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# STRONTIUM CONTENT AND Na/Ca RATIO IN RECENT MOLLUSK SHELLS AS SALINITY INDICATORS IN FLUVIAL WATER SYSTEMS

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**ABSTRACT**

Mollusk shells of C. (Corbicula) fluminalis (MULLER) and Unio tigridis (BOURGUIGNAT) were collected from 14 sites along the Euphrates River from Kerabla to Nassirriyah. The shells were analysed for their Sr, Ca and Na contents and examined by XRD for mineral composition.

The results were compared to the water main constituents of cations and anions as well as the total dissolved salts (TDS) reported in water samples collected from the same sites and in the same year. The results show that Sr and Na concentrations as well as Na/ Ca ratio are directly correlated with the TDS of the water and also with the water content of dissolved $Na^{+}$, $Ca^{2+}$, $SO_4^{2-}$ and $Cl^{-}$.

The nature of Sr and Na presence in these aragonite shells is believed to be in substitution for Ca in the aragonite structure. The Sr and Na contents of the Recent shells in the present study is much higher than that reported in ancient limestones. Strontium and sodium may be expelled and remobilized when aragonite alters to calcite during diagenesis.

## محتوى السترونتيوم ونسبة Ca / Na في أصداف الرخويات كمؤشرات على الملوحة في أنظمة المياه النهرية

**المستخلص**

يتناول هذا البحث تحليل أصداف الرخويات:

**C . ( Corbicula ) fluminalis (MULLER) و Unio tigridis (BOURGUIGNAT)**

التي تم جمعها من 14 محطة نمذجة على طول نهر الفرات من الكرابلة إلى الناصرية للعناصر Sr و Na و Ca وفحصها معدنيا بحيود الأشعة السينية. تمت مقارنة النتائج مع تحاليل مياه نهر الفرات للايونات الرئيسية الموجبة والسالبة فضلاً عن الأملاح الذائبة الكلية في العينات التي تم جمعها في نفس السنة ومن المحطات ذاتها.

بينت النتائج أن تراكيز Sr و Na فضلاً عن نسبة Ca / Na في أصداف الرخويات ذات علاقة ايجابية مباشرة مع تراكيز الأملاح الذائبة الكلية (TDS) وتراكيز $Na^{+}$ و $Ca^{2+}$ و $SO_4^{2-}$ و $Cl^{-}$ في المياه. أن طبيعة وجود Sr و Na في هذه الأصداف هي إحلال بلوري محل Ca في التركيب البلوري لمعدن الاراغونايت الذي تتكون منه هذه الأصداف. يلاحظ ايضاً من النتائج ان محتوى الأصداف الحديثة هذه من Sr و Na أعلى بكثير من محتواها في حجر الكلس في التكوينات القديمة ويعود ذلك إلى إمكانية طرد جزء كبير من هذه العناصر عند تحول الاراغونايت إلى كالسايت بفعل العمليات التحويرية.

## INTRODUCTION

The use of Sr in marine carbonate minerals and rocks as an environmental indicator has been a subject of research by many workers (Kinsman, 1969, Veizer et al., 1971, Veizer and Demovic, 1974, Al-Aasam and Al-Kufaishi, 1977 and Al-Bassam and Saeed, 1980, among others). It has been used in facies analysis and diagenetic studies of carbonates. The previous work on Sr as an environmental indicator, however, was concerned with marine carbonates only.

Sodium has been suggested as a possible index to salinity of diagenetic solutions in carbonate sediments and rocks (Land and Hoops, 1973). It has been

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also suggested as a paleosalinity indicator in ancient carbonate rocks; the hypersaline facies of the Lower Paleozoic carbonate sequence of Arctic Canada was found to contain higher soluble Na concentrations than their open - marine counterparts (Veizer et al., 1977).This is the first time that strontium and sodium in the carbonate shells are explored as potential indicators of salinity in fresh-water systems on the basis of actual and representative water analysis.

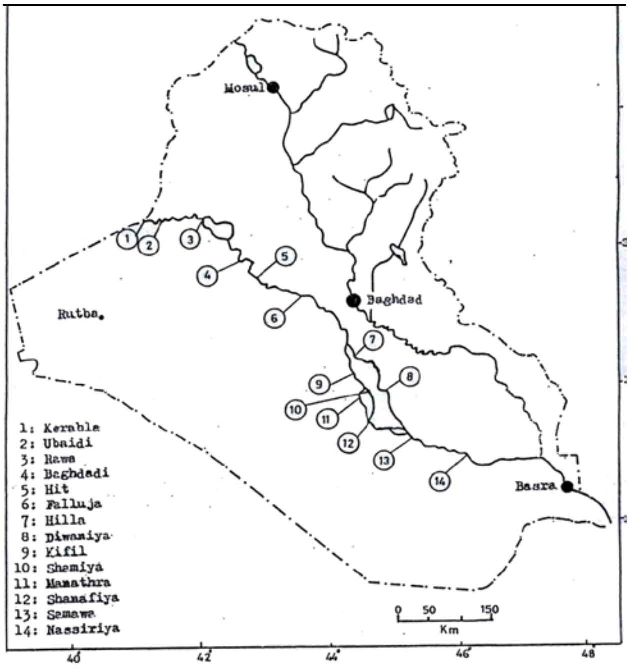


Fig. 1: Location map of the mollusk samples

In a recent study, samples of mollusk shells were collected from the Euphrates River banks and analysed for some heavy metals to study the environmental impact of the river on organisms (Al- Bassam, 1999). The Sr content of the shells of C. (Corbicula) fluminalis (MULLER) and Unio tigridis (BOURGUIGNAT) was compared to water salinity of the Euphrates River and found to be a useful indicator of TDS concentration (Al- Bassam, 2000).

The Euphrates River is 2940 km long and its length in Iraq is 1159 km. It enters the territory at Al- Qaim, descends southeast, following the general land gradient, separating the Iraqi desert from the Jezira and Mesopotamian Plain and finally joins the Tigris River at Qurna. One of the characteristic features of the water quality in the Euphrates River is the sudden increase of TDS from Shannafiya to Qurna. The TDS increases in this sector by about three folds. This phenomenon was noticed since 1978 when the observers started to document the changes in the water quality of the river (Banat et al., 1981).

## SAMPLING AND METHODS

Shell samples of dead mollusks were collected in 1998 from 14 stations along the Euphrates River; from Kerabla, near the entry of the river to Iraq, to Nassiriyah in the south (Fig. 1). Shells of C. (Corbicula) fluminalis (MULLER) and Unio tigridis (BOURGUIGNAT) were the most common among the mollusks in the sampled sites and were used in the present study for analysis (Fig. 2).

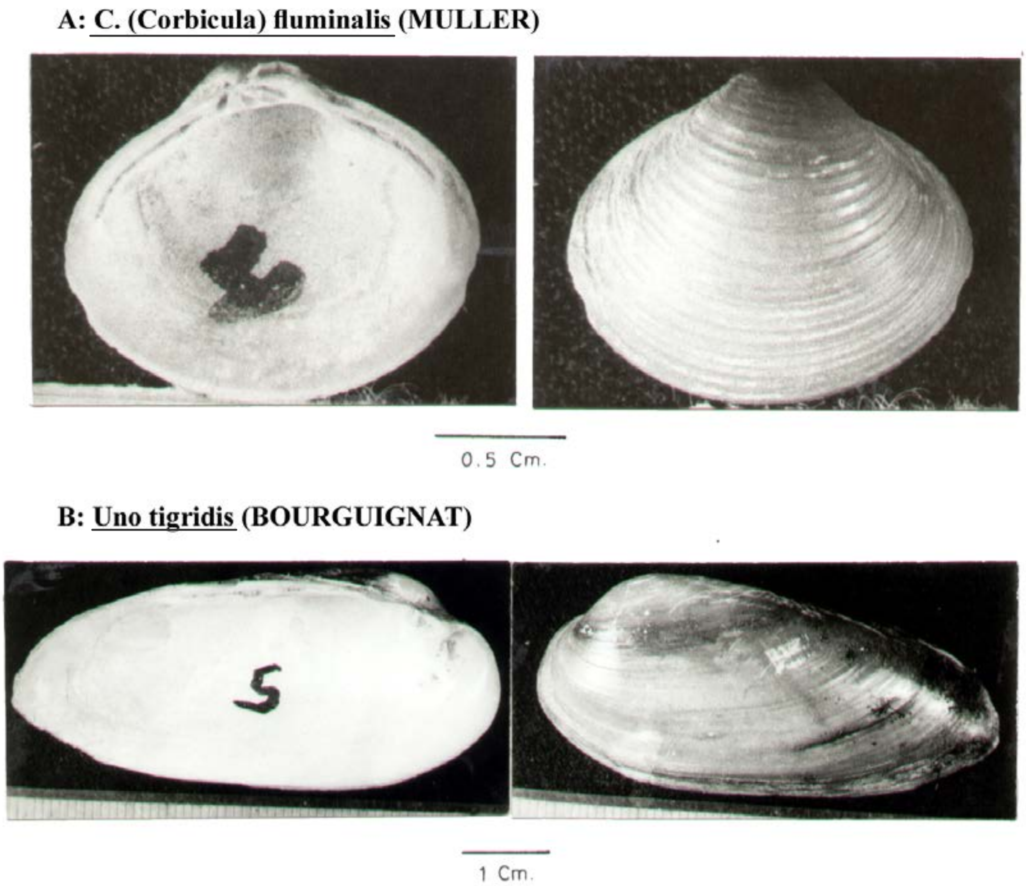


Fig. 2: Photographs of the mollusk shells

The shell samples were thoroughly cleaned by water to remove dirt and stains before analysis. The chemical analysis was carried out at the State Co. of Geological Survey and Mining. Dilute HCl was used for digestion, calcium was analysed by titration, strontium and sodium by atomic absorption spectrophotometry. The samples were examined by XRD for mineral constituents. Water analysis of the Euphrates River are annual mean values of the year 1998 (mean of bi-monthly collected samples analysed for the Euphrates Project/Ministry of Agriculture).

## RESULTS

The mollusk shells of the present study are composed of aragonite (Fig. 3). The chemical analysis of Sr, Na and Ca in the shells is shown in Table 1 together with the Euphrates water composition in the corresponding sites. The Sr concentration (Table 1) varies from 3366 ppm to 5665 ppm (mean 4119 ppm) in the shells of C. (Corbicula) fluminalis MULLER and from 3196 ppm to 5015 ppm (mean 3974 ppm) in the shells of Unio tigridis BOURGUIGNAT.Strontium concentration in the shells of both species increases significantly in the samples collected from the southern sector of the river, which includes Shanafiya, Samawa and Nassiriya stations.

Sodium content of the shells ranged from 2370 ppm to 3413 ppm (mean 2702 ppm) and calcium content ranged from 36.5% to 42.5% (mean 39.46%). Higher Na values were noticed in the samples from the southern sector of the river, whereas higher Ca values are more common in the northern sector. The shells of C. (Corbicula) fluminalis (MULLER) were found to contain higher concentrations of Na relative to those of Unio tigridis (BOURGUIGNAT) in the sites where both types of mollusks were sampled.

The water analysis of the Euphrates River showed gradual increase in the ionic concentrations from Kerabla to Hit (TDS: 424 - 475 ppm). Further increase in the concentration of total dissolved salts occurs between Falluja and Manathra (TDS: 579 - 600 ppm) and becomes remarkably higher between Shannafiya and Nassiriya (TDS: 1575 - 1640 ppm). The increase in TDS values is due to the increase of mainly $Na^+$, $Cl^-$ and $SO_4^{2-}$ concentrations in the southern sector which is expressed in the hydrochemical formulae and hypothetical salts combination of the water (Tables 2 and 3). On the bases of water chemistry the Euphrates River can be divided into three sectors: Northern (Kerabla - Hit), Central (Falluja - Manathra, including Shatt Al- Hilla) and Southern (Shannafiya - Nassirriya, which extends to Qurna).

The Sr content of the analysed shells is negatively correlated with the Ca content of these shells, but positively correlated with the major cations and anions of the water (Table 4).The Na content of the analysed shells is positively correlated with the main ionic constituents of the water and negatively correlated with the Ca content of the shells (Table 4). On the other hand, the Ca content of the shells is negatively correlated with all ionic constituents of the water. Consequently, the ratio Na/ Ca of the shells shows more pronounced positive correlation with the main constituents of water including TDS, $Na^+$, $Ca^{2+}$, $Cl^-$ and $SO_4^{2-}$.

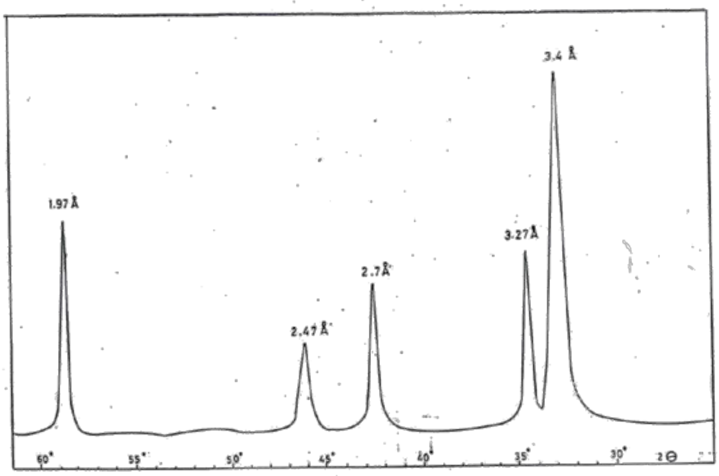


Fig. 3: X-ray diffractogram of a mollusk shell showing pure aragonite phase

Table 1: Analytical results of mollusk shells and the Euphrates water.

| Sampling site | | Na(S) | Ca(S) | Sr (S) | Na/Ca | Ca(W) | $SO_4$(W) | Cl(W) | Na(W) | TDS(W) | Mollusk |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | Kerabla | 3042 | 40.71 | 4651 | 74.70 | 54 | 97 | 71 | 50 | 424 | C |
| 1 | Kerabla | 2523 | 40.00 | 3196 | 63.10 | 54 | 97 | 71 | 50 | 424 | U |
| 2 | Ubaidi | 2300 | 41.25 | 3940 | 55.80 | 56 | 119 | 73 | 51 | 449 | U |
| 3 | Rawa | 2448 | 40.50 | 3932 | 60.40 | 55 | 97 | 71 | 50 | 424 | U &C |
| 4 | Baghdadi | 2448 | 42.50 | 4347 | 57.60 | 55 | 130 | 80 | 54 | 462 | C |
| 5 | Hit | 2374 | 39.50 | 4439 | 60.10 | 56 | 144 | 86 | 56 | 475 | U |
| 6 | Falluja | 2968 | 39.00 | 3366 | 76.10 | 70 | 185 | 101 | 69 | 579 | C |
| 7 | Hilla | 3042 | 38.50 | 3535 | 79.00 | 73 | 190 | 103 | 73 | 609 | C |
| 8 | Diwaniya | 3116 | 38.38 | 3662 | 81.20 | 75 | 195 | 116 | 75 | 640 | C |
| 9 | Kifil | 2448 | 39.75 | 4372 | 61.60 | 71 | 194 | 102 | 69 | 580 | C |
| 9 | Kifil | 2374 | 37.25 | 3856 | 63.70 | 71 | 194 | 102 | 69 | 580 | U |
| 10 | Shamiya | 2819 | 39.75 | 3873 | 70.90 | 73 | 195 | 105 | 70 | 600 | C |
| 10 | Shamiya | 2448 | 42.00 | 3839 | 58.30 | 73 | 195 | 105 | 70 | 600 | U |
| 11 | Manathra | 2523 | 40.25 | 3696 | 62.70 | 72 | 200 | 110 | 70 | 600 | C |
| 11 | Manathra | 2448 | 41.00 | 3535 | 59.70 | 72 | 200 | 110 | 70 | 600 | U |
| 12 | Shanafiya | 2894 | 39.50 | 4820 | 73.30 | 117 | 465 | 393 | 272 | 1640 | C |
| 13 | Samawa | 3265 | 36.50 | 4465 | 89.50 | 116 | 483 | 370 | 262 | 1575 | C |
| 14 | Nassiriya | 3413 | 36.63 | 5665 | 93.20 | 117 | 467 | 400 | 275 | 1640 | C |
| 14 | Nassiriya | 2448 | 36.75 | 5015 | 66.60 | 117 | 467 | 400 | 275 | 1640 | U |

**C: C. (Corbiculla) fluminalis (MULLER), U: Unio tigridis (BOURGUIGNAT). Analytical values in ppm except Ca (S) in %**
**(S): mollusk shells, (W): Euphrates water.**

Table 2: Hydrochemical formulae of the Euphrates River water (Al- Bassam, 1998). (epm %)

S 0.41 gm/1
$$\frac{SO_4\ 36.23 \quad HCO_3\ 34.95 \quad Cl\ 25.82 \quad CO_3\ 3.00}{Ca\ 43.81 \quad Mg\ 28.09 \quad Na\ 27.22 \quad K\ 0.87}$$
Northern sector, pH 8.15

S 0.72 gm/1
$$\frac{SO_4\ 42.90 \quad Cl\ 33.48 \quad HCO_3\ 20.45 \quad CO_3\ 3.17}{Ca\ 38.47 \quad Na\ 34.11 \quad Mg\ 26.54 \quad K\ 0.89}$$
Central sector, pH 8.23

S 1.44 gm/1
$$\frac{Cl\ 47.13 \quad SO_4\ 41.42 \quad HCO_3\ 9.93 \quad CO_3\ 1.80}{Na\ 47.52 \quad Ca\ 27.61 \quad Mg\ 24.25 \quad K\ 0.62}$$
Southern sector, pH 8.23

Table 3 : Hypothetical salt combination in the Euphrates River (Al- Bassam, 1998) (meq %)

| **Sector** | **$Ca(HCO_3)_2$** | **$CaSO_4$** | **$MgCO_3$** | **$MgSO_4$** | **$Na_2SO_4$** | **NaCl** | **KCl** |
|---|---|---|---|---|---|---|---|
| **Northern** | 34.95 | 8.86 | 3.00 | 25.09 | 2.28 | 24.94 | 0.88 |
| **Central** | 20.45 | 18.02 | 3.17 | 23.37 | 1.51 | 32.60 | 0.89 |
| **Southern** | 9.93 | 14.32 | 1.80 | 25.81 | 1.01 | 46.51 | 0.62 |

Table 4: Correlation coefficient values of the variables in shells and water.

| | **Sr (s)** | **Na (s)** | **Ca (s)** | **Na/Ca** | **Ca (w)** | **$SO_4$ (w)** | **Cl (w)** | **Na(w)** |
|---|---|---|---|---|---|---|---|---|
| Sr(s) | 1 | | | | | | | |
| Na(s) | 0.307 | 1 | | | | | | |
| Ca(s) | -0.371 | -0.525 | 1 | | | | | |
| Na/Ca | 0.368 | 0.971 | -0.709 | 1 | | | | |
| Ca(w) | 0.619 | 0.509 | -0.696 | 0.623 | 1 | | | |
| $SO_4$(w) | 0.658 | 0.481 | -0.686 | 0.600 | 0.994 | 1 | | |
| Cl(w) | 0.724 | 0.472 | -0.658 | 0.584 | 0.972 | 0.983 | 1 | |
| Na(w) | 0.723 | 0.478 | -0.661 | 0.590 | 0.969 | 0.981 | 0.999 | 1 |
| TDS(w) | 0.704 | 0.486 | -0.667 | 0.598 | 0.981 | 0.990 | 0.999 | 0.998 |

(s): shells, (w): water.

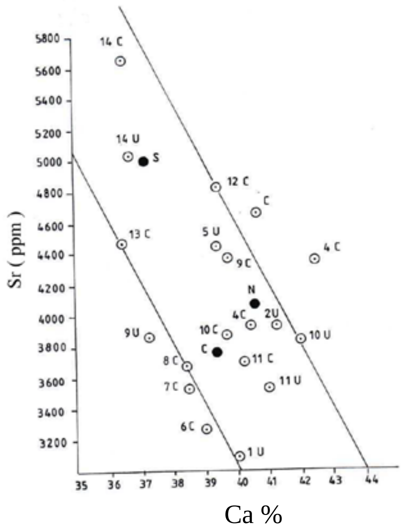


Fig. 4: Correlation between Sr and Ca in mollusk shells (solid circles are mean values for each sector of the river).

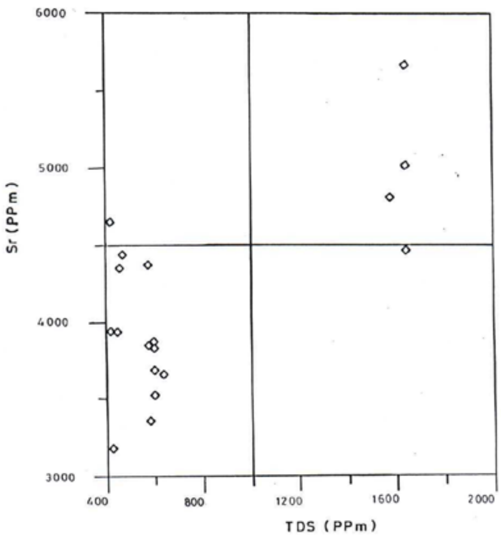


Fig. 5: Correlation diagram of Sr (mollusks) with TDS (water)

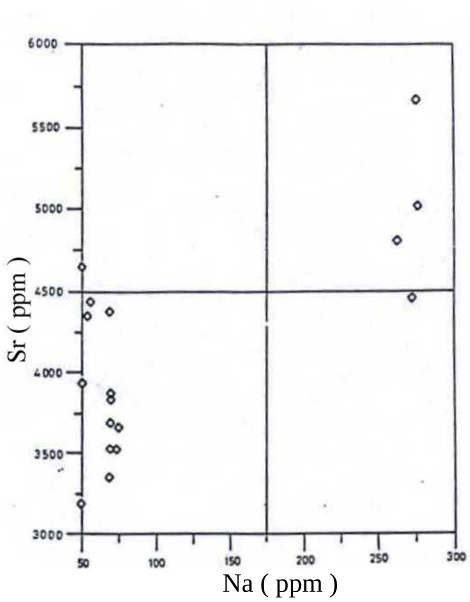


Fig. 6: Sr – Na correlation diagram

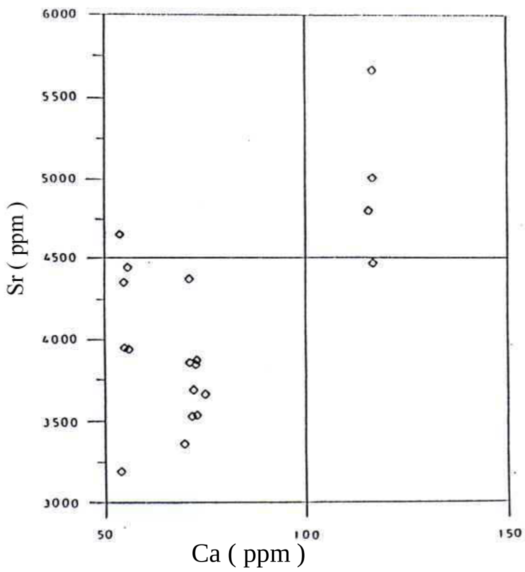


Fig. 7: Sr – Ca correlation diagram

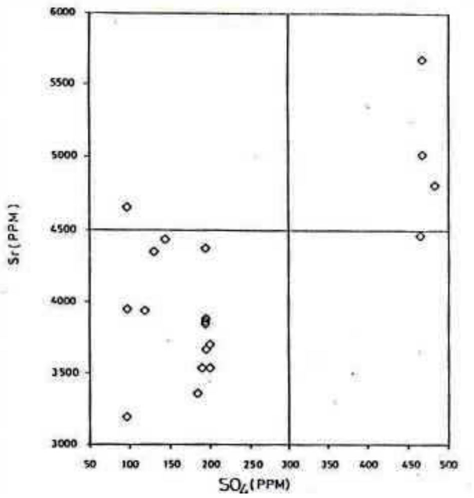

Fig. 8: Sr – $SO_4$ Correlation diagram

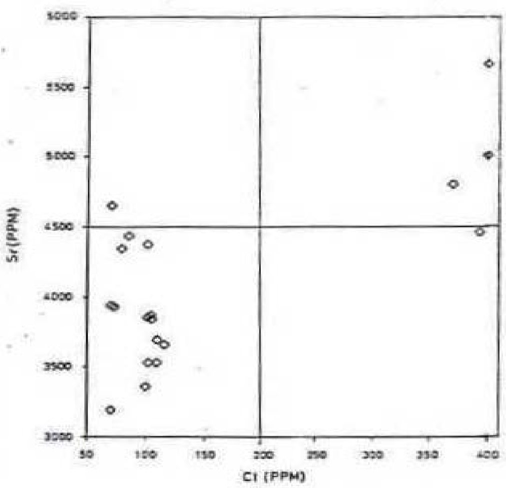

Fig. 9: Sr – Cl correlation diagram

## DISCUSSION

Mollusks build their shells from the inorganic components dissolved in the water where they live. Their mobility is limited and so is their living time. Hence, they can be useful indicators of water composition with regard to the type and concentration of dissolved salts. They have been successfully used in the detection and monitoring of heavy metal pollution in marine and fresh-water systems (Al-Aasam et al., 1997). The use of Sr as an environmental indicator in marine carbonate systems has been partially successful due to the diagenetic alteration of the original less stable carbonate minerals such as aragonite, and high-Mg calcite.

The present results indicate that $Sr^{2+}$ incorporation in the aragonite structure, evidenced by the negative correlate ion between Sr and Ca in the shells (Table 4 and Fig. 4) is proportional to the salinity of the water. Kolesar and Friedman (in Gavish and Friedman, 1969) clamed the existence of two types of fresh-water calcites: high-Sr and low-Sr calcites, depending on whether the water is rich in strontium. The water samples of the present study were not analysed for Sr, but it is expected that $Sr^{2+}$ concentration increases in the water as the concentration of other cations increases. The correlation diagrams of Sr in mollusk shells with TDS, $Na^{+}$, $Cl^{-}$, $SO_4^{2-}$ and $Ca^{2+}$ concentrations in the river water show very good separation of sample populations. The lower salinity water samples almost totally produced mollusk shells with less than 4500 ppm Sr (Figs. 5, 6, 7, 8 and 9).

The increase in Sr concentration in the analysed aragonitic shells is only evident when the salinity in the water, expressed as TDS, is increased by about three folds and the difference in TDS content becomes more than 1000 ppm. Minor changes in the salinity of the water were not felt in the Sr content of the shells. Furthermore, local sources of Sr may contribute to its increase in the water and consequently in the mollusk shells, irrespective of water salinity. Such sources may include Sr-rich groundwater, as those of Hit-Abu Jir, which discharge to the Euphrates and contaminate the river water near Hit by Sr and other metals. This is evident from the relatively high Sr-values in the shells collected from Baghdadi and Hit stations (Table 1).

The position of Na in calcium minerals can be attributed mainly to lattice substitution. The ionic radii of $Na^{+}$ and $Ca^{2+}$ in a six fold coordination are 0.97 Å and 0.99 Å respectively (Krauskopf, 1967) which permits substitution of $Na^{+}$ for $Ca^{2+}$ in calcite and aragonite (among other Ca- bearing minerals). The latter tolerates higher Na substitution due to its structural configuration, which provides more accommodation space for such ions including Sr and Pb (Deer et al., 1966).

In marine and hypersaline carbonates the possibility of solid or liquid inclusions of NaCl has been suggested as a minor mode of Na presence in these rocks (Fritz and Katz, 1972). Moreover, Na may be accommodated in the aluminosilicates present as impurities in carbonate rocks (Veizer et al., 1977). In the present study both of these possibilities can be ignored since the samples are fresh - water carbonates, not likely to include NaCl inclusions and they are single mineral samples, composed entirely of aragonite, clean of aluminosilicates or other Na - bearing impurities. They are recently formed and are not subjected to diagenetic alteration yet.

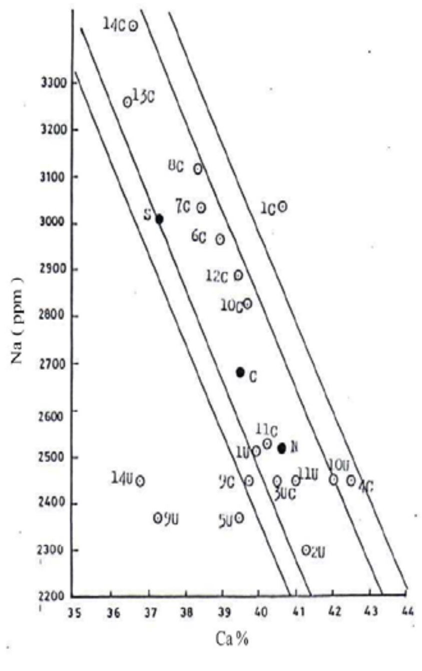


Fig. 10: Correlation between Na & Ca in mollusk shells (solid black circles are mean values for each sector of the river)

Hence, all the Na detected in the analysed mollusk shells may be attributed to structural substitution for Ca in aragonite. This conclusion is supported by the negative correlation between Na and Ca in the analysed samples (Fig. 10). In view of the positive correlation of Na (shells) with the water salinity components, and the negative correlation of Ca (shells) with these components, the Na/ Ca ratio was found more significant to illustrate the purpose of this paper.

The Na/ Ca ratio (shells) show relatively wider spread Values relative to the more homogeneous water constituents within individual sectors of the river. Mollusk shells are bound to exhibit wider range of their Na and Ca values in view of the individual nature of shell buildup and due to the local environmental factors that may influence the shell composition even within single sampling site. For this reason the results were dealt with collectively; they were grouped as three populations according to their position in the three sectors of the river.

The Na/ Ca ratio in the mollusk shells is positively correlated with the Na/ Ca ratio of the water (Fig. 11). It is also positively correlated with TDS (Fig. 12) and with the major ionic constituents of the water: $Na^+$, $Ca^{2+}$, $Cl^-$, and $SO_4^{2-}$ (Figs. 13, 14, 15 and 16). However, the variation in the Na/ Ca ratio in these shells seems to be mainly a function of $Na^+$ concentration in water and to a lesser extent to the Na/ Ca ratio of the water since Na/ Ca (shells) is positively correlated with $Ca^{2+}$ concentration of water (Table 4 and Fig. 14).

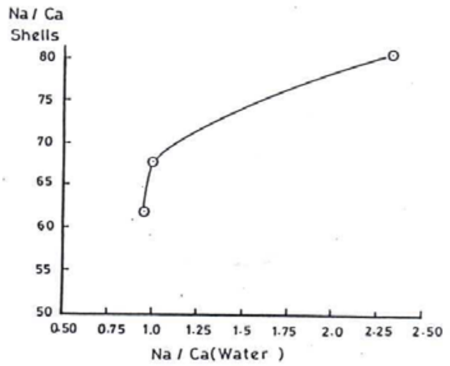


Fig.11: Correlation of Na / Ca in Shells and Na / Ca in water (mean values of river sectors)

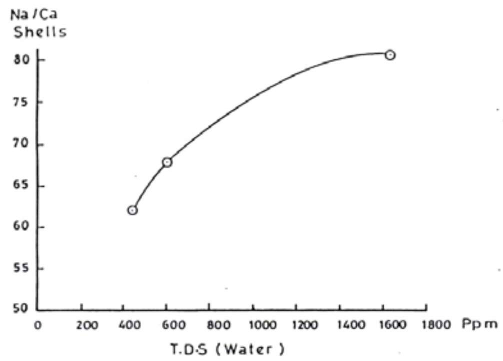


Fig.12: Correlation between Na / Ca in Shells and T.D.S of water (mean values of river sectors)

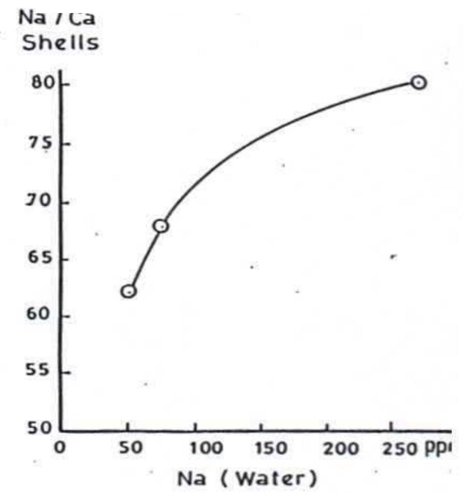


Fig.13: Correlation between Na / Ca in Shells and Na of water (mean values of river sectors)

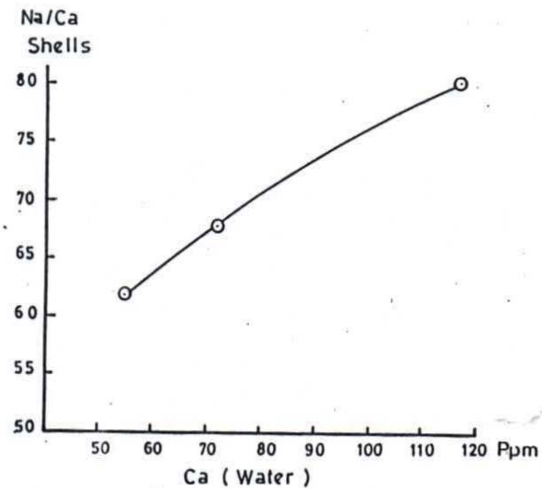


Fig.14: Correlation between Na / Ca in Shells and Ca of water (mean values of river sectors)

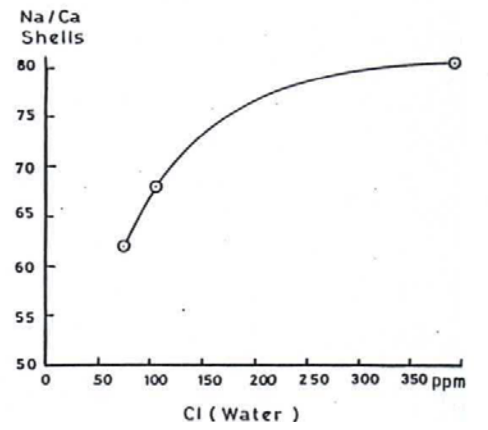


Fig.15: Correlation between Na / Ca in Shells and Cl of water (mean values of river sectors)

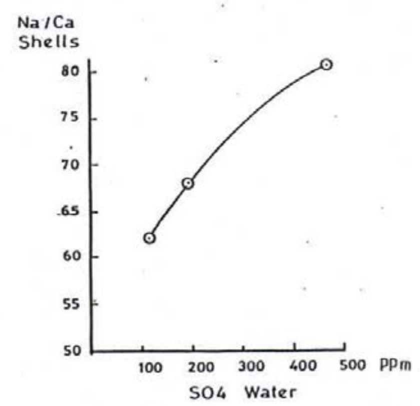


Fig.16: Correlation between Na / Ca in Shells and $SO_4$ of water (mean values of river sectors)

The role of other ionic constituents of the water on the rate of Na substitution in aragonite and consequently on the Na/ Ca ratio of the shells is unknown. However, the increase in the concentration of $Cl^-$ and $SO_4^{2-}$ together with $Na^+$ from North to South has influenced the water chemistry of the Euphrates River and the type of the major dissolved salts. The hypothetical dissolved salts contents derived from water chemistry in the three sectors of the Euphrates River (Table 3) show that $Ca(HCO_3)_2$ content decreases from about 35 meq % in the Northern sector to about 9.9 meq % in the Southern sector, accompanied by an increase in the NaCl content from about 24.9 meq % to 46.5 meq % in the two sectors respectively. This drastic change in water chemistry has its impact on the Na / Ca

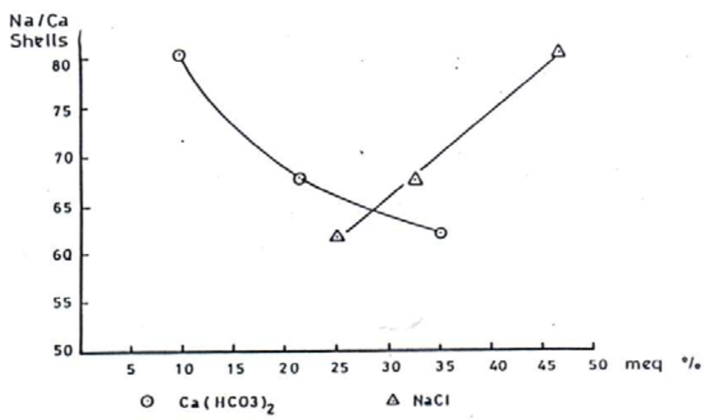


Fig. 17: Correlation between hypothetical dissolved salts of water with Na / Ca ratio of mollusk shells (mean values of river sectors)

ratio of the mollusk shells, which is negatively correlated with $Ca(HCO_3)_2$ and positively correlated with NaCl concentrations (Fig. 17). Consequently, the rate of Na substation for Ca in aragonitic fresh - water shells may be considered as an indicator of water salinity in fluvial systems. The fluctuation of the results may be dealt with by increasing the number of samples collected.

## CONCLUSIONS

Strontium content of recent mollusk shells can be a useful indicator of water salinity in fresh-water systems. In the Euphrates River, Sr content of C. (Corbicula) fluminalis MULLER and Unio tigridis BOURGUIGNAT is directly related to TDS, $Na^+$, $Cl^-$, $SO_4^{2-}$ and $Ca^{2+}$ concentrations in the river water. The relation is only significant when the difference in the TDS salinity is 1000 ppm or more. Fluctuations in the Sr content of the shells within a narrow range of water salinity may be attributed to local sources of Sr enrichment in the river system.

The results obtained in this study show that the rate of Na substitution in the mollusk shells is a function of water salinity in terms of $Na^+$ concentration in water. The rate is higher in NaCl- type of waters rather than in $Ca(HCO_3)^{2-}$ type. The Na/ Ca ratio of the mollusk shells can be used as a salinity indicator in fresh - water systems.

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