

Baltic Perspective on Early to early Late Ordovician $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ Records and its Paleoenvironmental Significance

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Key Points:

- New paired Baltic carbonate dataset improves Ordovician ^{18}O - and ^{13}C -record
- The new data supports Middle Ordovician climatic cooling
- Regional/intra-basinal consistency of oxygen isotope trends indicate primary nature of paleoenvironmental changes

1 **Abstract**

2 The current study presents new bed-by-bed brachiopod $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ records from Öland,
3 Sweden, which together with previously published data from the East Baltic region,
4 constitutes a high-resolution paired brachiopod and bulk rock carbon and oxygen isotope
5 archive through the Lower to Upper Ordovician of Baltoscandia. This new dataset refines the
6 temporal control on the global Ordovician $\delta^{18}\text{O}$ -trend considerably, improving
7 paleoenvironmental reconstructions through the main phase of the Great Ordovician
8 Biodiversification Event (GOBE). The new brachiopod carbon and oxygen isotope records
9 from Öland display strong similarity with the East Baltic records, elucidating the regional
10 consistency as well as global correlation utility of the ensuing composite Baltoscandian Early
11 to Middle Ordovician carbon and oxygen isotope record. The carbon isotope record from
12 Öland indicates that prominent carbon cycle perturbations are recorded in both brachiopods
13 and bulk carbonates, most notably the MDICE (Mid-Darriwilian Carbon Isotope Excursion).
14 The oxygen isotope record reveals a long-term Early to Late Ordovician trend of increasingly
15 heavier brachiopod $\delta^{18}\text{O}$ values, with a pronounced increase during the Middle Ordovician
16 Darriwilian Age. We interpret this trend as dominantly reflecting a paleotemperature signal
17 indicating progressively cooler Early to Middle Ordovician climate with glacio-eustasy. Our
18 Baltic $\delta^{18}\text{O}$ values are therefore consistent with postulations that the biotic radiations during
19 the GOBE and climatic cooling during the Darriwilian were strongly linked.

20 **Plain Language Summary**

21 Oxygen isotope values obtained from fossil brachiopod shells have traditionally been
22 used as a faithful paleoclimatic proxy to shed light on temperature trends in ancient oceans.
23 However, because brachiopod shells are susceptible to diagenetic overprint after burial,
24 secular oxygen isotope trends derived from these fossils are often questioned – notably the
25 farther one goes back in geological time. In this study, we present temporally well-resolved
26 oxygen isotope data from Early–early Late Ordovician rocks of Öland, Sweden. This interval
27 is important in Earth history as it brackets the greatest marine biodiversification event known
28 in the fossil record and coincides with a global climatic cooling phase (determined based on
29 proxies other than oxygen isotopes). The current study therefore provides an excellent test of
30 the spatial and temporal consistency of the secular Ordovician oxygen isotope trend. We find
31 that although our data is probably affected by diagenetic modification, primary paleoclimatic
32 signals are preserved. Furthermore, as current global Ordovician oxygen isotope records lack
33 sufficient resolution because they comprise data from geographically widely distributed low-
34 paleolatitudes, our new high-resolution dataset tied precisely to conodont
35 biostratigraphy on the bed-by-bed scale from one mid-paleolatitude region, provides
36 significant temporal insights that considerably improves our understanding of the
37 paleoclimatic development during the Ordovician.

38 **1 Introduction**

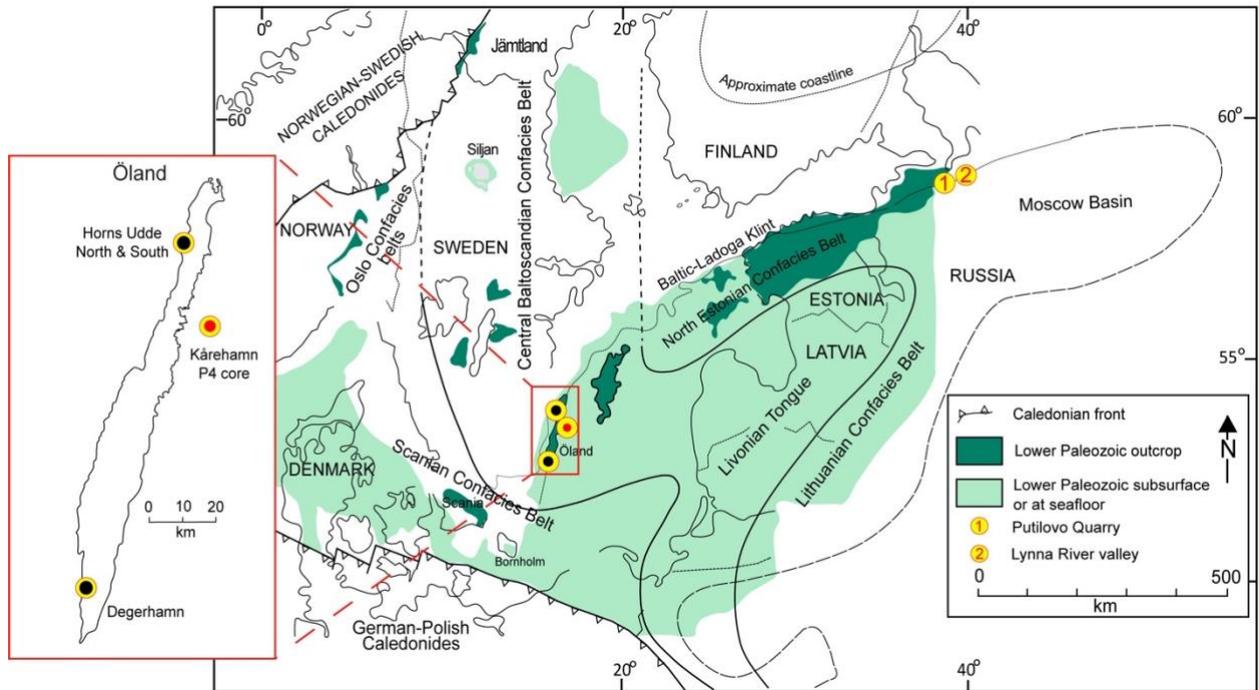
39 The Ordovician Period was characterized by drastic changes in biodiversity levels and
40 ecosystem engineering (Kröger, Franeck, & Rasmussen, 2019; C. M. Ø. Rasmussen, Kröger,
41 Nielsen, & Colmenar, 2019). Elevated changes in plate movements caused fundamental
42 reorganization of the global paleogeographic configuration as continents rifted off the major
43 continent Gondwana and towards lower latitudes (Cocks & Torsvik, 2005; McKenzie,
44 Hughes, Gill, & Myrow, 2014; Torsvik et al., 2012). This prominent dispersal of continents
45 may have constituted a first-order control on species richness as provincialism increased
46 (Valentine & Moores, 1970; Zaffos, Finnegan, & Peters, 2017) and contributed to significant

47 changes in global sea level (Hallam, 1992; Haq and Schutter, 2008), which were exacerbated
48 by transient climatic shifts (Barnes, 2004; Finnegan et al., 2011; Fortey & Cocks, 2005;
49 Quinton, Speir, Miller, Ethington, & MacLeod, 2018; C. M. Ø. Rasmussen et al., 2016; M. R.
50 Saltzman & Young, 2005; Trotter, Williams, Barnes, Lécuyer, & Nicoll, 2008;
51 Vandenbroucke et al., 2010). Furthermore, carbon isotope excursions hint at major
52 perturbations to the global carbon cycle at this time (Ainsaar et al., 2010; Bergström,
53 Saltzman, Leslie, Ferretti, & Young, 2015; Lindskog, Eriksson, Bergström, & Young, 2019;
54 M. M. Saltzman & Thomas, 2012; M. R. Saltzman & Young, 2005), further indicating a
55 coupling between Earth system changes and biodiversity trends during the Ordovician, the
56 most important of which was the Great Ordovician Biodiversification Event (GOBE).
57 Several hypotheses have been put forward regarding potential triggers of the GOBE including
58 environmental perturbations related to asteroid impact on Earth (Schmitz et al., 2019),
59 changes in weathering patterns and nutrient delivery to the oceans due to the Taconic orogeny
60 (Cárdenas & Harries, 2010; Miller & Mao, 1995) and increased ocean-atmosphere
61 oxygenation (Edwards, Saltzman, Royer, & Fike, 2017; Knoll & Carroll, 1999). However,
62 other evidence points to Middle Ordovician climatic cooling and subsequent reduction of
63 physiological stressors on marine organisms as a main driver (Goldberg, Present, Finnegan,
64 & Bergmann, 2021; C. M. Ø. Rasmussen et al., 2016; Trotter et al., 2008). The evidence for
65 long-term Ordovician climate change mainly emanates from oxygen isotope compositions of
66 fossil brachiopods and conodonts, which show a secular trend towards generally heavier
67 values from the Early to Late Ordovician. Although different views have been advanced to
68 explain this trend, such as changing seawater oxygen isotope composition, diagenesis, or
69 climate change (Bergmann et al., 2018; Shields et al., 2003; Trotter et al., 2008; Veizer et al.,
70 1999; Veizer, Godderis, & Francois, 2000; Veizer & Prokoph, 2015), sedimentological,
71 sequence stratigraphical and paleontological data have all supported the notion of shorter-
72 term cooling climate, particularly for the Middle Ordovician interval (Dabard et al., 2015;
73 Ghobadi Pour, Williams, & Popov, 2007; Le Hérisse, Al-Ruwaili, Miller, & Vecoli, 2007;
74 Lindskog & Eriksson, 2017; A. T. Nielsen, 2004; C. M. Ø. Rasmussen, Nielsen, & Harper,
75 2009; J. A. Rasmussen & Stouge, 2018; Turner, Armstrong, Wilson, & Makhlof, 2012).
76 To further test this view during the Early–Middle Ordovician, the current study presents new
77 high-resolution oxygen and carbon isotope data based on fossil brachiopod and bulk
78 carbonate samples from the island of Öland situated in the Baltic Sea (Figure 1). Particularly
79 in the context of the long-term Ordovician oxygen isotope trend, this interval has been
80 somewhat neglected probably due to the much larger perturbations in the $\delta^{18}\text{O}$ -record during
81 the earlier and later parts of the Ordovician (Shields et al., 2003; Veizer & Prokoph, 2015)
82 (Figure 2). Given that both of these intervals may well have been outside the optimal
83 temperature range for most organisms (either too warm or too cool), even a low amplitude
84 change in temperatures during the Dapingian–Darriwilian global stages could have been
85 significant enough for increasing the carrying capacity of ecosystems.
86 We test the regional consistency of previously reported Baltoscandian Ordovician oxygen
87 isotope trends, as well as the global correlation potential of current global C and O isotope
88 curves (e.g. Shields et al., 2003; Veizer et al., 1999; Veizer & Prokoph, 2015) which remain
89 based on sporadic sampling in the Ordovician interval. Thus, the current study enables, for
90 the first time, an intra-basinal comparison of fossil brachiopod carbon and oxygen isotope
91 compositions spanning the Early Ordovician (Floian) to Middle Ordovician (Darriwilian)
92 interval. Additionally, material is sampled through to the Late Ordovician (late Sandbian)
93 from Öland, Sweden, complementing the global Ordovician stable isotope record by adding
94 better temporal resolution tied precisely to conodont biostratigraphy.

95

96 2 Geological Setting

97 The paleocontinent of Baltica comprises most of northern Europe and consists of
98 Archaean and Proterozoic rocks forming the East European craton (Cocks & Torsvik, 2005).
99 The Ural Mountains in the East, the Trans-European Suture Zone to the south-west and the
100 Scandinavian Caledonides in the north-west border this paleocontinent. During the earliest
101 Cambrian, Baltica rifted off the continent of Gondwana, which opened the Tornquist Sea to
102 the southwest and separated Gondwana and Baltica. The drifting phase of Baltica was
103 associated with anticlockwise rotation starting in the mid-Cambrian and lasted into the
104 Middle Ordovician (Torsvik & Rehnström, 2003). Baltica moved from high southerly to
105 intermediate latitudes by the Middle Ordovician and continued towards the paleoequator
106 throughout the Ordovician (Cocks & Torsvik, 2005; Torsvik et al., 1992; Torsvik et al.,
107 2012). In this period, the passive margin was influenced by continental thermal submergence
108 and first and second order sea-level rises, which resulted in the generation of an extensive
109 epicontinental platform sea (A. T. Nielsen, 2004; Torsvik & Cocks, 2016). Baltica remained
110 tectonically calm until the late Middle Ordovician and was bounded by the Tornquist Sea to
111 the southwest and the Iapetus Ocean to the northwest. When the microcontinent Avalonia
112 reached Baltica, it first resulted in the development of an extensive hiatus across the platform
113 as well as Avalonian volcanism, which became evident in the Sandbian (early Late
114 Ordovician), during which a complex of bentonites appeared on Baltica as subduction
115 beneath Avalonia commenced (Huff, Bergström, & Kolata, 1992; Torsvik & Rehnström,
116 2003). This phase likely commenced already during the early Darriwilian as numerous
117 bentonites are found in Scanian shale deposits as well as in contemporaneous carbonate
118 successions in Sweden (Bagnoli & Stouge, 1999; Lindskog, Costa, Rasmussen, Connelly, &
119 Eriksson, 2017). The main feature of the paleocontinent, the Baltoscandian Paleobasin, trends
120 west-southwest (WSW) to east-northeast (ENE). The deposition of the Lower to Middle
121 Ordovician sedimentary successions in the basin took place under shallow to deep
122 epicontinental seawater conditions on the shelf of the stable craton. In this calm period, the
123 sedimentary deposits were extensive and covered the areas of Scandinavia, the east Baltic
124 countries and eastern Russia, Ukraine and northern Poland culminating during the sea-level
125 highstand in the Floian (*evae* transgression; Early Ordovician). Sediment accumulation was
126 slow and mid Cambrian and Lower–Middle Ordovician deposits were condensed, which
127 resulted in the fine clastic and organic-rich Alum Shale Formation that persisted from the
128 Cambrian into the Early Ordovician and the overlying carbonate blanket of the Lower to
129 Middle Ordovician Orthoceratite limestone.
130



131
 132 **Figure 1:** The confacies belts of the Ordovician Baltoscandian Paleobasin with lower
 133 Paleozoic outcrop and subsurface extent shown. Localities discussed in the text are
 134 highlighted. Insert map of Öland show an enlarged view of the current study's sampling sites.
 135 Modified after Jaanusson (1982a).
 136

137 The characteristic lithofacies arrangement of Männil (1966) was combined with the faunal
 138 distribution and divided into three broad confacies belts (Jaanusson, 1976, 1982a, 1995)
 139 (Figure 1). These belts differ from each other in types of sedimentation, faunal diversity and
 140 abundance. Öland experienced little burial, although there is a gradient from thermally
 141 immature rocks in the North (CAI: 1) to early mature rocks (CAI: ca. 1.5) in the South
 142 (Bergström, 1980; Tullborg, Larsson, Björklund, Stigh, & Samuelsson, 1995).
 143 Due to widespread Ordovician outcrops and well-preserved fossils, a relatively simple
 144 tectonic regime with absence of significant thermal alteration, the Baltoscandian Paleobasin
 145 in general, and Öland, represents an ideal site for investigating the Early–Middle Ordovician
 146 carbon and oxygen isotope record.
 147

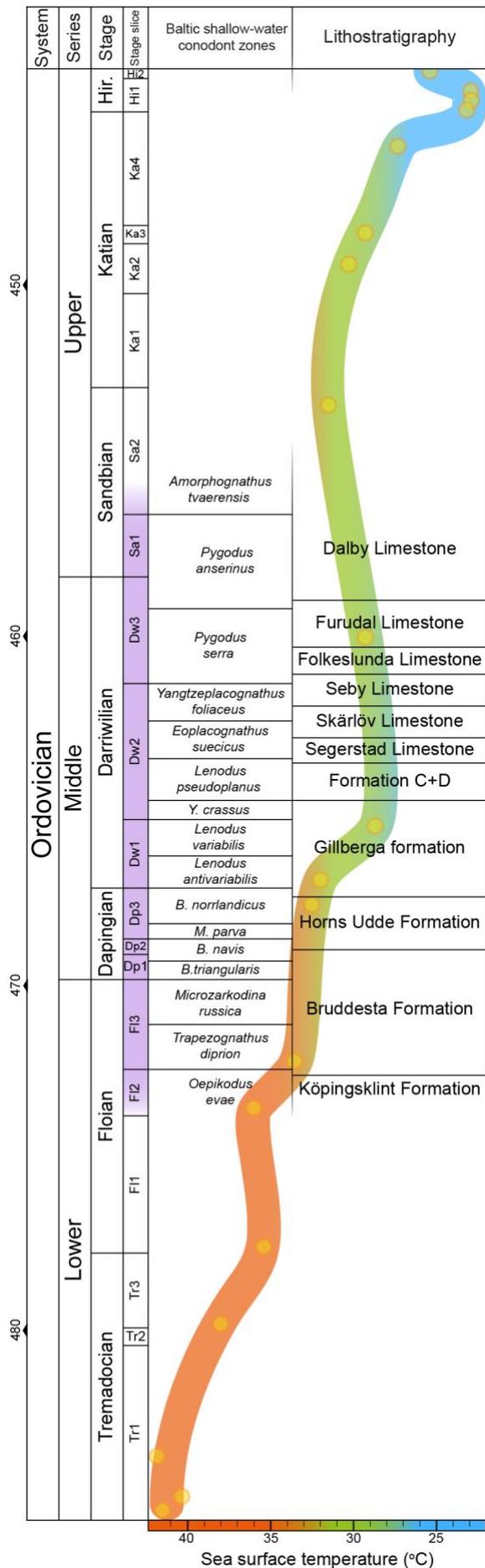
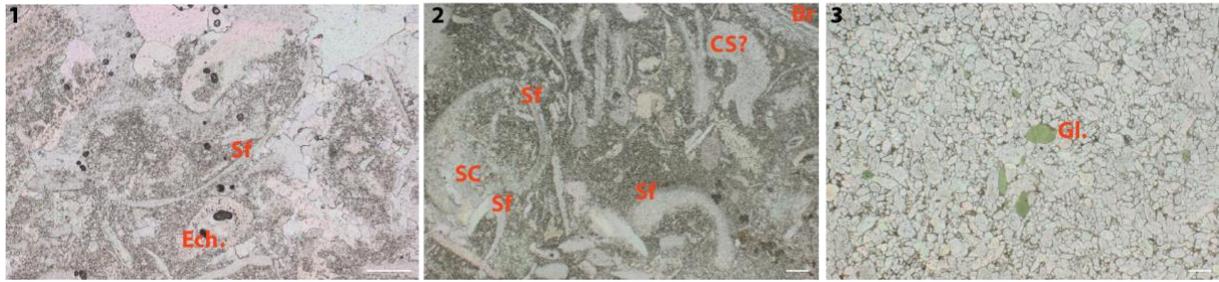


Figure 2: Ordovician chronostratigraphy and conodont $\delta^{18}\text{O}$ apatite paleothermometry. The studied interval is highlighted in purple with the litho- and conodont biostratigraphy of northern Öland shown. Numbers on far left are in million years. The oxygen isotope curve is modified from Trotter et al. (2008). Orange colors denote temperatures above present-day tropical sea surface average, green shading present-day window and blue shading below present-day levels. Note that the optimal temperature is reached during the uppermost Dapingian–lowermost Darriwilian Gillberga Formation on Öland.

3 Lithostratigraphy and biostratigraphy

The Lower to Middle Ordovician (Floian to mid-Darriwilian) Orthoceratite limestone on Öland is composed of green, grey- and red carbonate sedimentary rocks. The limestone is highly condensed, often glauconitic and with low-diversity skeletal composition (Figure 3). Many diastems occur in the succession and these are commonly marked by discontinuity surfaces (Jaanusson, 1961; Lindström, 1979) and less commonly, burrowed and mineralized corrosion hard ground/firm grounds are present (Ekdale & Bromley, 2001). The average carbonate accumulation rate and low siliciclastic input were in the order of 1–4 mm per 1000 years and sea-level fluctuation was a significant factor for the lithofacies development (Chen & Lindström, 1991; Jaanusson, 1982b; Lindskog et al., 2017; Lindström, 1984; Stouge, 2004). According to the confacies belts of Jaanusson (1976, 1995), the island of Öland lies within the Central Baltoscandia confacies belt, where the northern part is closer to the Estonian confacies belt than the southern part of the island (Figure 1).



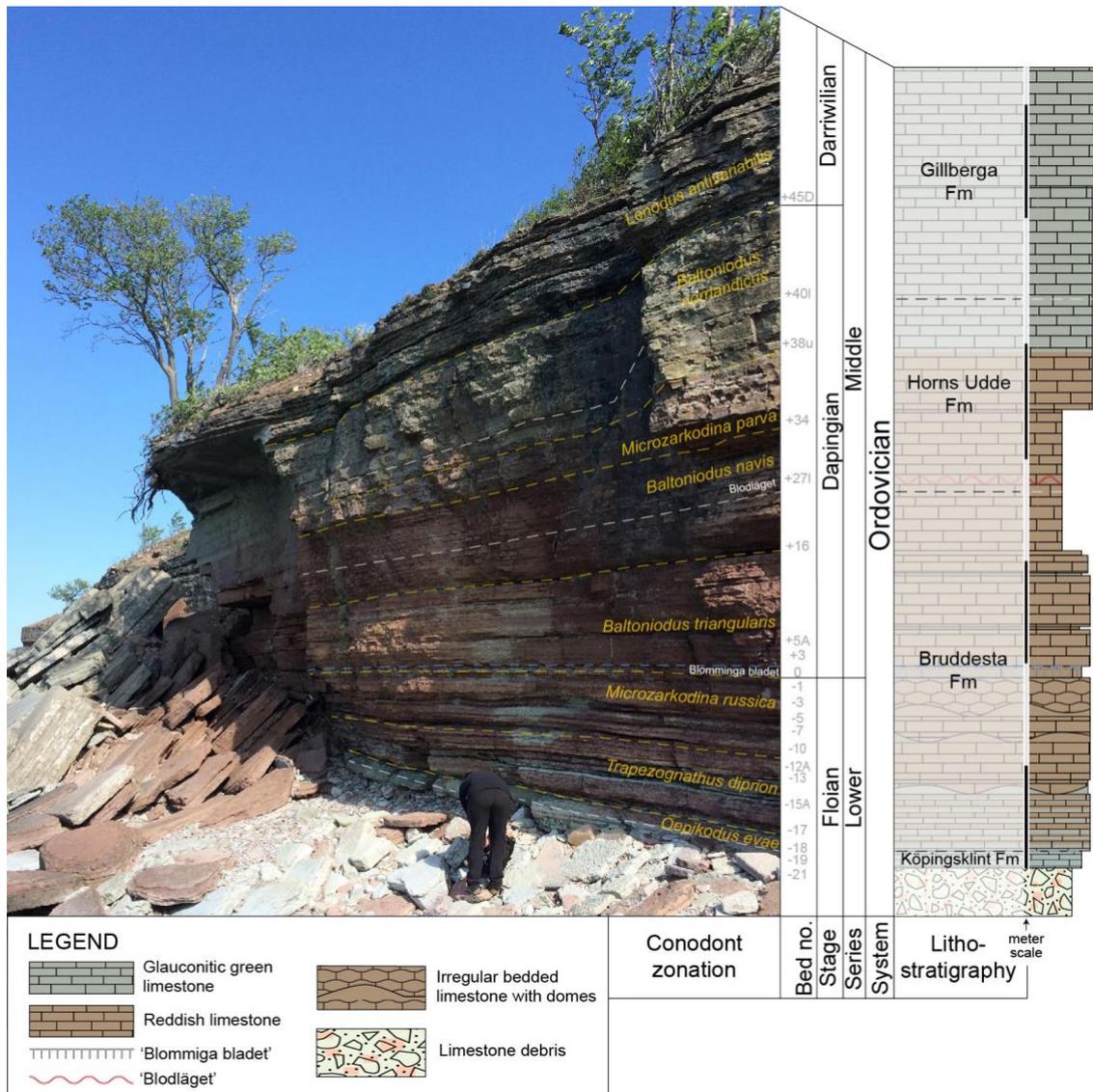
188
 189 **Figure 3:** Photomicrographs showing carbonate microfacies in the study area. Sf =
 190 undifferentiated skeletal fragments, Ech. = echinoderm, SC = sparry calcite, CS? =
 191 cephalopod shell, Br = brachiopod shell, Gl. = glauconite grain. 1. Sample OLK-16,
 192 bioclastic packstone, Sandbian. 2. Sample OLD-1, bioclastic packstone, Darriwilian. 3.
 193 Sample OLH-11, glauconitic grainstone, Floian.

194
 195 The Öland coastal cliff sections, onshore drill cores and the Kårehamn offshore drill core
 196 (Bohlin, 1949, 1953; Stouge, 2004; Wu, Calner, & Lehnert, 2017) are assigned to twelve
 197 lithostratigraphic units, although some of them are informal. The entire upper Lower to lower
 198 Upper Ordovician succession is further biostratigraphically well constrained based on
 199 conodonts. The litho-, bio- and chronostratigraphic schemes used here as reference are shown
 200 in Figure 2 and have been compiled based on information from several sources including
 201 (Bagnoli & Stouge, 1997; Bergström, 1971, 2007; Bergström, Chen, Gutiérrez-Marco, &
 202 Dronov, 2009; Lindström, 1971; Löfgren, 2000, 2003; Stouge, 2004; Stouge & Bagnoli,
 203 1990; Stouge, Bagnoli, & Rasmussen, 2020; van Wamel, 1974; Wu et al., 2017; Zhang,
 204 1998).

205 3.1 Sampled successions

206 3.1.1 Horns Udde North section

207 The Horns Udde North section lies ca. 1.5 km to the northeast of the Cape of Horns
 208 Udde (Figure 1). The complete sedimentary succession comprises Cambrian to lower Middle
 209 Ordovician (Darriwilian) sedimentary rocks. However, the Cambrian and the lowermost
 210 Ordovician (lower Tremadocian) strata in the beach zone are completely covered by rubble.
 211 The section has been described in detail by Lindström (1963), Tjernvik (1956), van Wamel
 212 (1974) and the most recent conodont zonation has been provided by Bagnoli and Stouge
 213 (1997) who sampled the section thoroughly in high resolution (Figure 4). The exposed
 214 succession consists of glauconitic siltstone, nodular grey to red mottled limestone, and grey
 215 to reddish-brown green mottled limestone with marly interbeds and several discontinuity
 216 surfaces. The succession is referred to the Köpingsklint, Bruddesta, Horns Udde and
 217 Gillberga formations respectively, with the Lower–Middle Ordovician boundary placed
 218 within the Bruddesta Formation (Stouge, 2004; van Wamel, 1974) (Figure 4). Dome-like
 219 structures and a pronounced hardground complex known as ‘Blommiga bladet’ (= Flowery
 220 sheet in English; see Figure 4; (Bohlin, 1949; Ekdale & Bromley, 2001) lies within the
 221 Bruddesta Formation. Just above the base of the Horns Udde Formation, three distinct
 222 horizons, collectively 0.2 m thick, and composed of prominent haematite-impregnated
 223 bacterial-like structures occur (named ‘Blodläget’ = ‘Bloody layer’ in English; (Bohlin, 1949;
 224 Stouge, 2004). The uppermost 2.3m of strata in the section belong to the lower sub-unit of the
 225 overlying Gillberga Formation (= Formation A of Stouge (2004)). These beds are composed
 226 of unevenly bedded, glauconitic limestone with some intervals of nodular limestone
 227 interbedded with minor green glauconite shale.



229

230

231 **Figure 4:** Field photo of the locality at Horns Udde North, northern Öland. Lithological
 232 units and boundaries are shown in white. Conodont biostratigraphical zonal boundaries
 233 based on Bagnoli & Stouge (1997) are shown in orange and tied to global stratigraphy (left).

234 The bed numbering system applied in the current study is also shown. The idealized log
 235 (right) shows main lithological features, unit names and thickness of profile. See legend for
 236 details.

237

238 The Lower to Middle Ordovician succession at the Horns Udde North section (Figure 4) was
 239 sub-divided into eight conodont zones by Bagnoli & Stouge (1997) and van Wamel (1974)

240 and partitioned into 101 beds, including sub-beds, from which brachiopods were collected.
 241 This has allowed for detailed bed-by-bed correlation tied accurately to the conodont
 242 biostratigraphy through the roughly nine-meter-thick section.

243

244 3.1.2 Horns Udde South section

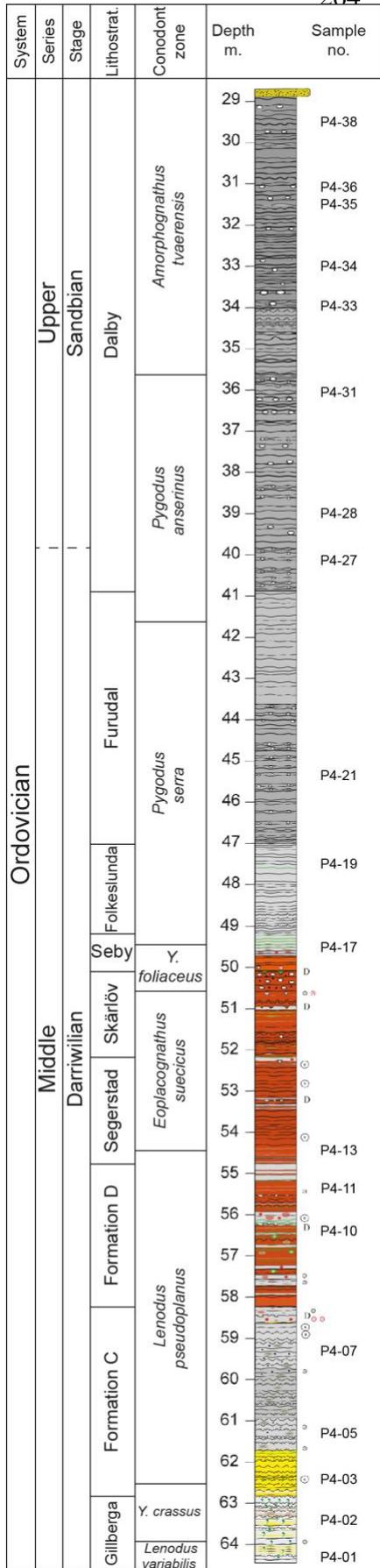
245 This locality is located at the Cape of Horns Udde. The succession covers the top of
 246 the Horns Udde Formation and reaches approximately two meters higher into the overlying
 247 Gillberga Formation than the section North of Horns Udde. The succession starts with the

248 reddish-colored Horns Udde Formation, of which the *B. navis*, *M. parva* and *B. norrlandicus*
 249 conodont zones are found (Figure 5). About midway through the latter conodont zone lies the
 250 formational boundary between the Horns Udde and the Gillberga formations.
 251



252
 253 **Figure 5:** Field photo of the auxiliary section, Horns Udde South, northern Öland.
 254 Lithological units and boundaries shown in white and conodont biozones of Bagnoli &
 255 Stouge (1997) in orange. Bed numbering system is also indicated at unit boundaries.
 256

257 The Gillberga Formation is lithologically similar to the Horns Udde South section (see
 258 above), being greyish–green and unevenly bedded. It is about 4.1 m thick at this locality, thus
 259 potentially yielding up to two meters of new section compared to the section North of Horns
 260 Udde. Towards the top of this section, abundant glauconite grains characterize the rocks.
 261 Although the upper beds did not yield diagnostic conodont elements to aid biostratigraphic
 262 assignment, the associated brachiopod fauna is very characteristic of the lowermost Kunda
 263 Regional Stage. We are therefore confident that these beds are coeval with the *L. variabilis*



Conodont Zone. In this section, the Gillberga Formation was sampled from its base and divided into 32 beds, all of which yielded brachiopods.

3.1.3 Kårehamn P4 drill core

The sampled Kårehamn P4 core was drilled ca. 5 km offshore and to the east of the Kårehamn harbor (Figure 1). The borehole penetrated about 40 m of Ordovician limestone covering Middle and lower Upper Ordovician strata (middle Darrivilian to upper Sandbian) and the succession is typical for the Baltoscandian Paleobasin (Figure 6). The detailed description of the core is not published and work on the succession is still in progress. The base of the drillcore (at -64.5 m below the sea floor) is characterized by the upper part of the glauconite-bearing Gillberga Formation (Figure 6) and is thus, stratigraphically very close to the interval covered by the section at Horns Udde South. It is within the upper *L. variabilis* Conodont Zone and is immediately followed by the *Yangtzeplacognathus crassus* Conodont Zone (Dw2, Middle Ordovician). The strata from -62.9 m to -54.9 m (Formations C and D) are referred to the *Lenodus pseudoplanus* Conodont Zone (with the *Microzarkodina hagetiana* and *M. ozarkodella* subzones, Dw2). The overlying Segerstad and Skärlov limestones (-54.8 to -50.2 m) are referred to the *Eoplacognathus suecicus* Conodont Zone *s.l.* The *Yangtzeplacognathus foliaceus* Conodont Zone (-51.1 to -49.6 m) is recorded from, and characterizes, the strata that are assigned to the Seby Limestone. There is a minor hiatus near the base of the *P. serra* Zone that is marked by a prominent discontinuity surface (D at -50.1 m on Figure 6). The *Eoplacognathus reclinatus* and *E. robustus* subzones of the *Pygodus serra* Conodont Zone are recorded from the Folkelslunda and Furudal limestones.

Figure 6: Lithology and sample levels of the Kårehamn P4 core tied to bio- and lithostratigraphy. The lithostratigraphical names are mainly informal with some of the units either being topo-formations (e.g. Jaanusson, 1960), traditional units (e.g. Bohlin, 1949, 1953), or informal units (Stouge, 2004). Conodont biozonation established by GB and SS. Note the sampling levels for the current study on the far right.

The lower 19.1 m of the core is assigned to the Orthoceratite limestone, which concludes with the Folkelslunda Limestone at -47 m (Figure 3; 6). The *Pygodus serra* Conodont Zone starting from the Folkelslunda Limestone represents the base of the

313 uppermost third of the Darriwilian Stage (Dw3, upper Middle Ordovician). The *Pygodus*
 314 *anserinus* Conodont Zone is recorded from -41.7 m near the base of the Dalby Limestone and
 315 is succeeded by the *Baltoniodus variabilis* Subzone of the *Amorphognathus tvaerensis*
 316 Conodont Zone.

317 The top of the core (-29 m) is within the lower upper Sandbian (Upper Ordovician) Dalby
 318 Limestone (Figure 6). The *Amorphognathus tvaerensis* Zone and the *Baltoniodus variabilis*
 319 Subzone encompass this unit and extend to the top of the core. The base of the Upper
 320 Ordovician Series (Sandbian Stage) is tentatively placed ca. at - 39.8 m in the core (Figure
 321 6). The core was sampled for brachiopods at one-meter resolution and nineteen beds yielded
 322 samples that could be analyzed (Figure 6).

323 3.1.4 Degerhamn Quarry

324 The Degerhamn Quarry (Figs. 1, 7) is an active limestone quarry, which is accessible
 325 by permission of the company that operates the quarry. The complete succession in the
 326 quarry extends down to the Cambrian Alum Shale Formation (Stouge, 2004), however, today
 327 this is covered by water. The exposed portion in the active quarry comprises the Gillberga
 328 Formation, which is composed of bedded, grey limestone superposed by various colors
 329 extending from green, red to violet. The overlying grey to green marker, locally known as
 330 ‘Sphaeronites’ bed, ca. 0.9 m thick, is composed of grainstone to packstone containing
 331 accumulations of ‘*Echinosphaeronites*’ in the middle of the unit (Stouge, 2004; see Figure 7).
 332 The coeval bed in south-central Sweden is referred to as the Täljsten (Eriksson et al., 2012).
 333 The upper part of the quarry consists of grey to mottled red or red-brown limestone.
 334



335 **Figure 7:** Field photo showing the studied outcrop in the Degerhamn Quarry, South Öland.
 336 Lithological units and informal names written in white with corresponding white punctuated
 337 lines. Conodont biozonation, based on Stouge (2004) and Stouge & Bagnoli (2014),
 338 highlighted in orange with corresponding punctuated lines. The stratigraphical position of
 339 the samples obtained from this section is shown in white. The section is ca. 5 meters thick.
 340
 341

342 The exposed succession in the active quarry is of Darriwilian age (Stouge, 2004; Stouge &
343 Bagnoli, 2014). The lower part, composed of grey multi-colored limestone, is referred to the
344 *Lenodus antivariabilis* and *L. variabilis* zones. The green-grey ‘Sphaeronites’ bed is referred
345 to the *L. variabilis* zone up to the accumulation of ‘*Echinospaeronites*’, which is in the *Y.*
346 *crassus* Conodont Zone. The upper part of the succession exposed in the quarry is assigned to
347 the *Microzarkodina hagetiana* subzone of the *Lenodus pseudoplanus* Zone. From this
348 locality, nine samples were collected through the *L. antivariabilis* to *L. variabilis* interval,
349 and they are thus, all placed within the Middle Ordovician Darriwilian Stage (Dw1).
350 Importantly, this locality provides samples from more deeply buried rocks ca. 100 km South
351 of the other three studied localities. Thus, enabling the assessment of potential diagenetic
352 impact by burial depth on carbon and oxygen isotope compositions within the *L.*
353 *antivariabilis* Conodont Zone across Öland.

354 **4 Materials and Methods**

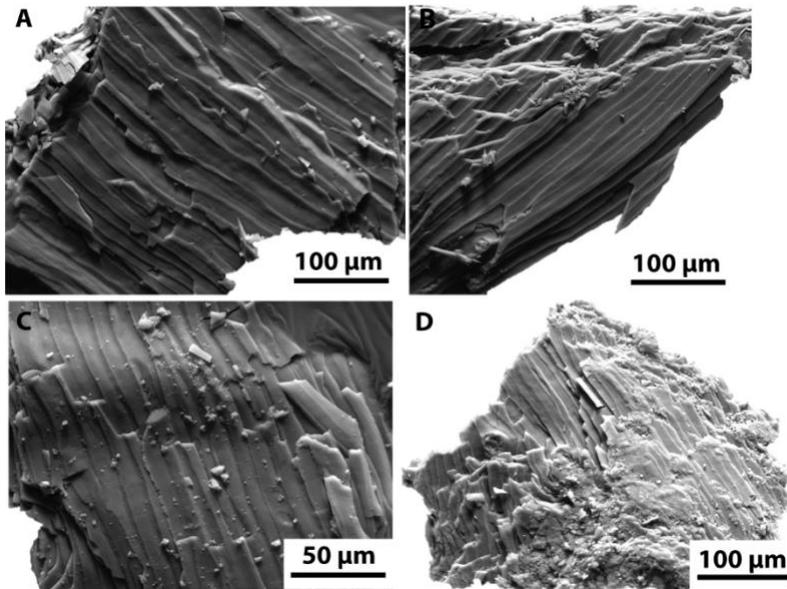
355 Fossil brachiopods (n = 185) and whole rock samples (n = 156) were collected from
356 the localities described above (Figs. 1; 4–7). Samples from the Horns Udde North and South
357 sections were collected bed-by-bed. The distance between the samples from the Degerhamn
358 Quarry section, South Öland, varies, but is about 40 cm. Fossil brachiopods, as well as the
359 bulk carbonate in which they were embedded, were collected at approximately one-meter
360 intervals from the Kärehamn P4 core (Figure 1). All analyzed material are stored at the
361 Natural History Museum of Denmark.

362 4.1 Sampling routines

363 Some brachiopods were sampled multiple times, hence, the total number of processed
364 samples for geochemical and isotopic analysis is 226 brachiopod and 169 bulk rock samples.
365 Brachiopods were cleaned using a brush and inspected for preservation under a binocular
366 microscope. Splinters from the secondary shell layer of brachiopods were collected using a
367 stainless-steel needle. Whole rock powder was extracted from the matrix adjacent to the
368 sampled brachiopod shells using a handheld drill with a diamond-coated steel bit of ca. 1 mm
369 diameter under a microscope. Rock surfaces were mechanically abraded, and powder was
370 extracted from the rock matrix, avoiding weathered parts and calcite veins.

371 4.2 Scanning electron microscopy (SEM)

372 Shell splinters from the secondary shell layer of brachiopods were checked for
373 textural preservation using SEM (Figure 8). Shell splinters were mounted on an adhesive stub
374 and subsequently gold coated before screening. SEM analysis was conducted at the Natural
375 History Museum of Denmark using a FEI Quanta 250 SEM in high vacuum mode.



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Figure 8: SEM images of Ordovician brachiopod shell material. *a)* Sample OLH-19, mid Floian, Köpingsklint Formation, Horns Udde North. *b)* OLH-64, mid Dapingian, Horns Udde Formation, Horns Udde North. *c)* Sample OLK-3, upper Darriwilian, Folkeslunda limestone, Kärehamn P4 core. *d)* OLH-98, lower Darriwilian, Gillberga Formation, Horns Udde North. Specimens *a*, *b* and *c* show texturally well-preserved secondary shell layers and specimen *d* is characterized by partial recrystallization.

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4.3 Carbon and oxygen isotopes

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Carbon and Oxygen isotope measurements of brachiopod shell material and whole rock powder were generated using an IsoPrime triple collector Gas Source Isotope Ratio Mass Spectrometer with a Multiflow unit at the University of Copenhagen following the procedures outlined by Ullmann et al. (2013). In summary, about 0.8 mg of sample material placed in glass vials were dissolved with ca. 0.05 ml of >100% concentrated phosphoric acid (H_3PO_4) and left to react for 90 minutes at 70°C. $\delta^{13}C$ and $\delta^{18}O$ compositions were measured from the resulting carbon dioxide. Data were corrected for weight-dependent biases by using the University of Copenhagen in-house reference standard – LEO (finely-crushed Carrara marble). Reproducibility of the measurements, as determined from the standard deviation of LEO is better than 0.1‰ for $\delta^{13}C$ and $\delta^{18}O$.

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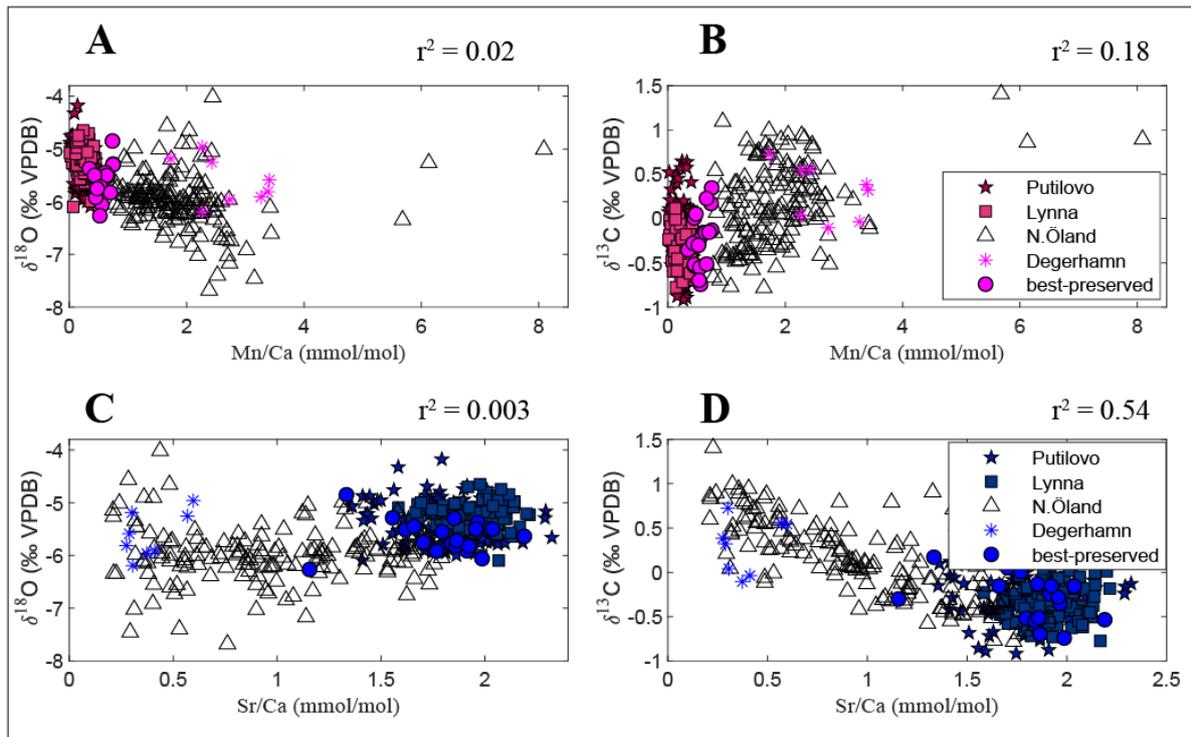
4.4 Element/Ca ratios

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Element/Ca ratios (Sr/Ca and Mn/Ca) were measured from the reacted carbonate remains of brachiopod subsamples using an Agilent 5110 VDV Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) at the University of Exeter, Penryn Campus following the procedure outlined in Ullmann et al. (2020). Accuracy of the analysis was controlled through multiple analyses of a synthetic quality control solution – BCQC (n = 4) and the international reference materials – AK (n = 8) and JLs-1 (n = 12). Analytical bias, determined via the deviation of measured element/Ca ratio from the expected value in the gravimetrically prepared quality control solution – BCQC, was $\leq 0.5\%$ for each of the reported element/Ca ratios.

406 **5 Results**

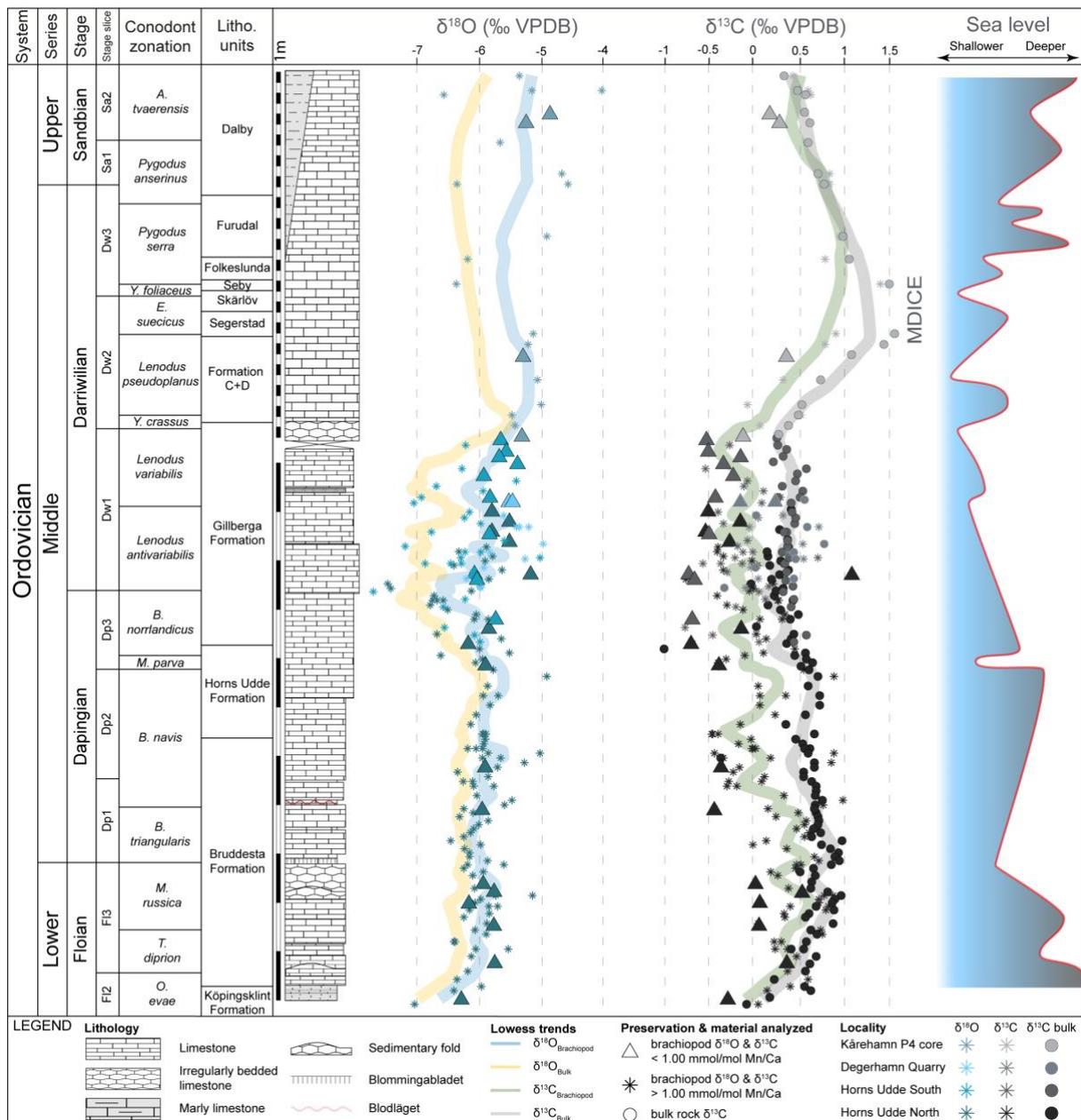
407 All carbon and oxygen isotope compositions of brachiopods and bulk rocks and their
 408 relations to their element/Ca ratios are summarized in Figure 9 and all data are presented in
 409 Supplementary information Figure 2, as well as the Supplementary datafile. In Figure 9, the
 410 new data are plotted together with literature data from well-preserved coeval samples from
 411 the eastern part of the Baltoscandian Paleobasin (St. Petersburg region, Rasmussen et al.,
 412 2016).
 413



414 **Figure 9:** Scatter plots showing correlation between element/Ca ratios and $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$
 415 values of investigated brachiopods in the current study and the east Baltic dataset (Putilovo
 416 and Lynna, C. M. Ø. Rasmussen et al., 2016). Element/Ca data interpreted as best-preserved
 417 if $\text{Mn}/\text{Ca} \leq 1$. N. Öland = North Öland. Values of r^2 only refer to the Öland dataset.
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419 **5.1 Carbon isotopes**

420 Carbon isotope values of brachiopods ($\delta^{13}\text{C}_{\text{brachiopod}}$) vary between -0.78‰ and
 421 +1.41‰ and whole rock carbonates ($\delta^{13}\text{C}_{\text{bulk}}$) between -1.01‰ and +1.57‰. Both datasets
 422 follow the same general temporal trend (Figure 10). $\delta^{13}\text{C}_{\text{brachiopod}}$ values are generally lighter
 423 than $\delta^{13}\text{C}_{\text{bulk}}$, being offset by approximately 0.5‰. Both $\delta^{13}\text{C}_{\text{brachiopod}}$ and $\delta^{13}\text{C}_{\text{bulk}}$ are
 424 characterized by an increasing trend during the Floian, showing a range of -0.3‰ to +1.0‰.
 425 This is followed by a decline of about 1.7‰ in $\delta^{13}\text{C}_{\text{brachiopod}}$ from the Floian–Dapingian
 426 transition to the end of the Dapingian (*B. triangularis* to the top of the *B. norrlandicus*
 427 conodont zones). For $\delta^{13}\text{C}_{\text{bulk}}$, the decrease is less severe as $\delta^{13}\text{C}$ values only plunge by ca.
 428 0.8‰ before stabilizing in the lower Darriwilian *L. antivariabilis* Conodont Zone to +0.3‰.
 429 Beginning in the *Y. crassus* Conodont Zone, a positive excursion takes place in both
 430 $\delta^{13}\text{C}_{\text{brachiopod}}$ and $\delta^{13}\text{C}_{\text{bulk}}$ which culminates in the middle Darriwilian *E. suecicus* Conodont
 431 Zone. Here, peak values of +1.4‰ and +1.6‰ are recorded for $\delta^{13}\text{C}_{\text{brachiopod}}$ and $\delta^{13}\text{C}_{\text{bulk}}$
 432 respectively. Subsequently, in the upper Darriwilian and Sandbian, $\delta^{13}\text{C}_{\text{brachiopod}}$ and $\delta^{13}\text{C}_{\text{bulk}}$
 433 values decrease by ca. 0.6‰ but remain heavier than during pre-excursion times, ranging
 434 between +0.2‰ and +1.0‰.



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Figure 10: Summary figure showing carbon and oxygen isotope compositions of investigated fossil brachiopods and bulk carbonates through the studied composite interval as well as our interpreted relative sea level curve up through the succession (based on lithology and conodont biofacies (see Bagnoli & Stouge (1996)). The four different localities studied are represented by their own shading. A 10-point Lowess smoothing has been applied to accentuate temporal trends in the dataset using the software OriginPro. Yellow Lowess curve represents bulk oxygen values shown in Supp. Fig 2. A pronounced positive $\delta^{13}\text{C}$ excursion is apparent in both bulk carbonates and fossil brachiopods during the Middle Darriwilian (MDICE), as well as a sustained Darriwilian increase in $\delta^{18}\text{O}$. Note that samples below the operational limit of preservation all fall within the range of the samples above the cutoff limit and that the dataset is not evenly scaled. The mid-Darriwilian to Sandbian part of the figure is vertically stretched due to reduced sampling resolution in this interval.

451 5.2 Oxygen isotopes

452 The oxygen isotope record exhibits a general long-term Ordovician increasing trend
453 and most notably, a sustained rise during the Darriwilian (Figure 10). Brachiopod $\delta^{18}\text{O}$ values
454 are typically offset by +0.6‰ relative to $\delta^{18}\text{O}_{\text{bulk}}$ (see supp. Figure 2) and vary by up to 1‰
455 within individual brachiopod beds. Bulk rock $\delta^{18}\text{O}$ composition ranges from -8.0‰ to -4.9‰
456 in the studied interval and between -7.7‰ and -4.0‰ for brachiopods. The Floian is
457 characterized by an increase of up to 1.8‰ in both brachiopods and bulk rocks, before
458 reverting to background values of about -6‰ at the Floian–Dapingian transition (*B.*
459 *triangularis* Conodont Zone). The Dapingian dataset is characterized by an initial increase of
460 ca. 1.5‰ in both $\delta^{18}\text{O}_{\text{bulk}}$ and $\delta^{18}\text{O}_{\text{brachiopod}}$, followed by a decrease of up to 2‰ reaching into
461 the lower Darriwilian. Within the lower Darriwilian (*L. antivariabilis* Conodont Zone), both
462 brachiopods and bulk rocks display a wide $\delta^{18}\text{O}$ range of 1.9‰ and 3.0‰ respectively.
463 Notably, the lowest $\delta^{18}\text{O}$ values in this interval correspond to samples from North Öland,
464 whereas those from Degerhamn, South Öland, are characterized by the heaviest $\delta^{18}\text{O}$ values.
465 Beginning in the *L. variabilis* Conodont Zone, brachiopod $\delta^{18}\text{O}$ values steadily increase by up
466 to 1.5‰ into the lower *E. suecicus* Conodont Zone. This is followed by a decrease of ca. 1‰
467 until the base of the *P. serra* Conodont zone (upper Darriwilian). Subsequently in the upper
468 Darriwilian to Sandbian (*P. serra* to *A. tvaerensis* Conodont Zone) interval, $\delta^{18}\text{O}$ values of
469 bulk carbonates and brachiopods show no clear pattern, but instead vary significantly by up
470 to 2.5‰.

471 5.3 Element/Ca ratios

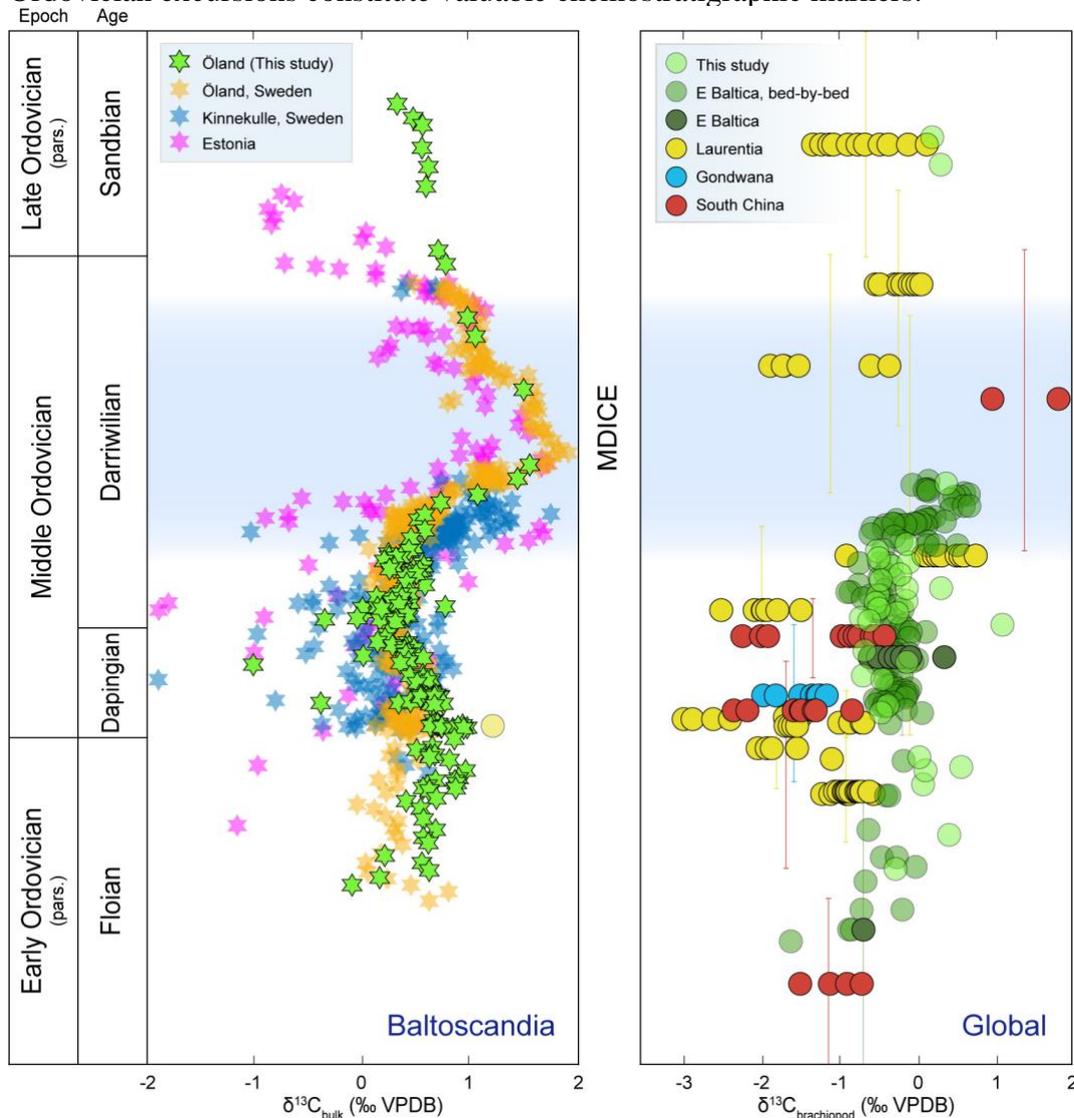
472 Element/Ca ratios of investigated brachiopods vary between 0.21 and 2.19 mmol/mol
473 for Sr/Ca and 0.34–8.09 mmol/mol for Mn/Ca. Element/Ca ratios do not vary distinctly
474 between localities and generally, and do not display discernible stratigraphic trends
475 (Supplementary information Figure 1). Correlation between element/Ca ratios and isotopic
476 compositions is observed in the case of Sr/Ca and $\delta^{13}\text{C}$ (Supplementary information Figure
477 3), with negative slope of $\Delta^{13}\text{C}$ (the difference of brachiopod and bulk carbonate $\delta^{13}\text{C}$) vs
478 Sr/Ca and $\delta^{13}\text{C}$ vs Sr/Ca suggesting a link between depletion in Sr/Ca and $\delta^{13}\text{C}$. These
479 element/Ca trends diverge from those documented for the Eastern Baltic (Figure 5; (C. M. Ø.
480 Rasmussen et al., 2016), hinting at variability in trace element patterns in sediments within
481 the Baltoscandian Paleobasin.

482 6 Discussion

483 6.1 Screening of samples

484 A combination of optical (Scanning Electron Microscopy), chemical (element/Ca ratios) and
485 statistical methods (correlation between element/Ca ratios and $\delta^{13}\text{C}$, $\delta^{18}\text{O}$) have been applied
486 to assess the fidelity of the carbon and oxygen isotope data presented herein (see
487 supplementary information).
488 Generally, brachiopods with Mn/Ca ratio of ≤ 1 mmol/mol show well-preserved shell
489 ultrastructure and usually exhibit Sr/Ca ratios ≥ 1.3 mmol/mol, which is comparable to values
490 reported for well-preserved early Paleozoic biogenic calcite (C. M. Ø. Rasmussen et al.,
491 2016; Steuber & Veizer, 2002). Therefore, these brachiopods probably represent the best-
492 preserved samples in our dataset. Importantly, the secular Ordovician carbon and oxygen
493 isotope trend herein recorded from Öland remains unchanged whether or not only samples
494 below a particular element/Ca ratio preservation limit (e.g. ≤ 1 mmol/mol Mn/Ca) are
495 considered (Figure 10), which is similar to the conclusion reached by Veizer et al. (1999) and
496 more recently by Goldberg, Present, Finnegan, & Bergmann (2021).

498 Over the last three decades, the carbon isotope stratigraphy of Baltoscandian
 499 Ordovician successions has been extensively documented, albeit mainly based on bulk rock
 500 carbonates (e.g. Ainsaar et al., 1999, 2004, 2007, 2010; Bauert et al., 2014; Calner et al.,
 501 2014; Kaljo et al., 2007; Lindskog et al., 2019; Wu et al., 2017) (Figure 11). This has enabled
 502 the identification of early Paleozoic carbon isotope excursions which have been used as
 503 important stratigraphic correlation tools, for instance the MDICE (Ainsaar et al., 2010;
 504 Ainsaar, Meidla, & Tinn, 2004; Ainsaar, Tinn, Dronov, Kiipli, & Radzevicius, 2020; Kaljo,
 505 Martma, & Saadre, 2007; M. R. Saltzman & Edwards, 2017; Schmitz, Bergström, &
 506 Xiaofeng, 2010; Young, Gill, Edwards, Saltzman, & Leslie, 2016). In one of the first studies
 507 to devote attention to the Early Ordovician interval in Baltoscandia, Calner et al. (2014)
 508 investigated the upper Tremadocian to middle Darriwilian $\delta^{13}\text{C}$ record of the Orthoceratite
 509 limestone of Öland, Sweden and made comparisons to that of the Argentine Precordillera.
 510 They reported a negative excursion (ca. 1‰) in the basal parts of the K pingsklint Formation
 511 and a marked positive excursion in the *O. evae* Conodont Zone and proposed that these Early
 512 Ordovician excursions constitute valuable chemostratigraphic markers.



513

514 **Figure 11:** Comparison of regional and global whole rock carbonate and brachiopod $\delta^{13}\text{C}$ 515 trends. Whole rock $\delta^{13}\text{C}$ data ($\delta^{13}\text{C}_{\text{bulk}}$) all originate from Baltoscandia (left figure): Öland516 (*Wu et al., 2017*); Kinnekulle (*Lindskog et al., 2019*); Estonia (*Kaljo et al., 2007*).

517 *Brachiopod $\delta^{13}\text{C}$ data (right figure) elucidate Baltoscandia $\delta^{13}\text{C}$ trends using best-preserved*
518 *brachiopods ($\text{Mn}/\text{Ca} \leq 1 \text{ mmol}/\text{mol}$) from this study in conjunction with data from C. M. Ø.*
519 *Rasmussen et al. (2016) compared to reported $\delta^{13}\text{C}$ trends from global compilations (Qing &*
520 *Veizer, 1994; Shields et al., 2003; Veizer et al., 1999; Veizer & Prokoph, 2015). Note the*
521 *marked MDICE trend in both curves, as well as the temporal resolution of the Baltic*
522 *brachiopod bed-by-bed dataset as compared to the global sites. Vertical bars in the right*
523 *diagram denote stratigraphical uncertainty of the global compilation samples highlighted in*
524 *corresponding colors. This temporal constraint is based on either the provided*
525 *lithostratigraphical information in the source references using C. M. Ø. Rasmussen et al.*
526 *(2019) or biostratigraphy, based on brachiopod species ranges where possible (St.*
527 *Petersburg: Egerquist (2004), Yangtze Platform: Zhan, Jin, & Chen (2007)).*
528

529 Lehnert, Meinhold, Wu, Calner, and Joachimski (2014) documented the presence of three
530 notable short-term negative $\delta^{13}\text{C}$ excursions in the Lower to Middle Ordovician and proposed
531 new names for $\delta^{13}\text{C}$ excursions encompassing the Cambrian–Ordovician boundary to the
532 lowermost Darriwilian interval. C. M. Ø. Rasmussen et al. (2016) reported $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$
533 records of the Floian to mid-Darriwilian interval in eastern Baltoscandia based on bed-by-bed
534 brachiopod samples from Russia. Subsequently, Wu et al. (2017) documented a complete
535 record of the MDICE (rising limb, peak interval and falling limb) and reported that it spans
536 the *L. pseudoplanus*, *E. suecicus*, *P. serra* and *P. anserinus* conodont zones. Thus, the Lower
537 to Middle Ordovician carbon isotope stratigraphy of Baltoscandia is well-constrained based
538 on bulk rock data.

539 In the present study, the significant carbon isotope excursion, discernible in both brachiopods
540 and bulk carbonate, is the MDICE with a magnitude of ca. 1‰ and peak values around
541 +1.5‰ (Figure 11). The peak of this isotope event is at -54.8 to -52.4 m of the Kårehamn
542 core, corresponding to the *E. suecicus* Conodont Zone (Figure 10). There is overall
543 agreement between bulk rock and brachiopod $\delta^{13}\text{C}$ trends (Figs. 10; 11), with about 0.5‰
544 lighter $\delta^{13}\text{C}_{\text{brachiopod}}$ values. The similarity of both trends is consistent with observations that
545 over specific time intervals, brachiopod and bulk rock $\delta^{13}\text{C}$ records show good correlation,
546 but some deviations exist (Brand, 2004; Munnecke, Calner, Harper, & Servais, 2010).
547 Primary inter- and intra-specific variability of $\delta^{13}\text{C}$ is documented in literature for ancient
548 (e.g. Korte and Hesselbo, 2011; Korte et al., 2005; Veizer et al., 1999) and modern
549 brachiopods (e.g. Takayanagi et al., 2013, 2015; Ullmann et al., 2017) and are linked to
550 pronounced seasonality of the shallow marine depositional environment, metabolism-
551 mediated or kinetic fractionation effects (Auclair et al., 2003; Korte et al., 2005, 2017;
552 Takayanagi et al., 2015). The smooth trend evident in the current $\delta^{13}\text{C}_{\text{bulk}}$ record, however,
553 can be explained by the mixing of microscopic-sized carbonate fragments in the micritic
554 carbonates, which yield homogenized $\delta^{13}\text{C}$ compositions and consequently, generated the
555 smoothed isotope curves that can display $\delta^{13}\text{C}$ high-frequency variability (cf. Korte et al.,
556 2017). In addition, variations in $\delta^{13}\text{C}$ values of dissolved inorganic carbon (DIC) in the open
557 ocean are relatively small (Swart, 2015) and the present Baltoscandia $\delta^{13}\text{C}$ record therefore
558 probably reflects the $\delta^{13}\text{C}$ composition of Ordovician open ocean DIC.

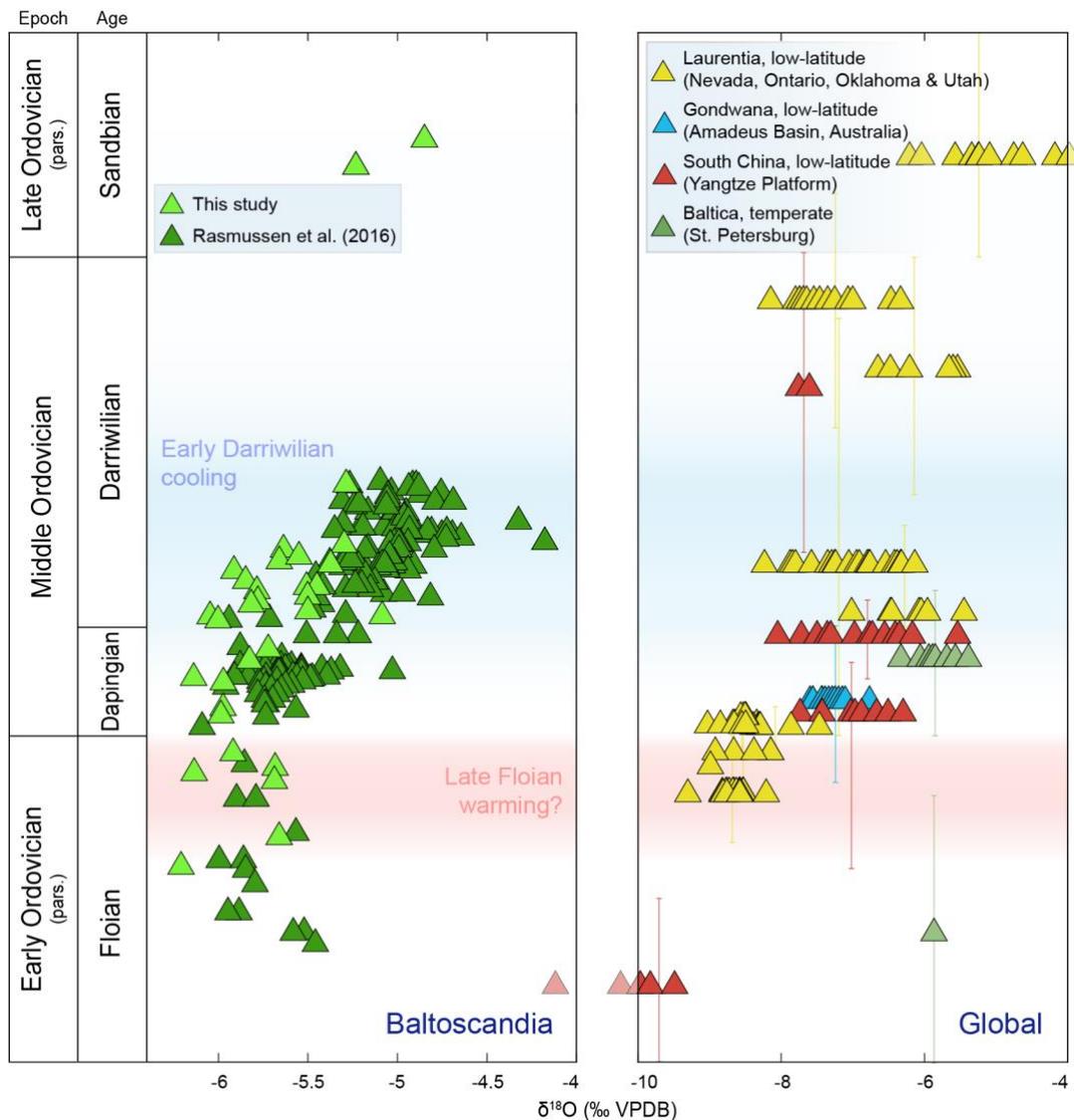
559 6.2.1 Regional and global comparison of the Baltoscandian $\delta^{13}\text{C}$ record

560 The 0.5‰ offset between $\delta^{13}\text{C}_{\text{brachiopod}}$ and $\delta^{13}\text{C}_{\text{bulk}}$ in the current study seems to be a
561 consistent pattern throughout the Baltoscandian Paleobasin. Specifically, Floian to Sandbian
562 $\delta^{13}\text{C}_{\text{bulk}}$ values from different parts of Baltoscandia (Figure 11) show a range between -1‰
563 and +2‰ (Kaljo et al., 2007; Lindskog et al., 2019; Wu et al., 2017). For fossil brachiopods,
564 values ranging between -1‰ and +1‰ have been reported for the eastern Baltoscandian
565 Paleobasin (e.g. Bergmann et al., 2018; C. M. Ø. Rasmussen et al., 2016) and these are

566 comparable with our observations (Figure 6). Floian–Sandbian $\delta^{13}\text{C}_{\text{bulk}}$ range between -1.0‰
567 and +1.6‰, and best preserved $\delta^{13}\text{C}_{\text{brachiopod}}$ values (with $\text{Mn}/\text{Ca} \leq 1$ mmol/mol) range
568 between -0.7‰ and +0.4‰. This offset may reflect a basin configuration where deeper
569 waters are ^{13}C -depleted and the upper parts are ^{13}C -enriched due to transport of organic
570 matter to deeper waters (Kroopnick, Margolis, & Wong, 1977; van de Schootbrugge, Föllmi,
571 Bulot, & Burns, 2000). However, this is unlikely to be the case as the Baltoscandian
572 Paleobasin was characterized by very low relief (Jaanusson, 1973) and thus, no significant
573 differences in water depth in the sea. The lighter $\delta^{13}\text{C}$ values in brachiopods compared to the
574 bulk rock data potentially reflects species-specific carbon isotope fractionation (vital effects).
575 Comparison of the new Baltic $\delta^{13}\text{C}_{\text{brachiopod}}$ record to published data and compilations (Shields
576 et al., 2003; Veizer et al., 1999; Figure 11) reveals that the Baltoscandian record is
577 characterized by generally heavier $\delta^{13}\text{C}_{\text{brachiopod}}$ values. This disparity can be attributed to
578 local/regional differences in C isotope compositions, and this is in concert with data from
579 several other sedimentary basins (Shields et al., 2003; Veizer et al., 1999) which have
580 depositional environments and tectonic regimes different from those of the Baltoscandian
581 Paleobasin (see supplementary information). Nevertheless, the apparent similarity in the
582 trends of both $\delta^{13}\text{C}_{\text{brachiopod}}$ records strengthens the view that the Baltoscandian $\delta^{13}\text{C}_{\text{brachiopod}}$
583 reflect a near-primary record and a global trend.

584 6.3 Oxygen isotopes

585 The composite Baltic $\delta^{18}\text{O}$ record (Figure 12) reveals a secular Floian to Sandbian
586 $\delta^{18}\text{O}$ increase of ca. 1.4‰. This is most apparent during the Darriwilian, where an increase of
587 ca. 0.8‰ is recorded (Figure 10), mirroring the Early to Middle Ordovician $\delta^{18}\text{O}_{\text{brachiopod}}$
588 record from eastern Baltoscandia (C. M. Ø. Rasmussen et al., 2016; Figure 12). The
589 similarity between these oxygen isotope records (Figure 12) thus suggests that the
590 Baltoscandian Early to Middle Ordovician oxygen isotope record is spatially consistent and a
591 primary geochemical signature. Although the late Darriwilian to Sandbian portion of this
592 record is less constrained, data from the best-preserved brachiopods suggests that $\delta^{18}\text{O}$ values
593 in that period remained at least as heavy as during middle Darriwilian times. This is
594 congruent with published records indicating a transient cooling event at this time (Saltzman
595 and Young, 2005; Vandenbroucke et al., 2010).
596 Furthermore, the Early to Late Ordovician Baltoscandian $\delta^{18}\text{O}$ record of the present dataset is
597 consistent with the trend of increasing $\delta^{18}\text{O}$ values with decreasing age, and this has long
598 been documented for the Ordovician (Grossmann and Joachmiski, 2020; Qing and Veizer,
599 1994; Shields et al., 2003, Veizer et al., 1999; Veizer and Prokoph, 2015). However, the
600 current Baltic dataset provides improved temporal resolution compared to the global
601 compilations, which have limited resolution in the Ordovician (see Figure 12 for comparison
602 of datasets).



603
 604 **Figure 12:** Long-term comparison between Early Ordovician (Floian) to early Late
 605 Ordovician (Sandbian) brachiopod $\delta^{18}\text{O}$ values in Baltoscandia (based on pristine
 606 brachiopods) and global brachiopod $\delta^{18}\text{O}$ compilations. Both the Baltoscandian and global
 607 compilation datasets elucidate a general pattern of increasing brachiopod $\delta^{18}\text{O}$ values
 608 upwards, which is most prominent during the Middle Ordovician (compare with Figure 2
 609 showing a similar trend based on the conodont apatite-derived curve of Trotter et al. (2008).
 610 Vertical bars in the right diagram denote stratigraphical uncertainty of the global
 611 compilation samples highlighted in corresponding colors. Note the well-resolved temporal
 612 resolution of the Baltic brachiopod bed-by-bed dataset as compared to the global data points.
 613 Global compilation dataset obtained from Qing & Veizer (1994), Veizer et al. (1999), Shields
 614 et al. (2003), Veizer & Prokoph (2015). Samples are temporally constrained as in Figure 11.

615

616 6.3.2 Regional and global comparison of the Baltoscandian $\delta^{18}\text{O}$ record

617 The comparison of the Baltoscandian $\delta^{18}\text{O}$ record with global compilation data
 618 (Figure 12) indicates that the Baltoscandian record is characterized by heavier $\delta^{18}\text{O}$ values.
 619 This may be attributable to the paleogeographical position of Baltica during the studied
 620 interval, as well as the relatively shallow sedimentary burial – thus, reduced susceptibility to

621 burial diagenesis – which largely characterized the Baltoscandian Paleobasin. The mid
622 latitudinal positions, which Baltica occupied during the Early to Middle Ordovician (Torsvik
623 et al., 2012) suggests that Baltoscandian brachiopods lived in cooler waters compared to their
624 counterparts in more equatorial paleocontinents, which constitute the majority of the
625 Ordovician global compilation data (Shields et al., 2003; Veizer et al., 1999) (Figure 12). For
626 instance, the paleocontinent of Laurentia was located in equatorial realms throughout the
627 Ordovician while Baltica only approached equatorial paleolatitudes during the Late
628 Ordovician (Kaljo et al., 2007).

629 6.4 Paleoenvironmental significance of the Baltoscandian oxygen isotope record

630 Temporal variation in $\delta^{18}\text{O}$ of biogenic calcite can be influenced by several factors
631 including changes in seawater $\delta^{18}\text{O}$ composition, temperature of calcite precipitation, vital
632 effects, pH changes and diagenetic alteration of near-primary brachiopod $\delta^{18}\text{O}$ (Bruckschen
633 & Veizer, 1997; Munnecke et al., 2010; Qing & Veizer, 1994; Swart, 2015). Climatic cooling
634 has been invoked as an explanation for the Ordovician $\delta^{18}\text{O}$ trend observed in both
635 brachiopods and conodonts, and cooling, in turn, has been associated with the coinciding
636 GOBE (Qing and Veizer, 1994; C. M. Ø. Rasmussen et al., 2016; Trotter et al., 2008).
637 Alternative interpretations have also been postulated for this trend including changes in the
638 oxygen isotope composition of seawater (Jaffrés et al., 2007; Shields et al., 2003) and
639 diagenetic alteration (Bergmann et al. 2018).

640 In the current Baltoscandia record, vital effects are unlikely to explain the secular $\delta^{18}\text{O}$ trend
641 because the secondary layer of brachiopod shells, which are more likely to have been
642 secreted in isotopic equilibrium with or very close to the seawater were utilized (Carpenter &
643 Lohmann, 1995; C. Ullmann, Frei, Korte, & Lüter, 2017). Although the $\delta^{18}\text{O}$ values during
644 the early Darriwilian and late Darriwilian to late Sandbian portion of the Öland data show a
645 wide range (Figure 10) suggesting that diagenesis has modified some of the primary $\delta^{18}\text{O}$
646 compositions, the best-preserved samples (Figure 10, 12), however, show a long-term trend
647 which is consistent with the $\delta^{18}\text{O}$ LOWESS smoothing line and narrow $\delta^{18}\text{O}$ range (Figure
648 10), indicating that the near-primary $\delta^{18}\text{O}$ trends are preserved (see also section 6.1).

649 Besides the influence of significant ice-volume changes, seawater $\delta^{18}\text{O}$ composition may
650 become heavier through high-temperature reactions of seawater with silicate minerals in
651 hydrothermal systems associated with oceanic ridges and their flanks (Veizer and Prokoph,
652 2015; Verard and Veizer, 2019 and references therein). Results based on modelling efforts
653 show that the maximum rate of change of the $\delta^{18}\text{O}$ composition of seawater due to high-
654 temperature reactions is ca. 1‰ per 100 million years (Jaffrés et al., 2007; Veizer and
655 Prokoph, 2015). However, the time-period covered by the current Baltic dataset is ca. 19
656 million years (Gradstein & Ogg, 2020), thus making it unlikely that seawater-silicate rock
657 interactions could have been rapid enough to generate the observed $\delta^{18}\text{O}$ change.

658 Furthermore, clumped isotope results based on Middle Ordovician brachiopods have been
659 reported to yield seawater $\delta^{18}\text{O}$ compositions between -0.9‰ and -1.2‰ (Bergmann et al.,
660 2018), and this is comparable to $\delta^{18}\text{O}$ compositions of modern seawater.

661 Consequently, we interpret the long-term Baltoscandia $\delta^{18}\text{O}_{\text{brachiopod}}$ trend as dominantly
662 reflecting a near-primary paleotemperature signal, in agreement with previous studies
663 (Goldberg, Present, Finnegan, & Bergmann, 2021; C. M. Ø. Rasmussen et al., 2016; Trotter
664 et al., 2008). In this scenario, the $\delta^{18}\text{O}$ trend represents a transition from warmer climatic
665 conditions during the Early Ordovician to less-warm conditions during the Early to Middle
666 Ordovician transition and a cooling episode during the Darriwilian which may have persisted
667 into the Sandbian.

668 6.4.1 Middle Ordovician cooling

669 The composite Baltoscandia record (Figure 12) indicates a ca. 1.5‰ $\delta^{18}\text{O}_{\text{brachiopod}}$
670 increase between the Floian and mid-Darriwilian. Assuming a $\sim 4\text{ }^{\circ}\text{C}$ temperature change for
671 1‰ $\delta^{18}\text{O}$ shift (Epstein and Mayeda, 1953), this is suggestive of a 6–7 $^{\circ}\text{C}$ relative cooling
672 within a period of ca. 8 million years in an ice-free world even during the period with cooler
673 temperatures is assumed (Gradstein & Ogg, 2020). This relative cooling is comparable to bio-
674 apatite- and brachiopod-based estimates of sea surface temperature evolution during the same
675 interval (Goldberg, Present, Finnegan, & Bergmann, 2021; Grossman and Joachimski, 2020;
676 Trotter et al., 2008). The northward drift of Baltica towards the equator during the Ordovician
677 (Torsvik et al., 2012) can be expected to have resulted in progressively lighter $\delta^{18}\text{O}$ values
678 due to warming. However, the opposite trend is apparent in the $\delta^{18}\text{O}$ record. Therefore, the
679 1.5‰ amplitude of change in $\delta^{18}\text{O}_{\text{brachiopod}}$ potentially represents an under-estimation of the
680 actual global seawater temperature change. Moreover, short-term sea level fall occurs in the
681 upper part of the Dapingian, and a pronounced and long-lasting sea-level fall is observed both
682 at the regional and global scale starting within the earliest Darriwilian, suggesting a good
683 correspondence with the pronounced shift to heavier $\delta^{18}\text{O}$ values during the Mid-Ordovician
684 (Figure 13). These observations support glacio-eustasy driven by climatic cooling and this
685 would suggest that a portion of the $\delta^{18}\text{O}_{\text{brachiopod}}$ increase is related to ice volume effect and
686 the temperature decline on Baltica was less than 6–7 $^{\circ}\text{C}$.

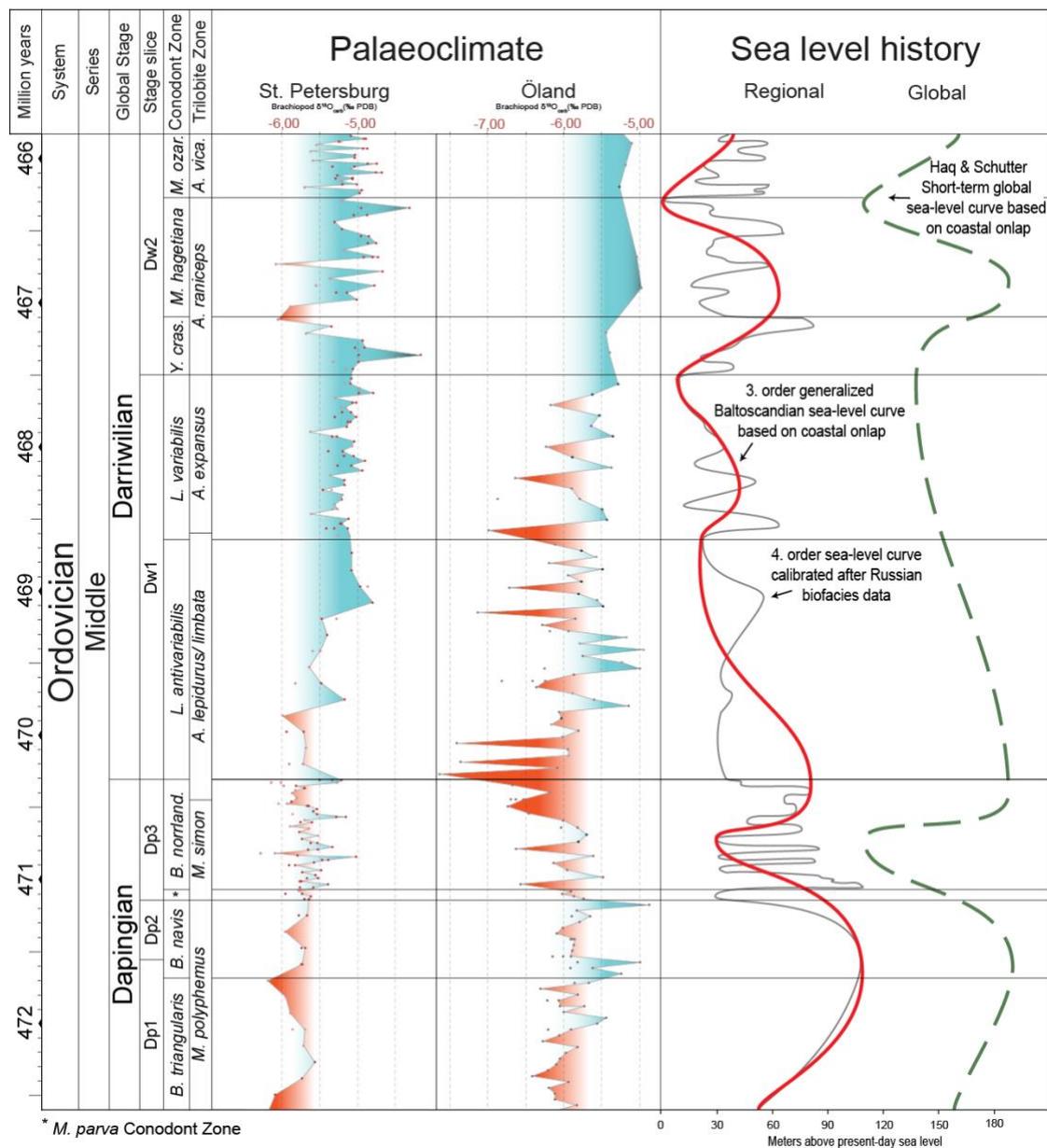
687 Biofacies analyses of brachiopods, conodonts and trilobites have all demonstrated that
688 shallow-water faunas became ubiquitous in Baltoscandia during the early Darriwilian,
689 indicative of falling sea level in the order of 150 m from the late Floian to the early
690 Darriwilian (A. T. Nielsen, 1995; C. M. Ø. Rasmussen et al., 2016; J. A. Rasmussen &
691 Stouge, 2018; Stouge et al., 2020). Also, prior to the Middle Ordovician, lighter $\delta^{18}\text{O}_{\text{brachiopod}}$
692 values during the late Floian (Figure 12) coincide with the late Floian migration of Laurentian
693 warm-water conodont taxa into Baltoscandia (i.e. an influx of species typical of low-latitude
694 regions), interpreted to denote a relatively brief warming episode. Hereafter follows the
695 Middle Ordovician influx of temperate-water taxa denoting cooler waters (Bagnoli & Stouge,
696 1997; Stouge et al., 2020).

697 In addition to paleontological evidence, several sedimentological and cyclostratigraphical
698 studies have argued for a global Darriwilian cooling (Cherns et al., 2013; Dabard et al., 2015;
699 Fang et al., 2019; Lindström, 1984; Lindskog et al., 2014, 2019; Turner et al., 2012).
700 Therefore, the discussion of whether the Middle Ordovician oxygen isotope trend is climate-
701 related, or reflecting either diagenetic overprint (Bergmann et al., 2018) or changing seawater
702 composition (Veizer & Prokoph, 2015) seems to overlook the point that climate is invoked as
703 a main driver for this secular trend based on a whole suite of *other* proxies that independently
704 from the oxygen isotope record indicate sea level fluctuations, and, in many cases, on a bed-
705 by-bed scale (Figure 13). Of equal importance is the rapidity of these sea level oscillations
706 such as those observed in the late Dapingian and in the *L. variabilis*–*Y. crassus* interval of the
707 Darriwilian. As they occur in a stable intra-cratonic setting on Baltica, it is difficult to invoke
708 other causal mechanisms than glacio-eustasy. We note, however, that high-frequency
709 fluctuations in 4th order sea-level changes are only expressed in the late Dapingian and then
710 again in the *L. variabilis* Zone to the *M. ozarkodella* Conodont subzones of the Darriwilian
711 (Figure 13). Perhaps, this change in the expression of sea-level change is linked to distinct
712 phases of ice-sheet growth, with high-frequency fluctuations particularly well-expressed in a
713 late Dapingian phase that saw the onset of growth of continental ice with small volume ice-
714 changes paced by orbital changes. The *L. antivariabilis* Zone, instead, may have been
715 characterized by the establishment of a larger but stable ice sheet, less sensitive to high-

716 frequency fluctuations. The return to more prominent high-frequency fluctuations in 4th order
 717 sea level changes in the *L. variabilis* Zone, and within the rest of the studied succession,
 718 could be due to a third phase of ice-sheet growth with a new increase in volume of
 719 continental ice occupying new areas that were particularly sensitive to ice growth and decay
 720 via orbital changes. Although this interpretation is highly speculative, it presents the merit to
 721 reconcile observed trends in sea-level change and $\delta^{18}\text{O}_{\text{brachiopod}}$ data.

722 A similar interpretation was suggested based on a sequence stratigraphical analysis of early
 723 Darriwilian sections from southern Jordan (Turner et al., 2012), as did model simulations
 724 which suggest that the climatic threshold for glacial onset was reached during the Darriwilian
 725 (Pohl et al., 2016 and references therein), in agreement with reported contemporaneous 3rd
 726 order eustatic cycles.

727



728 * *M. parva* Conodont Zone
 729 **Figure 13:** Middle Ordovician $\delta^{18}\text{O}_{\text{brachiopod}}$ and sea level evolution across the Baltoscandian
 730 Paleobasin. Note that even though the 4th-order sea level curve (right) is based on a biofacies
 731 framework from the St. Petersburg region, individual excursions in the $\delta^{18}\text{O}$ -Öland curve is
 732 still mirrored. $\delta^{18}\text{O}$ -St. Petersburg curve and sea level curves as in C. M. Ø. Rasmussen et al.

733 (2016) but here calibrated to the time domain. Original sea level data modified from Hansen
734 & Nielsen (2003), Haq & Schutter (2008), A. T. Nielsen (1995, 2004, 2011); C. M. Ø.
735 Rasmussen et al., 2009).
736

737 6.4.2 Deciphering orders of sea level change

738 In successions lacking a properly calibrated astrochronological framework –
739 something still in its infancy when it comes to early Paleozoic rocks – only a high-resolution
740 sequence- or ecostratigraphical analysis can resolve sea level changes at a sufficient temporal
741 resolution to recognize glacio-eustasy. This has been done, in detail, on lowermost
742 Darriwilian rocks of Baltica (A. T. Nielsen, 1995; C. M. Ø. Rasmussen et al., 2009, 2016)
743 and Armorica (Dabard et al., 2015), revealing similar magnitude 3rd order sea level
744 oscillations potentially at the kyr-scale. This interval precisely correlates to the interval where
745 the current Baltic $\delta^{18}\text{O}$ record shows the strongest positive trend, in accordance with the
746 inferred early Middle Ordovician sea level drop (Figure 13).
747 The trend towards relatively heavier late Darriwilian–Sandbian brachiopod $\delta^{18}\text{O}$ values
748 reported here occurs at the start of an interval where global and regional sea level estimates
749 suggest the start of a sea level rise that eventually peaked during the early–mid Katian
750 (Hallam, 1992; Haq & Schutter, 2008; A. T. Nielsen, 2004; C. M. Ø. Rasmussen et al., 2019).
751 This therefore seems to oppose a climatic driver for the oxygen isotope trend in this interval.
752 However, as with the early Darriwilian (where there is a discordance between the actual
753 brachiopod $\delta^{18}\text{O}$ trend and the expected trend based on the paleolatitudinal location of
754 Baltica), there is a discordance between the $\delta^{18}\text{O}$ -signal and inferred eustatic sea level rise
755 during the later Darriwilian–Sandbian times as the latter would suggest a warming pulse.
756 It is therefore expedient to distinguish between 1st-order plate tectonic-induced changes and
757 the dramatic amplitudes of 4th and 3rd-order sea level changes suggestive of the waxing and
758 waning of ice sheets (Hallam, 1992; Haq and Schutter, 2008; Nielsen, 2004; 2011). Whereas
759 climatic cooling likely accelerated faster than the expected latitudinal temperature gradient
760 during the early Darriwilian, the 1st-order sea level rise subsequently outpaced the 3rd-order
761 sea level fall from the later Darriwilian onwards.

762 7 Conclusions

763 This study presents biostratigraphically well-resolved brachiopod and bulk carbonate
764 carbon and oxygen isotope data spanning the Early (Floian) to early Late Ordovician
765 (Sandbian) from Baltoscandia. The temporal scale of this Baltoscandian dataset allows for
766 considerable refinement from a mid-latitude perspective of previously published global
767 carbon and oxygen isotope data, which historically have been characterized by spot sampling
768 across several paleoplates in low-latitude settings.

769 Several lines of evidence indicate that, while the carbon and oxygen isotope dataset may have
770 been affected by diagenetic alteration, long-term trends in isotopic compositions which are
771 useful for paleoenvironmental interpretation, are preserved. Our $\delta^{18}\text{O}$ record from Öland
772 reveals that previously reported Early to Middle Ordovician $\delta^{18}\text{O}$ trends from eastern
773 Baltoscandia are spatially consistent and together, the composite Baltoscandian $\delta^{18}\text{O}$ record is
774 concordant with global Ordovician $\delta^{18}\text{O}$ compilations, which intimate an Early to Late
775 Ordovician $\delta^{18}\text{O}$ increasing trend. We interpret the Baltoscandian $\delta^{18}\text{O}$ record as being
776 dominated by a paleotemperature signal indicating a transition from warmer
777 paleotemperatures during the Early Ordovician to cooler conditions in the Middle
778 Ordovician.

779 Thus, the Baltoscandian $\delta^{18}\text{O}$ record is compatible with previous studies which suggest that
780 present-day seawater temperatures were attained during the Darriwilian. This optimal
781 temperature window may have sparked the GOBE.

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