

Tectonic Forcing of Early to Middle Jurassic seawater Sr/Ca

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ABSTRACT

The Jurassic Period (~201–145 Ma) is marked by fundamental reorganizations of palaeogeography, palaeoceanography, ecosystems and the progressive shift from aragonite to calcite as the favored marine biogenic carbonate polymorph. Sr/Ca ratios of well-preserved Jurassic oysters and belemnites from sections in the UK and Poland demonstrate that the Sr/Ca ratio of seawater varied systematically throughout the Early and Middle Jurassic in parallel with already documented seawater ⁸⁷Sr/⁸⁶Sr. The Sr flux from increased Mid Ocean Ridge activity in the Early Jurassic outbalanced the input of riverine Sr, leading to gradually lower seawater ⁸⁷Sr/⁸⁶Sr, associated with the parallel and strong decrease in seawater Sr/Ca ratios. This downward trend was reversed by enhanced continental input in the Toarcian and Aalenian, but resumed in the Bajocian and continued to the Callovian. Parallel changes of seawater ⁸⁷Sr/⁸⁶Sr and Sr/Ca suggest a common cause for these long-term variations and are best explained by changes in the balance of continental weathering and hydrothermal fluxes. These findings underline the strong control of global tectonic processes on the evolution of bio-mineralization and downplay the role of biomineral evolution in influencing strontium chemistry of seawater in the mid-Mesozoic.

INTRODUCTION

The chemical composition of coeval seawater is of great importance to understanding past environmental perturbations such as occurred from the latest Triassic to the Middle Jurassic. As well as a mass extinction and Oceanic Anoxic Event (OAE), the interval was marked by substantial change in seawater composition, evidenced by the change from Aragonite to Calcite seas (e.g., Stanley and Hardie, 1998; Zhuravlev and Wood, 2009). Important questions remain regarding the extent to which different facets of the Earth System contributed to this chemical evolution, namely the balance of physical and chemical versus palaeobiological factors. Here we present an extensive Sr/Ca data set of Early to Middle Jurassic ostreoids and belemnites, covering the aftermaths of the End Triassic Mass Extinction (ETME) and the Early Jurassic (Toarcian) OAE. This new record is used to identify the predominant controls on the Sr/Ca ratios of Early and Middle Jurassic seawater.

Reconstruction of past seawater Sr/Ca ratios (and Mg/Ca ratios) has mostly been based on analyses of fossil biogenic carbonates (e.g., Steuber and Veizer, 2002; Lear et al., 2003), calcium carbonate veins (CCVs) precipitated in Mid Ocean Ridge basalts (e.g., Coggon et al., 2010; Rausch et al., 2013) and evaporites (Lowenstein et al., 2001). Estimates of seawater Sr/Ca ratios from fossil biogenic calcite can be calibrated through analysis of modern analogues and detailed investigations of the co-variation of geochemical proxies in shell material. Screening for the preservation of the original geochemical signatures is most advanced for fossil materials and large data sets with high fidelity and biostratigraphic resolution can be generated from these with confidence.

MATERIALS AND METHODS

For the present study we discuss 411 oyster values and 665 belemnite values (Fig. 1; supplemental item 1, 2). These comprise ~550 new analyses from stratigraphically well-defined Hettangian to Bajocian successions of the UK, together with published data for these successions and from the Bathonian to Callovian of Poland. Sedimentology, stratigraphy and biozonation for the UK (supplemental item 1) are described in Taylor (1995) and Hesselbo et al., (1998), and for Poland in Wierzbowski et al. (2012). Analytical results were pooled for ammonite biozones to compute statistically meaningful mean values at a temporal resolution of ~1 Myr (Fig. 2a; supplemental item 1,2).

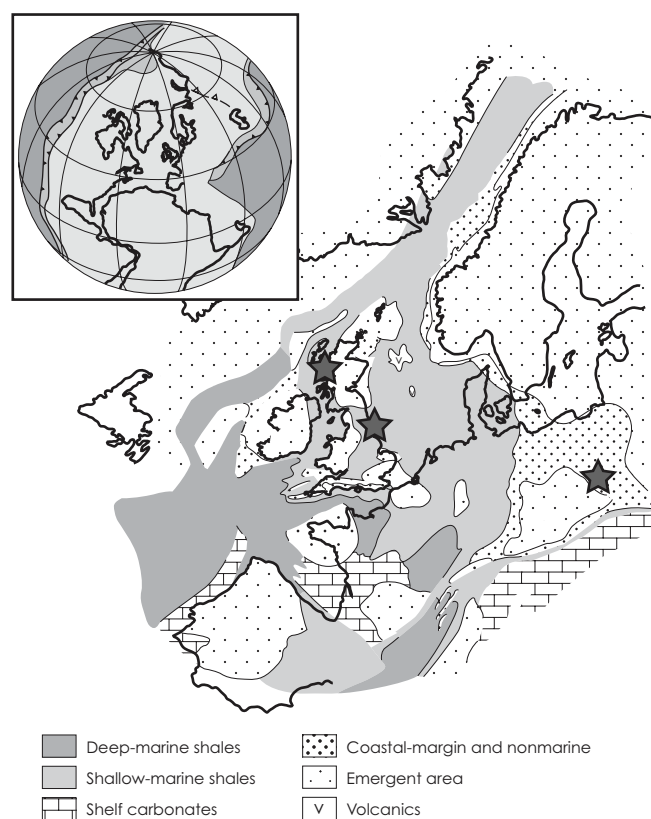


Figure 1. Palaeogeography for the UK and Polish localities (stars represent sampled localities). Inset: Global paleogeography of the Early Jurassic. Modified from Coward et al. (2003).

Shells of the bivalve families Ostreidae and Grypheididae, and the rostra of Belemnitida were screened for the preservation of their ultrastructure using optical and scanning electron microscopy and Mn/Ca ratios. An upper limit for Mn/Ca ratios of 0.46 mmol/mol was adopted from Korte and Hesselbo (2011). Belemnite rostra were broken and sample powders were drilled from translucent, light brown calcite, avoiding regions close to the rim and apical area as well as cloudy, fractured or discolored areas. Shell surfaces of ostreoids were removed with a stainless steel needle and small fragments of deeper layers sampled by

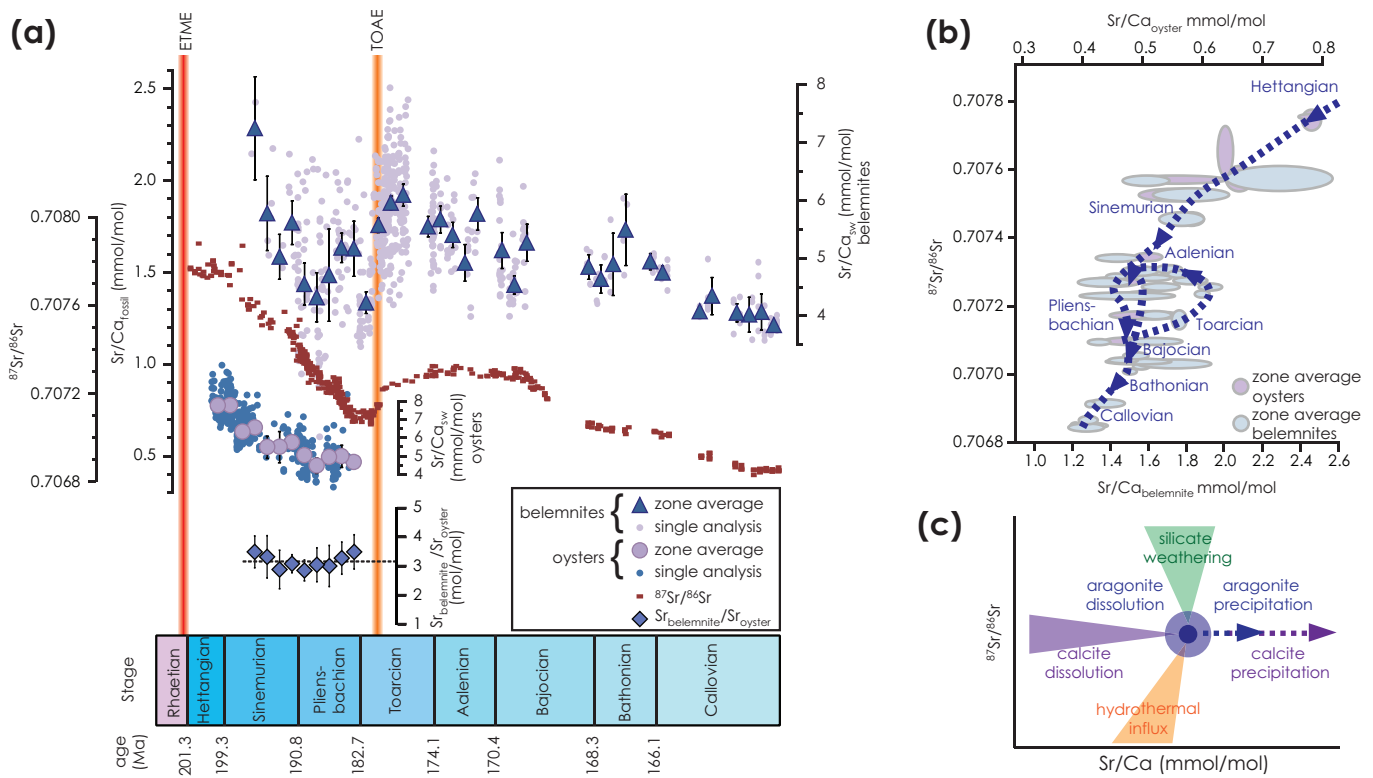


Figure 2. (A) Sr/Ca ratios in oysters and belemnites ($\text{Sr}/\text{Ca}_{\text{fossil}}$), and reconstructed palaeoseawater Sr/Ca ($\text{Sr}/\text{Ca}_{\text{sw}}$) for the Hettangian to Callovian. The ratio of average Sr/Ca ratios in belemnites and oysters in nine ammonite zones is stable within uncertainty. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are from Jenkyns et al. (2002) (Hettangian to Bajocian) and Wierzbowski et al. (2012) (Bathonian and Callovian), and numerical ages from Gradstein et al. (2012). ETME: End-Triassic mass extinction; TOAE: Toarcian oceanic anoxic event. (B) Co-variation of fossil Sr/Ca ratios and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. A long-term decrease for both ratios from the Hettangian to Callovian, interrupted in the Toarcian and Aalenian is visible. (C) Conceptual model of major controlling factors for seawater Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. While the balance of the carbonate fluxes may have a significant influence on the seawater Sr/Ca ratios, strong changes in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios can only be generated by a changing balance of hydrothermal and weathering fluxes.

flaking off shell chips. Sr/Ca and Mn/Ca ratios of the samples were analysed at the University of Copenhagen using a Perkin Elmer Optima 7000 DV ICP-OES. Repeat analyses of JLs-1 and JDo-1 (Imai et al., 1996) yield an accuracy of better than 2% and a 2 sd reproducibility of better than 2.4% for Sr/Ca ratios, 2.8% for Mn/Ca in JDo-1 (0.113 mmol/mol) and 8.1% for Mn/Ca in JLs-1 (0.029 mmol/mol).

RECONSTRUCTION OF PALAEOSEAWATER SR/CA

Sr/Ca in Calcite Fossils

Fossil calcites are promising substrates for reconstruction of past seawater Sr/Ca, because the variability of Sr/Ca within single shells is comparatively low, e.g., ~20% (2 rsd) in modern oysters (Almeida et al., 1998), and sample sizes < 1 mg suffice for analysis. This allows for the generation of large data sets and precise average values. The most important factors influencing Sr/Ca ratios in biogenic carbonates are (I) the composition of the liquid from which they are precipitated, (II) the calcium carbonate polymorph, (III) the species specific fractionation of the Sr/Ca ratio, (IV) metabolic controls on this fractionation factor and (V) water temperature. While the first parameter is the object of this study and the second excluded through the focus on fossil low Mg calcite, the latter three require detailed investigation and are addressed below.

Metabolic Control and Temperature

A temperature control on Sr/Ca ratios of shell carbonates has been proposed on the grounds of inverse correlations of $\delta^{18}\text{O}$

and Sr/Ca ratios (e.g., McArthur et al., 2007; Li et al., 2012 for belemnites). These empirical relationships are in contrast to the positive relationship between $\delta^{18}\text{O}$ and Sr/Ca predicted by theoretical and experimental work (Tang et al., 2008; DePaolo, 2011), but temperature-induced changes in growth rate remain a plausible explanation (e.g., Stoll and Schrag, 2001; Tang et al., 2008; Sosdian et al., 2012).

To address the magnitude of this possible growth rate effect, Sr/Ca and $\delta^{18}\text{O}$ on individual ammonites zones - our chosen temporal resolution - are compared. Only weak correlations are observed for belemnites and ostreoids (Figs. DR1, DR2). Slopes of correlations of Sr/Ca with $\delta^{18}\text{O}$ from $\square 0.2$ to $+0.2$ (mmol/mol)/‰ are not uniform and too small to permit a strong temperature or growth rate control on the observed Sr/Ca variability, and reported past climatic changes are in contrast to observed trends (Jenkyns et al., 2002). For Early Jurassic oysters no relation between Sr/Ca ratios and $\delta^{18}\text{O}$ has been observed (Korte and Hesselbo, 2011). The mean Sr/Ca ratios in well-preserved oysters and belemnites in the overlapping Sinemurian–Pliensbachian period (Fig. 2a) show a constant relative offset despite the contrasting ecology of the two different taxonomic groups (filtrating, benthic and sessile versus predatory and nekto-benthic). We consider it an unlikely coincidence that growth rate or evolutionary effects in both fossil groups caused coeval and equal changes of their Sr/Ca ratios.

Species Specific Fractionation of the Sr/Ca Ratio

From the extensive data set ($n = 267$) of Almeida et al. (1998) for *Crassostrea gigas* an average Sr distribution coef-

efficient of $Sr/Ca_{\text{oyster}} = 0.10 * Sr/Ca_{\text{seawater}}$ was derived. Values of 0.09–0.12 for other modern oysters (Lerman, 1965; Ohde and Kitano, 1984) and the Jurassic bivalve *Praexogyra* (0.12) (Holmden and Hudson, 2003) calculated from smaller data sets are in very good agreement with this value. We therefore adopt a Sr distribution coefficient of 0.10 for modern oysters and use this to estimate seawater Sr/Ca ratios from oyster material. From the relation $Sr/Ca_{\text{belemnite}} = 3.16 \pm 0.16 * Sr/Ca_{\text{oyster}}$ (Fig. 2a) a Sr distribution coefficient of ~ 0.32 can be derived for belemnites. Such a high factor is plausible, considering that Middle Jurassic, aragonitic, ammonites from the Polish Basin have Sr/Ca ratios that are only $\sim 50\%$ higher than those of coeval belemnites from the same localities (Wierzbowski et al., 2012).

LATE TRIASSIC AND JURASSIC SEAWATER SR/CA TREND

The calculated Early to Middle Jurassic seawater Sr/Ca values range from 3.8 to 7.8 mmol/mol (Fig. 3), $\sim 10\text{--}60\%$ lower than the modern counterpart of 8.54 mmol/mol (de Villiers, 1999). Our new late Hettangian Sr/Ca ratio of 7.8 mmol/mol suggests a pronounced decrease in seawater Sr/Ca, when compared to the computed Rhaetian brachiopod seawater value of 9.7 ± 1.1 mmol/mol (Steuber and Veizer, 2002) (Fig. 3). This decreasing trend continued until the Early Toarcian, where it was interrupted by a steep rise, which is also observed in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. Through the Aalenian, both the $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr/Ca ratios remained relatively stable. The Bajocian to Callovian Stages are then characterized by a continuous decrease in Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, the latter reaching their Phanerozoic minimum at the end-Callovian. The positive correlation of the Sr/Ca trends with the coeval seawater $^{87}\text{Sr}/^{86}\text{Sr}$ evidences the close resemblance of the two curves (Fig. 2b); a strong similarity with sea level, however, is not observed (Fig. 3). In general our data agree well with the few estimates for Middle Jurassic seawater Sr/Ca that have been published so far (Holmden and Hudson, 2003; Rausch et al., 2013; Fig. 3).

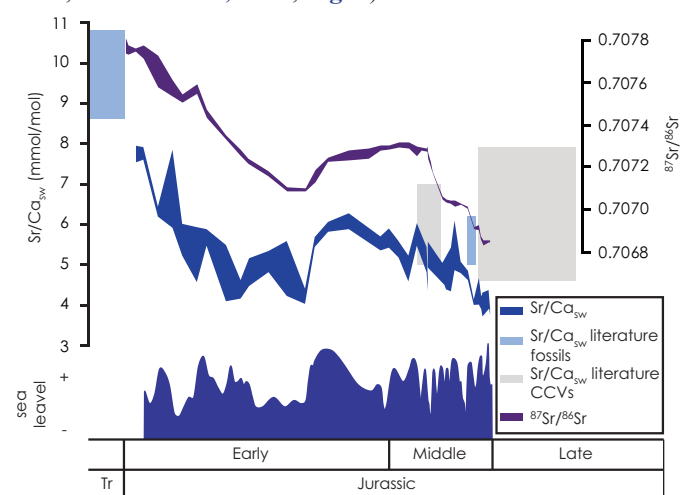


Figure 3. Reconstructed high-resolution seawater Sr/Ca for the investigated period (blue) in comparison with a Rhaetian value from Steuber and Veizer (2002) and an early Callovian estimate from Holmden and Hudson (2003) (blue boxes) as well as data from calcium carbonate veins from Rausch et al. (2013) (gray boxes). The regional relative sea-level curve is from Hesselbo (2008); major rises and falls correspond to global changes. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are from Jenkyns et al. (2002) (Hettangian to Bajocian) and Wierzbowski et al. (2012) (Bathonian and Callovian).

Controls on the Seawater Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ Ratios

Sr and Ca fluxes to the oceans are dominated by hydrothermal circulation at the mid ocean ridges (MOR), the weathering of silicates, and dissolution of carbonates (Fig. 2c), while the fluxes out of the ocean are primarily regulated by carbonate burial (Steuber and Veizer, 2002; Krabbenhöft et al., 2010). The Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the sources and the balance of the associated fluxes determine the Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and the absolute concentrations of Sr and Ca of the oceans (Fig. 2c). Since aragonite has a similar Sr/Ca ratio as the liquid from which it was precipitated and the same $^{87}\text{Sr}/^{86}\text{Sr}$, its formation and dissolution have little impact on the Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of seawater. The net burial flux of calcite can potentially strongly affect the seawater Sr/Ca ratio. Sr is an incompatible element in the calcite lattice, leading to a relative enrichment in the residual liquid when calcite is formed and a relative depletion when it is dissolved. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the seawater, however, is not changed during calcite formation and responds only slightly to the dissolution of calcite, which in general has $^{87}\text{Sr}/^{86}\text{Sr}$ ratios similar to seawater. Significant changes in the burial of (biogenic) calcite, therefore, have the potential to decouple the trends of seawater Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. A significant coupled change of the $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr/Ca in seawater, on the other hand, is induced through the change in the balance of hydrothermal activity and continental weathering or a change in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the weathering flux.

Early and Middle Jurassic Sr Cycle Dynamics

In the absence of a major orogeny in the Early and Middle Jurassic, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the global weathering flux probably remained relatively stable (Jones et al., 1994). Changes in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios can therefore be understood as defined by the change in the relative importance of weathering and hydrothermal inputs of Sr and Ca into the oceans. The ETME is closely linked in time to the onset of basaltic volcanism of the Central Atlantic Magmatic Province (CAMP) (Pálffy, 2003; Ruhl and Kürschner, 2011). Protoatlantic rifting was initiated on the continents, and is manifested later through enhanced mid-ocean ridge activity. This rifting provided less radiogenic strontium to the oceans, leading to a steady decrease of seawater $^{87}\text{Sr}/^{86}\text{Sr}$ until the Pliensbachian–Toarcian boundary (Jones et al., 1994; Jenkyns et al., 2002), accompanied by decreasing Sr/Ca ratios (Figs. 2,3). The reversal of these trends in the Early Toarcian (Fig. 2a) can be explained by enhanced silicate weathering on the continents at this time. This weathering peak was attributed to climatic effects, perhaps induced by the Karoo-Ferrar volcanism (Cohen and Coe, 2007).

The parallel overall downward trends of $^{87}\text{Sr}/^{86}\text{Sr}$ and Sr/Ca ratios in marine biogenic calcite throughout the Early and Middle Jurassic (Fig. 2a) can hardly be associated with prominent dissolution of carbonates with low Sr/Ca and low $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 2c). Only Middle and Late Permian carbonates would have delivered Sr with slightly lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratio than Pliensbachian seawater to the oceans. The Jurassic is marked by a gradual shift to predominant calcite formation with the commencing Jurassic Calcite Sea and the rise of pelagic calcite production (Blättler et al., 2012). The biological imprint on the seawater chemistry through increased calcite production and pelagic burial should have caused rising seawater Sr/Ca ratios during the Early and Middle Jurassic. A strong biological imprint on the observed long-term trends of Sr/Ca ratios over the Hettangian to Callovian can therefore not be postulated. The lack of this biological signature in the evolution of seawater Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios

implies that calcifying organisms of this period did not actively induce the chemical changes in the seawater that ultimately led to the transition of the Triassic Aragonite Sea to Jurassic Calcite Sea.

CONCLUSION

Well-preserved fossil biogenic calcite can be used to estimate the chemical composition of seawater in deep time. The co-variation of Sr/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in fossil carbonates suggests that the strongest forcing on the Sr and Ca cycles in the Early and Middle Jurassic was exerted by the fundamental reorganization of the plate tectonic setting commencing in the latest Triassic. The associated change in the chemical composition of seawater played a strong role in the evolutionary adoption of calcite by Jurassic calcifying organisms, but was likely not caused by a changing style of bio-mineralization and increasing pelagic calcite production.

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