

OSTRACOD SHELL CHEMISTRY: A NEW PALAEOENVIRONMENTAL INDICATOR APPLIED TO A REGRESSIVE/TRANSGRESSIVE RECORD FROM THE GULF OF CARPENTARIA, AUSTRALIA

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Abstract

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We demonstrate the feasibility of determining a continuous record of chemical changes occurring during marine transgressive/regressive phases, using the Mg/Ca and Sr/Ca ratios measured in some 300 single valves of the euryhaline ostracod *Cyprideis* from two cores from the Gulf of Carpentaria, Australia. The molar Mg and Sr distribution coefficients $K_D[\text{Mg}]$ and $K_D[\text{Sr}]$ for modern Australian *Cyprideis* specimens (Chivas et al. 1986b),

$$K_D[\text{Mg}] \text{ at } 25^\circ\text{C} = (\text{Mg/Ca})_{\text{shell}}/(\text{Mg/Ca})_{\text{water}} = 0.00458 \pm 0.00072$$

$$K_D[\text{Sr}] = (\text{Sr/Ca})_{\text{shell}}/(\text{Sr/Ca})_{\text{water}} = 0.475 \pm 0.057$$

are used here to reconstruct the palaeoenvironmental record of the gulf. The two cores studied show similar patterns during the past 40,000 yr for equivalent hydrologic phases recognized in the cores. The Mg/Ca ratios of the water inferred from ostracod shell chemistry indicate that Carpentaria was a fresh or slightly saline lake between $\approx 40,000$ and $\approx 13,000$ yr B.P., before being transgressed by the sea during sea level rise. Around 26,000 yr B.P., a phase of substantial precipitation of authigenic calcite caused a dramatic increase in the Sr/Ca and Mg/Ca ratios of the lake water, and this was registered by the ostracod shell chemistry. From $\approx 26,000$ to $\approx 13,000$ yr B.P., the lake remained fresh or slightly saline despite the Ca depletion of the water which caused the Sr/Ca and Mg/Ca of the ostracods to be unusually high.

Introduction

Until now, it has proven difficult to determine fairly accurately past changes of water salinity such as those that occur at sites of marine transgression–regression and evaporative concentration. Identification of past

environmental changes has usually relied on data gathered from the fossil biota, combined with interpretation of sedimentary facies. Attempts have been made to determine past salinities from carbonate chemistry. Veizer et al. (1977) determined that in a Lower Palaeozoic carbonate sequence of Arctic Canada a hypersaline facies is recognizable by high soluble sodium concentrations in comparison

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with the open-marine facies which has lower values. Similarly, Land and Hoops (1973) attempted to establish an index for the salinity of diagenetic solutions by studying the bulk sodium in carbonates. Veizer and Demovič (1974) also examined the possibility of distinguishing different facies (from hypersaline to littoral neritic and shallow bathyal) by measuring the distribution of Sr in Mesozoic carbonate rocks of the central western Carpathians. These investigators were unable to obtain accurate water salinity values.

Since most organisms living in estuaries can and must tolerate a broad range of salinity, it has rarely been possible to precisely define past salinities simply by using fossil material, unless a complete assemblage of species could be matched against well documented modern examples. Even then, only broad salinity ranges can be commonly estimated. In addition, since typically marine organisms (e.g. Foraminifera) and freshwater organisms (e.g. charophytes) have been found in continental saline waters (Burne et al., 1980; De Deckker and Geddes, 1980; Cann and De Deckker, 1981), confusion has occurred in the interpretation of palaeoenvironments, and more particularly with regard to palaeosalinities in trying to identify marine and freshwater facies in marine transgression-regression sequences.

Recently, Chivas et al. (1983, 1985, 1986a, b) conducted a series of laboratory and field investigations to determine environmental factors controlling the uptake of Sr, Mg, Ba in the calcareous shells of ostracods. This work so far is restricted to lacustrine material. Consideration is given here to using the amount of Mg and Sr in ostracod shells to determine the relationship between seawater composition and salinity and the Mg/Ca and Sr/Ca in the shells of ostracods. The site chosen to test this salinity indicator is the shallow (<69 m) Gulf of Carpentaria in northern Australia which, during the late Quaternary, was at times isolated from the ocean by sea-level fluctuations.

Ostracods

We chose to study ostracods as possible hydrochemical and salinity indicators because they are ubiquitous bivalved organisms found in continental, estuarine, marine and hypersaline waters, and because they have low-Mg calcite valves which readily fossilize. Their fossil record spans the entire Phanerozoic. In contrast to molluscs and foraminifers, which during growth continue to secrete additional layers or chambers respectively, ostracods, being arthropods, have to moult and build new valves after shedding the old ones. They usually undergo this process up to 9 times before reaching adulthood. The components of the shells are taken directly from the host water after moulting (Turpen and Angell, 1971); there is no storage of material within the animal prior to moulting. It is therefore accepted that ostracod valve chemistry should reflect the ambient conditions of the aquatic environment at the time of moulting and new valve formation. Chivas et al. (1983, 1986a, b) have demonstrated with in vitro experiments that uptake of Mg in lacustrine ostracods is affected by water temperature and the Mg/Ca of the host water. They showed for the same ostracods that Sr uptake is directly related to the Sr/Ca of the water, apparently with little temperature effect over the range 10–25°C (Chivas et al., 1983, 1985, 1986b).

The principal ostracod chosen for this study belongs to the well known euryhaline genus *Cyprideis*. It is cosmopolitan and tolerates a wide range of salinities; it can be found in fresh waters and in salinities up to ~100‰ (De Deckker, 1981; Vesper, 1972a; Rosenfeld and Vesper, 1977). *Cyprideis* has been the subject of numerous investigations (e.g. general: (Sandberg, 1964); ecological: (Vesper, 1972a; Heip, 1976; Herman and Heip, 1982; Herman et al., 1983); palaeontological and shell morphology: (Vesper, 1972b; Kilenyi and Whittaker, 1974; Rosenfeld and Vesper, 1977; Carbonel, 1980; Rosenfeld, 1982; Bodergat, 1983), the latter reference with chemical analyses). *Cyprideis* is

known to occur in brackish to hypersaline conditions and consequently was used to identify particular events such as the "Messinian salinity crisis" in the Mediterranean (Benson, 1975, 1976). *Cyprideis* is also present in nearly all the samples examined from the Gulf of Carpentaria cores. In addition, *Ilyocypris australiensis* which occurs in both cores, is basically a freshwater ostracod species that tolerates slightly saline waters up to 10‰ (De Deckker, 1982). Unfortunately, there are no modern sites yet found in Australia where this species is known to cohabit with *Cyprideis* to permit palaeoecological comparison.

Distribution coefficients and relation to seawater

Following the discovery that the molar distribution coefficient (K_D) of congeneric ostracod species and of species belonging to related genera are similar (Chivas et al., 1983, 1985, 1986b), it has been possible to determine the $K_D[\text{Mg}]$ and $K_D[\text{Sr}]$ of *Cyprideis* using live specimens from several parts of the world, and to apply the K_D values to fossil material despite morphological differences among species. The molar distribution coefficient for strontium $K_D[\text{Sr}]$ in *Cyprideis* was measured by collecting live specimens and relating their shell composition to that of their host water.

Samples used for this calibration derive from sites in Australia (Fig.1) (Little Dip Lake, South Australia, a pool near Hutt Lagoon, Western Australia, a pool near Weipa on the edge of the Gulf of Carpentaria, Queensland; and from Belgium (Dievengat), France (Étang de Galabert, Camargue) and Spain (Santa Pola). European specimens belong to *Cyprideis torosa*; the Australian material is usually referred to *C. australiensis*. However, we consider that all Australian specimens discussed here (Recent or fossil) are synonymous with *C. torosa*. Specimens which were cultivated and produced new shells in the laboratory in different types of waters were also analysed (Chivas et al., 1986b and in prep.). All these results indicate that:

$$K_D[\text{Sr}]_{\text{Cyprideis}} = \frac{(\text{Sr}/\text{Ca})_{\text{shell}}}{(\text{Sr}/\text{Ca})_{\text{water}}} = 0.475 \pm 0.057.$$

(All ratios are in moles throughout the text.)

Because temperature variation affects Mg uptake in ostracod shells (Chivas et al., 1983, 1986a), in addition to the effect the Mg/Ca of the host water has on shell composition, the $K_D[\text{Mg}]$ of *Cyprideis* could only be calculated from shells formed under monitored temperature conditions. To obtain reliable analyses, only fully calcified shells were used because of the high Mg uptake by ostracods during the early stages of calcification. Only one experiment was performed using waters of different salinities and of different Mg/Ca. This permitted the calculation of

$$\begin{aligned} K_D[\text{Mg}]_{\text{Cyprideis}} \text{ at } 25^\circ\text{C} &= \frac{(\text{Mg}/\text{Ca})_{\text{shell}}}{(\text{Mg}/\text{Ca})_{\text{water}}} \\ &= 0.00458 \pm 0.00072 \end{aligned}$$

The K_D values for a given ostracod genus or species can be used to calculate the molar distribution coefficient (D) for the same ostracod living in seawater:

$$D_{[\text{Me}]} = \frac{(\text{Me}/\text{Ca})_{\text{shell}}}{(\text{Me}/\text{Ca})_{\text{seawater}}} = K_D[\text{Me}] \times (\text{Me}/\text{Ca})_{\text{seawater}}$$

(where Me is either Sr or Mg.)

Similar calculations for foraminifers have already been published (Graham et al., 1982) and were also compared with those of other organisms. By using the molar ratios for seawater, $\text{Mg}/\text{Ca} = 5 \pm 0.8$ (Renard, 1985) and $\text{Sr}/\text{Ca} = 0.0086 \pm 0.0004$ (Kinsman, 1969), we establish for *Cyprideis* that $D[\text{Mg}] = 0.0229$ at 25°C and $D[\text{Sr}] = 0.0041$.

We use the widely accepted value of 0.0086 ± 0.0004 for the Sr/Ca molar ratio of seawater although different values have been obtained from semi-restricted environments: eastern Mediterranean: 0.00785; western Mediterranean: 0.00819; the English Channel: 0.00938 (Renard, 1985). We have no data for the Gulf of Carpentaria.

Since the Sr/Ca of seawater, within the salinity range of up to $\sim 100\%$ (as would be

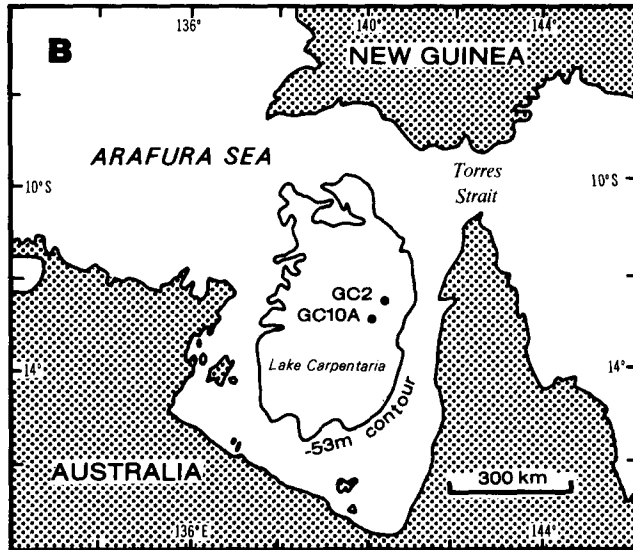
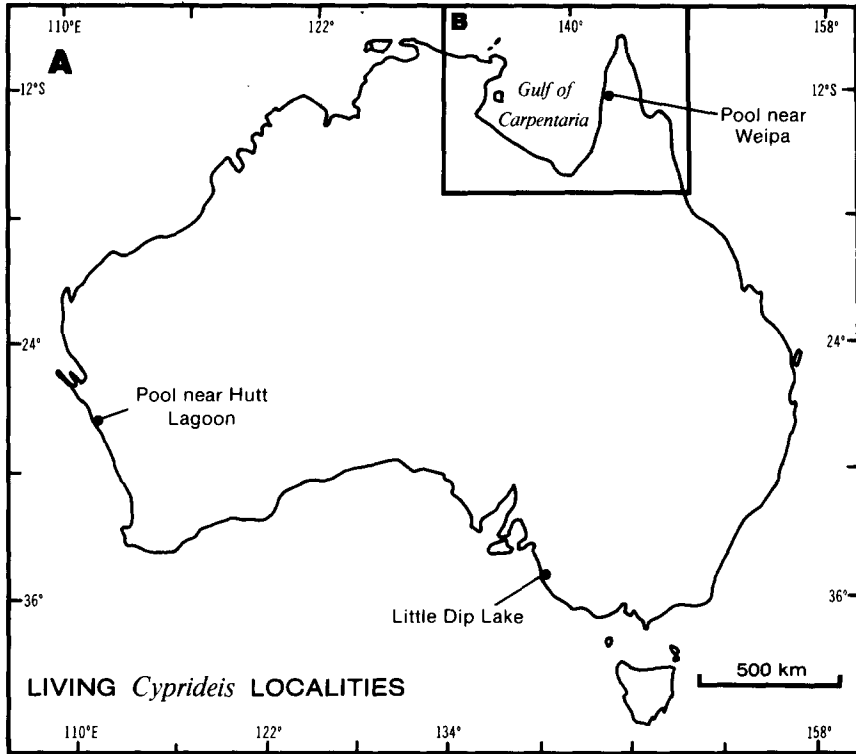


Fig.1A. Map of Australia showing the location of the Gulf of Carpentaria and the three localities where live *Cyprideis* was collected to establish the Mg and Sr distribution coefficients of this ostracod. B. Map of the Gulf of Carpentaria showing the location of cores GC2 and GC10A and the extent of Lake Carpentaria when sea level dropped below - 53 m.

caused by evaporation) or even for values below 35‰ (achieved by dilution), remains approximately constant (Kinsman, 1969), Sr/Ca values of *Cyprideis* shells grown in these

waters also should be constant. The upper salinity tolerance of *Cyprideis* is ~100‰ (De Deckker, 1981). Consequently, Sr/Ca values of *Cyprideis* shells near 0.0041 indicate that the

ostracod grew in water connected to the sea (e.g. in an estuary or even the sea itself), or in water with an oceanic Sr/Ca ratio, but provide no information about water salinity. From our observations made at two sites in Australia (a pool on the edge of the Gulf of Carpentaria near Weipa, probably affected by sea spray, and a pool at the edge of Hutt Lagoon in Western Australia, probably in contact with a marine-derived brine) (see Fig.1), and also from an experiment designed to grow *Cyprideis* in ocean water (water collected at Robe in South Australia), we determined that the Sr/Ca of the ostracod shell is independent of salinity (Table I).

With a Sr/Ca value for *Cyprideis* close to 0.0041 and with a Mg/Ca value in the vicinity of 0.0229 for the same shell, it would be obvious that the *Cyprideis* grew in normal seawater. This water could not have been greatly evaporated nor diluted otherwise its Mg/Ca would have departed from the commonly accepted value of 5 ± 0.8 for average seawater. In the case of evaporation, depletion of Ca would occur due to CaCO_3 precipitation, and in the case of dilution, supply of Ca would mostly occur via fresher water. However, if the Mg/Ca ratio of the water calculated from measurements of *Cyprideis* shells gave a value close to 1, it would be possible to conclude, with some confidence, that the waters in which the ostracods grew were fresh. This conclusion follows from field measurements for non-marine waters of all possible salinities which indi-

cate that as soon as water becomes slightly saline ($> 3\text{‰}$) the Mg/Ca deviates from unity (Müller et al., 1972; Eugster and Hardie, 1978; De Deckker, Chivas and Shelley, unpublished data). Note also that fresh waters can also have Mg/Ca values different from 1.0.

Gulf of Carpentaria

The Gulf of Carpentaria is a shallow epicontinental sea situated between Australia and New Guinea (Fig.1). Its maximum depth is 69 m. Bathymetric studies (Nix and Kalma, 1972; Torgersen et al., 1983) indicate that it is connected to the Arafura Sea to the west by a -53 m sill. When sea level dropped -53 m below the present level, the Gulf became a closed depression called Lake Carpentaria by Torgersen et al. (1983) (see Fig.1b). Smart (1977) and Torgersen et al. (1983) described the Late Quaternary physiography of the Gulf/Lake Carpentaria and Torgersen et al. (1985) discussed its hydrological changes during the last 40,000 years. Five basic phases (Units I-V) (see Fig.3) based on facies, lithology and faunal assemblages have been recognized (Torgersen et al., 1985).

Cores

Two cores (GC2, GC10A) 60 km apart collected in 67 m of water (Fig.1) were selected for ostracod analyses for information on water salinity when the sea level dropped below

TABLE I

Distribution coefficient, $K_D[\text{Sr}]$, for *Cyprideis* from three environments with various salinities and ocean-water conservative chemistry

Locality	Salinity (‰)	(Sr/Ca) water	(Sr/Ca) shell	(n)	$\frac{(\text{Sr/Ca})_{\text{shell}}}{(\text{Sr/Ca})_{\text{water}}}$
Pool near Weipa, Queensland	17.6	0.0081	0.00412 ± 0.00019	(10)	0.509
Pool-seep near Hutt Lagoon, Western Australia	49.1	0.0082	0.00413 ± 0.00009	(8)	0.504
Experiment using ocean water from Robe, South Australia	40.7	0.0080	0.00394	(2)	0.493

Analyses are presented as atomic ratios; errors are 1σ , (n) is number of valves analysed. The township of Robe is located near Little Dip Lake (Fig.1.A).

–53 m and to duplicate the data to confirm that the ostracod shell composition controlled by water chemistry at the time of shell formation is not modified by variable and local diagenesis.

Material preparation and analytical procedure

Ostracod valves, selected from core samples, were treated with 10% H_2O_2 , washed with distilled water over a 60 μm sieve and dried at room temperature. Care was taken to use only very clean, single ostracod valves in the range 20–70 μg . Individual valves were transferred into 5 ml of 2% HCl in 25 ml scintillation vials in batches of about 50 and allowed to stand for at least 24 h before analysis. The solutions were analysed for Ca, Mg and Sr with a very high-resolution Inductively-Coupled Argon-Plasma Emission Spectrometer (ICPAES) (Shelley and Taylor, 1981) with limits of detection of Ca^{2+} 0.02 ppb, Mg^{2+} 0.04 ppb, and Sr^{2+} 0.03 ppb in solution. Merck Suprapur HCl acid, Millipore MQ water and SPEX Hipure standard solutions were used. Solutions of giant clam (*Tridacna gigas*) aragonite shell (ANU-P3) provided a comparative reference for each batch analysed, and confirmed a precision and accuracy of better than 2%.

Results

Figure 2 shows the range of Mg/Ca and Sr/Ca values from analyses of single *Cyprideis* valves from cores GC2 and GC10A. Since the rates of sedimentation appear different in the two cores (GC2 is 2.2 m long and GC10A is 1.4 m long), the represented length of each of the 5 sedimentary facies (Units I–V) for GC10A has been proportionately adjusted in Fig.3 to match those in GC2 to facilitate visual comparison of Mg/Ca and Sr/Ca data points. However, it is important to note that corresponding Units I–V in the two cores are not necessarily chronologically equivalent.

Figure 3 shows plots of the mean Mg/Ca and Sr/Ca values obtained from each stratigraphic horizon in both cores. Inferred water Mg/Ca

and Sr/Ca ratios from ostracod Mg/Ca and Sr/Ca values respectively are based on the previously calculated K_D numbers for *Cyprideis*. The distribution in both cores of the fossil biota used to reconstruct the palaeoenvironmental history of Carpentaria is presented in Fig.2.

Figure 3 provides a brief palaeoenvironmental interpretation for the last 40,000 years of Carpentaria which is expanded below. This reconstruction could not have been derived only with knowledge of the salinity tolerance of living equivalents of the fossil biota (namely ostracods and foraminifers) and is mainly based on information gained from ostracod shell chemistry. The interpretation is supplemented by "traditional" palaeoecological interpretation of the fossil data. However, anoxic events near the water–sediment interface could only have been inferred from the presence of dissolution marks, preferentially occurring in the sieve pore areas of *Cyprideis* specimens (De Deckker, 1988), and accretions of pyrite crystals on some of the benthic ostracods, including *Cyprideis*.

During the deposition of Unit V, and most of Unit IV, Carpentaria was a lake. Freshwater conditions are postulated where the water Mg/Ca values were close to unity. At other times, the water was slightly saline. The Sr/Ca values for the water, being close to seawater composition at the beginning of the lacustrine phase (GC10A in Fig.3 — no adequately well-preserved *Cyprideis* specimens occur in Unit V in GC2) indicate that the lake water, although very dilute, contained solutes of marine derivation. Where the range of Mg/Ca values in individual horizons is narrow (Fig.2), a substantial water depth is postulated that would minimize temperature variations. A change of water composition (perhaps caused by $CaCO_3$ precipitation) and perhaps a salinity increase, accompanied by shallowing of the lake is postulated for levels 185–175 cm (true depth in GC2 and its facies equivalent in GC10A). A return to freshwater conditions and Sr/Ca values of sea water derivation is detected in both cores before the end of this phase. Note

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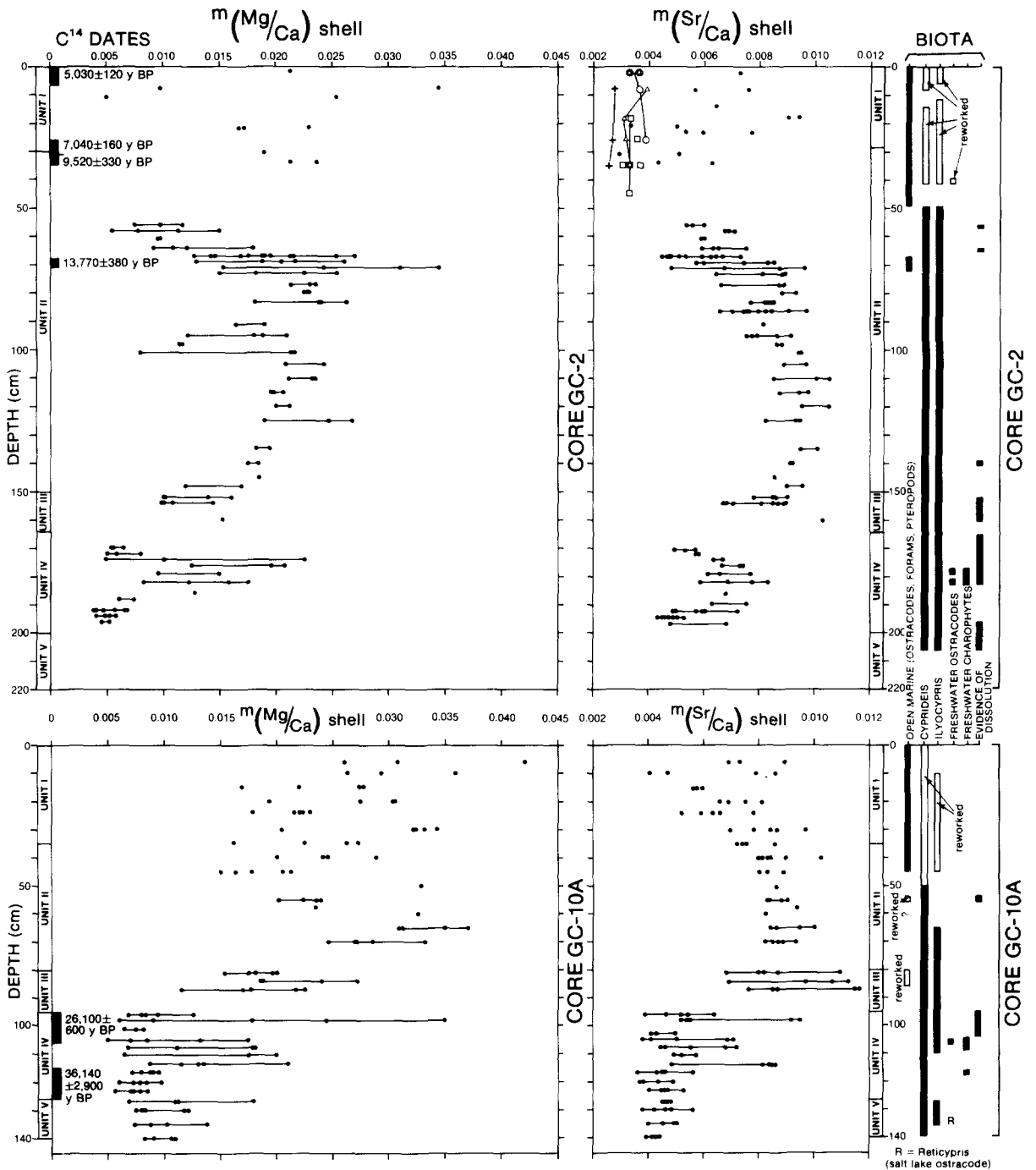


Fig. 2. Mg/Ca and Sr/Ca values of individual valves of *Cyprideis* ostracods (●) and open-marine ostracods (+ = *Cytherella*, ○ = *Bicornucythere*, △ = *Pterygocythereis*, □ = bairdiid) for 48 horizons in core GC2 and for 32 horizons in core GC10A. The horizontal bars represent the range of values obtained for only non-reworked *Cyprideis* specimens. On the right, the distribution of the biota indicative of different palaeoenvironments is presented; evidence of partial dissolution of ostracod specimens (inferring anoxia) is also indicated.

that the site of core GC2 nearly always had more dilute waters than the GC10A site. We suggest that substantial freshwater flowed into the lake from the north, or that site GC2 received more rain.

Unit III and the upper part of Unit IV are characterized by authigenic calcite laminations formed under episodic anoxic conditions and probably under shallow water (as indicated by the ostracods; Fig.3). The substantial amount of calcite precipitation caused a relative enrichment of Sr and Mg in the lake water.

After this event, the lake waters remained enriched in Sr and Mg, as recorded by the twofold increase in Sr/Ca and a twofold to threefold increase in the Mg/Ca ratio of *Cyprideis*. Part of the observed increase in Mg/Ca may have been due to an increase in temperature. Nevertheless, these Mg/Ca ratios obtained in the lower portion of Unit II indicate fresh or slightly saline water conditions, allowing for the effect of the 2 to 3-fold increase previously caused by the Mg enrichment.

The consistent Sr/Ca values in Unit II up to and near 80 cm indicate no further enrichment of Sr, hence no substantial calcite precipitation. The Mg/Ca fluctuations within the same portion of Unit II therefore result from salinity and/or temperature changes. The continuous difference between the Mg/Ca values in GC2 and GC10A suggests that site GC10A was still more saline than site GC2.

Above ≈ 80 cm, a connection to seawater is postulated at site GC10A based on the presence of an open marine fauna. In addition, for both sites, *Cyprideis* Mg/Ca ratios are within the range of seawater values (no great precision can be expected because of the additional possible temperature effect on Mg/Ca ratios).

Between levels 65 and 55 cm in core GC2, a

definite dilution of the water, for it to become close to fresh, is recorded. This suggests that this site was once again part of a lake (caused by a minor regression), or that it was a significantly diluted estuary.

Finally, after that phase, Carpentaria was transgressed by the sea as indicated by the presence of pteropods and truly marine genera of ostracods and foraminifers. *Cyprideis* is still present in both cores and there is evidence of its having been reworked; some shells are cracked, others broken or worn. Chemical analyses of these ostracod valves confirm the probability of diverse origins for these *Cyprideis* specimens by the broad range of Mg/Ca and Sr/Ca values. It is possible that *Cyprideis* specimens were reworked from the shallow margins of the Gulf during this phase, since population densities for one species, *Cyprideis torosa*, can be extremely high (up to 1.8 million individuals m^{-2} (Heip, 1976; Herman et al., 1983)). Note that for the upper 45 cm of core GC2, the Sr/Ca values for open marine ostracods (Fig.2) are fairly constant, showing they are not reworked from environments with varying compositions, and that open marine conditions prevailed. Note also that three of the open-marine genera (Fig.2) have similar K_D [Sr], although different from *Cyprideis*, whereas the fourth (*Cytherella*) appears to have a lower K_D [Sr].

Discussion

The study of Mg/Ca and Sr/Ca of the shells of the euryhaline *Cyprideis* from the Gulf of Carpentaria illustrates how palaeoenvironments, including (in some cases) palaeosalinities, can be determined. Where Mg/Ca and Sr/Ca values cover a broad range for individuals from a single sedimentary horizon,

Fig.3. Plots of the mean values of Mg/Ca and Sr/Ca data points presented in Fig.2 for individual stratigraphic horizons, and only for non-reworked *Cyprideis* specimens. The thickness of each facies in GC10A has been proportionately adjusted to allow direct lithological correlation with GC2. The column on the right indicates environmental interpretation based on the ostracod shell chemistry data, and the biota. From the K_D values of modern *Cyprideis*, the inferred Mg/Ca and Sr/Ca ratios of the waters of Carpentaria in which the ostracods lived are indicated by the lower horizontal scale.

either a broad range of salinities or chemical changes can be postulated, or it indicates that specimens have been mixed or reworked from several environments. Where Mg/Ca values cover a broad range and Sr/Ca values a narrow one, temperature fluctuations can be postulated. Where ranges for both Mg/Ca and Sr/Ca values are narrow, salinity and temperature fluctuations are small, implying a substantial depth of water above the benthic ostracods.

At present we cannot provide accurate salinity values based on ostracod shell chemistry, especially from Mg/Ca ratios because the uptake of Mg is controlled by both the water Mg/Ca ratio and temperature. Perhaps by combining chemical analyses of ostracod shells and of authigenic, non-biogenic carbonates within the same samples, it might be possible to obtain a better definition of the aquatic environments under which the carbonates (ostracods and others) were formed. In the present study, we have been able to provide a good estimate of the water salinity, but once the temperature dependence of Mg uptake in *Cyprideis* is known, we should be able to refine our palaeoenvironmental interpretation of Lake Carpentaria.

A micropalaeontologist examining the fossil biota recovered in cores from the Gulf of Carpentaria would be unable to provide information on past salinities and temperatures for when *Cyprideis* and *Ammonia* lived. These organisms are characteristically found over a broad range of salinities. Hence trace-element analyses of these organisms can provide a better definition of the aquatic environment. With knowledge of the life cycles of any ostracod species it would become possible to establish seasonal variations by analysing separately juvenile and adult forms, which moult during different periods of the year.

The reconstruction of palaeoenvironments by analysing Mg/Ca and Sr/Ca of ostracod shells has wide applications once the K_D of various species is established. The K_D for extinct species could be calculated for those extinct genera that are found accompanying extant representatives with known K_D . This

technique therefore has potential to solve some of the long-standing controversies on the nature (e.g. marine vs. non-marine) of some ancient environments (e.g. the Mesozoic Purbeck/Wealden Beds in the British Isles), provided the ostracod shells have not suffered diagenetic effects, and should be very informative on marine transgressions/regressions in general and on sea-level changes. Similarly, a combination of Mg/Ca and Sr/Ca analyses of fossil *Cyprideis* should provide precise information on the hydrochemical evolution of sites such as the Azov and Caspian Seas, where today several hundred thousand living specimens m^{-2} occur (Caspers, 1957 and Zenkevitch, 1957, respectively), and other Paratethyan sites and especially the Mediterranean during its "Messinian salinity crisis" (when *Cyprideis* thrived; see Benson 1975, 1976; De Deckker et al., in press).

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