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ABSTRACT

The partition coefficients ($K_{\rm D}$) guiding the uptake of Mg and Sr in the low-Mg calcite valves of ostracodes in Australian saline lakes have been reviewed in line with new information on the bicarbonate content and the $\alpha Mg/\alpha Ca$ of the ambient waters. Strongly alkaline and very high $\alpha Mg/\alpha Ca$ (>>20) waters cause the partition coefficients of ostracodes to differ significantly from those found in other lakes with lower $\alpha Mg/\alpha Ca$ values. Water salinity does not affect the uptake of Mg or Sr in ostracode valves, whereas the αHCO_3^- of the water strongly correlates to $K_{\rm D}[{\rm Sr}]$, with high αHCO_3^- resulting in a significant $K_{\rm D}[{\rm Sr}]$ decrease.

INTRODUCTION

Trace metal substitution of Ca in the calcium carbonate system as proxies for reconstructing paleoenvironmental variables has been used wherever biogenic calcium carbonate is preserved. This technique has been used in studies of marine fossils to determine changes in temperature, salinity, terrestrial input, and many other factors, and has been utilized for a great number of calcium carbonate-secreting organisms, including foraminifera (for reviews see Eggins et al., 2003; Lea, 2003; Cusack and Freer, 2008), coccolithophores, molluscs, brachiopods (Cusack and Freer, 2008), corals (for reviews see Lea, 2003; Corrège, 2006; Cusack and Freer, 2008), fish otoliths (Bath et al., 2000), ostracodes (Corrège and De Deckker, 1997; Dwyer et al., 2002; for a review see Lea, 2003), and others. Trace metal substitution for calcium in calcite has been utilized in lacustrine environments using ostracodes (for reviews see Holmes and Chivas, 2002; Ito et al., 2003).

The uptake of bivalent cations replacing some Ca atoms during the calcification process in ostracodes is only beginning to be understood. The pioneering work of Turpen and Angell (1971) showed, using ⁴⁵Ca as a tracer, that ostracodes obtain Ca for their valves directly from the ambient water, and do not utilize Ca from old (molted) valve material. Following this, Chivas et al. (1983, 1985, 1986a, 1986b) and De Deckker et al. (1988) developed the use of trace metal substitution of Ca in the calcitic lattice as a tool for determining environmental change from limnic ostracodes.

Chivas et al. (1986b) showed that Mg incorporation into ostracode valves is primarily driven by the Mg/Ca_{water} and ambient water temperature, and that Sr incorporation is only related to the Sr/Ca_{water}. However, De Deckker et al. (1999) showed that there may be a slight temperature effect guiding the uptake of Sr in ostracode valves, and concluded that water salinity is not the controlling factor in trace element uptake.

Assuming that ostracode calcite forms in thermodynamic equilibrium from a large volume of solution containing low concentrations of trace metals (Katz et al., 1972), then trace metal (Mg, Sr) incorporation at a given temperature (*T*) is dictated by the partition coefficient (K_D) between the molar trace metal ([Me] = any divalent cation [Mg, Sr]) to Ca ratio (Me/Ca) of the water and the valves (Chivas et al., 1986a). However, in highly saline water, the ions bond temporarily, forming ion complexes that are not available for carbonate production (Hem, 1985). Consequently, it is assumed that the true concentration of ions available is the concentration of the active ions in the solution (activities, α), and follows the relationship

$$K_{\rm D} [Me]_{\rm T} = (Me/Ca)_{\rm ostracodevalve} / (\alpha Me/\alpha Ca)_{\rm water}.$$
 (1)

Several $K_D[Mg]$ and $K_D[Sr]$ have been determined for several lacustrine, estuarine, and marine ostracode species (for summary, see Holmes and Chivas, 2002), but only the thermodependence with respect to Mg was established for one species, *Cyprideis australiensis* (De Deckker et al., 1999). Other cations have been used variously to reconstruct environmental variables such as Fe and Mn (see Holmes, 1998) and U (see Ricketts et al., 2001).

Chivas et al. (1986a) proposed that cogeneric species have identical $K_{\rm D}[{\rm Sr}]$, and that closely related genera, such as *Australocypris* and *Mytilocypris* from Australian lacustrine environments, share a similar $K_{\rm D}[{\rm Sr}] = 0.208$; however, they found that $K_{\rm D}[{\rm Sr}]$ for *Australocypris robusta* in Lake Keilambete, a volcanic maar lake in western Victoria, has a very different $K_{\rm D}[{\rm Sr}] = 0.082$ (Chivas et al., 1985).

Wansard (1996) and Wansard et al. (1998) showed that *Cyprideis* torosa from European lakes have very different $K_D[Sr]$ with a high degree of variation (0.602–0.83) from the cogeneric *C. australiensis* ($K_D[Sr] =$ 0.475) from Australian waters (De Deckker et al., 1988). De Deckker et al. (1999) showed some variation in the $K_D[Sr]$ of *C. australiensis* ($K_D[Sr] =$ 0.438) from their previous work (Chivas et al., 1986a). Wansard et al. (1999) showed that in *Herpetocypris intermedia*, a freshwater ostracode, the D(Mg) and D(Sr) (D[Me] implies a constant solution composition during mineral precipitation) varied with respect to the Mg/Ca of the water. Ito and Forester (2009) urged caution in using ostracode partition coefficients in paleoenvironmental reconstructions, stating a range of hydrological and physiological issues that affect the uptake of both Mg and Sr in ostracode valves.

Our investigation seeks to clarify the Chivas et al. (1985, 1986a) paradox between invariant congeneric Australian lacustrine ostracode partition coefficients and the very different partition coefficients of conspecifics in terms of hydrochemical variability. We reanalyze the Chivas et al. (1985, 1986a) partition coefficients from southeastern Australia (SEA) in light of new data from southwestern Western Australia (SWA) lakes.

METHODS

Because the methods presented in this paper are standard procedures, the methods are presented in the GSA Data Repository.¹

RESULTS

The $K_D[Mg]$ and $K_D[Sr]$ for the ostracode species belonging to the nektonic *Australocypris*, *Mytilocypris*, *Diacypris*, and *Reticypris* collected from SEA and SWA lakes are presented in Table DR1 in the Data Repository.

For all *Australocypris* and *Diacypris* species collected from SWA and SEA lakes (Table DR1), the $K_D[Mg]$ from the SWA lakes is larger, as is the variation in $K_D[Mg]$. Most *Mytilocypris* species, however, have similar $K_D[Mg]$ values and variation in both SEA and SWA lakes (Table DR1), with the exception of *M. mytiloides*. The $K_D[Mg]$ values of *Reticypris* spe-

¹GSA Data Repository item 2010091, a description of the sites sampled and the methods used, and Table DR1 (the Kd[Mg] and Kd[Sr] determined from a number of nektonic ostracode species from southeastern and southwestern Australian lakes), is available online at www.geosociety.org/pubs/ft2010.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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cies do not vary at all between regions, and have similar means and variances (Table DR1).

Determination of species $K_D[Sr]$ for SEA and SWA demonstrates that there is a regional difference in the $K_D[Sr]$ between *A. insularis* collections (Table DR1). Little variability occurs between Western Australia species of *Mytilocypris*, although a high variability occurs within species from SEA lakes (Table DR1). The overall means of $K_D[Sr]$ for all *Mytilocypris* species in the two regions are very similar (SEA $K_D[Sr] = 0.3045 \pm 0.2309$ and SWA $K_D[Sr] = 0.2924 \pm 0.113$).

The $K_D[Sr]$ values of *Diacypris* species determined from SWA lakes are systematically higher than those from SEA lakes (Table DR1). Individual species comparisons show that *D. spinosa* differs significantly between regions; however, there is no significant regional difference in *D. compacta* $K_D[Sr]$ (Table DR1). The reverse is true of the *Reticypris* species analyzed between the two regions, the specimens analyzed from SEA having systematically, although not significantly, higher $K_D[Sr]$ values than the SWA species (Table DR1).

DISCUSSION

The variation in $K_D[Mg]$ and $K_D[Sr]$ suggests that the mechanism of trace metal incorporation in ostracodal calcite may vary between SEA and SWA. Several authors (Xia et al., 1997, for *Candona rawsoni*; Wansard et al., 1998, for several other *Candona* species and *Cyprideis torosa*; Wansard et al., 1999, for *Herpetocypris intermedia*) found that the $K_D[Mg]$ and $K_D[Sr]$ varied with water chemistry. Xia et al. (1997) found that Mg/Ca_{valve} of *C. rawsoni* varied most with respect to temperature and that $K_D[Sr]$ increased with higher Sr/Ca_{water} and Mg/Ca_{valve}. Wansard et al. (1998),

however, showed that $K_D[Mg]$ decreased as Mg/Ca_{water} increased for three *Candona* species, and that the $K_D[Mg]$ for *Cyprideis torosa* differed significantly from De Deckker et al.'s (1999) work on *Cyprideis australiensis*. Wansard et al. (1999) also showed that the $K_D[Mg]$ of *Herpetocypris intermedia* (a freshwater species) was constant as Mg/Ca_{water} increased above 1, and that the variability and value of $K_D[Mg]$ increased when Mg/Ca_{water} decreased. De Deckker et al. (1999) clearly showed that the $K_D[Mg]$ and $K_D[Sr]$ of *C. australiensis* did not vary as a function of salinity.

Chivas et al. (1985, 1986a) suggested that the unique water chemistry of Lake Keilambete in SEA with high salinity (ranging from 71.5 to 74 g/L), high Mg/Ca_{water} (89), high HCO₃⁻ (2180 meq/L), and negligible SO₄²⁻ may be related to the lower K_D[Sr] of *A. robusta* found in this lake (0.082 \pm 0.0051) compared with the K_D[Sr] of *Australocypris* and *Mytilocypris* species found in other SEA lakes (0.208 \pm 0.048).

Figure 1 shows the variation in water chemical parameters for both SWA and SEA lakes, with very little variation in α Mg/ α Ca_{water} (Figs. 1A and 1B) and α Sr/ α Ca_{water} (Figs. 1C and 1D), with salinity for both regions. Similarly, comparison of Mg/Ca_{valve} (Figs. 1E and 1F) and Sr/Ca_{valve} (Figs. 1G and 1H) with salinity shows no significant relationship. This confirms that, for Australian lacustrine ostracodes, De Deckker et al.'s (1999) suggestion for *C. australiensis* that Sr/Ca_{valve} (Figs. 1K and 1L); similarly, Mg/Ca_{valve} is not a function of α Sr/ α Ca_{water} (Figs. 1K and 1L); similarly, Mg/Ca_{valve} is not a function of salinity, but is weakly a function of α Mg/ α Ca_{water} (Figs. 1I and 1J) and the high variability is due to the temperature effect on Mg incorporation (Chivas et al., 1986a, 1986b).

As mentioned previously, however, the $K_D[Mg]$ and $K_D[Sr]$ of ostracodes vary in both value and amplitude for both species and genera

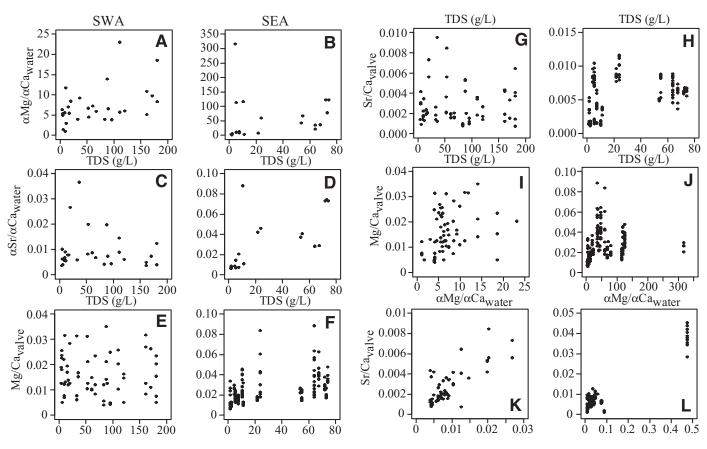


Figure 1. Plots showing various water chemistry and ostracode shell chemistry relationships. Southwestern Western Australia (SWA) lakes are on left, southeastern Australia (SEA) lakes are on right. A, B: α Mg/ α Ca_{water} vs. TDS (g/L) (TDS—total dissolved solids equivalent to salinity). C, D: α Sr/ α Ca_{water} vs. TDS (g/L), E, F: Ostracode Mg/Ca_{valve} vs. TDS (g/L). G, H: Ostracode Sr/Ca_{valve} vs. TDS (g/L). I, J: Ostracode Mg/Ca_{valve} vs. α Mg/ α Ca_{water}. K, L: Ostracode Sr/Ca_{valve} vs. α Sr/ α Ca_{water}.

between SWA and SEA. Figure 2 shows combined $K_D[Mg]$ and $K_D[Sr]$ for both SWA and SEA versus salinity (g/L), $\alpha Mg/\alpha Ca_{water}$, and αHCO_3^- (Figs. 2C–2F). Both $K_D[Mg]$ and $K_D[Sr]$ for all Australian lacustrine species examined here show a weak decrease at first as salinity increases (Figs. 2A and 2B). At low salinity values (<20 g/L), the variability in partition coefficients is high (Figs. 2A and 2B), but then values are scattered. The variability in $K_D[Mg]$ and $K_D[Sr]$ is relatively uniform above a salinity of 20 g/L. This suggests that uniform $K_D[Mg]$ and $K_D[Sr]$ can be applied to reconstruct paleoenvironmental variables from ostracode valve chemistry (Chivas et al., 1986a). However, the values and variability of $K_D[Mg]$ decrease as $\alpha Mg/\alpha Ca_{water}$ increases (Fig. 2C).

The decrease in $K_D[Mg]$ with increasing $\alpha Mg/\alpha Ca_{water}$ suggests that the physiological mechanism for secreting low-Mg calcite in Australian lacustrine ostracodes is a tradeoff between the physiological control of Mg uptake in high $\alpha Mg/\alpha Ca_{water}$ and the thermodependence on the Mg uptake in low $\alpha Mg/\alpha Ca_{water}$. This implies that the influence of temperature on Mg uptake diminishes as $\alpha Mg/\alpha Ca_{water}$ increases, and is replaced by physiological control. A similar result has been found for the freshwater species *Herpetocypris intermedia* in low (<4) Mg/Ca_{water} cultures (Wansard et al., 1999) and *Candona* species in a range of Mg/Ca_{water} (<15) (Wansard et al., 1998).

The value and variability of $K_{\rm D}[\rm Sr]$ also decrease with increased $\alpha Mg/\alpha Ca_{\rm water}$ as $\alpha Mg/\alpha Ca_{\rm water}$ approaches 110 (Fig. 2D). In the two lakes with $\alpha Mg/\alpha Ca_{\rm water} > 110$ (Lake Purdigulac $\alpha Mg/\alpha Ca_{\rm water} = 115$; Lake Coragulac $\alpha Mg/\alpha Ca_{\rm water} = 316$, both in western Victoria, SEA), both of which

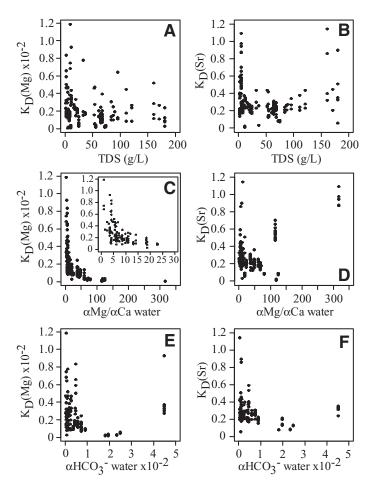


Figure 2. Plots of combined southeastern Australia and southwestern Western Australia data showing relationship between ostracode K_p[Mg] (on left) and K_p[Sr] (on right). A, B: Against TDS (total dissolved solids equivalent to salinity, g/L). C, D: Against α Mg/ α Ca_{water}. (Inset in C enlarges 0–30 α Mg/ α Ca_{water} range). E, F: Against α HCO₃⁻ water.

have highly evolved waters, the $K_D[Sr]$ increases substantially ($K_D[Sr] = 0.566 \pm 0.06$ for Lake Purdigulac and $K_D[Sr] = 0.956 \pm 0.092$ for Lake Coragulac). This suggests that $\alpha Mg/\alpha Ca_{water}$ is not the primary controlling factor in the variability in $K_D[Sr]$. Wansard et al. (1998) examined $K_D[Sr]$ variability with respect to Mg/Ca_{water} and found no systematic variation; however, they only analyzed waters with Mg/Ca < 15. Australian lacustrine ostracodes show a similar non-relationship for $\alpha Mg/\alpha Ca_{water} < 15$.

Figures 2E and 2F show that with increasing α HCO₃⁻, the variability in K_D[Mg] and K_D[Sr] decreases. SWA analyses are all from waters with a low α HCO₃⁻ and display very high variability, whereas SEA analyses vary from low to high α HCO₃⁻ and the variability decreases with increasing α HCO₃⁻. Exceptions to this are the analyses of K_D[Mg] and K_D[Sr] of *M*. *splendida* in very shallow Lesser Twin Lake in SEA (α HCO₃⁻ = 0.045) that show a high degree of variability. East Basin Lake and Lake Keilambete in SEA also recorded high α HCO₃⁻ concentrations on two of the three sampling occasions (East Basin Lake = 0.0195 and 0.0247; Lake Keilambete = 0.018 and 0.0232), and the K_D[Sr] variability was low in both lakes. This suggests that if the K_D[Sr] analyses of Lesser Twin Lake are ignored, the α HCO₃⁻ water appears to influence the K_D[Sr]. Lesser Twin Lake had only 10 cm of water at the time of collection and the water's ionic ratio may have varied between the time of ostracode calcification and water and ostracode collection.

Mechanism for HCO₃-Induced Partition Coefficient Variability

A mechanism behind the systematic variation in K_{p} [Me] with α HCO₂ water documented here was presented by Paquette and Reeder (1995), who described a model of calcite growth in which crystal growth occurs on the $\{10\overline{1}4\}$ face by the spiral mechanism. The spiral mechanism causes a microtopographic relief with randomly distributed pyramidal hillocks that comprise two characteristically similar pairs (a and b, a' and b') of vicinal faces that vary slightly in orientation ($\leq 1^{\circ}$) from the $\{10\overline{1}4\}$ face; Paquette and Reeder (1995) found that Mg enrichment always occurred on the a' and b' vicinal faces, and Sr enrichment on the a and b vicinal faces. This enrichment in trace metals of each vicinal face is related to kink structures that are caused by the Ca^{2+} and CO_{3}^{2-} coordination in the crystal structure. Paquette and Reeder (1995) also found that the degree of supersaturation of calcite in the precipitating solution (ranging from 8 to 20 times calcite saturation) and, more important, the $\alpha Ca^{2+}:\alpha CO_3^{2-}$ of the precipitating solution (ranging from 100 to 30,000 in their experiments) influenced the shape of these hillocks, with increasing αCO_3^{2-} in the precipitating solution resulting in less differentiation between the pairs of vicinal faces. The αCO_3^{2-} increase in the solution also caused a decrease in the relative growth of a' and b' vicinal faces, thus reducing the uptake of Sr. All of the Paquette and Reeder (1995) experiments were conducted at 28 ± 2 °C, and so comment on the temperature influences of Mg uptake into the a' and b' vicinal faces cannot be made.

This microstructural analysis of trace metal incorporation in calcite has fundamental implications for ostracodal $K_D[Me]$ in lacustrine systems in SEA lacustrine systems. In pH-buffered lacustrine systems with elevated αHCO_3^- , such as SEA lakes (Radke et al., 2003), increases in αCO_3^{2-} are expected, and as Paquette and Reeder (1995) found, elevated αCO_3^{2-} suppresses trace metal, in particular Sr, incorporation into the calcitic lattice during crystal growth. The implication for ostracodal $K_D[Me]$ is that as αHCO_3^- increases, trace metal incorporation into ostracode calcitic valves will be suppressed, regardless of the $\alpha Me/\alpha Ca$ water. This results in decreased $K_D[Me]$ in HCO_3^- -enriched waters.

The biomineralization of several ostracode species was investigated by Keyser and Walter (2004); they showed that calcium mobilization from inside epidermal cells into the calcite valves changed morphology and that calcium underwent at least one chemical transformation, from calcium phosphate to calcium carbonate. The microenvironment in which these chemical and morphological transformations occur must be the result of subtle chemical variations. Comprehension on how ostracodes secrete their valves and the biophysical and biochemical changes during this calcification is still in its infancy and more research is required before a holistic system-based understanding is gained.

CONCLUSION

The $K_D[Mg]$ and $K_D[Sr]$ of several Australian saline lacustrine ostracode species are shown to vary between southeast Australia and southwest Australia, both in terms of mean values and in the degree of variability. As the $\alpha Mg/\alpha Ca_{water}$ increases above ~50, the Mg uptake by ostracodes has a reduced thermodependence, and so $K_D[Mg]$ can be utilized as an indicator of $\alpha Mg/\alpha Ca_{water}$. When $\alpha Mg/\alpha Ca_{water} < 50$, $K_D[Mg]$ depends both on $\alpha Mg/\alpha Ca_{water}$ and temperature at the time of calcification. In low αHCO_3^- waters, the $K_D[Sr]$ is higher and more variable than in waters with high αHCO_3^- . This suggests that at low αHCO_3^- water, the $K_D[Sr]$ does not accurately reflect the $\alpha Sr/\alpha Ca_{water}^-$. At the opposite, at greater $\alpha HCO_3^$ water, the $K_D[Sr]$ variability decreases, suggesting that it can be used as an accurate indicator of $\alpha Sr/\alpha Ca_{water}^-$. Across all salinities, the $Sr/Ca_{valve}^$ is clearly a poor indicator of salinity in Australian lacustrine ostracodes.

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