Paleoceanography of the Late Cretaceous (Maastrichtian) Western Interior Seaway of North America: evidence from Sr and O isotopes

J. Kirk Cochran [a,*], Neil H. Landman [b], Karl K. Turekian [c], Annie Michard [d], Daniel P. Schrag [e]

[a] *Marine Sciences Research Center, State University of New York, Stony Brook, NY 11794-5000, USA*
[b] *American Museum of Natural History, New York, NY 10024, USA*
[c] *Department of Geology and Geophysics, Yale University, New Haven, CT 06520-8109, USA*
[d] *CEREGE, Europôle Méditerranéen de l'Arbois, BP 80, 13545 Aix-en-Provence, France*
[e] *Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA 02138, USA*

Abstract

Well-preserved fossils of the Late Cretaceous Western Interior Seaway (WIS) of North America have been analyzed for Sr concentration and Sr and O isotopes in order to decipher paleosalinities and paleotemperatures. The samples are from four biofacies within the Seaway (late Maastrichtian): offshore Interior (Pierre Shale), nearshore Interior (Fox Hills Formation), brackish (reduced salinity; Fox Hills Formation) and freshwater (Hell Creek Formation). Samples were also obtained from the Severn Formation of Maryland (considered to be representative of the open ocean). All biofacies (except the freshwater) are demonstrably within the *Jeletzkytes nebrascensis* ammonite zone (< 1 Ma duration). The $^{87}Sr/^{86}Sr$ ratios show significant and systematic decreases from marine (mean $\pm$ 1 S.D. $= 0.707839 \pm 0.000024$) to brackish facies (mean $\pm$ 1 S.D. $= 0.707677 \pm 0.000036$), consistent with dilution by freshwater with a lower $^{87}Sr/^{86}Sr$ ratio than seawater. Such variation disallows using the $^{87}Sr/^{86}Sr$ ratios of fossil shell material to assign ages to fossils from the Late Cretaceous WIS without knowledge of the salinity in which the organism grew. The Sr isotope ratios for scaphitid ammonites within a single biofacies are similar to each other and different from those for scaphites in other biofacies, implying that these organisms are restricted in their distribution during life. The $^{87}Sr/^{86}Sr$ values of freshwater unionid mussels range widely and are not compatible with the freshwater endmember $^{87}Sr/^{86}Sr$ ratio required by the trend in $^{87}Sr/^{86}Sr$ vs. biofacies established from the other samples. Paleosalinities for the biofacies are estimated to range from 35‰ in the open marine to a minimum of 20‰ in the brackish, based on the presence of cephalopods in all four facies and the known salinity tolerance of modern cephalopods. Producing reasonable $^{87}Sr/^{86}Sr$ values for the freshwater endmember of a $^{87}Sr/^{86}Sr$ vs. 1/[Sr] plot requires a Sr concentration 0.2–0.5 that of seawater for the dominant freshwater input to the WIS. Such high Sr concentrations (relative to seawater) are not observed in modern rivers, and we suggest that the brackish environment in the WIS arose through the mixing of freshwater and seawater in a nearshore aquifer system. Reactions of the solution with aquifer solids in this 'subterranean estuary' [Moore, Mar. Chem. 65 (1999) 111–125] produced brackish water with the Sr concentration and isotopic composition recorded in the brackish biofacies. $\delta^{18}O$ values of the fossils

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* Corresponding author. Fax: +1-631-632-3066.   *E-mail address:* kcochran@notes.cc.sunysb.edu (J.K. Cochran).

show decreases from the marine to brackish biofacies consistent with increasing temperatures (from ~ 13 to $23°C$) or, if temperatures were relatively constant, to a decrease in the $\delta^{18}O$ of the water in which the shell formed. The latter interpretation is consistent with less-than-fully marine salinities in the nearshore biofacies, but both changes in temperature and the isotopic composition of the water may have occurred in this environment.

1. Introduction

The Cretaceous Period was characterized by the existence of broad continental seaways that connected the world oceans. In North America, the Western Interior Seaway (WIS) connected the Boreal and Tethys oceans. During the late Maastrichtian this seaway was closed to the north and disappeared altogether by the end of the Cretaceous. Because these epicontinental seas have no modern analogs as far as magnitude and setting are involved, very little is known about their oceanography. In particular, distributions of water masses as defined by temperature and salinity variations are poorly characterized although reconstructions of WIS paleoceanography and paleocirculation have been attempted (e.g. Hay et al., 1993). One approach to reconstructing paleotemperatures and paleosalinities in such environments is through the record of geochemical tracers that are proxies for these parameters and are incorporated in the calcium carbonate shells of organisms. For example, the determination of $\delta^{18}O$ in shell material as an index of paleotemperatures has been applied to a variety of systems since Urey (1947) initially proposed that the $\delta^{18}O$ values of carbonates are a function of both the temperature and $\delta^{18}O$ of the water in which the carbonate formed. In the WIS, Tourtelot and Rye (1969) and Wright (1987) among others measured $\delta^{18}O$ values in well-preserved shell material. Calculated paleotemperatures for benthic organisms ranged up to 40°C and were considered unrealistically high. Moreover, Wright (1987) noticed isotopic differences in epifauna compared with nekton and infauna and argued that the WIS had an unusual salinity structure, with warm saline water formed by evaporation in the shallow eastern portion of the sea and subsequently transported into the basin interior underlying colder, less saline intermediate waters.

An additional paleoenvironmental indicator is the ratio of ^{87}Sr to ^{86}Sr in shell material. The $^{87}Sr/^{86}Sr$ ratio in the open ocean reflects dominantly the input of Sr from continents via rivers and mantle-derived sources such as hydrothermal vents (Brass, 1976; Albarède et al., 1981; Palmer and Edmond, 1989). Variations in the $^{87}Sr/^{86}Sr$ ratio over geologic time are due to variations in the relative importance of these Sr sources, as well as evolution in the Earth's $^{87}Sr/^{86}Sr$ ratio due to ^{87}Rb decay. Such variations are recorded in the $^{87}Sr/^{86}Sr$ of marine carbonates, which are characteristically low in Rb, and the Phanerozoic evolution of the $^{87}Sr/^{86}Sr$ ratio in seawater has been well documented using these deposits (Peterman et al., 1970; Dasch and Biscaye, 1971; Brass, 1976; Veizer and Compston, 1974; Burke et al., 1982; Hess et al., 1986, DePaolo, 1986; Martin and Macdougall, 1991; Veizer et al., 1999). In modern ocean–river systems, the $^{87}Sr/^{86}Sr$ ratio of rivers can be significantly different from that of seawater (0.70916), ranging from values much lower (e.g. rivers entering San Francisco Bay; Ingram and Sloan, 1992) to much greater than that of seawater (e.g. rivers entering the Baltic Sea; Andersson et al., 1992, 1994). In the estuarine and coastal environment, the mixture of fresh and salt water produces a gradient in $^{87}Sr/^{86}Sr$ ratios that can be recorded by organisms living in a given salinity. Such gradients in $^{87}Sr/^{86}Sr$ vs. salinity are seen in San Francisco Bay and the Baltic Sea, for example (Ingram and Sloan, 1992; Andersson et al., 1992). Thus the $^{87}Sr/^{86}Sr$ ratio may be used to reconstruct paleosalinities in areas where salinity gradients, as well as differences in Sr isotope ratios between freshwater and seawater exist.

We present data for oxygen, carbon and strontium isotopes and strontium concentrations in well-preserved shell material from within the WIS and from coeval material collected from the Atlantic Coastal Plain of North America. The samples were collected in the *Jeletzkytes nebrascensis* ammonite zone, representing ≤ 1 Ma in the late Maastrichtian, around 67 Ma BP (Waage, 1968; Landman and Waage, 1993; Kauffman et al., 1993; Kennedy et al., 1998), from the Pierre Shale, Fox Hills and Hell Creek Formations in South Dakota. Because of the extensive paleoenvironmental reconstructions for this region (Gill and Cobban, 1966; Waage, 1968), it is possible to separate the samples a priori into environments corresponding to offshore Interior, nearshore Interior and brackish (less than fully marine salinity). Samples were collected from the same biozone in the Severn Formation (Maryland; Kennedy et al., 1997) on the Atlantic Coastal Plain and are assumed to represent fully marine, open-ocean conditions.

2. Methods

2.1. Sample localities

Samples were collected from the Pierre Shale and Fox Hills Formation in South Dakota and the Severn Formation in Maryland, all within the *Jeletzkytes nebrascensis* ammonite biozone (Table 1, Fig. 1). The samples were divided into paleoenvironments according to the reconstruction of Waage (1968). We have labeled these environments as follows: 'offshore Interior', corresponding to an open-water biofacies representative of the central seaway with a maximum depth of ~ 200 m; 'nearshore Interior', representative of a more nearshore facies; and 'brackish' biofacies, representative of an estuarine, nearshore environment inhabited by organisms with lower salinity tolerances. Samples from the Severn Formation from the same biozone (Kennedy et al., 1997) are considered to represent a fully 'open-ocean' biofacies and include only nektonic organisms (ammonites and nautiloids).

Within the WIS, gastropods, pelecypods, am-

monites, nautiloids and belemnites are represented in the fauna. Specimens of two ammonite species (*J. nebrascensis* and *Sphenodiscus lobatus*) were analyzed from all four paleoenvironments. Additionally, freshwater unionid bivalves from the basal Hell Creek Formation were analyzed in an effort to constrain the $^{87}Sr/^{86}Sr$ ratio in freshwater at that time. However, the Hell Creek samples lack the biostratigraphic control represented by the other samples. All samples are deposited in the collections of the Yale Peabody Museum, American Museum of Natural History, and the US National Museum. Three samples of the rock making up the presumed ambient terrain also were taken for determination of Sr isotope ratio and concentration. These samples included the Elk Butte and Mobridge members of the Pierre Shale, which lie stratigraphically below the Fox Hills Formation and may have been exposed along the paleo-shoreline.

2.2. Sample preservation

Samples were taken from single specimens, except K15, in which two specimens were combined; samples of cephalopods were taken from the outer shell, rather than the septa. The shell material of the ammonites and nautiloids showed a distinctive iridescence characteristic of nacreous shell structure. However, prior work has shown that authigenic deposits on the surfaces of samples can potentially bias isotopic and concentration data (Turekian and Armstrong, 1961; Landman et al., 1983). Thus, all samples were sonified for several minutes in distilled water to remove loosely adhering surface phases. Samples were air-dried and the best pieces were selected following examination with a light microscope. If necessary, the sonification step was repeated. Samples were then examined by scanning electron microscopy (SEM) to confirm both the absence of surface phases and the presence of original shell structure (Plate I). In the cephalopod samples, the characteristic nacreous microstructure was preserved. Excellent preservation quality was indicated when SEM photos showed nacreous tablets of even thickness, without evident alteration or deposition of diagenetic interlayer material. *Je-*

Table 1
Sample identification and localities[a]

Sample ID	Museum ID[b]	Formation	Member[c]	Species	Organism
Freshwater					
K6	YPM 38033	Hell Creek		*Unio* sp.	bivalve
K12	YPM 38034	Hell Creek		*Unio* sp.	bivalve
K16	YPM 38029	Hell Creek		*Unio* sp.	bivalve
K17	YPM 38005	Hell Creek		*Unio* sp.	bivalve
K22	YPM 38044	Hell Creek		Unionoida	bivalve
Open-ocean					
K3	AMNH 45406	Severn		*Eutrephoceras dekayi*	nautiloid
K4	AMNH 45404	Severn		*Sphenodiscus lobatus*	ammonite
K35	USNM 520476	Severn		*Jeletzkytes nebrascensis*	ammonite
Offshore Interior					
K2	USNM 486542	Pierre		*J. nebrascensis*	ammonite
K23	USNM 486528	Pierre		*Belemnitella* cf. *bulbosa*	belemnite
K34	AMNH 47462	Pierre		*S. lobatus*	ammonite
K37	USNM 520477	Pierre		*J. nebrascensis*	ammonite
Nearshore Interior					
K5	AMNH 45463	Fox Hills	Timber Lake	*S. lobatus*	ammonite
K8	YPM 44980	Fox Hills	Timber Lake	*S. lobatus*	ammonite
K11	YPM 162363	Fox Hills	Trail City, C, IC	*E. dekayi*	nautiloid
K10	YPM 162434	Fox Hills	Timber Lake	*Serrifusus dakotensis*	gastropod
K1	AMNH 45405	Fox Hills	Timber Lake	*J. nebrascensis*	ammonite
K13	YPM 162433	Fox Hills	Timber Lake, CT	*J. nebrascensis*	ammonite
K21	YPM 202291	Fox Hills	Timber Lake, CT	*Cymbophora warreni*	bivalve
K33	YPM 32588	Fox Hills	Trail City, C	*Belemnitella* cf. *bulbosa*	belemnite
Brackish					
K7	YPM 46036	Fox Hills	Timber Lake, TO	*Tancredia americana*	bivalve
K9	YPM 162385	Fox Hills	Iron Lightning, CL	*Corbicula* sp.	bivalve
K14	YPM 162374	Fox Hills	Iron Lightning, BL	*J. nebrascensis*	ammonite
K15	YPM 162397,8	Fox Hills	Iron Lightning, CL	*Lunatia* sp.	gastropod
K18	YPM 162437	Fox Hills	Iron Lightning, BL	*J. nebrascensis*	ammonite
K19	YPM 200285	Fox Hills	Hell Creek	*Lunatia* sp.	gastropod
K20	YPM 162402	Fox Hills	Hell Creek	*J. nebrascensis*	ammonite
K36	YPM 44661	Fox Hills	Hell Creek	*S. lobatus*	ammonite
Rock					
K30	YPMIP loc. D4083[c]		Elk Butte		
K31	YPMIP loc. C1872		Mobridge		
K32	YPMIP loc. C479		Mobridge		

[a] Detailed descriptions of localities are available upon request.

[b] YPM = Yale Peabody Museum, AMNH = American Museum of Natural History, USNM = US National Museum, YPMIP loc. = Yale Peabody Museum Invertebrate Paleontology locality.

[c] C, *Cucullaea* Assemblage Zone; CT, *Cymbophora–Tellina* Assemblage Zone; TO, *Tancredia–Ophiomorpha* biofacies; CL, Colgate lithofacies; BL, Bullhead lithofacies; IC, Irish Creek lithofacies.

letzkytes nebrascensis samples were available from all four biofacies (Plate IA–D), and of these samples, the best preservation was evident in the nearshore Interior and brackish facies (Plate IC,D). However, the *J. nebrascensis* samples from the open-ocean and offshore Interior facies (Plate IA,B) showed no evident overgrowths indicative of diagenetic alteration. Preservation in the nautiloid *Eutrephoceras dekayi*, from the open-ocean biofacies, was excellent (Plate IE). A final screen-
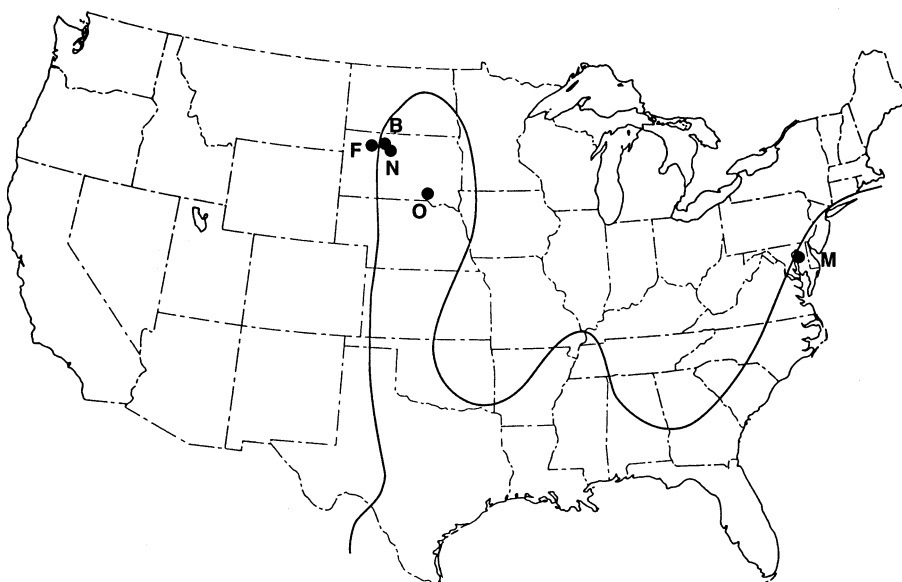
Fig. 1. Map showing biofacies sampled relative to the paleo-shoreline (heavy line) of the WIS during the late Maastrichtian zone of *Jeletzkytes nebrascensis* (modified from Kennedy et al., 1998). Key: F = freshwater, B = brackish Interior, S = nearshore Interior, D = offshore Interior, M = open-ocean.

ing involved X-ray diffraction to confirm that the samples were 100% aragonite, with the exception of the belemnites, which were 100% calcite.

2.3. Sample analyses

2.3.1. Strontium isotopes

Cleaned pieces of sample (2–6 mg) were dissolved in 1 N HCl and any undissolved residue (largely organic) was separated by centrifugation. Approximately 2 ml of sample was loaded onto a cation exchange column (AG50WX8) calibrated for Sr. After passing 22 ml of 2 N HCl through the column, the Sr fraction was collected in 4 ml of 2 N HCl and evaporated to dryness. Total blank Sr was less than 200 pg compared with $\sim$ 2 µg Sr in the samples.

The matrix samples from the Elk Butte and Mobridge Members were subjected to a simple experiment to determine the Sr isotope ratio that might be released during chemical weathering by freshwater. Samples of 0.1–0.2 g were leached in 10–20 ml distilled water for several hours at room temperature. The solution was centrifuged and the leachate was separated from the residue,

spiked with a Sr tracer and evaporated. Subsequent separation of Sr followed the procedure outlined above.

Strontium was loaded with tantalum oxide (Birk, 1986) onto a single rhenium filament and analyzed in the dynamic mode on a Finnigan MAT 262 mass spectrometer. The static mode was used for the matrix samples. Repeated analyses of NBS 987 during the time of sample analyses gave a mean $^{87}Sr/^{86}Sr$ value of 0.710238 ± 0.000011 (16 runs) in the dynamic mode and 0.710271 ± 0.000012 (10 runs) in the static mode. The $^{87}Sr/^{86}Sr$ values of the spiked matrix samples (static mode) have been corrected to the NBS standard $^{87}Sr/^{86}Sr$ value of 0.710248. The $^{87}Sr/^{86}Sr$ values in all samples are normalized to a $^{86}Sr/^{88}Sr$ value of 0.1194.

2.4. Strontium concentrations

Strontium concentrations were measured on separate aliquots of the samples following the procedure of Schrag (1999). Samples were dissolved in 2% HNO_3, diluted and Sr was measured by inductively coupled plasma emission spectros-
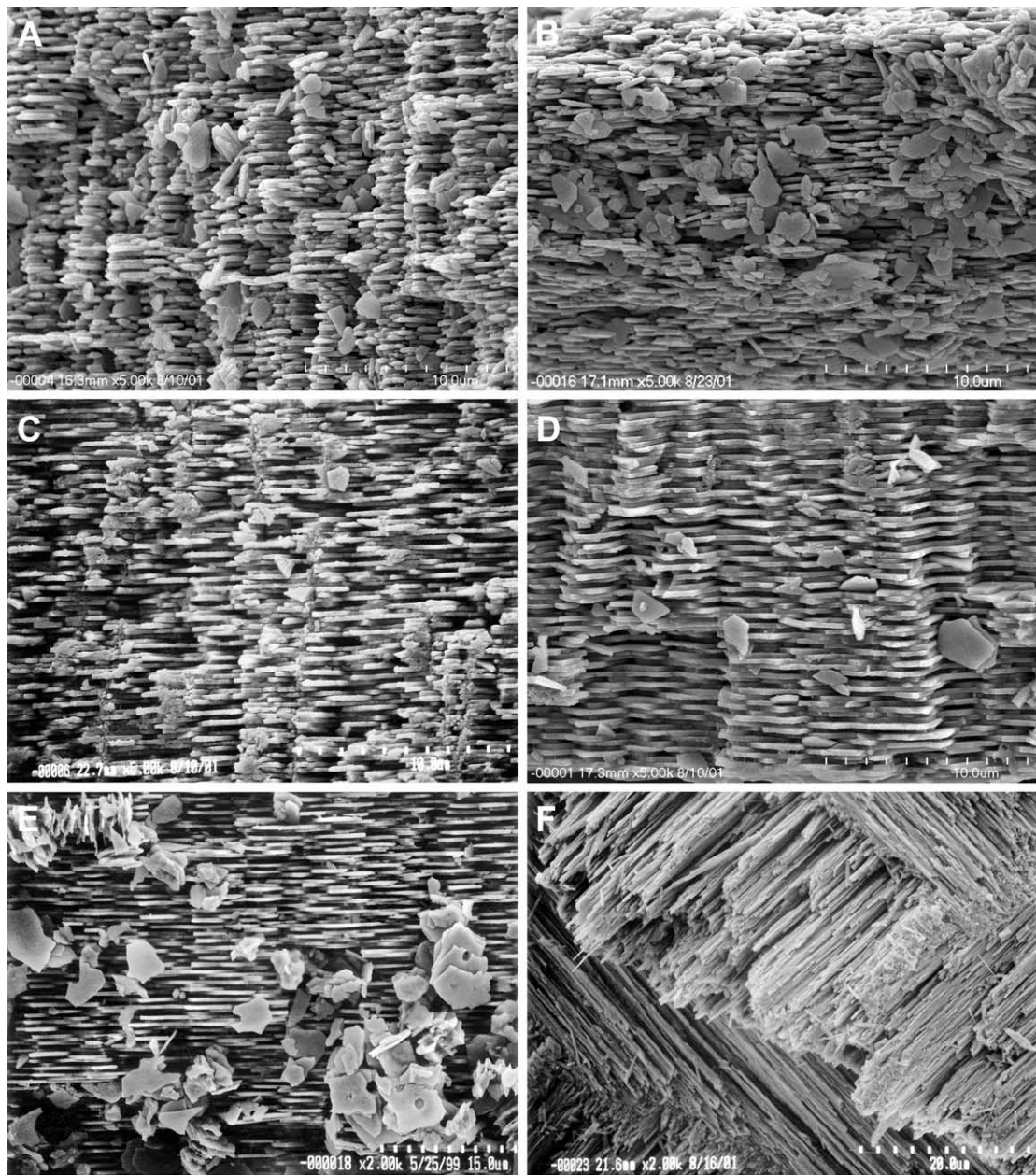
Plate I. Shell microstructure in fractured cross section of six samples. (A–D) *Jeletzkytes nebrascensis* from four different biofacies. The nacreous microstructure is preserved although some platelets have broken off in fracturing. (A) Fully marine, sample K35 (USNM 520476, Severn Formation). (B) Offshore Interior, sample K37 (USNM 520477, Pierre Shale). (C) Nearshore Interior, sample K1 (AMNH 45405, Fox Hills Formation). (D) Brackish Interior, sample K20 (YPM 162402, Fox Hills Formation). (E) *Eutrephoceras dekayi*, fully marine, sample K3 (AMNH 45406, Severn Formation), showing the original nacreous microstructure. (F) *Serrifusus dakotensis*, nearshore Interior, sample K10 (YPM 162434, Fox Hills Formation) with cross-lamellar microstructure.

copy on a Jobin-Yvon simultaneous instrument, model 46-P. Analytical precision was determined to be better than $\pm 0.2\%$, based on replicate analyses of multiple dilutions of solutions of single samples.

2.5. Oxygen and carbon isotopes

Oxygen and carbon isotopic analyses were made at the University of Michigan Stable Isotope Laboratory by L. Wingate. Carbonate samples weighing a minimum of 10 μg were placed in stainless steel boats. Calcite samples were roasted at 380°C in vacuo for 1 h to remove volatile contaminants. Aragonite was roasted at a lower temperature, 200°C, to minimize effects of thermal decarboxylation that can alter the isotopic composition of the carbonate. Samples were then placed in individual borosilicate reaction vessels and reacted at $76° \pm 2°C$ with three drops of anhydrous phosphoric acid for 8 min in a Finnigan Kiel preparation device coupled directly to the inlet of a Finnigan MAT 251 triple-collector isotope ratio mass spectrometer. Isotopic enrichments were corrected for acid fractionation and oxygen-17 contribution by calibration to a best-fit regression line defined by two NBS standards, NBS 18 and NBS 19. Precision of data was monitored through daily analysis of a variety of powdered carbonate standards. At least six standards were reacted and analyzed daily, bracketing the sample suite at the beginning, middle, and end of the day's run. Measured precision was maintained at better than 0.1‰ for both carbon and oxygen isotope compositions. Data are reported in ‰ notation relative to VPDB (Vienna PeeDee Belemnite).

3. Results

3.1. Sr concentrations

Concentrations of Sr in the shell material range from 355 to 4705 ppm (Table 2). The lowest values are recorded in the freshwater bivalves and the highest values are from ammonites (*Sphenodiscus lobatus*, *Jeletzkytes nebrascensis*) from the

Seaway. Specimens of the same species from the same environment have similar Sr concentrations, varying within $\sim 10\%$ (e.g. *S. lobatus* collected in the nearshore Interior and *J. nebrascensis* from both the nearshore Interior and brackish environments). There is a range of Sr concentrations for different organisms from a single environment, presumably reflecting interspecific differences in the factors affecting Sr incorporation into shell material (see discussion below). In particular, the benthic gastropods *Lunatia* and *Serrifusus* and bivalve *Cymbophora* have lower Sr concentrations than those of the ammonites and nautiloid samples in any given environment. The bivalve *Corbicula* is conspicuous in having the lowest Sr concentration of the brackish group. This low value, together with its unusually light $\delta^{18}O$ value (see below), suggests that this specimen was transported into the brackish assemblage from freshwater. Indeed, present-day *Corbicula* can live in completely fresh water (Counts, 1986; Mackie, 1986). The Sr concentrations of ammonite and nautiloid fossils measured in this study are comparable to values measured by Turekian and Armstrong (1961) in material with negligible-to-low percentages of calcite (and thus judged to be minimally altered).

Total Sr concentrations in the Elk Butte and Mobridge Members are greater than those measured in the fossils and range from 5823 to 7434 ppm. The Sr released by the leaching experiments was 646 ppm for K30, 708 ppm for K31 and 20 ppm for K32. In the latter case only 0.3% of the Sr was leached, whereas the fraction was 9% for K30 and 12% for K31.

3.2. Sr isotope ratios

With the exception of the freshwater bivalves, the $^{87}Sr/^{86}Sr$ ratios of the samples cluster into discrete groups with minimal overlap: 0.707815–0.707862 for the open-ocean group, 0.707796–0.707827 for the offshore Interior group, 0.707704–0.707795 for the nearshore Interior group and 0.707605–0.707712 for the brackish group (Table 2, Fig. 2). In contrast, the $^{87}Sr/^{86}Sr$ values in the freshwater bivalves range from 0.707615 to 0.707986, although the large range

Table 2
Isotopic and concentration data

	Species	$^{87}Sr/^{86}Sr$	Sr (ppm)	$\delta^{18}O$ (‰)	$\delta^{13}C$ (‰)	Remarks
Freshwater						
K6	*Unio* sp.	0.707940 ± 0.000009	405	−20.29	−4.69	
K12	*Unio* sp.	0.707903 ± 0.000011	504	−19.71	−4.99	
K16	*Unio* sp.	0.707942 ± 0.000016	561	−19.11	−1.73	
K16	*Unio* sp.	0.707986 ± 0.000012	na	na	na	Powdered sample
K17	*Unio* sp.	0.707628 ± 0.000015	355	−16.87	−2.56	
K17	*Unio* sp.	0.707615 ± 0.000014	na	na	na	Powdered sample
K22	Unionoida	0.707754 ± 0.000016	na	na	na	
K22	Unionoida	0.707869 ± 0.000013	na	na	na	Powdered sample
Open-ocean						
K3	*Eutrephoceras dekayi*	0.707815 ± 0.000007	3261	−0.66	−1.51	
K4	*Sphenodiscus lobatus*	0.707839 ± 0.000011	3018	0.28	−2.95	
K35	*Jeletzkytes nebrascensis*	0.707862 ± 0.000006	1890	na	na	
Offshore Interior						
K2	*J. nebrascensis*	0.707810 ± 0.000008	na	1.43	1.94	
K23	*Belemnitella* cf. *bulbosa*	0.707801 ± 0.000009	1319	0.31	0.36	
K34	*S. lobatus*	0.707827 ± 0.000008	3859	na	na	
K37	*J. nebrascensis*	0.707796 ± 0.000007	2520	na	na	
Nearshore Interior						
K5b	*S. lobatus*	0.707777 ± 0.000010	4705	−0.92	−3.64	
K5g	*S. lobatus*	0.707768 ± 0.000012	na	na	na	Duplicate sample
K8	*S. lobatus*	0.707781 ± 0.000009	4329	−1.61	−3.34	
K11	*E. dekayi*	0.707785 ± 0.000015	3867	0.18	0.21	
K10	*Serrifusus dakotensis*	0.707773 ± 0.000014	2297	−0.49	−0.13	
K1	*J. nebrascensis*	0.707740 ± 0.000010	3963	−0.04	0.99	
K13	*J. nebrascensis*	0.707750 ± 0.000015	4247	−0.06	−1.83	
K21	*Cymbophora warreni*	0.707704 ± 0.000012	1898	−0.23	1.36	Same locality and concretion as K13
K33	*Belemnitella* cf. *bulbosa*	0.707795 ± 0.000008	1772	−1.28	1.91	Same locality as K11
Brackish						
K7	*Tancredia americana*	0.707695 ± 0.000011	2412	−1.34	1.16	
K9	*Corbicula* sp.	0.707688 ± 0.000015	667	−17.91	−4.79	
K14	*J. nebrascensis*	0.707654 ± 0.000014	2545	−0.34	1.29	
K15	*Lunatia* sp.	0.707605 ± 0.000010	1589	−0.47	0.49	Same locality as K9
K18	*J. nebrascensis*	0.707688 ± 0.000008	2680	−2.44	3.10	
K19	*Lunatia* sp.	0.707699 ± 0.000014	2043	−2.40	0.33	
K20	*J. nebrascensis*	0.707703 ± 0.000012	na	−0.31	1.63	Same locality and concretion as K19
K36	*S. lobatus*	0.707712 ± 0.000007	na	na	na	
Rock						
K30	Elk Butte member	0.707165 ± 0.000018[a]	7434	na	na	
K31	Mobridge member	0.707507 ± 0.000016[a]	5823	na	na	
K32	Mobridge member	0.710998 ± 0.000016[a]	5859	na	na	

na = not analyzed.

[a] Sr isotope ratio released to solution during leaching experiments. See text.

tends to be driven by a single sample. There is generally close correspondence in the $^{87}Sr/^{86}Sr$ values of multiple specimens of the same species collected in a given environment; the $^{87}Sr/^{86}Sr$ ratios of specimens of the same species within a biofacies are often within the analytical uncertainty of one another. This is evident in the multiple specimens of *J. nebrascensis* in the offshore Interior, nearshore Interior and brackish groups as well as *S. lobatus* in the brackish group (Table 2). In contrast, the values are significantly different for the same species in different biofacies.

The $^{87}Sr/^{86}Sr$ ratios leached from the Elk Butte and Mobridge Member rocks bracket the entire range of values seen in the fossil shell material and vary from 0.707165 to 0.710998 (Table 2). The highest ratio corresponds to sample K32, in which only 0.3% of the Sr was released to solution.

3.3. Oxygen isotopes

$\delta^{18}O$ values range from $-20.29‰$ to $1.43‰$ (Fig. 3) and $\delta^{13}C$ values range from $-4.99‰$ to $3.10‰$. In contrast to the Sr isotope results, there is less consistency among the paleoenvironmental groups, although the data for the WIS samples show a tendency toward lighter $\delta^{18}O$ values proceeding from offshore Interior to brackish environments: 0.3 to 1.4‰ for the offshore Interior,
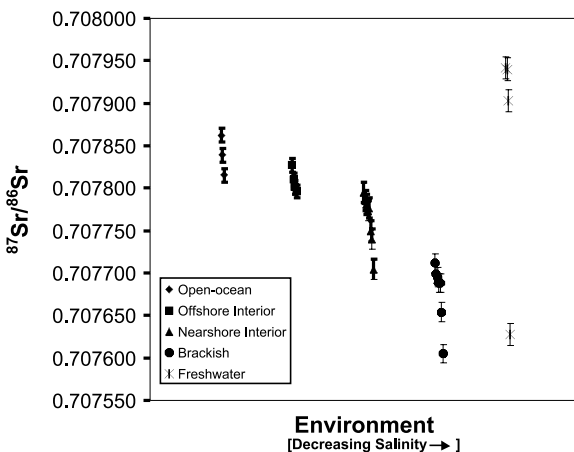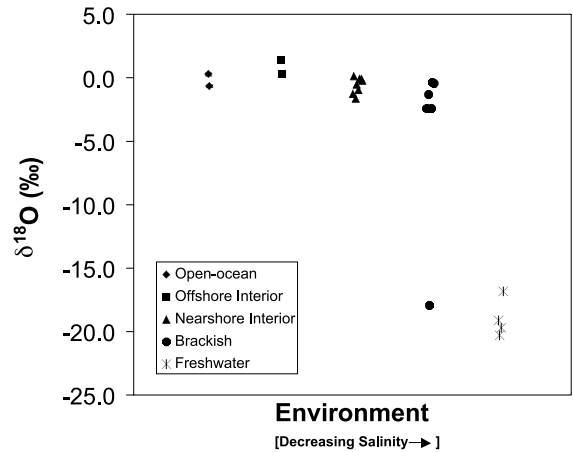
Fig. 3. $\delta^{18}O$ (‰ VPDB) of shell carbonate vs. biofacies (relative scale: decreasing salinity to the right; values in each group are slightly offset to improve readability).

0.0 to $-1.6‰$ for the nearshore Interior and -0.3 to $-2.4‰$, excluding the value of specimen K9, for the brackish group. The $\delta^{18}O$ for K9, a sample of *Corbicula sp.*, is quite light, $-17.9‰$ and is similar to values obtained for the freshwater bivalves (-16.9 to $-20.3‰$). We take the oxygen isotope composition of *Corbicula*, as well as its low Sr concentration, to indicate that the shell grew in freshwater. Given that both the $\delta^{18}O$ and Sr concentration in a co-occurring specimen of *Lunatia* are more consistent with the other brackish fauna, we believe the sample of *Corbicula* was transported into the brackish environment after death.
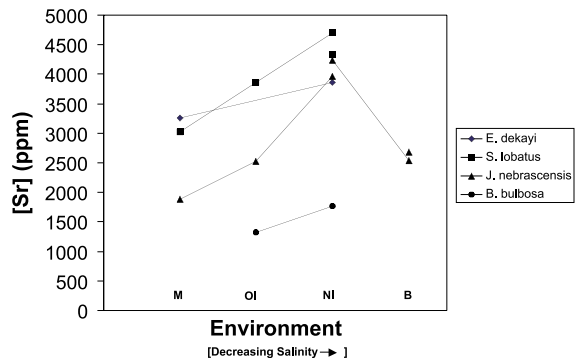
Fig. 2. $^{87}Sr/^{86}Sr$ vs. biofacies (relative scale: decreasing salinity to the right; values in each group are slightly offset to improve readability).

Fig. 4. Shell Sr concentration vs. biofacies for cephalopod species. M — Open-ocean; OI — Offshore Interior; NI — Nearshore Interior; B — Brackish.

4. Discussion

4.1. Strontium concentrations in shells

There is considerable interspecific variation in the Sr concentrations for organisms collected in a single paleoenvironment. Despite these differences in absolute concentration, the trends from open-ocean to brackish biofacies are consistent for the cephalopod species (Fig. 4). The Sr concentration progressively increases from open-ocean to offshore Interior to nearshore Interior. Only a single species (*J. nebrascensis*) from the brackish environment was analyzed, and the data show a decrease in Sr concentration relative to the nearshore Interior.

The Sr concentration in biogenic carbonate reflects the Sr/Ca ratio in the water in which the carbonate formed as well as the physiological factors involved in biomineralization. The latter are dependent upon water temperature, salinity, and organism growth rate (Smith et al., 1979; Stoll and Schrag, 2000; Stoll et al., 2002a,b; Weinbauer et al., 2000). Factors such as water temperature and growth rate also may be interdependent (Yatsu et al., 1998; Stoll et al., 2002a). With respect to the variation in the Sr/Ca ratio of the water, the present-day ocean shows a difference between the value in the open ocean and in world-average river water, with the former being approximately three times greater than the latter (Palmer and Edmond, 1989; Turekian, 1969). A simple trend of decreasing shell Sr concentrations from the marine to brackish environments is not seen in our data, suggesting that variation of the Sr/Ca ratio in the water is not the first-order control on Sr/Ca in the shell. Although the exact fractionation of Sr/Ca in the shell vs. water will vary among species, the similarity in trends among all the cephalopod specimens suggests that fundamental differences in biomineralization related to environmental factors such as temperature, salinity or pCO_2 are causing the trends.

The effects of temperature and other factors on the Sr/Ca ratio of biogenic carbonates have been documented for coral skeletons, fish otoliths, squid statoliths and coccolithophorids (Smith et al., 1979; Yatsu et al., 1998; Stoll et al., 2002b).

The dependence of Sr/Ca on temperature is not consistent among different organisms: in hermatypic corals (*Porites*), the carbonate Sr/Ca decreases by $\sim 10\%$ as the temperature increases from 10 to 25°C (Alibert and McCulloch, 1997) while in coccoliths the ratio increases by 25% over approximately the same temperature range (Stoll et al., 2002b). It is possible that the water temperature of the nearshore WIS environments sampled by us was greater than that of the offshore Interior (see discussion below), and the increase in shell Sr from the offshore to nearshore biofacies is consistent with the effect of temperature seen in the coccolith data (Stoll et al., 2002b). However, the magnitude of the increase predicted from the coccoliths is less than we observe.

4.2. The $^{87}Sr/^{86}Sr$ ratio as a paleosalinity indicator

The Sr isotopic evolution of seawater over geologic time has been well-documented (Peterman et al., 1970; Veizer and Compston, 1974; Brass, 1976; Spooner, 1976; Hess et al., 1986; DePaolo, 1986; Martin and Macdougall, 1991; McArthur et al., 1994; Veizer et al., 1999). In the Late Cretaceous, values of open ocean water increased from 0.7073 to 0.7078 (Hess et al., 1986) with a maximum of ~ 0.7079 at the Cretaceous–Tertiary boundary (Martin and Macdougall, 1991). Seawater $^{87}Sr/^{86}Sr$ increased from ~ 0.7077 to ~ 0.7079 in the last ~ 3 Ma of the Cretaceous, and was ~ 0.70784 at the time represented by our sampling (Martin and Macdougall, 1991). Indeed, within the analytical error, $^{87}Sr/^{86}Sr$ was constant at ~ 0.70784 in the ~ 2 Ma preceding the K/T boundary (Martin and Macdougall, 1991). This value agrees well with the average $^{87}Sr/^{86}Sr$ of the 'open-ocean' group: mean ± 1 S.D. = 0.707839 ± 0.000024, and suggests that this group is accurately reflecting open marine conditions. Values in the offshore Interior group are only slightly less than the open-ocean group: mean ± 1 S.D. = 0.707808 ± 0.000014. The ratio trends toward lower values proceeding from offshore to nearshore Interior (mean ± 1 S.D. = 0.707763 ± 0.000030) to the brackish biofacies (mean ± 1 S.D. = 0.707677 ± 0.000036). Although values within adjacent groups overlap within the error,

the overall trend is toward progressively lower values such that the brackish group is significantly different from the open-ocean group.

We note that the trend in $^{87}Sr/^{86}Sr$ exists even though each group includes nektonic organisms that might have ranged from one environment to another. The ammonite *Jeletzkytes nebrascensis* is a good example in that this ammonite occurs in all four environments yet specimens from a given paleoenvironment possess a significantly different isotopic signature from the other environments. This is strong evidence that these organisms were living in the environment in which they were collected. We conclude that although these animals were nektonic, their swimming ability was limited or the distances were too great to permit much migration between biofacies. In contrast, it is interesting to note that the two belemnite specimens analyzed (K23 and K33) have nearly identical $^{87}Sr/^{86}Sr$ ratios and Sr concentrations although they were collected in two different biofacies (offshore and nearshore Interior; Table 2). This may indicate that belemnites were more active swimmers, like the present-day squid, and were mobile between various WIS habitats (W. Christensen, Univ. of Copenhagen, personal communication, 2000).

The trend in $^{87}Sr/^{86}Sr$ ratios observed among the paleoenvironments is consistent with a salinity variation caused by mixing with a freshwater $^{87}Sr/^{86}Sr$ endmember that is lower than the seawater value during late Maastrichtian time. In this respect, the trend is similar to that observed in modern San Francisco Bay (Ingram and Sloan, 1992) and in many geologically older systems (Ingram and DePaolo, 1993; Holmden et al., 1997; Schmitz et al., 1991; Vonhof et al., 1998; Vaiani, 2000).

An alternative explanation is that the trend represents a temporal difference in sample age. Our samples were collected within a single ammonite zone, estimated to span ∼1 Ma of time (Kauffman et al., 1993). We reject the possibility that the trend in $^{87}Sr/^{86}Sr$ ratios is due to a monotonic change in the age of the samples reflecting a change in seawater values of the ratio because the brackish fauna is either coeval or younger (but still within the same ammonite biozone)

than the offshore Interior samples. The brackish samples were collected stratigraphically up-section or, in a paleogeographic sense, landward of the nearshore Interior samples. Variation in the open ocean $^{87}Sr/^{86}Sr$ ratio for the late Maastrichtian documented by Martin and Macdougall (1991) shows that the ratio remains the same (∼0.70784) or increases slightly toward the end of the Cretaceous, with a maximum at the K/T boundary. Thus, if the brackish samples are not coeval, but are younger, then their $^{87}Sr/^{86}Sr$ ratios would be greater than that in the other samples, not lower. On the basis of both stratigraphic relationships and the temporal pattern of Sr isotopes in the Late Cretaceous, we conclude that the $^{87}Sr/^{86}Sr$ ratios in the samples reflect the paleoenvironments.

Our efforts to use unionids to establish the freshwater endmember of the $^{87}Sr/^{86}Sr$ vs. habitat trend were not successful. The values span the range observed in the samples and are generally greater than values recorded in organisms from the WIS (Fig. 2, Table 2). This situation is similar to that observed by Ingram and Sloan (1992) in San Francisco Bay. Their analyses of the rivers draining into the Bay, coupled with the trend in $^{87}Sr/^{86}Sr$ vs. salinity, show that considerable variation in $^{87}Sr/^{86}Sr$ is observed among the different freshwater inputs, with only the dominant input forming the appropriate endmember for the $^{87}Sr/^{86}Sr$ vs. salinity trend. Albarède and Michard (1987) noted a strong dependence of watershed lithology on the $^{87}Sr/^{86}Sr$ of French rivers draining into the Mediterranean, with significant variation in the ratio (0.708719–0.712765) in a relatively small geographic area. We conclude that the samples of unionids analyzed for this study are not representative of the dominant freshwater inputs to the WIS during the Late Cretaceous.

A relatively high $^{87}Sr/^{86}Sr$ ratio (0.709926) in a freshwater unionid bivalve from the late Maastrichtian was measured by McArthur et al. (1994) in their detailed investigation of Sr isotope systematics in the last 30 Ma of the Cretaceous Western Interior of North America. McArthur et al. (1994) did not have the distribution of environments within a single ammonite zone that characterizes the present study, and concluded that

Table 3
Paleosalinities in the WIS

	Open-ocean	Offshore Interior	Nearshore Interior	Brackish	Freshwater
$[Sr]_{sw}:[Sr]_{fw} = 10:1$					
$^{87}Sr/^{86}Sr$	0.707839[a]	0.707808[a]	0.707763[a]	0.707677[a]	0.705517[b]
Salinity (‰)	35[c]	30.8[e]	26.2[e]	20[d]	0
$[Sr]_{sw}:[Sr]_{fw} = 5:1$					
$^{87}Sr/^{86}Sr$	0.707839[a]	0.707808[a]	0.707763[a]	0.707677[a]	0.706597[b]
Salinity (‰)	35[c]	31[e]	26.4[e]	20[d]	0
$[Sr]_{sw}:[Sr]_{fw} = 2:1$					
$^{87}Sr/^{86}Sr$	0.707839[a]	0.707808[a]	0.707763[a]	0.707677[a]	0.707245[b]
Salinity (‰)	35[c]	31.5[e]	27.4[e]	20[d]	0

[a] Mean $^{87}Sr/^{86}Sr$ value in indicated biofacies (Table 2).
[b] Calculated from linear trend of $^{87}Sr/^{86}Sr$ vs. 1/[Sr] using marine and brackish values, assuming surficial estuarine mixing.
[c,d] Assumed marine and brackish salinities.
[e] Equivalent salinities determined from values of 1/[Sr] calculated from $^{87}Sr/^{86}Sr$ vs. 1/[Sr] relationship.

freshwater entering the seaway had a higher $^{87}Sr/^{86}Sr$ ratio than seawater. They further concluded that the analytical uncertainty in the Sr isotope ratios precluded seeing a perceptible effect of less-than-fully-marine salinity on the $^{87}Sr/^{86}Sr$ ratios. We conclude in contrast that the difference between the $^{87}Sr/^{86}Sr$ ratios of the open-ocean and brackish assemblages (ΔSr of $\sim 160 \pm 40 \times 10^{-6}$) is significant.

4.3. Estimating paleosalinities in the Late Cretaceous Western Interior Seaway

Using $^{87}Sr/^{86}Sr$ ratios to estimate paleosalinities requires (1) a significant difference between marine and freshwater Sr concentrations and isotope ratios, (2) knowledge of the endmember values and (3) conservative behavior of Sr with respect to freshwater–saltwater mixing. In such conditions, a plot of $^{87}Sr/^{86}Sr$ vs. 1/[Sr] will yield a straight line between the marine and freshwater endmember values.

In this study, we are unable to define the freshwater endmember using the unionids and the freshwater and marine Sr concentrations are unknown. In the present ocean, the Sr concentration (~ 90 μmol/kg) is 10–100 times greater than riverwater Sr concentrations (Brass and Turekian, 1974; Palmer and Edmond, 1989). Owing to the large difference in Sr concentration of the endmembers, the $^{87}Sr/^{86}Sr$ ratio in water of less than fully marine salinity should approach the

seawater ratio even in water of fairly low salinity (Schmitz et al., 1991). If we use as the marine $^{87}Sr/^{86}Sr$ the value obtained from the average of the open-ocean biofacies samples (0.707839), the lowest $^{87}Sr/^{86}Sr$ measured in the rock leachates as the freshwater $^{87}Sr/^{86}Sr$ (0.707165) and take the difference between the marine and freshwater Sr concentration to be 10:1, within the range of values in the Recent, we obtain a salinity of $\sim 9‰$ for the brackish facies.

The brackish biofacies contains abundant cephalopods that presumably were living in this environment, as suggested by the similarity of their Sr isotope ratios with other organisms from this same environment. Modern cephalopods poorly tolerate salinities below about 20‰ (Forsythe and Van Heukelem, 1987; S. von Boletzky, personal communication 1999). If we take this value (20‰) as a lower limit on the salinity of the brackish biofacies, the dominant freshwater input to the WIS must have had a $^{87}Sr/^{86}Sr$ of ~ 0.70552 if the ratio of marine/freshwater Sr concentration was 10:1 (Table 3). This $^{87}Sr/^{86}Sr$ ratio for the freshwater input is considerably lower than the lowest value observed in the unionid samples (0.707615) and is also lower than the lowest value measured in the rock leachates (0.707165).

Indeed, if we take the value of 0.707165 as representative of the freshwater input to the WIS, the ratio of marine/freshwater Sr concentration required to produce 20‰ salinity in the brackish
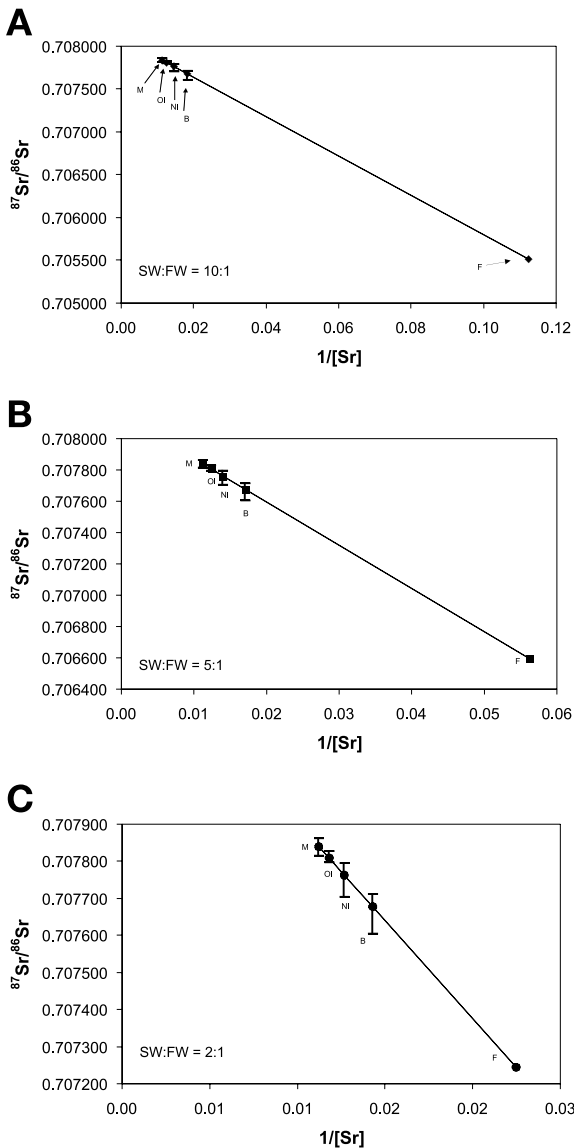
Fig. 5. $^{87}Sr/^{86}Sr$ ratio vs. 1/[Sr] for mixtures of seawater and freshwater with different Sr concentrations. All plots use the present-day Sr concentration of ∼90 μmol Sr/kg in 35‰ seawater (Brass and Turekian, 1974) and a $^{87}Sr/^{86}Sr$ ratio of 0.707839, the average measured value of the open-ocean samples (Table 2). The plots are generated for varying Sr concentrations of freshwater relative to seawater, and lines are fit through the marine (M) and brackish (B) data, assuming that the latter represents 20‰ salinity. (A) [Sr]$_{sw}$:[Sr]$_{fw}$ = 10:1. (B) [Sr]$_{sw}$:[Sr]$_{fw}$ = 5:1. (C) [Sr]$_{sw}$:[Sr]$_{fw}$ = 2:1. The offshore Interior (OI) and nearshore Interior (NI) data are plotted on the lines as well. The plotted points represent the means of the $^{87}Sr/^{86}Sr$ in a given biofacies and the vertical bars denote the range of values for each group.

facies is ∼2:1. By fixing the brackish salinity at 20‰, the estimated salinities for the nearshore and offshore Interior facies are 26.2–27.4‰ and 30.8–31.5‰, respectively (Table 3). Fig. 5a–c shows the extrapolated values of the freshwater $^{87}Sr/^{86}Sr$ ratios for mixing lines that assume different ratios of marine/freshwater Sr and use our observed average $^{87}Sr/^{86}Sr$ of the open-ocean biofacies (mean = 0.707839, Table 2) and the constraint that the brackish biofacies (mean $^{87}Sr/^{86}Sr$ = 0.707677, Table 2) must be at least 20‰ salinity. Values for the freshwater $^{87}Sr/^{86}Sr$ range from 0.707245 to 0.705517 for marine/freshwater Sr ratios of 2:1 to 10:1 (Table 3). We note that even lower freshwater $^{87}Sr/^{86}Sr$ values would be required if the brackish biofacies corresponded to salinities greater than 20‰.

The explanation most consistent with the data is that the mixing curve suggested by the trend in $^{87}Sr/^{86}Sr$ in the different biofacies has a freshwater endmember that is Sr-rich compared with modern river water. One possibility is that the rocks being weathered at that time were carbonate-rich, with abundant marine fossils and Sr concentrations and isotope ratios much like those of the rock samples analyzed (5800–7000 ppm Sr; Table 2). However, as noted above, if the Sr carried by rivers entering the Late Cretaceous WIS had a $^{87}Sr/^{86}Sr$ typical of the lowest value added in our leaching experiments of the matrix samples (0.707165, Table 2), the riverine Sr concentration must have been about half that of seawater to produce brackish salinities of at least 20‰. There is no evidence in the Recent of such Sr-rich freshwaters.

An alternative explanation is that the brackish WIS environment is produced by the subterranean mixing of groundwater and seawater, with the Sr concentration and isotope ratio of the brackish water resulting from both mixing and reaction (i.e. leaching or dissolution of the aquifer solids). Basu et al. (2001) have documented substantial elevations (factors of 10) of Sr even in low-salinity groundwater from the Bengal Basin, relative to concentrations in the Ganges and Brahmaputra rivers. Similar elevations were seen in Ra (Sarin et al., 1990). Groundwater inputs are being increasingly recognized as important in af-

fecting coastal waters of the modern ocean. Moore (1996), for example, documented high activities of Ra in shallow shelf waters of the South Atlantic Bight (eastern USA), relative to open-ocean values. Although the various Ra isotopes can be introduced to groundwater via recoil associated with their production (Krishnaswami et al., 1982), Shaw et al. (1998) documented that Ba, which has no comparable input mechanism, is also enriched in the South Atlantic Bight. Indeed, Basu et al. (2001) argued that groundwater inputs of Sr are important in the oceanic mass balance of Sr.

Moore (1999) suggested the concept of a 'subterranean estuary' in which groundwater and saltwater mix and interact with aquifer solids to elevate concentrations of elements such as Ra and Ba. Bryant et al. (1995) documented $^{87}Sr/^{86}Sr$ values in mollusks collected in the Mississippi Sound that fell off the mixing line determined by seawater and the Mississippi River. They attributed these results to local input of groundwater whose Sr isotope composition was imprinted by reactions with older, carbonate-rich rocks. We speculate that such a process may have been operating in the Late Cretaceous WIS as well. By this mechanism, the brackish facies represents an environment in which Sr-rich groundwater and seawater mixed in the subterranean estuary and the water was discharged with a greater-than-expected Sr concentration, relative to classical estuarine mixing of freshwater and seawater in a surficial estuary.

We can use our data to test the possibility that reactions in the subterranean estuary can produce the Sr concentration and isotopic composition of the brackish biofacies (Fig. 6). We assume that the rocks supplying Sr to solution during mixing of seawater and groundwater have a Sr concentration and isotopic composition typical of the Elk Butte and Mobridge members. The Sr concentrations are ~ 7400 ppm (85 μmol Sr/g) in the Elk Butte and ~ 5800 ppm (66 μmol/g) in the Mobridge (Table 2). The leaching experiments that released significant quantities of Sr from this material yielded $^{87}Sr/^{86}Sr$ in solution of 0.707165 for the Elk Butte and 0.707500 for the Mobridge material (Table 2). Although we do not

know the Sr concentration or isotopic composition of fresh groundwater that might have mixed with seawater in the subterranean estuary of the WIS, we take as a starting point for calculation [Sr] of 1 μM (average of modern rivers) and $^{87}Sr/^{86}Sr$ of 0.707848 (average of unionid values, Table 2). If we mix this water with seawater of 90 μM Sr and $^{87}Sr/^{86}Sr$ of 0.707839 (average of open-ocean biofacies, Table 2) to produce 20‰ brackish water, we can calculate the amount of Sr that must have been added to solution to produce the isotopic signature recorded in the brackish biofacies ($^{87}Sr/^{86}Sr = 0.707681$, Table 2). The $^{87}Sr/^{86}Sr$ of the brackish mixture is given by:

$$R_{mix} = \frac{[Sr]_{fw} \cdot f_{fw} \cdot R_{fw} + [Sr]_{sw} \cdot (1-f_{fw}) \cdot R_{sw} + [Sr]_{rock} \cdot R_{rock}}{f_{fw} \cdot [Sr]_{fw} + (1-f_{fw}) \cdot [Sr]_{sw} + [Sr]_{rock}} \quad (1)$$

where $R_{mix,fw,sw,rock}$ are the $^{87}Sr/^{86}Sr$ in the brackish water, freshwater, seawater and released from aquifer solids, respectively, f_{fw} is the fraction of freshwater in the brackish mixture (0.43 for a salinity of 20‰), and $[Sr]_{fw,sw,rock}$ are the μmol Sr in freshwater, seawater and released from aquifer solids per kg of water. Solving for $[Sr]_{rock}$ using the leached $^{87}Sr/^{86}Sr$ from Elk Butte or Mobridge Member material produces the $^{87}Sr/^{86}Sr$ observed in the brackish facies by adding ~ 16 μmol Elk Butte Sr or ~ 45 μmol Mobridge Sr per kg of groundwater. If the freshwater endmember has a lower $^{87}Sr/^{86}Sr$ ratio than the average of the unionids used in the calculation, the amount of Sr released from the rocks to yield the $^{87}Sr/^{86}Sr$ of the brackish water would be less. However, the difference is not large (only $\sim 4\%$ if the freshwater $^{87}Sr/^{86}Sr$ is taken to be 0.707165) because the 'seawater' and 'rock' terms dominate in Eq. 1.

The Elk Butte and Mobridge Member rocks are Sr-rich, and as a consequence the calculated additions of Sr from them to the groundwater require leaching from only about 0.2–0.6 g of rock per kg of groundwater, using the rock Sr concentrations of Table 2. An aquifer with about 20% water content (porosity ~ 0.4) would have a rock/water ratio of about 4000 g rock/kg water, and the calculated release of Sr using our data thus represents dissolution of only 0.005–0.015% of the aquifer solids. The resulting Sr concentration in the brackish mixture is ~ 68 μM if Elk Butte

rocks are reacting or ∼ 96 μM if Mobridge material is supplying the Sr. For comparison, simple mixing of freshwater (with 1 μM Sr) with seawater (with 35‰ salinity and 90 μM Sr) to attain 20‰ would yield a Sr concentration of 52 μM.

We note that the processes supplying Sr to the waters of the subterranean estuary need not have been (and likely were not) in steady state. Indeed, the subterranean estuary has been viewed as a dynamic zone that responds to forcing factors such as sea-level change (Moore, 1999). The rocks comprising the coastal aquifer system in the Late Cretaceous WIS were deposited earlier and acquired a lower $^{87}Sr/^{86}Sr$ ratio typical of the ocean when the carbonates they contain were formed. Subsequent diagenenesis, for example during times of rising sea level when the aquifer was progressively invaded by seawater, could have added Sr to the aquifer solids. During periods of lowering sea level, such as the Late Cretaceous, the aquifer salinities lower as the saltwater lens retreats and Sr can be released through dissolution, leaching or desorption.

The application of the subterranean estuary concept to producing brackish salinities in the Late Cretaceous WIS has several implications. First, because the freshwater endmember has little influence on the Sr concentration or isotopic composition of the mixture, it can readily have the isotopic composition suggested by our analyses of the unionids (Table 2, Fig. 6) and indeed, we have used the average unionid $^{87}Sr/^{86}Sr$ value in the calculations above. This implies that the $^{87}Sr/^{86}Sr$ of the freshwater entering the WIS by rivers could well be greater than that of the fully marine waters of the Interior, as suggested by some of our unionid analyses and as postulated by McArthur et al. (1994). We suggest, however, that the Late Cretaceous WIS brackish environment was not created by normal, surficial estuarine mixing of freshwater and seawater, but rather that such mixing took place in the subterranean estuary and the resultant brackish water was discharged to the nearshore environment.

Secondly, mixing in the subterranean estuary produces brackish water that has an isotopically lighter $\delta^{18}O$ signature than fully marine waters (see discussion below). Epifauna and infauna
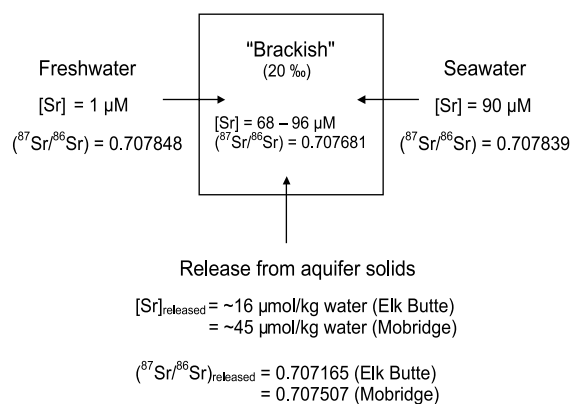

Fig. 6. Schematic diagram showing the production of 20‰ brackish water by mixing of seawater and groundwater in the WIS. The Sr concentrations of the freshwater and saltwater endmembers are assumed to be the present world-average values (Palmer and Edmond, 1989) and the $(^{87}Sr/^{86}Sr)$ are the mean values measured in the unionids and the marine biofacies, respectively. The $(^{87}Sr/^{86}Sr)$ released from the aquifer solids is from our leaching experiments that released significant Sr to solution (Table 2) and the [Sr] released from the aquifer solids is that necessary to produce the observed $(^{87}Sr/^{86}Sr)$ in the brackish biofacies. See text for discussion.

that are most exposed to the seepage of brackish groundwater may thus be isotopically lighter than the nekton (depending on mixing in the water column), as observed by Wright (1987).

Finally, the concept of the discharge of brackish water produced in the subterranean estuary may explain both the periodic kill-offs of fauna and the formation of concretions that preserve them in the WIS (Waage, 1968). Submarine groundwater discharge has the potential to locally alter salinity or dissolved oxygen concentrations and lead to mortality of organisms. Moreover, as noted by Berner (1971) and others, the alkaline putrefaction of decomposing organic matter may cause the precipitation of calcium soaps or salts that subsequently convert to calcitic concretions. Reactions with aquifer solids can release the dissolved Ca necessary for such precipitation, and groundwater flow increases the rate of concretion formation.

We now compare our results with the determinations of strontium isotopes in fossils from the WIS made by Whittaker and Kyser (1993) and McArthur et al. (1994). Whittaker and Kyser (1993) observed that the Sr isotope ratio of the

WIS was similar to the contemporary ocean value except for certain specimens younger than 80 Ma. Such specimens had ^{87}Sr/^{86}Sr ratios (as well as δ^{18}O values; Whittaker et al., 1987) that were lower than open-ocean seawater, and Whittaker and Kyser (1993) attributed these values to dilution with freshwater. In contrast, as noted above, McArthur et al. (1994) analyzed freshwater fauna, including *Unio* sp., from the late Maastrichtian and found higher ^{87}Sr/^{86}Sr values than those of fossils from the WIS. Their interpretation of the variation of ^{87}Sr/^{86}Sr vs. salinity was that the isotope ratio was increasing toward lower-salinity water. McArthur et al. (1994) concluded that, for ratios of marine/freshwater Sr concentrations of ∼100:1 and ∼30:1, variation in the ^{87}Sr/^{86}Sr ratio from 35 to 20‰ salinity could not be detected within the analytical uncertainty of the isotope ratio. This analysis depended on the value of the freshwater ^{87}Sr/^{86}Sr ratio. However, McArthur et al. (1994) did not have the paleoenvironmental control of the present study such that variation in ^{87}Sr/^{86}Sr within a single time period in the WIS could be seen. Indeed, the variation in ^{87}Sr/^{86}Sr observed among the cephalopod fossils collected in the different biofacies sampled in the present study is equivalent to approximately 6 Ma in the Late Cretaceous trend of marine ^{87}Sr/^{86}Sr vs. time as defined by McArthur et al. (1994). Thus our results suggest that the use of the ^{87}Sr/^{86}Sr ratio to assign ages with an accuracy of less than several Ma to fossils from the Late Cretaceous WIS must take into account the variation in paleoenvironment. Similar cautionary notes on the use of the ^{87}Sr/^{86}Sr ratio in chronostratigraphy in settings such as the WIS have been cited by Bryant et al. (1995) and Vaiani (2000).

4.4. Oxygen and carbon isotopes in the WIS

The δ^{18}O values of the samples show significant variation among the different biofacies, with lighter values in the brackish environment (Fig. 3). There is no correlation between δ^{13}C and δ^{18}O ($R^2 = 0.002$). Assuming equilibrium fractionation, the δ^{18}O in the specimens is dependent on both the temperature of formation and the δ^{18}O of the water in which the carbonate forms. Thus the
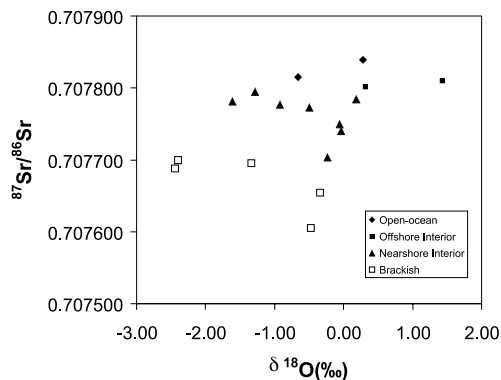

Fig. 7. ^{87}Sr/^{86}Sr vs. δ^{18}O of shell carbonate from the open-ocean and WIS biofacies ($R^2 = 0.14$).

observed trend is consistent with increases in temperature or decreases in δ^{18}O of the water (or both) from marine to brackish environments. There is also a weak trend ($R^2 = 0.14$) of lower ^{87}Sr/^{86}Sr ratios with lighter δ^{18}O (Fig. 7) that is consistent with mixing of seawater and freshwater. The δ^{18}O values of the unionids are indeed light, ∼ −17 to −20‰, although, based on the Sr isotope data, we have argued that these shells were not recording the dominant freshwater input to the WIS.

Earlier studies of δ^{18}O in Cretaceous fossil mollusks (Tourtelot and Rye, 1969; Rye and Sommer, 1980; Wright, 1987) found anomalously high temperatures when paleotemperatures were calculated from oxygen isotope data. If we assume that the marine-to-brackish trend in δ^{18}O solely reflects water temperature changes, we can calculate the water temperature provided the δ^{18}O of the water is known. We have calculated temperatures using a δ^{18}O$_{water}$ = −1‰, a value that is often taken as typical of the Late Cretaceous open ocean (e.g. Fatherree et al., 1998). The aragonite–water equation of Grossman and Ku (1986) was used for all samples except the belemnites, which are calcite and require the calcite–water equation (Craig, 1965; O'Neil et al., 1969). The results show values of 10.4–11.6°C in the offshore Interior waters, 16.3–24.7°C in the shallow, nearshore Interior and 18.7–28.6°C in the brackish environment (Fig. 8). None of the calculated temperatures is as anomalously high as those obtained by Tourtelot and Rye (1969) and Wright (1987),
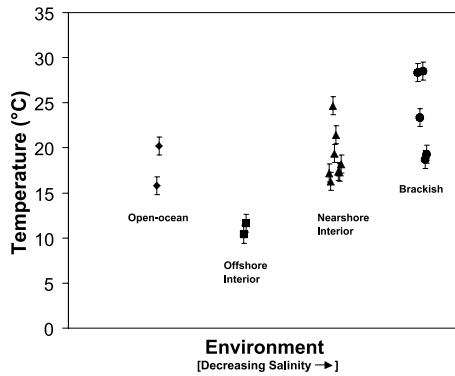
Fig. 8. Water temperatures determined from $\delta^{18}O$ analyses of shell material. Calculations assume that the $\delta^{18}O$ of the water is $-1\%o$ (Fatherree et al., 1998). Values are slightly offset in each group to improve readability.

although the temperature anomalies in their studies generally were found in inoceramid bivalves rather than cephalopods or other mollusks.

Although the calculated oxygen isotope temperatures are reasonable within and between the different habitats, salinity variations in the WIS would likely result in geographic variation of $\delta^{18}O_{water}$. For example, the temperature calculated for the brackish group averages about 23°C, using the estimated open-ocean Cretaceous $\delta^{18}O_{water}$ value of $-1\%o$ (Fig. 8). If the Late Cretaceous WIS had temperatures throughout that were comparable to those calculated for the offshore biofacies ($\sim 12°C$) we estimate that the brackish water had a $\delta^{18}O$ of $\sim -3.5\%o$. In contrast, if the $\delta^{18}O$ of the freshwater that mixed with seawater to produce the water characteristic of the brackish biofacies was as light as the values observed in the unionids (e.g. $-17\%o$), then $\delta^{18}O_{water}$ values of $\sim -8\%o$ can be estimated for salinities of $\sim 20\%o$. This value, when used as $\delta^{18}O_{water}$ in the temperature equation, yields unrealistic (negative) temperatures for the brackish group.

We have suggested that the brackish water of the WIS was produced by groundwater–seawater mixing. To the extent that the carbonate fraction of the aquifer solids was comprised of the fossils of carbonate-secreting organisms, the $\delta^{18}O$ of such material likely reflected equilibrium with seawater at temperatures characteristic of the earlier

WIS, with values ranging, for example, from ~ 0 to $-2\%o$. Because the rock/water ratios are typically high in aquifers (see above), isotopic exchange between the solids and solution in the subterranean estuary would have imprinted the water with the $\delta^{18}O$ characteristic of the dominant oxygen reservoir (i.e. the solids) and the $\delta^{18}O$ values of the brackish mix would be greater than predicted from simple mixing of freshwater with $\delta^{18}O = -17\%o$ and seawater with $\delta^{18}O = -1\%o$. Thus, mixing in the subterranean estuary, coupled with reaction between solution and aquifer solids as implied by the Sr isotope data, produced $\delta^{18}O_{water}$ values that were heavier than those resulting from normal estuarine mixing. We conclude that, as with the Sr isotope ratios, the $\delta^{18}O$ values of the unionid mussels sampled are not representative of the $\delta^{18}O$ of the dominant freshwater source to the WIS.

In her study of $\delta^{18}O$ in different habitat groups in the early Maastrichtian, Wright (1987) noticed that the epifauna was characterized by lighter $\delta^{18}O$ relative to nektonic cephalopods and to infauna. She explained this as the result of a dense, warm water mass formed in the eastern portion of the basin that sank into the basin interior. We do not have sufficient detail in the samples analyzed to represent epifauna and infauna significantly. However, we note that, much like the variation in $^{87}Sr/^{86}Sr$ ratios, the benthos and nekton do not display significant offsets in their $\delta^{18}O$ values in any given habitat group where both types are represented (i.e. nearshore Interior and brackish). It is possible that the water column in these locations was too shallow to show much stratification with respect to temperature or salinity.

5. Conclusions

This study demonstrates that well-preserved, stratigraphically controlled fossils from the Late Cretaceous Western Interior Seaway of North America preserve a record of environmental variation recorded in Sr and O isotopes. The $^{87}Sr/^{86}Sr$ ratios in such samples display significant variation, with values decreasing from 0.707839 ± 0.000024 in fully marine to 0.707677 ± 0.000036

in brackish environments. When compared with the well-documented temporal variation in seawater $^{87}Sr/^{86}Sr$ in the Late Cretaceous, the observed range suggests that using the open-ocean $^{87}Sr/^{86}Sr$ curve to date fossils from the Late Cretaceous WIS should be applied with caution and must take into account the paleoenvironments sampled.

It is not possible to evaluate the freshwater $^{87}Sr/^{86}Sr$ endmember by direct measurement of freshwater mussels, either because the specimens analyzed do not represent the dominant freshwater input to the seaway, or do not correspond to the same time because they lack the biostratigraphic control of the specimens from the Seaway. If a lower limit of $\sim 20\%o$ salinity is set for the brackish biofacies based on salinity tolerances of modern cephalopods (Forsythe and Van Heukelem, 1987), the $^{87}Sr/^{86}Sr$ in the dominant freshwater input to the WIS is estimated to be 0.707245–0.705517 for ratios of the marine/freshwater Sr concentration of 2:1 to 10:1, respectively. The former is more consistent with the likely $^{87}Sr/^{86}Sr$ ratios of rocks being weathered at the time, but requires freshwater with a very high Sr concentration compared with modern rivers. We conclude that the trend in $^{87}Sr/^{86}Sr$ in our samples can be best explained by the mixing of freshwater and seawater in a coastal aquifer and seepage of the brackish water into the nearshore WIS. Through reactions with aquifer solids in this 'subterranean estuary', the resultant brackish water has a Sr concentration enhanced relative to the conservative behavior of Sr seen in the surficial estuarine mixing of freshwater and seawater. Moreover, the $^{87}Sr/^{86}Sr$ ratio of the brackish mix reflects contributions from both seawater and aquifer solids and thus is lower than might be expected from surficial estuarine mixing.

Values of $\delta^{18}O$ decrease from the offshore Interior to brackish biofacies, consistent with a decrease in $\delta^{18}O$ with decreasing salinity and/or increasing water temperature. If the effect is entirely due to temperature, values increase from ~ 10–12°C in the offshore Interior waters of the seaway to 19–28°C in the brackish environment. If the $\delta^{18}O$ of the brackish water is isotopically lighter than the values in Late Cretaceous seawater, as

might be expected from mixing of seawater with isotopically light freshwater, then both the calculated temperatures in the brackish group and the temperature gradient from the offshore Interior to the brackish environment will be decreased. However, as with the Sr isotopes, simple mixing of freshwater with the $\delta^{18}O$ seen in the freshwater mussels with open-ocean seawater to produce 20‰ brackish water yields unrealistic temperatures for the brackish environment. Either the dominant freshwater entering the seaway is heavier than that suggested by the freshwater mussels or the $\delta^{18}O$ of the brackish water has been modified by isotopic exchange with aquifer solids in the subterranean estuary.

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