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Demise of Ediacaran dolomitic seas marks widespread biomineralization on the Siberian Platform

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ABSTRACT

The trigger for biomineralization of metazoans in the terminal Ediacaran, ca. 550 Ma, has been suggested to be the rise of oxygenation or an increase in seawater Ca concentration, but geochemical and fossil data have not been fully integrated to demonstrate cause and effect. Here we combine the record of macrofossils with early marine carbonate cement distribution within a relative depth framework for terminal Ediacaran to Cambrian successions on the eastern Siberian Platform, Russia, to interrogate the evolution of seawater chemistry and biotic response. Prior to ca. 545 Ma, the presence of early marine ferroan dolomite cement suggests dominantly ferruginous anoxic "aragonite-dolomite seas", with a very shallow oxic chemocline that supported mainly soft-bodied macrobiota. After ca. 545 Ma, marine cements changed to aragonite and/or high-Mg calcite, and this coincides with the appearance of widespread aragonite and high-Mg calcite skeletal metazoans, suggesting a profound change in seawater chemistry to "aragonite seas" with a deeper chemocline. By early Cambrian Stage 3, the first marine low-Mg calcite cements appear, coincident with the first low-Mg calcite metazoan skeletons, suggesting a further shift to "calcite seas". We suggest that this evolution of seawater chemistry was caused by enhanced continental denudation that increased the input of Ca into oceans so progressively lowering Mg/Ca, which, combined with more widespread oxic conditions, facilitated the rise of skeletal animals and in turn influenced the evolution of skeletal mineralogy.

INTRODUCTION

The appearance and diversification of diverse animal skeletons in the late Ediacaran to early Cambrian (550–520 Ma) suggest an external trigger such as a change in seawater chemistry or rise in predation (Knoll, 2003). Abiotic factors proposed include the increased availability of oxygen (Towe, 1970) or a rise in the concentration of calcium in seawater (Brennan et al., 2004). Uncertainty persists, however, as to both the record of shallow marine oceanic redox during this interval and the relationship to changes in seawater chemistry.

Most early metazoan skeletons were calcium carbonate ($CaCO_3$), forming as aragonite, low-Mg calcite, and high-Mg calcite (Zhuravlev and Wood, 2008), which were also major abiotic precipitates (e.g., Corsetti et al., 2006). By contrast, dolomite [$CaMg(CO_3)_2$] has a highly ordered crystal lattice with slow kinetic growth rates, does not readily form in modern oceans despite supersaturation, and has never been documented as a biomineral. This is of note because early metazoan skeletal clades commonly co-opted carbonate minerals in concert with ambient ocean chemistry driven mainly by inferred changing seawater Mg/Ca (Porter, 2007; Zhuravlev and Wood, 2008).

Here we analyze an underutilized proxy for seawater chemistry—the mineralogy and trace element chemistry of early marine carbonate cements. This avoids bulk sampling which can lead to an averaging or contamination of redox signal, and also allows analysis of shallow carbonate settings where Ediacaran–Cambrian skeletal metazoan biodiversity was highest. Mimetic preservation by dolomite (i.e., retention of original crystallographic orientation) of originally aragonite and/or high-Mg calcite grains (Tucker, 1982; Corsetti et al., 2006) and dolomite cements (Hood and Wallace, 2015) provides evidence that early marine dolomite precipitation dominated Cryogenian to early Ediacaran oceans (ca. 740 to ca. 630 Ma). This is inferred to be due to widespread low-oxygen oceans or stratified oceans and high-Mg/Ca seawater (Hood et al., 2011). The presence of high iron (ferroan) concentrations in early dolomite cements (Hood and Wallace, 2015) and ferroan dolomite concretions in shales further indicates that these oceans were anoxic and ferruginous (Spence et al., 2016). These so-called "aragonite-dolomite seas" (Hood et al., 2011) are thought to have been largely replaced by "aragonite seas" by the early Ediacaran (Corsetti et al., 2006; Hardie, 2003). Here we present evidence, however, for the continuation of aragonite-dolomite seas very close to the Ediacaran–Cambrian boundary on the Siberian Platform. We use the stratigraphic distribution of carbonate minerals and macrofossils, and combine this with petrography, cathodoluminescence (CL) and electronprobe element analysis.

GEOLOGICAL SETTING

We consider three Ediacaran–Cambrian sections on the Yudoma River, Uchur-Maya region, located at (1) the Yudoma River–Maya River confluence, (2) the Nuuchchalakh valley, and (3) the Kyra-Ytyga River (Fig. 1). These sections encompass the entire Ediacaran Yudoma Group, which is unconformably overlain by the lower Cambrian Pestrotsvet Formation (Fig. 1) (see the GSA Data Repository[1]). The Yudoma Group is subdivided into the Aim (stratigraphic thickness 45–95 m) and Ust′-Yudoma (150–205 m) Formations (Khomentovsky, 1985).

The sections record a shelf-edge transect from shore-proximal (Yudoma-Maya confluence) to increasingly distal settings toward the northeast. Sequences are dominated by clastics proximally and carbonates distally (Khomentovsky, 1985). We use sequence stratigraphy to place lithological and macrofossil distribution within a framework of changing

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[1]GSA Data Repository item 2017007, locality information, stratigraphy and correlation, Figure DR1 (stratigraphy and early dolomitzation), methods, elemental compositions, is available online at http://www.geosociety.org/pubs/ft2016.htm or on request from editing@geosociety.org.

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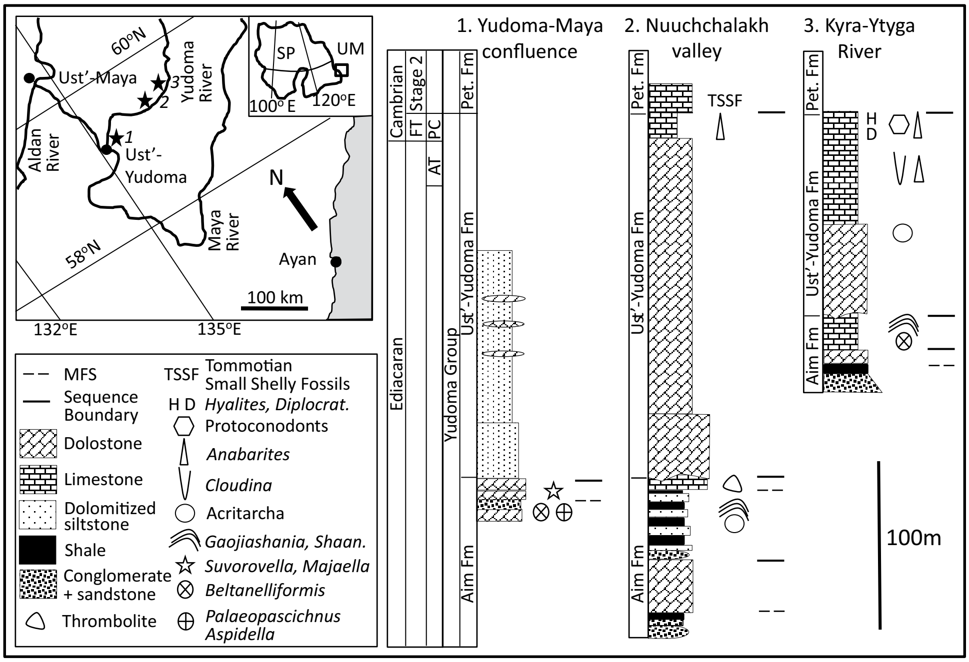

Figure 1. Sections of Ediacaran–Cambrian Yudoma Group on Yudoma River (Russia), with inset map of Uchur-Maya region (UM) within Siberian Platform (SP) (1—Yudoma River–Maya River confluence; 2—Nuuchchalakh valley; 3—Kyra-Ytyga River) and stratigraphic logs and fossil distribution with inferred zones and international stages. Fm—Formation; Pet.—Pestrotsvet; AT—*Anabarites trisulcatus* Zone; PC—*Purella cristata* Zone; FT—Fortunian; MFS—maximum flooding surfaces; *Diplo.—Diplocraterion*; *Shaan.—Shaanxilithes*.

relative sea level (Fig. 1). Aim Formation sequences are composed of transgressive systems tracts (TSTs) of mainly dolomitized siltstones and shales (Fig. DR1 in the Data Repository) followed by thick (up to ~150 m) highstand systems tracts (HSTs) of shallow-marine dolostones and subordinate limestones. Dolomite dominates these successions, except in the mainly HSTs of the Aim Formation and the uppermost 10–70 m of the Ust′-Yudoma Formation where the lithology switches to limestone. Limestone then persists into the lower Cambrian Pestrotsvet Formation (Fig. 1) and continues to dominate throughout the Cambrian over the entire Siberian Platform (Ashtashkin et al., 1991).

MACROFOSSIL DISTRIBUTION

The distribution of fossils is closely related to lithology in these successions (Fig. 1). Macrofossils in the Aim Formation are restricted to TST and early HST facies and consist almost exclusively of soft-bodied biota: *Gaojiashania* (Fig. 2A), *Aspidella terranovica* (Fig. 2B), *Beltanelliformis brunsae, Shaanxilithes*, and *Palaeopascichnus* (Zhuravlev et al., 2009, 2012; Ivantsov, 2016). Thrombolites occur in the HST limestones in the Aim Formation, and microbial structures are abundant throughout the succession, particularly in the dolomitic parts of the Ust′-Yudoma Formation.

Calcareous macrofossils are restricted to carbonate lithologies. *Suvorovella aldanica* (Fig. 2C) and *Majaella verkhojanica* (Khomentovsky, 1985) occur in the latest HST dolostone at the top of the Aim Formation. *Suvorovella* is very similar in size and morphology to *Aspidella* found in the underlying sandstones. The terminal Ediacaran *Cloudina* (Fig. 2E) and *Anabarites* (Fig. 2F) both appear 50 m below the top of the Ust′-Yudoma Formation within the HST limestones (Zhuravlev et al., 2012). Rare dolomitized megasphaeromorph acritarchs occur just below this level (Fig. 2D). In the uppermost 8 m of light-gray dolomitic limestone of the Ust′-Yudoma Formation at Kyra-Ytyga River, a characteristic upper Nemakit-Daldynian *Purella cristata* Zone (i.e., Fortunian, lowermost Cambrian) skeletal assemblage appears, including protoconodonts, anabaritids (Zhuravlev et al., 2012), the hyolithid *Allatheca,* and the trace fossil *Diplocraterion.*

EVIDENCE FOR EARLY DOLOMITIZATION

Several observations suggest that dolomitization of these sections was very early, and rapid. First, siltstone clasts from shallower depths are dolomitized within a non-dolomitized matrix (Fig. DR1C) suggesting dolomitization occurred prior to reworking. Second, organic-walled acritarchs are preserved in folded, but uncompressed, form by encrustations of very fine crystalline dolomite (Fig. 2D). Third, cavities in dolostones are lined with dolomite cements. In a subtidal-intertidal dolomitic grainstone, grains and *Suvorovella* skeletal material are preserved as molds with a pronounced micrite envelope with no breakage or compaction features (Fig. 3A). These molds are now filled with, and encrusted by, isopachous rims of fibrous dolomite and a later generation of dolomite rhombs (Fig. 3B). We infer an originally aragonitic or high-Mg calcite mineralogy for *Suvorovella* and other grains, with rapid dissolution occurring post-micritization as evidenced by the symmetrical growth of early marine cement crusts from micrite substrates.

Dolomite cements are fibrous and radial, with a length-slow character (a high angle between the *c*-axis and the greatest growth direction), and have abundant inclusions which define steep rhombic patterns that follow the crystal form (Fig. 3B). Under CL, crystals show a well-preserved primary growth zonation of multiple dull and bright zones of rhombic patterns confined to individual crystals that do not extend across crystal boundaries (Figs. 3C and 3D). These features confirm the primary marine nature of these dolomite cements (Hood et al., 2011) and are distinct from burial cements and the coarsely recrystalline, dolomitic replacements of primary calcite and aragonite cements characteristic of early Cambrian carbonates (Whittaker et al., 1994; Corsetti et al., 2006).

By contrast, earliest marine intergranular cements from the limestone intervals of the Aim, latest Ust′-Yudoma, and Pestrotsvet Formations are exclusively low-Mg calcite. In the Aim and latest Ust′-Yudoma Formations, these are present as recrystallized spar, but in the Pestrotsvet Formation cements occur as fascicular optic fibrous and prismatic crystals (Fig. 3E), which are largely non-luminescent or with some blotchy bright patches, with limited preservation of primary growth zonation (Fig. 3F).

REDOX OF EARLY CEMENTS

Seawater Mn and Fe content is mainly controlled by redox, and cathodoluminescence zonation follows this chemical variation. Non-luminescent cements have low Fe and Mn; bright luminescent cements have high Mn but low Fe; and dull luminescent cements have moderate values of both Fe and Mn. A non-bright-dull progression is caused by carbonate precipitation in waters with decreasing Eh (Barnaby and Rimstidt, 1989).

Dolomitic isopachous crusts reveal moderate to high levels of Fe, up to 3624 ppm (mean: 1393 ppm, n = 68), and variable levels of Mn, up to

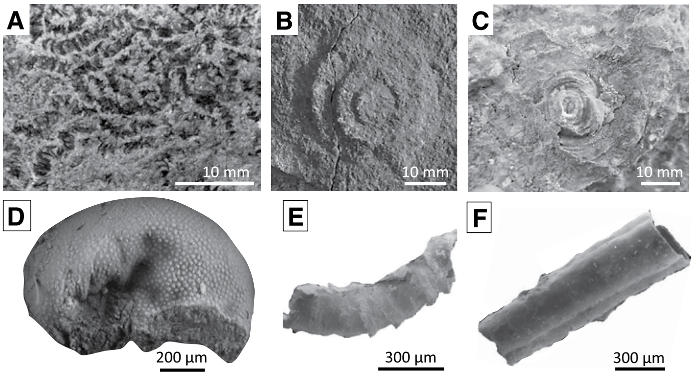

Figure 2. Ediacaran macrofossils of Yudoma Group (Russia). A: *Gaojiashania,* Aim Formation, Nuuchchalakh valley. B: *Aspidella terranovica*, Aim Formation, Yudoma River–Maya River confluence. C: Dolomitized, skeletal *Suvorovella aldanica,* Aim Formation, Yudoma-Maya confluence. D–F: Scanning electron photomicrographs. D: Dolomitized megasphaeromorph acritarch, Ust′-Yudoma Formation, Kyra-Ytyga River. E: *Cloudina* ex gr. *C. riemkeae*, Ust′-Yudoma Formation, Kyra-Ytyga River (photo: Aleksander Fedorov). F: *Anabarites trisulcatus,* Ust′-Yudoma Formation, Kyra-Ytyga River (photo: Aleksander Fedorov).

552 ppm but with the mean below the detection limit (n = 68) (see the Data Repository). Later dolomite rhombs show higher levels of Fe, up to 6127 ppm (mean: 2733 ppm, n = 68), and Mn up to 218 ppm (mean: 113 ppm, n = 68).

By contrast, the recrystallized low-Mg calcite cements from the Aim Formation show far lower levels of Fe (up to 1044 ppm; mean: 501 ppm, n = 10) but moderate to high levels of Mn (up to 602 ppm; mean: 237 ppm, n = 10). Recrystallized low-Mg cements from the Pestrotsvet Formation show very low levels of Fe (mean: 148 ppm) and moderate levels of Mn (mean: 213 ppm) except for a thin, very early Fe- and Mn-enriched crust (Fe, 4084 ppm; Mn, 1219 ppm), and later, burial, ferroan calcite zones.

Such Fe-Mn abundance and behavior indicate that these cements were precipitated in variable redox conditions: dolostones with early dolomite cements under ferruginous, anoxic conditions, but limestones under dominantly non-ferruginous, sub-oxic to oxic conditions.

DISCUSSION

Within any one conformable sequence and traced laterally across equivalent tracts, we see evidence for exclusively aragonite or high-Mg calcite early cement precipitation, as inferred from recrystallized low-Mg calcite spar, only in very shallow proximal settings, but extensive early marine dolomite precipitation at other all water depths. The stratigraphic distribution of early dolomitization with changes in relative sea level suggests a very shallow chemocline below the upper Aim Formation. Limestone is found when accommodation space was decreasing and the sedimentary system switched to dominantly carbonate production (Figs. 1 and 4B). The HST of the Ust′-Yudoma Formation is extensively dolomitized, suggesting that the originally aragonitic sediments were rapidly bathed in anoxic, ferruginous seawater. In such a setting, the originally aragonitic *Suvorovella* was rapidly dolomitized. The oxic layer was therefore restricted to proximal and very shallow coastal waters, where wave action aerated and oxygenated the seawater, or where oceans received oxidized continental waters (Fig. 4B). In the latest Ust′-Yudoma Formation (ca. 545 Ma), all localities show a change from dolostone to limestone lithologies with aragonite or high-Mg calcite early cement precipitation, but no attendant changes in facies. This implies a change in local seawater chemistry rather than another environmental factor such as water depth or hydrodynamic regime.

Given the continued absence of early dolomite in the Siberian Cambrian record, we interpret this as a change in seawater chemistry from aragonite-dolomite seas to aragonite seas, probably controlled by a marked expansion

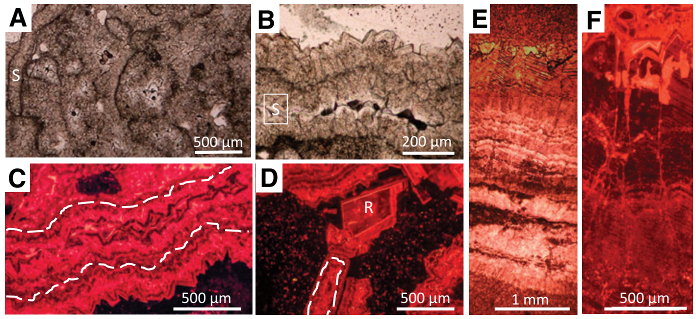

Figure 3. Early marine cements from Ediacaran Yudoma Group (A–D) and Cambrian Pestrovset Formation (E,F) (Russia). A–D: Photomicrographs of isopachous crusts of radial-slow fibrous dolomite, Aim Formation, Yudoma River–Maya River confluence. A: Uncompacted grains and *Suvorovella aldanica* fragment (S). B: Plane-polarized image of symmetrical growth from micrite envelope around *Suvorovella aldanica* (S) fragment, showing rhombic inclusion–rich early zones and clear latest zones. C,D: Cathodoluminescent images showing primary alternating thin dull and bright zones. Dotted line shows approximate outline of *Suvorovella aldanica* fragment. C: Radial-slow isopachous crust. D: Radial slow isopachous crust and later rhombic cements (R). E,F: Recrystallized fibrous calcite cements from Nuuchchalakh valley. E: Multiple generations interlayered with sediment. F: Cathodoluminescent image showing dominant non-luminescence except in latest generations.

of the oxic zone associated with a lowering of the chemocline (Fig. 4A). There is a globally documented general decrease in ferruginous dolomite during the Ediacaran to early Cambrian interval (Corsetti et al., 2003; Spence et al., 2016). Individual basins probably responded variably, with the expansion and contraction of the local oxic chemocline manifest in an oscillation between dominant early dolomite and limestone shallow marine lithologies.

The precise controls on Neoproterozoic dolomite formation are unclear, as there are few experimental data to infer conditions under which mixed Fe-Ca-Mg carbonates form (Spence et al., 2016), but precipitation was probably promoted by ocean anoxia, high Mg/Ca and ferruginous conditions, and

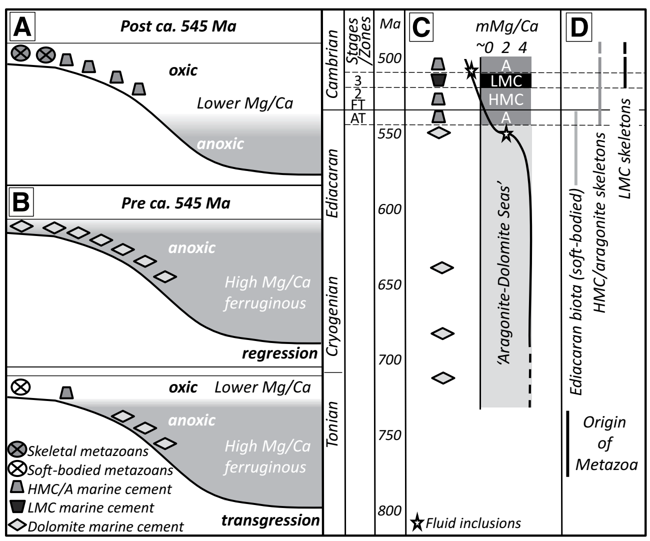

Figure 4. Relationship between evolution of Ediacaran–Cambrian seawater chemistry and skeletal metazoans. A: Post–ca. 545 Ma. B: Pre–ca. 545 Ma during relative sea-level changes, transgression and regression (highstand), with inferred Mg/Ca and redox state. C: Inferred changes in oceanic *molar* Mg/Ca. FT—Fortunian; AT—*Anabarites trisulcatus* Zone; HMC—high-Mg calcite; A—aragonite; LMC—low-Mg calcite. C: Inferred changes in oceanic *molar* Mg/Ca from fluid inclusions (Brennan et al., 2004). D: Biotic response showing first appearance of HMC and A and then LMC skeletal metazoans.

possibly organic matter enrichment, concentrated organic surface carboxyl groups, low marine sulfate, and high pH and alkalinity (Vasconcelos et al., 1995; Hardie, 2003; Roberts et al., 2013; Hood and Wallace, 2015; Spence et al., 2016). These conditions coupled with unstable and reactive phases such as aragonite would further enhance the dolomitization potential of fluids sourced from seawater (Corsetti et al., 2006).

Dramatically enhanced continental weathering occurred during the Neoproterozoic, creating a marked increase in carbonate deposition inferred to be due to a substantial input of Ca^{2+} into seawater (Peters and Gaines, 2012). Fluid inclusion data confirm that seawater Ca^{2+} increased markedly and Mg^{2+} declined slightly during the Ediacaran to early Cambrian, so progressively lowering Mg/Ca by the early Cambrian (Brennan et al., 2004). This reduction in seawater Mg/Ca is also supported by models (Hardie, 2003).

We propose that the Neoproterozoic anoxic aragonite-dolomite seas may have ceased due to increasing oxygenation potentially combined with a reduction in organic matter preservation, as well as an increase in Ca which lowered Mg/Ca. Further decrease in Mg/Ca would have favored another switch to low-Mg calcite marine precipitates, and indeed this is documented in lower Cambrian Stage 3 (525–520 to 514 Ma) (Zhuravlev and Wood, 2008) (Fig. 4C).

We note a biotic response to this proposed progressive decline of Mg/Ca from the Ediacaran to early Cambrian in the Yudoma successions (Fig. 4D). When ferruginous dolomite dominates, we record mainly soft-bodied macrofossils in exclusively very shallow settings inferred to be above the chemocline. The rarity of skeletal biota may be due to restricted habitable space with a seawater chemistry permissive for biomineralization: the slow kinetics of dolomite make it unsuitable to be co-opted as a biomineral. Additionally, the low-Mg calcite cements of the Aim Formation have comparatively high concentrations of Mn and Fe compared to both modern marine cements (Barnaby and Rimstidt, 1989) and those in the Pestrotsvet Formation, suggesting a lower oxidation state than in the earliest Cambrian.

Most skeletal macrofossils (*Cloudina, Anabarites*) appear with the succeeding limestones at the top of the Ust′-Yudoma Formation. This is coincident with the appearance of widespread high-Mg calcite and/or aragonite marine cements. We infer that biomineralization was facilitated by a rise in oxygenation and increased concentration of Ca, which is known to enhance biologically induced calcification (Brennan et al., 2004). Moreover, all known Ediacaran and lowermost Cambrian (Fortunian and Stage 2) metazoans have either aragonitic or high-Mg calcitic skeletal mineralogy. But synchronous with the first low-Mg calcitic marine cements in Cambrian lower Stage 3, we record the first metazoans with low-Mg calcite skeletons (Fig. 4D; Zhuravlev and Wood, 2008).

CONCLUSIONS

We propose that the late Ediacaran to early Cambrian early diagenetic setting underwent step changes coincident with the rise of skeletal metazoans. We document a succession of early marine cements on the Siberian Platform from dominantly dolomite (pre–545 Ma) to aragonite and/or high-Mg calcite (ca. 545 to ca. 525 Ma) to low-Mg calcite (525–520 to 514 Ma). This coincides with the first appearance of aragonite and/or high-Mg calcite skeletons at ca. 545 Ma and low-Mg calcite skeletons at 525–520 Ma. These events may have been facilitated by the rising oxygenation state of the oceans, enabling irrigation of the shallow diagenetic environment, as well as an input of Ca driven by enhanced continental weathering.

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