Coincident Boron Profiles of Bivalves from the Gulf of California: Implications for the Calculation of Paleosalinities

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The constant relationship between seawater salinity and boron concentration suggests that boron can be used as a measure of salinity. Previous workers have suggested, on the basis of analyses of the boron content of shells belonging to the bivalved mollusc Mytilus edulis, *that salinity could be reconstructed from biomineral records of boron. Molluscan shells grow by incremental accretion and preserve within them geochemical records of their environments. Therefore, if boron concentration is being controlled by an external factor, the ontogenetic boron profiles of different shells, contemporaneous and sympatric, should be similar. This paper reports on the analysis of three shells belonging to two species of bivalves,* Chionopsis gnidia *and* Chione californiensis, *collected from the Gulf of California. Ontogenetic profiles constructed using a spectrophotometric method are quantitatively similar among the three specimens. The range of boron concentrations in each shell coincide with estimated ambient salinity variation, but also seems to be influenced by aperiodic regional rainfall, and the resulting influx of boron-poor freshwater. The similarity among the shells demonstrates that boron concentration is under external, environmental control, and directly reflects boron concentration in the surrounding water. This conclusion raises the possibility of reconstructing ambient boron concentrations from molluscan shells, and perhaps also reconstructing local salinity values. Further research is required to determine if boron is deposited in equilibrium with ambient concentrations, the nature of any dependence of incorporation on temperature, and, therefore, if paleosalinity reconstruction is possible.*

INTRODUCTION

Geochemical analysis of biogenic minerals has become one of the most powerful tools available for paleoecological reconstruction. An increasing number of approaches and organisms are being utilized for the estimation of such variables as marine paleotemperatures, pH, productivity, and salinity. The present paper reports on the use of boron concentrations in molluscan shells as a potential tool for the reconstruction of ancient salinities.

Procedures that have been used to calculate paleosalinities in fossils and ancient sediments include analyses of boron concentration (Schopf, 1980), stable oxygen and carbon isotopic ratios (see review in Dodd and Stanton, 1990; Ingram et al., 1996a, b), strontium isotopic ratios (Ingram and Sloan, 1992; Ingram and DePaolo, 1993), and magnesium and strontium to calcium ratios (Klein et al., 1997). The latter non-boron methods appear to be quite successful in regimes of substantial freshwater-seawater mixing (large salinity gradients), such as estuarine conditions, or when the relevant chemical composition of the waters are known or can be estimated. On the other hand, boron concentration in clay particles is directly proportional to ambient seawater salinity and has long been a standard method for measuring salinity in sedimentary environments (Couch, 1971; Schopf, 1980). There are, however, several problems with this approach. Analyses of sedimentary particles are subject to substantial error due to post-depositional alterations (Perry, 1972; Cook, 1977). They are measurements of "average" salinity and are insensitive to seasonal and other short-term temporal fluctuations of salinity, such as can occur in coastal marine environments. Knowledge of temporal variability is important to many of the purposes of paleoecological reconstruction, notably the occurrence of abrupt short-term changes (Klein et al., 1997). These problems would be resolved if boron concentration could be measured in a medium that incorporated boron in chemical equilibrium with the ambient environment, and did so in a fashion reflective of environmental variation in ambient concentration.

Molluscan shells grow by terminal accretion, and the growth of coastal species occurs typically in daily or subdaily increments, the frequency and thickness of which vary seasonally (Lutz and Rhoads, 1980; Tanabe, 1988; Ohno, 1989; Jones et al., 1990). Though the bulk of the shell mineral is calcium carbonate (either aragonite or calcite), other environmental elements are often incorporated (for example, boron, magnesium, strontium, and sulphur; Rosenberg and Jones, 1975; Furst et al., 1976; Lowenstam and Weiner, 1989) during mineralization. The resulting shell is an ontogenetic geochemical record of its own environment, a detail that is often utilized for the reconstruction of annual and seasonal environmental variation utilizing stable isotopic ratios (e.g., Bemis and Geary, 1996; Steuber, 1996; Klein et al., 1997; see Wefer and Berger, 1991 for a summary). If the chemicals are incorporated randomly, or incorporation is only partially controlled by the animal, then temporal variations in environmental concentration are captured by growth of the shell. Therefore, it is proposed in this paper that measurement of the boron concentration in molluscan shells may provide a method for calculating sub-annual salinity variation.

Furst et al. (1976), building on earlier research by Leutwein and Waskowiak (1962), demonstrated that shells of the blue mussel, *Mytilus edulis*, from the Baltic Sea contain concentrations of boron correlated to the salinity regimes of the waters in which they were mineralized. These authors analyzed both aragonite and calcite samples (both are present in *M. edulis* shells) using neutron activation and a subsequent count of α-particle tracks to determine boron concentration. They concluded that boron is enriched in aragonite relative to calcite (see also Kitano et al., 1978), and that its concentration approximates average annual salinity values but varies significantly within individual shells. As discussed above, detecting seasonal

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variation in the chemical composition of a shell is a direct consequence of the manner in which the shell carbonate is sampled. Furst et al.'s (1976) sampling was along a transverse, ontogenetic section of the shell. Although Furst et al. (1976) do not conclude this, we hypothesize that much of the individual variation observed by them could represent seasonal salinity fluctuations, which should be expected in a restricted, low-salinity sea such as the Baltic (annual averages range from 5 to 23 ppt). The variation of boron concentration would correspond to seasonal variation of salinity determined by, for example, river freshwater influx or salinity "events" (e.g., rainfall). Furst et al. (1976) interpreted the variation as fluctuations about a mean value (the average annual salinity), but it is possible that the variation is temporally based and is reflecting variation of environmental salinity.

Boron in Seawater

Validity of a boron-salinity relationship relies on the known behavior of boron in seawater. Boron is present in seawater in substantial concentrations as boric acid (0.027 g/kg; Schopf, 1980) and undergoes a dissociation similar to that of carbonic acid:

$$H_3BO_3 + 2H_2O \leftrightharpoons H_3O^- + B(OH)_4^-$$

The concentration of the borate anion is directly proportional to chlorinity (a proxy measure of salinity) as

$$C_{\Sigma B}(\text{moles/l})/\text{Cl}(\text{ppt}) = 2.1 \times 10^{-5}$$

where $C_{\Sigma B}$ = boron concentration, and Cl (ppt) = chlorinity (Riley and Chester, 1981; Millero and Sohn, 1992). The success of the method relies upon two criteria: (1) the shell's incorporation of boron is under environmental and not physiological or genetic control; and (2) boron is being incorporated in chemical equilibrium (or some factor thereof) with the surrounding seawater. The present paper tests the first criterion, reporting on the measurement of boron concentration in the valves of three molluscan bivalves from the Gulf of California, and discusses the potential for the development of a tool that measures historical environmental boron concentrations and, subsequently, paleosalinities.

We analyzed the ontogenetic boron concentrations of two species of venerid bivalves with aragonitic shells, *Chione californiensis* and *Chionopsis gnidia*. The specimens were collected within the same year, in the same region, and were presumably exposed to the same range of salinities. The premise is that if molluscan shells are recording ambient boron concentrations, then the boron profiles of contemporaneous and sympatric specimens should be similar. Moreover, these profiles will exhibit variation, and the variation will allow the correlation of profiles among different shells. This paper demonstrates that sympatric shells do indeed record similar boron profiles. Further research is required to determine the empirical nature of the relationship between salinity and shell boron concentration, if boron incorporation into aragonite is temperature dependent, and subsequently if boron concentrations in bivalve shells can be used to calculate salinity.

METHODS

Specimen Collection

Specimens were collected from two sites on opposite sides of the Gulf of California. One specimen of *Chione californiensis* was collected near Punta Concepcion (27°N, 112°W) in January 1990, and two specimens of *Chionopsis gnidia* were collected near Guaymas (28°N, 111°W) in September 1990. All specimens were collected near shore, alive, and in less than 6 m of water. The animals were sacrificed shortly after collection, and the shells cleaned without the use of chemicals.

Analysis of Boron Concentration

Chionine species are well known for their elaborate concentric sculpture (Roopnarine, 1996), and this was utilized to obtain samples of sufficient mass (>20 mg, see below) for analysis. Each concentric ornament on these species represents one discrete growth period of the shell, and samples extracted from anywhere along the length of a concentric element are, therefore, temporally homologous. The shell material composing concentric elements was removed sequentially with a low-speed drill beginning at the growing margin of the shell; sampling was stopped when elements had been removed completely.

The boron content of each sample was measured using a modification of the method presented by Lopez et al. (1993) for the measurement of boron concentrations in solution. In preparation for analysis, samples were pulverized, weighed (to the nearest mg), and dissolved in 5 ml 0.6N HCl. Next, 4 ml of a mercaptoacetic acid buffer solution and 2 ml of Azomethine-H dye were added, and the mixture reacted for 60 minutes. After 60 minutes, and again after 90 minutes, the absorbance of the mixture was measured spectrophotometrically versus a blank solution (distilled water plus reactants) at 410 nm. Boron concentration in the sample mixture was determined by comparison to a standard calibration curve constructed using the analysis of known concentrations of boric acid. The procedure is capable of measuring standard concentrations as low as 0.1 ppm, but it was discovered that at least 20 mg of shell carbonate are necessary for successful detection. This unfortunately prevented replicate analyses of single samples. After analysis, boron profiles were constructed for each shell by recording the level of boron measured in a particular sculptural ornament, as well as that ornament's maximum distance from the shell's umbo (direction of maximum growth).

An attempt was made to relate the resulting boron concentrations and profiles to temporal salinity variation in the Gulf of California, by utilizing data available in the Levitus and Boyer (1994) database. The data used represent regional monthly salinity variation derived from averages of the years 1900 to 1992 (see below). Actual salinity data for the collection sites and times are not available.

RESULTS AND DISCUSSION

Chione californiensis

Six samples were obtained from the single specimen of *Chione californiensis*. Sample masses, boron concentra-

TABLE 1—Data for each shell analyzed. Sample masses of carbonate were extracted from individual sculptural elements; note the lack of correlation between sample masses and boron concentration, and the similarity among average concentrations. Data are illustrated in Figures 1 and 2.

Species	Distance from margin (mm)	Sample mass (mg)	Boron concentration (ppm)
Chione californiensis	0	16	20.62
	1.26	39	14.23
	2.21	26	9.23
	3.15	20	18.75
	4.37	36	50.42
	6.35	22	29.32
Mean concentration			23.7
Chionopsis gnidia	0	36	13.47
	5.8	36	23.05
	16.36	74	16.69
	24.16	43	19.3
	34.64	21	54.76
	47.24	19	15.53
Mean concentration			23.8
Chionopsis gnidia	5.05	32	31.56
	15.26	161	12.22
	22.76	90	20.07
	29.27	85	27.81
	35.1	63	42.86
	42.73	33	16.09
Mean concentration			25.1

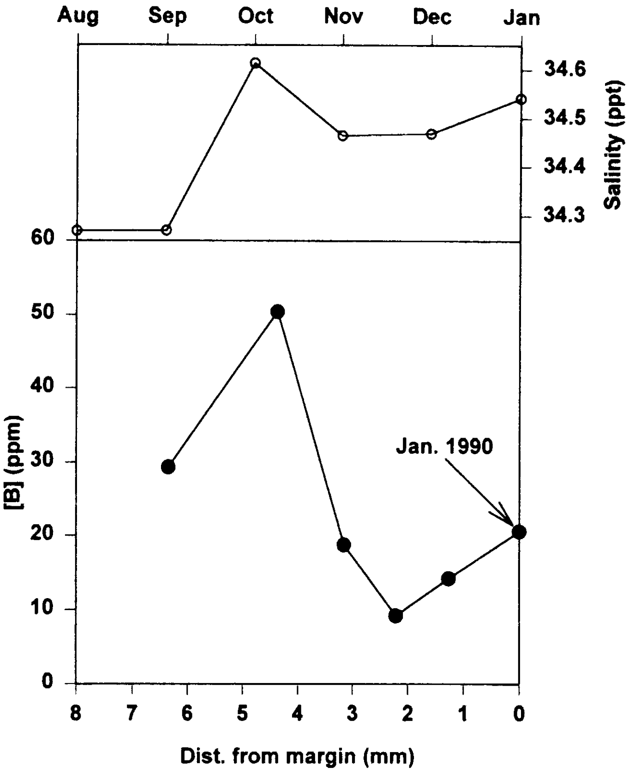

FIGURE 1—Boron profile for specimen of *Chione californiensis* from Punta Concepcion. The upper graph is an illustration of average annual salinity variation at this location (Levitus and Boyer, 1994). The lower graph plots boron concentration against distance from the growing margin of the shell. Note the maximum concentration of 50.42 ppm. The upper and lower curves were aligned by the month of specimen collection, linking maximum boron concentration to maximum salinity, and then spacing the months equidistantly.

tion, and distance from the growing margin are presented in Table 1. Note that there is no correlation between sample mass and boron concentration. Boron concentration ranges from 9.23 to 50.42 ppm, with an average of 23.76 ppm (Fig. 1). While this single profile by itself is not a sufficient test of the environmental control of boron incorporation, the profile is a single wavelength and could be representative of seasonal salinity cyclicity at Punta Concepcion . Moreover, the range of the values reflects substantial variation of the boron incorporation rate of the shell. The boron concentrations observed are likely to be underestimates of actual local maxima and minima because of the low sampling resolution.

Chione gnidia

Six samples were also obtained from each specimen of *Chione gnidia*. Sample masses ranged from 19 to 90 mg and, again, there is no correlation between sample mass and boron concentration. Boron concentrations range from 13.47 to 54.76 ppm in one shell and from 12.22 to 42.86 ppm in the other. While the ranges are similar for both shells, more striking is the comparison of their profiles (Figs. 1 and 2). Both shells are aligned at the ventral margin, where shell material was last deposited before death. If roughly equivalent growth rates are assumed, which is not an unreasonable assumption for two conspecific bivalves inhabiting the same locality, the implication is that both animals were incorporating similar amounts of boron at the same time. The maximum values, 54.76 and 42.86 ppm, coincide very well on the shell transects (34.64 and 35.10 mm from the margin, respectively). These data indicate that the animals were responding in similar fashion to an extraneous environmental variable.

Seasonal Profiles

The profile from the specimen of *Chione californiensis* also records a high maximum, in this case 50.42 ppm, a value well above the mean of 23.76 ppm (which, incidentally, is very close to the mean values of the *Chionopsis gnidia* specimens, 23.80 and 25.10 ppm). Bearing in mind the earlier death of this specimen compared to the *C. gnidia* specimens (January versus September), the measured profile probably ends somewhere just prior to the minima of the *C. gnidia* profiles. The growth rates of the two species are undoubtedly different, given the great disparity in maximum heights (for example, ~50 mm for *Chione californiensis* and 75 mm for *Chionopsis gnidia*; Roopnarine, 1996), so an exact correspondence between the distance axes should not be expected. Nevertheless, the extreme maximum of all three profiles acts as a marker at which to compare the profiles. The similarity of the profiles, despite the fact that two different species are under consideration,

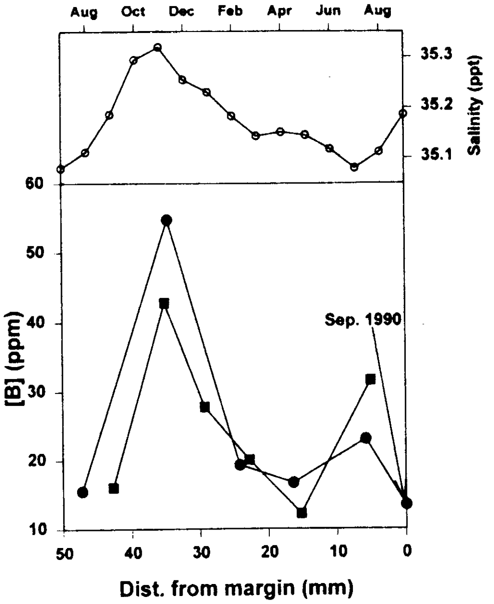

FIGURE 2—Boron profiles for two specimens of *Chionopsis gnidia* from Guaymas. The upper graph illustrates average annual salinity at Guaymas, and the lower plots boron concentration against distance from growing margin for each specimen. The upper and lower graphs were aligned as explained in Figure 1. Note the coincidences of the maxima and minima, and the overall similarity of the curves. One specimen (filled squares) was not sampled at the growing margin. Both specimens were sympatric and contemporaneous.

is further evidence that boron concentration is being determined by an external factor.

Average annual salinity variation in the Gulf is low, ranging on average from 34.14 to 34.61 ppt at Punta Concepcion and 35.08 to 35.32 ppt at Guaymas (Levitus and Boyer, 1994). Fluctuations coincide with seasonal rainfall and water temperature. In the absence of river input (water from the Colorado River reaches the Gulf only rarely), these factors are the primary determinants of salinity in shallow coastal areas via evaporation (concentration) and dilution. Sea-surface temperatures (SST) at the two localities vary annually between 18 and 28°C (Levitus and Boyer, 1994), while rainfall, though seasonal, is sporadic. The patterns of boron concentration in all specimens covary with local salinity variation (Figs. 1 and 2), but exhibit seasonal ranges of 30.64 (difference between maximum and minimum values) and 41.29 ppm in *Chionopsis gnidia,* and 41.19 ppm in *Chione californiensis.* These are compared to corresponding seasonal salinity ranges of 0.214 and 0.344 ppt, at each of the two localities, respectively. It should be borne in mind, however, that the salinity values employed here are multi-year averages and geographic interpolations provided by the Levitus and Boyer (1994) dataset. Therefore, they do not reflect actual salinity variation for the years or places under consideration, and probably underestimate true salinity ranges. Rainfall, on the other hand, may serve to explain some of the discrepancy. There was exceptionally heavy precipitation during the periods June–September 1989 (average 11.3 mm), June-July 1990 (average 23.7 mm), and again in September, 1990 (average 9.5 mm); these episodes would be expected to lower salinity below the monthly averages. The localities conversely experienced very little precipitation during similar periods in 1988 (June–September 1988, average <0.1 mm) and the latter quarter of 1989 (average <0.3 mm). Thus, salinity may have varied more during the period of growth sampled from the specimens than would be expected on the basis of multi-year averages. Without actual measurements of ambient salinity and boron concentration in seawater and runoff, it is impossible to equate the shell boron concentrations directly to salinity.

Furst et al. (1976) observed mean levels of boron as high as 15.1 ± 2.5 (one standard deviation) ppm in the aragonite portion of *Mytilus edulis* shells from Roscoff (France) and equated these to an annual average salinity of 35 ppt. The specimens analyzed in this paper yielded average values of 23.76 ± 14.69 ppm corresponding to a mean salinity of 34.36 ppt (Punta Concepcion) and 23.80 ± 15.52 ppm and 25.10 ± 11.29 ppm corresponding to a mean salinity of 35.19 ppt (Guaymas). The Gulf of California specimens appear to be enriched relative to the North Atlantic shells. However, this could be an artifact of the lack of contemporaneous salinity data for the Gulf, or a result of the greater seasonality of Gulf waters (note the much higher standard deviations). Vengosh et al. (1991) observed concentrations of 11.0 and 15.0 ppm in aragonitic portions of *Tridacna* shells from the Gulf of Elat and the Great Barrier Reef, respectively; corals from the same locations yielded values as high as 58.7 and 79.7 ppm, respectively. They attributed the lower values of molluscs compared to corals to vital effects of preferential exclusion or inclusion. However, these authors apparently used bulk samples that would then represent seasonal averages, ignoring annual maxima and minima. Moreover, vital effects do not dominate the incorporation of other chemical species such as strontium and stable oxygen isotopes into molluscan shells.

Implications for the Calculation of Paleosalinity

The consistent relationship between boron concentration in seawater and salinity suggests the utility of this element as a recorder of salinity. Demonstrating that boron is incorporated into bivalve shells (Furst et al., 1976; Vengosh et al., 1991; this paper), that the incorporation is controlled by an external (environmental) factor, and that the factor is temporally variable, suggests that the factor is salinity. The additional implication is that salinity conditions at the time of shell mineralization can be reconstructed from the boron concentration of the shell. Seasonal variation can also be reconstructed because of the accretionary nature of molluscan shell growth. This logic is directly applicable to fossil material and the derivation of paleosalinities.

The immediate limitation of the study is the need to establish an empirical relationship between shell-boron concentration and ambient salinity, and always a consideration is the currently unknown effect(s) of diagenetic alteration of fossil shells. An empirical relationship between shell boron content and ambient boron concentration cannot be derived from the present study because neither ambient boron concentration nor contemporaneous salinity data are available for the areas and times in which the specimens were collected. The solution lies in the analysis of specimens from localities that have been monitored with respect to salinity and boron concentration for at least one year prior to specimen collection.

Other potential problems concern the dependence of boron incorporation on temperature, and the range of environments to which the technique can be applied. Borate-boron concentrations in seawater do not vary with temperature, but rather only with salinity. Nevertheless, it is possible that the coefficient of incorporation into aragonite could be temperature dependent, and this needs to be investigated. The constancy of the relationship between boron and salinity also suggests, however, that the approach outlined in this paper may be applicable to any environment that experiences variation of salinity. Total boron concentrations (boric acid plus borate anion) in rivers and rain (average 10 µg/l) are negligible compared to those in seawater (average 4.5 mg/l) (Riley and Chester, 1981), and, while they might contribute significantly to ambient concentrations in restricted basins (e.g., the Baltic Sea), are not expected to do so in most coastal settings.

CONCLUSIONS

(1) Contemporaneous and sympatric specimens of two bivalve species from the Gulf of California, *Chione californiensis* and *Chionopsis gnidia*, exhibit similar ontogenetic profiles of shell boron concentration. This suggests that the incorporation of boron into these shells was under external environmental control.

(2) Boron concentration in the shells ranged from 9.23–50.52, 13.47–54.76 and 12.22–42.86 ppm. The patterns of variation coincide with previously documented average patterns of seasonal salinity variation in the Gulf of California, suggesting that shell boron concentrations reflect ambient seawater salinity.

(3) Average annual salinity at the two localities sampled ranges between 34.14–34.61 and 35.08–35.32 ppt. Seasonal boron turnover, as measured in the shells, may not coincide closely with these averages because of the effects of heavy seasonal rainfall. High levels of rainfall during the lives of the sampled specimens may account for the large ranges of boron concentrations observed in the shells.

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