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Citation for published version:

Wood, R, Zhuravlev, AY, Sukhov, SS, Zhu, M & Zhao, F 2016, 'Demise of Ediacaran dolomitic seas marks widespread biomineralization on the Siberian Platform' *Geology*. DOI: 10.1130/G38367.1

Digital Object Identifier (DOI):

[10.1130/G38367.1](https://doi.org/10.1130/G38367.1)

Link:

[Link to publication record in Edinburgh Research Explorer](#)

Document Version:

Peer reviewed version

Published In:

Geology

Publisher Rights Statement:

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

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13 **ABSTRACT**

14 The trigger for biomineralization of metazoans in the terminal Ediacaran, ~550
15 million years (Ma) ago has been suggested to be the rise of oxygenation or an increase in
16 sea water Ca concentration, but geochemical and fossil data have not been fully
17 integrated to demonstrate cause and effect. Here we combine the record of microfossils
18 with early marine carbonate cement distribution within a relative depth framework for
19 terminal Ediacaran to Cambrian successions on the eastern Siberian Platform, Russia to
20 interrogate the evolution of sea water chemistry and biotic response. Prior to ~545 Ma the
21 presence of early marine ferroan dolomite cement suggests dominantly ferruginous
22 anoxic ‘aragonite-dolomite seas’, with a very shallow oxic chemocline that supported

23 mainly soft-bodied macrobiota. After ~545 Ma, marine cements changed to
24 aragonite/high-Mg calcite, and this coincides with the appearance of widespread
25 aragonite and high-Mg calcite skeletal metazoans suggesting a profound change in sea
26 water chemistry to ‘aragonite seas’ with a deeper chemocline. By early Cambrian Stage
27 3, the first marine low-Mg calcite cements appear coincident with the first low-Mg calcite
28 metazoan skeletons suggesting a further shift to ‘calcite seas’. We suggest that this
29 evolution of sea water chemistry was caused by enhanced continental denudation that
30 increased the input of Ca into oceans so progressively lowering Mg/Ca which, combined
31 with more widespread oxic conditions, facilitated the rise of skeletal animals and in turn
32 influenced the evolution of skeletal mineralogy.

33 **INTRODUCTION**

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

41 Most early metazoan skeletons were calcium carbonate (CaCO_3), forming as
42 aragonite, low-Mg calcite and high-Mg calcite (Zhuravlev and Wood, 2008), which were
43 also major abiotic precipitates (e.g. Corsetti et al., 2003). By contrast, dolomite
44 ($\text{CaMg}(\text{CO}_3)_2$), has a highly ordered crystal lattice with slow kinetic growth rates, does
45 not readily form in modern oceans despite supersaturation, and has never been

46 documented as a biomineral. This is of note because early metazoan skeletal clades often
47 co-opt carbonate minerals in concert with ambient ocean chemistry driven mainly by
48 inferred changing seawater Mg/Ca (Porter, 2007; Zhuravlev and Wood, 2008).

49 Here we analyze an underutilized proxy for seawater chemistry – the mineralogy
50 and trace element chemistry of early marine carbonate cements. This avoids bulk
51 sampling which can lead to an averaging or contamination of redox signal, and also
52 allows analysis of shallow carbonate settings where Ediacaran-Cambrian skeletal
53 metazoan biodiversity was highest. Mimetic preservation by dolomite (i.e. retention of
54 original crystallographic orientation) of originally aragonite/high-Mg calcite grains
55 (Tucker, 1982; Corsetti et al., 2006) and dolomite cements (Hood and Wallace, 2015)
56 provide evidence that early marine dolomite precipitation dominated Cryogenian to early
57 Ediacaran oceans (~740- ~630 Ma). This is inferred to be due to widespread low oxygen
58 or stratified oceans and high Mg/Ca seawater (Hood et al., 2011). The presence of high
59 iron (ferroan) concentrations in early dolomite cements (Hood and Wallace, 2015), and
60 ferroan dolomite concretions in shales further indicates that these oceans were anoxic and
61 ferruginous (Spence et al., 2016). These so-called ‘aragonite-dolomite seas’ (Hood et al.,
62 2011) are thought to have been largely replaced by ‘aragonite seas’ by the early
63 Ediacaran (Corsetti et al., 2006; Hardie, 2003). Here we present evidence, however, for
64 the continuation of ‘aragonite-dolomite seas’ very close to the Ediacaran/Cambrian
65 boundary on the Siberian Platform.

66 **GEOLOGICAL SETTING**

67 We consider the stratigraphic distribution of carbonate minerals and macrofossils
68 across three Ediacaran-Cambrian sections on the Yudoma River, Uchur–Maya Region

69 –1) Yudoma-Maya confluence, 2) Nuuchchakh valley, and 3) Kyra-Ytyga river (Fig.
70 1). These sections encompass the entire Ediacaran Yudoma Group which is
71 unconformably overlain by the lower Cambrian Pestrotsvet Formation (Fig. 1) (*see*
72 *Supplementary Information*). The Yudoma Group is subdivided into the Aim
73 (stratigraphic thickness, 45 - 95 m) and Ust'-Yudoma (150 - 205 m) formations
74 (Khomentovsky, 1985).

75 The sections record a shelf-edge transect from proximal to the shore (Yudoma-
76 Maya confluence), to increasingly distal settings toward the northeast. Sequences are
77 dominated by clastics proximally and carbonates distally (Khomentovsky, 1985). We use
78 sequence stratigraphy to place lithological and macrofossil distribution within a
79 framework of changing relative sea level (Fig. 1). Aim Formation sequences are
80 composed of Transgressive Systems Tracts (TST) of mainly dolomitised siltstones and
81 shales (Fig. DR1), followed by thick (up to ~150 m) Highstand Systems Tracts (HST) of
82 shallow marine dolostones and subordinate limestones. Dolomite dominates these
83 successions, except in the mainly HSTs of the Aim Formation and the uppermost 10–70
84 m of the Ust' - Yudoma Formation where the lithology switches to limestone. Limestone
85 then persists into the lower Cambrian Pestrotsvet Formation (Fig. 1), and continues to
86 dominate throughout the Cambrian over the entire Siberian Platform (Ashtashkin et al.,
87 2003).

88 **MACROFOSSIL DISTRIBUTION**

89 The distribution of fossils is closely related to lithology in these successions (Fig.
90 1). Macrofossils in the Aim Formation are restricted to TST and early HST facies, and
91 consist almost exclusively of soft-bodied biota: *Gaojiashania* (Fig. 2A), *Beltanelliformis*

92 *brunsae* (Fig. 2B), *Aspidella terranovica*, *Beltanelliformis brunsae*, *Shaanxilithes* and
93 *Palaeopascichmus* (Zhuravlev et al., 2009, 2012; Ivantsov, 2016). Thrombolites occur in
94 the HST limestones in the Aim Formation, and microbial structures are abundant
95 throughout, particularly in the dolomitic parts of the Ust'-Yudoma Formation.

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

107 **EVIDENCE FOR EARLY DOLOMITIZATION**

108 Several observations suggest that dolomitization of these sections was very early,
109 and rapid. First, siltstone clasts from shallower depths are dolomitized within a non-
110 dolomitised matrix (Fig. DR1C) suggesting dolomitization occurred prior to reworking.
111 Second, organic-walled acritarchs are preserved in folded, but uncompressed, form by
112 encrustations of very fine crystalline dolomite (Figs. 2D). Third, cavities in dolostones
113 are lined with dolomite cements. In a subtidal-intertidal dolomitic grainstone, grains and
114 *Suvorovella* skeletal material are preserved as molds with a pronounced micrite envelope

115 with no breakage or compaction features (Fig. 3A). These molds are now filled with, and
116 encrusted by, isopachous rims of fibrous dolomite and a later generation of dolomite
117 rhombs (Figs. 3B). We infer an originally aragonitic or high-Mg calcite mineralogy for
118 *Suvorovella* and other grains, with rapid dissolution occurring post-micritisation as
119 evidenced by the symmetrical growth of early marine cement crusts from micrite
120 substrates.

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

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

137 **REDOX OF EARLY CEMENTS**

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

144 Dolomitic isopachous crusts reveal moderate to high levels of Fe, up to 3624 ppm
145 (mean: 1393 ppm, n = 68) and variable levels of Mn up to 552 ppm, but with the mean
146 below the detection limit (n = 68) (*see Supplementary Information*). Later dolomite
147 rhombs show higher levels of Fe (up to 6127 ppm; mean: 2733 ppm, n = 68) and Mn up
148 to 218 ppm, mean 113 ppm (n = 68).

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

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

159 **DISCUSSION**

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

177 Given the continued absence of early dolomite in the Siberian Cambrian record we
178 interpret this as a change in seawater chemistry from 'aragonite-dolomite seas' to
179 'aragonite seas' probably controlled by a marked expansion of the oxic zone associated
180 with a lowering of the chemocline (Fig. 4A). There is a globally documented general
181 decrease in ferruginous dolomite during the Ediacaran to early Cambrian interval
182 (Corsetti et al., 2003; Spence et al., 2016). Individual basins probably responded variably,

183 with the expansion and contraction of the local oxic chemocline manifest in an oscillation
184 between dominant early dolomite and limestone shallow marine lithologies.

185 The precise controls on Neoproterozoic dolomite formation is unclear as there is little
186 experimental data to infer conditions under which mixed Fe–Ca–Mg carbonates form
187 (Spence et al., 2016), but precipitation was probably promoted by ocean anoxia, high
188 Mg/Ca and ferruginous conditions, and possibly by organic matter enrichment,
189 concentrated organic surface carboxyl groups, low marine sulfate, high pH and alkalinity
190 (Vasconcelos et al., 1995; Hardie, 2003; Roberts et al., 2013; Hood and Wallace, 2015;
191 Spence et al., 2016). These conditions coupled with unstable and reactive phases such as
192 aragonite would further enhance the dolomitization potential of fluids sourced from
193 seawater (Corsetti et al., 2006).

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

201 We propose that the Neoproterozoic anoxic ‘aragonite-dolomite seas’ may have
202 ceased due to increasing oxygenation potentially combined with a reduction in organic
203 matter preservation, as well as an increase of Ca which lowered Mg/Ca. Further decrease
204 in Mg/Ca would favor another switch to low-Mg calcite marine precipitates, and indeed

205 this is documented in lower Cambrian Stage 3 (525-520 to 514 Ma) (Zhuravlev and
206 Wood, 2008) (Fig. 4C).

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

217 Most skeletal macrofossils (*Cloudina*, *Anabarites*) appear with the succeeding
218 limestones at the top of the Ust'-Yudoma Formation. This is coincident with the
219 appearance of widespread high-Mg calcite/aragonite marine cements. We infer that
220 biomineralisation was facilitated by a rise in oxygenation and increased concentration of
221 Ca, which is known to enhance biologically-induced calcification (Brennan and
222 Lowenstein, 2004). Moreover, all known Ediacaran and lowermost Cambrian (Fortunian
223 and Stage 2) metazoans have either aragonitic or high-Mg calcitic skeletal mineralogies.
224 But synchronous with the first low-Mg calcitic marine cements in Cambrian lower Stage
225 3, we record the first metazoans with low-Mg calcite skeletons (Fig. 4D; Zhuravlev and
226 Wood, 2008).

227 **CONCLUSIONS**

228 We propose that the late Ediacaran to early Cambrian early diagenetic setting
229 underwent step changes coincident with the rise of skeletal metazoans. We document a
230 succession of early marine cements on the Siberian Platform from dominantly dolomite
231 (pre-545 Ma), to aragonite/high-Mg calcite (~545 to ~525 Ma), to low-Mg calcite (525-
232 520 to 514 Ma). This coincides with the first appearance of aragonite/high-Mg calcite
233 skeletons at ~545 Ma, and low-Mg calcite skeletons at 525-520 Ma. These events may
234 have been facilitated by the rising oxygenation state of the oceans enabling irrigation of the
235 shallow diagenetic environment, as well as an input of Ca driven by enhanced continental
236 weathering.

237 **ACKNOWLEDGMENTS**

238 We thank the Director of Biological Resources and Protected Natural Territories
239 of the Ministry of Nature Protection of the Republic of Sakha (Yakutia), led by Semen
240 Terekhov and the administration of the Ust'Maya Region for logistical support, and
241 Nikolay Atlasov and Elena Aleksandrova for fieldwork support. MZ and FZ received
242 grant support from the National Natural Science Foundation of China, the Ministry of
243 Science and Technology of China (2013CB835006). We thank Aleksander Fedorov,
244 Mike Hall, Chris Haywood, Donald Herd, Amena Al Harthi, Mariam Al Blooshi, Amelia
245 Penny and Fred Bowyer.

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310

311 FIGURE CAPTIONS

312

313 Figure 1. Sections of the Ediacaran-Cambrian Yudoma Group on the Yudoma River, with
314 inset map of Uchur–Maya region (UM) within the Siberian Platform (SP), Russia. 1,

315 Yudoma-Maya confluence. 2, Nuuchchalakh valley. 3, Kyra–Ytyga river. Stratigraphic
316 logs and fossil distribution with inferred zones and international stages. Fm, Formation;
317 Pet., Pestrotsvet. AT = *Anabarites trisulcatus* Zone; PC = *Purella cristata* Zone; FT =
318 Fortunian. MFS = Maximum Flooding Surfaces. *Diplo.* = *Diplocraterion*; *Shaan.* =
319 *Shaanxilithes*.

320

321 Figure 2. Ediacaran macrofossils of the Yudoma Group. **A:** *Gaojiashania*, Aim
322 Formation, Nuuchchalakh valley. **B:** *Aspidella terranovica*, Aim Formation, Yudoma-
323 Maya confluence. **C:** Dolomitised, skeletal *Suvorevella aldanica*, Aim Formation,
324 Yudoma-Maya confluence. **D-F**, SEM photomicrographs. **D:** Dolomitised
325 megasphaeromorph acritarch, Ust-Yudoma Formation, Kyra-Ytyga river. **E:** *Cloudina* ex
326 gr. *C. riemkeae*, Ust'-Yudoma Formation, Kyra-Ytyga river (photo: Aleksander
327 Fedorov). **F:** *Anabarites trisulcatus*, Ust'-Yudoma Formation, Kyra-Ytyga river (photo:
328 Aleksander Fedorov).

329

330 Figure 3. Early marine cements from A-D Ediacaran Yudoma Group and E,F Cambrian
331 Pestrovset Formation. A - D, Photomicrographs of isopachous crusts of radial-slow
332 fibrous dolomite, Aim Formation, Yudoma-Maya confluence. A: Uncompacted grains
333 and *Suvorevella aldanica* fragment (S). B: Plane polarized image of symmetrical growth
334 from a micrite envelope around *Suvorevella aldanica* (S) fragment, showing rhombic
335 inclusion-rich early zones and clear latest zones. C, D, Cathodoluminescent images
336 showing primary alternating thin dull and bright zones. Dotted line shows approximate
337 outline of *Suvorevella aldanica* fragment. C: Radial slow isopachous crust. D: Radial

338 slow isopachous crust and later rhombic cements (R). E,F, Recrystallized fibrous calcite
339 cements from Nuuchchalahk valley. **E:** Multiple generations interlayered with sediment.
340 **F:** Cathodoluminescent image showing dominant non-luminescence except in latest
341 generations.

342

343 Figure 4. Relationship between the evolution of Ediacaran-Cambrian seawater chemistry
344 and skeletal metazoans. **A:** post- ~545 million years (Ma). **B:** pre-~545 Ma during
345 relative sea level changes: transgression, and regression (highstand), with inferred Mg/Ca
346 and redox state. **C:** Inferred changes in oceanic *m*Mg/Ca. FT = Fortunian. AT =
347 *Anabarites trisulcatus* Zone. HMC/A = high-Mg calcite/aragonite; LMC = low-Mg
348 calcite. Fluid inclusion *m*Mg/Ca (Brennan and Lowenstein, 2004). **D:** Biotic response
349 showing the first appearance of HMC/A, then LMC skeletal metazoans.

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351 ¹GSA Data Repository item 2016xxx, xxxxxxxx, is available online at

352 <http://www.geosociety.org/pubs/ft2016.htm> or on request from editing@geosociety.org.