-bedded alternations of marl and limestone with diatomites and sapropels incorporated cyclically in higher parts of the succession (Krijgsman et al., 1999a). These lithological characteristics are thought to be a response to orbitally-driven changes in insolation (Hilgen et al., 1995) under conditions of gradual progressive restriction of Atlantic–Mediterranean exchange (Krijgsman et al., 1999b; Krijgsman et al., 2000). Astronomical tuning of these successions using magnetostratigraphy and foraminiferal bioevents permits correlation on a bed-by-bed basis across the region (Krijgsman et al., 1999a) and from a central Mediterranean, relatively deep water position in Sicily to a marginal setting in the North Adriatic at Monte del Casino (Krijgsman et al., 1997).

Major faunal changes during most of this Late Tortonian to Lower Messinian period are restricted to changes in the deep water benthic foraminiferal species

(Kouwenhoven et al., 1999). Only the ~200 kyr period immediately preceding the onset of evaporite precipitation contains evidence of faunal variability (e.g. Blanc-Valleron et al., 2002). Planktic foraminiferal bioevents are therefore used for correlation throughout this 3 m.y. period because they are synchronous right across the Mediterranean and the assemblage appears largely unaffected by an impending salinity crisis (Krijgsman et al., 1999a).

This indicates that, despite the lithological evidence for increasing isolation during the 3 m.y. leading up to evaporite precipitation, surface water salinity remained at typical marine-Mediterranean levels (Bryden et al., 1994) until around 200 kyr before the first salt (Blanc-Valleron et al., 2002). The hypothesis to be tested therefore is that all areas of the Mediterranean were equally affected by a gradual, progressive reduction in Atlantic exchange, but that the connection was never severed. Any model for this restriction must explain why, despite increasing restriction, Mediterranean salinity did not increase.

The Sr isotope records of pre-evaporite Late Miocene foraminifera provide more detailed information about the changing hydrologic fluxes during this period. Successions can be divided into two geographical groups on the basis of their Sr isotope history (Fig. 4). Those from geographically central areas of the Mediterranean (Table 1, Flecker et al., 2002; Sprovieri et al., 2003) have Sr isotope ratios that plot within error of coeval ocean water values (Fig. 4A, McArthur et al., 2001). This suggests that more than half the water entering the Mediterranean was oceanic. Those located in northern sub-basinal areas (Müller et al., 1990; Montanari et al., 1997; Flecker and Ellam, 1999) have Sr isotope records that at times, diverge from coeval ocean water values (Fig. 4B).

Without visual or chemical evidence for diagenetic alteration, the successions containing anomalously low $^{87}Sr/^{86}Sr$ are assumed to record the Sr isotope evolution of the Mediterranean's marginal sub-basins. These records indicate that on two occasions prior to salt precipitation (e.g. 7–8 and 5.96–6.5 Ma; Fig. 4B), the proportion of ocean water entering these marginal areas dropped below ~50% and the threshold at which river water influence on the Sr isotope ratio can be detected was exceeded. Calculating the proportion of ocean and river water from these low Sr isotope data assuming present day river fluxes indicates that at their most restricted, these marginal sub-basins received <10% of their water from the Atlantic (Fig. 5). In order to maintain normal marine salinities from such a dilute solution, the proportion of water lost by evaporation must have been greater than net Atlantic–Mediterranean exchange. The balance of the two main hydrologic fluxes controlling water loss, outflow and evaporation must therefore have shifted such that either Mediterranean outflow was significantly reduced or the evaporation flux increased. Fig. 5 illustrates the proportion of water lost by evaporation calculated for the Turkish and Tyrrhenian sub-basins during the period immediately preceding the salinity crisis.

Mediterranean records containing non-oceanic $^{87}Sr/^{86}Sr$ corroborate interpretations of faunal and lithological data that suggest Atlantic exchange, though restricted, continued throughout the early Messinian and Late Tortonian. However, the hypothesis that all areas of the Mediterranean were equally affected by a gradual, progressive reduction in Atlantic exchange can be shown to be false. Contrasting $^{87}Sr/^{86}Sr$ histories from different areas of the Mediterranean indicate that, despite a similar lithological and faunal response (Fig. 6, Hilgen and Krijgsman, 1999; Sierro et al., 2001), the northern sub-basins were more restricted than central areas. The Sr isotope records from the northern sub-basins also demonstrate that the Mediterranean experienced two discrete restriction events, punctuated by an increase in oceanic exchange between 7 and 6.5 Ma. The older of these may correspond to the 7.2 Ma change in deep water faunal assemblages identified by Kouwenhoven et al. (1999). The younger event has not been previously identified in Mediterranean faunal or lithological records.

This example of the pre-evaporite marginal Mediterranean successions suggests that when net-evaporation is positive Sr isotope records can pick-up changes in ocean exchange that do not affect fauna and lithology. Combining these datasets in a hydrologic budget model constrains the degree of restriction suffered by different areas and indicates that, even prior to salt precipitation, marginal sub-basins received <10% ocean water. Despite this, there is no indication of a salinity change in any area of the Mediterranean. The reasons for this are closely linked to the trigger for salt precipitation and are therefore explored in the next section.

3.2. The onset of evaporite precipitation in the Mediterranean

As well as providing temporal and spatial control on the oceanic/riverine composition of marginal basin water when salinity remains static, Sr isotope data can also be used to discriminate palaeohydrologic flux changes that occurred during salinity transitions. The onset of evaporite precipitation in the Mediterranean is a good example. Faunal, lithological and isotopic data suggest a rapid change in environment during the last few

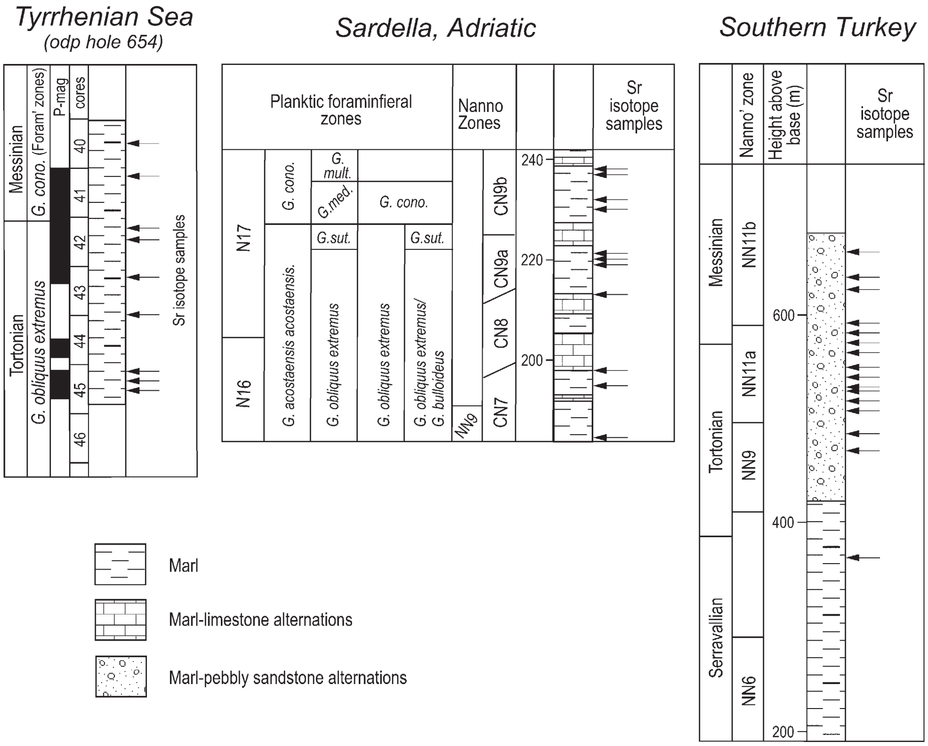

Fig. 6. Simplified logs of the three northern sub-basins sections that contain non-oceanic Sr isotope data (Tyrrhenian Sea ODP 654, Glaçon et al., 1990; Sardella, Montanari et al., 1997; Southern Turkey, Flecker and Ellam, 1999). All three of the age models for these sections currently rely on biostratigraphic data (Glaçon et al., 1990; Montanari et al., 1997; Flecker and Ellam, 1999 and references therein).

precessional cycles (e.g. Bellanca et al., 2001) followed by an abrupt, basin-wide switch to hypersaline conditions at 5.96 Ma (Krijgsman et al., 1999a). The main models advocate a major reduction in Atlantic–Mediterranean exchange over and above that sustained prior to salt precipitation. Desiccation hypotheses (Hsü et al., 1972, 1973, 1977; Cita, 1982) argue that both inflow and outflow are limited or terminated. Connected basin hypotheses (Fabricius and Hieke, 1977) argue for continuing inflow and imply reduced outflow. At root however, the faunal and lithological response is to a change in salinity which is not exclusively controlled by any aspect of Atlantic exchange. If it were possible to generate a basin-wide increase in salinity by some process other than reduced Atlantic exchange, neither faunal nor lithological data would be able to discriminate the mechanism. Since ^{87}Sr/^{86}Sr is unaffected by salinity change, but directly controlled by simple mixing of ocean and river water (e.g. Ingram and Sloan, 1992; Flecker et al., 2002), Sr isotope records can help test whether the salinity change resulted from changes in inflow, outflow, or both.

At present no single succession provides a complete Sr isotope record before, during and after the MSC although exposed and drill-holes on Sicily (Fig. 3) get close. Partly this is the result of the sedimentation–erosion dynamics of the Mediterranean margins and the accessibility of deep basin successions; the problem could be addressed with IODP drilling. However, for the purposes of examining the onset of evaporite precipitation, the Sr isotope dataset for the Mediterranean is temporally well-constrained and spatially sufficiently extensive to monitor the behaviour of the Mediterranean as a whole.

Typically, the earliest Lower Evaporites have oceanic ^{87}Sr/^{86}Sr (Fig. 7). Divergence from the oceanic Sr isotope curve occurs during the Lower Evaporite succession reaching a maximum in the Upper Evaporites and Lago Mare sediments before returning to the curve in the Lower Pliocene (Fig. 7). This implies that the earliest evaporites precipitated from water dominated by an oceanic source. These oceanic Sr isotope ratios therefore preclude the possibility that severe reduction or termination of Atlantic inflow triggered evaporite precipitation as proposed by the desiccation hypothesis. On the contrary, analysis of Sr isotope records suggests that the Mediterranean received increased Atlantic inflow coincident with the onset of evaporite precipitation (Flecker et al., 2002). The pre-evaporite northern

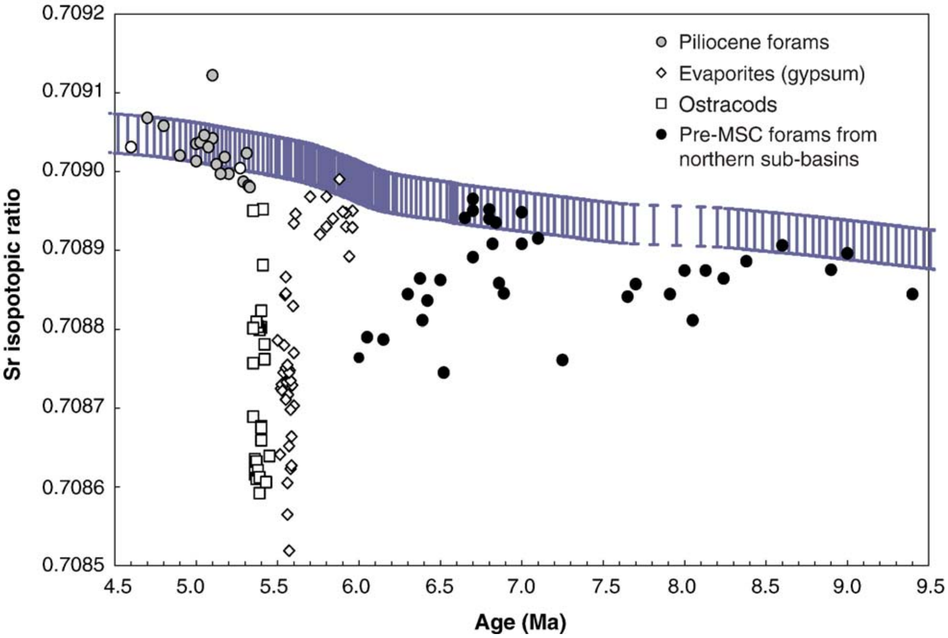

Fig. 7. Sr isotope records from pre-evaporite marginal sub-basins, Messinian Salinity Crisis evaporites and brackish-water deposits and Pliocene foraminifera are plotted against the ocean Sr isotope record (McArthur et al., 2001). Data derived from Table 1 and the following published sources (McKenzie et al., 1988; McCulloch and De Deckker, 1989; Müller et al., 1990; Müller and Mueller, 1991; Müller, 1993; Fortuin et al., 1995; Flecker and Ellam, 1999; Flecker et al., 2002; Sprovieri et al., 2003). Error bars for the data are as shown for the ocean water curve.

sub-basins which received a sufficiently low proportion of ocean water to show fluctuations in the inflow fluxes, track an abrupt increase in $^{87}Sr/^{86}Sr$, returning from anomalously low Sr isotope ratios to ratios within error of the oceanic Sr isotope curve at the transition from marine to evaporitic sediments (Figs. 5 and 7). Calculation of the proportions of river and ocean water demanded by this Sr isotope evolution indicates that, in the northern sub-basins at least, initial evaporite precipitation was accompanied by a sudden and dramatic increase in the proportion of ocean water (Fig. 5). On Sicily the possibility that this was induced by a reduction in river inflow can be excluded because the pollen spectra indicate aridity before, during and after the Messinian Salinity Crisis (Suc and Bessais, 1990). Further north, pollen records indicate cyclic changes in climate reflecting fluctuating humidity (e.g. Northern Apennines, Bertini, 1994), but there is no evidence of a sudden reduction in humidity coincident with the onset of evaporite precipitation that could account for the change in the ocean–river water balance.

The desiccation hypothesis for triggering evaporite precipitation can therefore be disregarded on the basis of Sr isotope information. The alternative connected basin hypothesis which explains rising salinity through increased evaporative loss and reduced outflow can also be tested.

The salinity and Sr isotope anomaly graph (Fig. 5) divides the hydrologic budget into its two components:

- the inflow parameter which monitors the proportion of total water entering the Mediterranean that derives from the ocean or rivers. This is derived directly from the Sr isotope ratios and plots along the *x*-axis.
- the water loss parameter which monitors the proportion of total water loss to the Mediterranean that is lost by evaporation or outflow to the Atlantic. This is calculated from the ocean/fresh water composition of the water and the evaporative water loss required by the salinity of the sample. The water loss parameter is shown as contours across the Sr isotope–salinity plot (Fig. 5).

Sr isotope data from Tyrrhenian Sea and Turkish foraminifera that immediately predate evaporite precipitation are plotted assuming typical foraminiferal salinity tolerance range (Stanley, 1972). The ocean-like Sr isotope ratios of the earliest evaporites are also plotted assuming typical salt saturations required for gypsum precipitation (130 – > 150 g/l). The simplest pathway between the two situations parallels the 0.8 water loss parameter contour (Fig. 5). This means that while the transition to evaporite precipitation required an increase in the proportion of ocean water, the outflow conditions may have been unchanged with evaporation strongly dominant over outflow (Fig. 5). As a result the connected basin hypothesis which advocates reduced outflow and increased evaporative loss as the trigger for salt precipitation must also be discarded.

This example examining the onset of evaporite precipitation in the Mediterranean indicates that Sr isotopes combined with faunal and lithological salinity constraints are a powerful tool for investigating both inflow and outflow components of palaeohydrologic budgets. Testing the two main hypotheses for triggering evaporite precipitation using this approach demonstrates that neither provides a description of the hydrologic flux changes compatible with the data. Instead, the model output data permits a new hypothesis to be constructed. Flecker et al. (2002) argue that the strongly evaporation-dominated conditions required in the northern sub-basins to sustain marine salinities in water heavily dominated by freshwater, were unable to generate hypersaline conditions because there was not enough salt in the system. The sudden influx of oceanic water at 5.96 Ma (Krijgsman et al., 1999a) demonstrated by the Sr isotope record (Figs. 5 and 7) increased the mass of the salt in the basin. The same strongly evaporative conditions (water loss parameter 0.7–0.85; Fig. 5) were then able to raise salinity sufficiently to produce basin-wide precipitation of evaporites. This new hypothesis therefore proposes that the trigger for salt precipitation was an increase in the proportion of ocean water entering the Mediterranean, probably in the form of a marine transgression rather than the regression advocated by earlier hypotheses.

In addition to the different hydrologic flux scenarios, one of the key distinctions between the two end-member models tested here is the associated sea-level change. Desiccation hypotheses clearly require periods of sea level fall, while the connected basin hypotheses may imply a more constant water level. The new model proposed here also has implications for vertical fluctuations in Mediterranean water level. This model requires an influx of Atlantic water; we envisage this in terms of a transgression. This could be the result of global sea level rise, or a lowering of sill levels controlling Atlantic–Mediterranean exchange. The inflow of ocean water is assumed to have had an immediate impact on Mediterranean water levels although it is less clear how long this raised level was maintained. Subsequently, the Sr isotope data indicate that inflow from the global oceans was reduced implying lower water levels during the MSC. Even when the Sr isotope anomaly is at its largest (e.g. during deposition of the Upper Evaporites and Lago Mare sediments, Fig. 7) the data indicate neither complete desiccation nor complete isolation (Flecker et al., in prep).

One of the advantages of this model is that it helps to explain why evaporite precipitation was both widespread and synchronous across both east and west Mediterranean basins and in both basinal and marginal settings (Krijgsman et al., 1999a, 2002). The desiccation models with their associated draw-down should result in a marginal to basinal evaporite chronology. Connected basin models imply the reverse i.e. that basinal evaporites should predate those precipitated in marginal settings. By contrast, this new model accounts for the observed synchronous precipitation in both basinal and marginal settings.

3.3. Mediterranean–Marmara connectivity (ostracod and Sr isotope data)

The previous examples investigate Atlantic–Mediterranean connectivity. It is also possible to use Sr isotopes to examine connectivity between water bodies where neither supports an oceanic Sr isotope record, in this case Paratethys and the Mediterranean during the MSC.

The timing and nature of connection between the Mediterranean and Paratethys is an important, poorly understood and controversial component of the MSC. For example, some authors invoke an inundation of low salinity Paratethyan water to dilute the hypersaline environment and generate brackish-water Lago Mare conditions in the Mediterranean (Hsü et al., 1973; Archambault-Guezou, 1976). The inflow of Mediterranean water to Paratethys during the MSC has also been inferred principally from horizons containing marine nannofossils (Snel et al., 2001). Romanian examples of these nannofossil layers appear to coincide with the onset of evaporite precipitation in the Mediterranean. The nature and timing of Mediterranean–Paratethys exchange described in both these hypotheses could be elucidated with Sr isotope data which are currently lacking. However, a study was undertaken to evaluate the connectivity history of the Sea of Marmara (Fig. 3) with the Mediterranean and Paratethys during the MSC. This is described in full by Çaðatay et al. (this volume). Only the Sr isotope results from Intepe, adjacent to the Sea of Marmara are discussed here.

Late Miocene Paratethyan sections typically contain ostracod fauna which are interpreted as indicating brackish water salinities (Fig. 1, Hsü et al., 1977; Cita et al., 1978). Episodic changes to the salinity are reflected in increases in different species with fresh or marine salinity tolerance (Çagatay et al., this volume). The occurrence of marine species in Paratethys and adjacent areas has been attributed to the inflow of Mediterranean waters (Sengör et al., 1985; Görür et al., 1997; Snel et al., 2001). The Sea of Marmara which today links the Mediterranean and Black Sea may also have been a gateway joining the Mediterranean and Paratethys during the Late Miocene (Çagatay et al., this volume). Testing for the influence of oceanic, Mediterranean or Paratethyan waters on Late Miocene Marmara sediments is one way to establish whether such connectivity existed.

Well preserved ostracods from the Intepe succession east of the Sea of Marmara are age equivalent with the MSC (Çagatay et al., this volume). Brackish ostracods dominate the lower part of the section (Fig. 8). Higher up the section, species tolerant of marine salinities occur and dominate some samples (Fig. 8). The Sr isotope analysis performed on ostracod valves are also plotted on Fig. 8. All the ^{87}Sr/^{86}Sr are lower than contemporaneous ocean water and there is a trend towards lower values with stratigraphic height (Fig. 8).

Fossil assemblages which contain species tolerant of different salinity conditions are presumed to be reworked to some extent. In this instance, where fresh, brackish and marine species are found in the same horizon, either the fresh and limited brackish water species were transported down depositional slope into a higher salinity environment, or the marine species were reworked from earlier, deposits. Assuming the simplest interpretation, that the marine species are in situ, this suggests that while salinities fluctuated, there was a general increase in salinity with time at Intepe.

We can consider the impact of inflow from each of the water bodies on this change in salinity and Sr isotope ratio separately:

- If the increase in salinity were caused by the influx of oceanic salty water, the Sr isotopes would respond by migrating back towards ocean values. The opposite occurs (Fig. 8).
- If the increase in salinity were caused by influx from a hypersaline Mediterranean in the middle of the salinity crisis, we would expect the Sr isotopes to move towards values reflecting the Sr isotope ratio of the evaporites (e.g. ranging from oceanic to values approaching 0.70865; Figs. 7 and 8). In fact, the three Sr isotope analyses from the highest part of the section have Sr isotope ratios much lower than this lowest recorded ratio from the MSC.
- The impact of inflow from Paratethys is more difficult to ascertain since there is little evidence of its Sr isotope ratio in the Late Miocene. However, assuming that it was dominated by inflow from the Danube, as the Black Sea is today (Shimkus and Trimonis, 1974), the Sr isotope ratio of Paratethyan water is likely to have reflected the Danube ratio which is close to ocean water values (Palmer and Edmond, 1992). As a result an influx of brackish water from Paratethys is unlikely to account for either the rise in salinity or the lowering of Sr isotope ratios seen at Intepe.

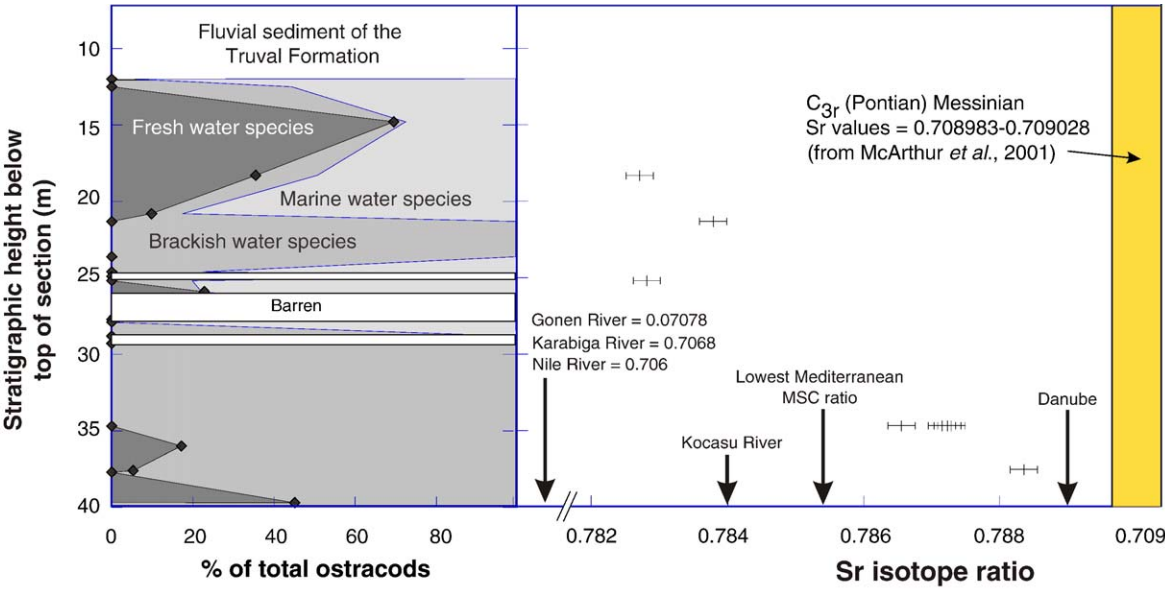

Fig. 8. Log of the Intepe Section with the salinity implications of the ostracods sampled there. Sr isotope values measured on ostracod valves are also plotted along with Late Miocene ocean water (McArthur et al., 2001) and local river water values (Palmer and Edmond, 1992; Major et al., 2004).

Since none of the three possible water bodies (Mediterranean, Paratethys or global ocean) have Sr isotope ratios that can account for the Sr isotope evolution at Intepe, some other water source must be implicated. Rivers flowing into the Sea of Marmara have sufficiently low Sr isotope ratios (Major et al., 2004) to generate the Intepe record (Fig. 8). Although hydrothermal fluids cannot be excluded, river water is the most probable source of water with low Sr isotope ratio.

If an influx of fresh, river water is assumed to control the Sr isotope evolution at Intepe, then the concomitant rise in salinity cannot reflect simple mixing of different salinity water sources. Instead Marmara Sea water salinity is likely to have been controlled by an increase in the dominance of evaporation over water loss by outflow. Decreasing exchange with the Mediterranean, global ocean and/or Paratethys is therefore the most likely explanation of both rising salinities and lowered Sr isotope ratios in the Sea of Marmara. Restriction would not only have increased the proportion of river water feeding the basin leading to lower ^{87}Sr/^{86}Sr, but would also have reduced the amount of outflow from the basin, concentrating salt by evaporation and resulting in more marine-like salinities. Just as in the example of pre-evaporite foraminifera in the northern Mediterranean sub-basins, this illustrates the danger of assuming oceanic connection on the basis of faunal salinity tolerance.

In this Sea of Marmara example then, although it is not possible to exclude all connection with Paratethys or the Mediterranean, the changes in salinity are demonstrably not the result of increased inflow from these water bodies.

4. Conclusions

The hydrologic flux scenarios required for major salinity changes preserved in the geological record have previously been difficult to test. An innovative approach which combines ^{87}Sr/^{86}Sr data with paired palaeosalinity estimates from faunal and lithological evidence allows past outflow and inflow components of the hydrologic budget of a marginal basin to be separated and examined. This permits robust interrogation of hypotheses put forward to explain major salinity fluctuations. Where hypotheses fail to satisfy the Sr isotope data they can be ruled out definitively unless the ^{87}Sr/^{86}Sr data are discounted altogether.

This approach has been applied to the end-member models for triggering salt precipitation in the Mediterranean: desiccation and connected basin hypotheses. The results suggest that the desiccation model which requires reduced Atlantic inflow is inconsistent with Sr isotope data from the pre-evaporite–Lower Evaporite transition. These data indicate an increase in the proportion of ocean water entering the Mediterranean coincident with the onset of evaporite precipitation. The combined Sr isotope–salinity approach also indicates that there is no sudden change in the proportion of water lost to the Mediterranean by evaporation or outflow with initiation of salt. This suggests that connected basin hypotheses are also unlikely to be correct because they imply an associated increase in evaporative water loss and/or reduction in outflow. Neither of these hypotheses can survive this robust test of their hydrologic flux scenarios unless all the Sr isotope variation is attributed to diagenesis.

An alternative hypothesis which is consistent with all currently available datasets suggests that salt saturation in the Mediterranean was raised from marine to hypersaline levels at the onset of the MSC as a result of an Atlantic transgression. Faunal and Sr isotope records from the northern sub-basins show that prior to salt precipitation, the Mediterranean was highly diluted with river water, but maintained marine salinities because the evaporation flux was the dominant method of water loss. With an increase in the mass of salt made available by a transgression, the same proportion of fresh-water loss by evaporation resulted in a much higher salinity of the remaining water. This new model has implications for the water-level history of the Mediterranean. In particular, the relative rise in Mediterranean water-level associated with the transgression, helps to explain why the onset of evaporite precipitation appears to be synchronous across the Mediterranean in both basinal and marginal settings; something that challenges earlier desiccation or connected basin models.

In mid-latitude marginal marine basins where net evaporation exceeds precipitation, changes in the degree of oceanic exchange may not have a simple relationship with salinity. Since fauna and lithology respond to salinity and not directly to the degree of restriction, an alternative approach is needed to test connectivity histories and hypotheses for large changes in salinity. Sr isotopes can be a more robust way of testing for episodes of connection or isolation either between basins or with the ocean than traditional approaches reliant on faunal salinity tolerance. This approach could be used to test whether the nannofossil-bearing layers in Late Messinian sections in Romania indicate Mediterranean–Paratethyan connection.

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