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Lower–Middle Ordovician carbon and oxygen isotope chemostratigraphy at Hällekis, Sweden: implications for regional to global correlation and palaeoenvironmental development

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A high-resolution chemostratigraphical (coupled $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) study of the topmost Floian through the middle Darriwilian (Ordovician) succession at the Hällekis quarry, Kinnekulle, southern Sweden, shows relatively steady isotopic values with overall minor changes, although some notable short- and long-term shifts are discernible. A pronounced positive shift in $\delta^{13}C$ in the uppermost part of the study succession is identified as the Middle Darriwilian Isotopic Carbon Excursion (MDICE), representing the only named global isotopic excursion in the data set. Regional and global comparisons suggest that few details in the different carbon and oxygen isotope curves can be confidently correlated, but longer-term patterns appear quite consistent. Trends in the isotope data are in agreement with palaeogeographical reconstructions. Differences in stratigraphical patterns of both carbon and oxygen isotopes between localities suggest strong secular development at several spatiotemporal scales; any global signal involving relatively minor isotopic shifts is often masked/subdued by local and regional overprinting and care should be taken not to overinterpret data sets. Collectively, the data suggest rising sea levels and cooling climates through the studied time interval, but detailed interpretations remain problematic. □ *'Orthoceratite limestone', correlation, Dapingian, Darriwilian, $\delta^{13}C$, $\delta^{18}O$.*

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The Ordovician System has been the subject of intense palaeoenvironmental, palaeontological and stratigraphical studies over the last few decades. These investigations have led to a more detailed understanding of the complexities of the faunal and environmental perturbations that occurred during this interval of the early Palaeozoic (e.g. Bergström *et al.* 2009). The Ordovician was a time of high global sea level, with expansive epeiric seas covering large parts of the palaeocontinents. In these shallow water regions, there were remarkable changes in the global marine biota during the so-called Great Ordovician Biodiversification Event (GOBE), which resulted in numerous new taxa and the establishment of new ecological niches (e.g. Webby *et al.* 2004; Servais *et al.* 2009; Algeo *et al.* 2016; Edwards & Saltzman 2016; Servais & Harper 2018). By contrast, the last portion of the Ordovician Period is marked by the first of the big five mass-extinction events, during which approximately 85% of all marine species are estimated to have disappeared (Jablonski 1991; Sheehan 2001; Harper *et al.* 2014).

In recent years, the overall view of the Ordovician climatic state has changed rather dramatically, from a 'greenhouse world' to one with a climate that transitioned into conditions more similar to those of today (e.g. Frakes *et al.* 1992; Trotter *et al.* 2008). Detailed geochemical studies using refined palaeotemperature proxies and modelling have led to a much more complex picture of Ordovician climate, including the recognition of several time intervals potentially representing ice ages (although glaciogenic deposits older than the latest Ordovician remain to be found; e.g. Trotter *et al.* 2008; Finnegan *et al.* 2011; Pohl *et al.* 2016; Rasmussen *et al.* 2016). Collectively, a surge of new biotic, sedimentological and geochemical data in recent decades has greatly improved our knowledge about this system,

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resulting in, among other things, the establishment of a robust and detailed chronostratigraphical framework with three global series and seven stages (e.g. Bergström *et al.* 2009; Cooper & Sadler 2012; Lindskog *et al.* 2017).

Various geochemical proxies, notably carbon isotopes ($\delta^{13}C$), have proven to be important for the development of global and regional stratigraphical correlations (e.g. Bergström *et al.* 2009). It is of crucial importance that the $\delta^{13}C$ chemostratigraphical framework is biostratigraphically well constrained, but distinctive isotopic shifts and excursions have proven to be important also for the correlation of strata in which biostratigraphically important fossils are rare or missing. However, great caution is needed in carrying out such correlations even at the regional scale (see Fanton & Holmden 2007; Schiffbauer *et al.* 2017). Despite the wealth of newly available stable isotopic data, more studies are needed in order to improve our understanding of their relations to palaeoenvironmental conditions and developments during the Ordovician, as well as their significance to the establishment of correlations at local, regional and global scales. For instance, problems persist in disentangling isostatic and eustatic signals in the global sea level history (and by extension any associated climatic history). Accurate and detailed correlations are keys to the understanding abiotic and biotic events and processes at any larger spatiotemporal perspective.

The aim of the present study was to analyse the latest Floian through the middle Darriwilian carbon and oxygen stable isotopic records at the Hällekis quarry in the Province of Västergötland in south-central Sweden. The Hällekis section is well studied in terms of biostratigraphy, sedimentology and event stratigraphy, and is relatively expanded compared to most other coeval sections in Baltoscandia. This provides excellent possibilities to place the isotope data within local and regional palaeoenvironmental contexts of the Baltic palaeobasin, and its evolution throughout this time interval of the Ordovician. Here, we discuss and compare our results with previously studied stable isotopic profiles from coeval strata at both the regional and global scales.

Geological setting

During most of the early Palaeozoic, considerable parts of present-day northern Europe were covered by widespread epeiric seas (e.g. Torsvik & Cocks 2017). In Sweden and elsewhere on the Baltoscandian craton, a thin cover of sedimentary rocks records a long-term transgression (first- or second-order sea level cycle) that locally began in late Precambrian time (Fig. 1). During the Ordovician, much of Baltoscandia was covered by marine water in which net depositional rates were low (generally a few millimetres per thousand years; e.g. Lindström 1971). Surrounding landmasses were largely peneplained, with low relief resulting in limited transport of weathered terrigenous material into the basin. Bergström *et al.* (2011) calculated a net depositional rate for the 'orthoceratite limestone' (see below) at Kinnekulle of ~3.3 mm/ka, and Lindskog *et al.* (2017) calculated ~4 mm/ka.

Throughout the Ordovician, Baltica drifted from high southern latitudes into subtropical–tropical latitudes (Torsvik & Cocks 2017). On the Baltoscandian platform, most of the Ordovician was characterized by carbonate deposition (e.g. Lindström 1971; Jaanusson 1973). The 'orthoceratite limestone', a condensed cool-water carbonate facies, comprises the main rock type in the Lower–Middle Ordovician succession on the Swedish craton. Today, it occurs only in places where it has been protected from post-Ordovician erosion. Typically, the 'orthoceratite limestone' is reddish, brownish or grey in colour and consists of calcareous mud mixed with sand-sized skeletal grains and very limited amounts of terrigenous matter (e.g. Lindskog & Eriksson 2017; and references therein). Although nautiloid cephalopods, including orthoceratids, are common locally and lend their name to the informal rock unit, many parts of the 'orthoceratite limestone' do not contain abundant macrofossils. However, microfossils (notably conodonts) occur throughout the succession. The many cratonic outcrops in Sweden appear largely homogenous at the macroscopic scale and in Västergötland most outcrops are typically devoid of conspicuous evidence of tectonic activity (cf. Lindskog & Eriksson 2017; and references therein).

In Västergötland, there are numerous outcrops of lower Palaeozoic strata and, in part because of past extensive quarrying, exposures of the 'orthoceratite limestone' are abundant. This is the case on Kinnekulle, an approximately 300-m high table mountain on the southeastern shore of Lake Vänern (e.g. Holm & Munthe 1901; Munthe 1905; Priem *et al.* 1968). Here, the lower Palaeozoic marine succession, which rests on peneplained Precambrian gneiss, ranges from strata of the Cambrian Series 2 to the lower Llandovery, which is overlain by Permian dolerite. Kinnekulle can be regarded as the type area of the 'orthoceratite limestone' in Sweden (Hisinger 1828; Jaanusson 1982).

Fig. 1. Map of the Baltoscandian region (modified from Lindskog & Eriksson 2017). Geographical areas (e.g. localities, provinces) referred to in the main text and succeeding figures are shown. The distribution of lower Palaeozoic rocks is shown by green shading; areas with significant outcrops of Ordovician strata are indicated by darker shading. Generalized, the shelf became deeper from east to west and north to south. [Colour figure can be viewed at wileyonlinelibrary.com]

The Hällekis study interval ranges from the uppermost Floian Stage to the middle Darriwilian Stage (for stage terminology, see Bergström *et al.* 2009) and includes strata from the uppermost Lower to the middle part of the Middle Ordovician. The study succession begins in the uppermost Tøyen Shale, which ranges across the boundary between the Billingen and Volkhov Baltoscandian stages and corresponds to the uppermost part of Stage slice Fl3 of Bergström *et al.* (2009). The Tøyen Shale is overlain by the 'Lanna' and 'Holen' limestones, which are obsolete topoformations (i.e. historical units defined by a combination of lithology and fossil content; see Jaanusson 1960). Hence, these limestones are not lithostratigraphically defined and in lack of a revised lithostratigraphical framework they are herein retained as informal and provisional units (cf. Kumpulainen 2017; Lindskog & Eriksson 2017). The boundary between the Tøyen Shale and the 'Lanna Limestone' closely coincides with that between the Floian and Dapingian global stages (e.g. Bergström & Löfgren 2009). A conspicuous ~1.5-m-thick interval of grey limestone, informally known as the 'Täljsten', occurs across Kinnekulle in the lower part of the 'Holen Limestone' (see Lindskog & Eriksson 2017; and references therein). Disconformably overlying the 'Holen limestone' is the decimetre-thick 'Skövde Limestone', which belongs to the Lasnamägi Baltoscandian Stage and the Dw3 Stage slice of Bergström *et al.* (2009). The gap below the 'Skövde Limestone' corresponds to 2–3 conodont zones and some four million years of time (Zhang 1998a,b; Lindskog *et al.* 2017). This unit is overlain by the Gullhögen and Ryd formations of Uhaku age, which corresponds to the upper part of Stage slice Dw3. The basal beds of the Gullhögen Formation form the top of our study succession.

For this study, we collected a composite section at the Hällekis quarry on the northwestern slope of Kinnekulle (WGS 84 coordinates N58°36′33″,

E13°23′42″; Figs 1, 2). The same section and sample series were recently described in close detail for carbonate microfacies analyses and relative sea-level reconstructions by Lindskog & Eriksson (2017), allowing for direct comparison between data sets.

Materials and methods

A total of 220 limestone samples were collected through the measured Hällekis succession, spanning the uppermost Tøyen Shale through the basal Gullhögen Formation (Fig. 3; Lindskog & Eriksson 2017). The average space between the sample levels was ~15 cm. To obtain $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values, the carbonate matrix of samples was preferentially targeted using a micro-drill to generate ~1 g of powder, which was stored in clean micro-centrifuge tubes. Approximately 100–300 μg of carbonate powder were weighed into glass exetainer vials and then placed into a drying oven at 80°C for 8 h, in order to remove H_2O-vapour and any trace volatile organic compounds. These samples were then acidified with 3–5 drops of anhydrous phosphoric acid (H_3PO_4) in He purged, airtight exetainer vials. Samples were reacted with H_3PO_4 at 25°C for 24 h, and stable carbon and oxygen isotopic ratios of the evolved CO_2 were then measured using a Gas Bench II Autocarbonate device connected to a Thermo Delta Plus XP stable isotope ratio mass spectrometer (IRMS) at the National High Magnetic Field Laboratory at Florida State University. Carbon isotope ratios are corrected for ^{17}O contribution and reported in standard delta notation relative to the Vienna Pee Dee Belemnite (V-PDB) standard:

$$\delta^{13}C = [(^{13}C/^{12}C)_{sample}/(^{13}C/^{12}C)_{V\text{-}PDB} - 1] \times 1000$$

$$\delta^{18}O = [(^{18}O/^{16}O)_{sample}/(^{18}C/^{16}O)_{V\text{-}PDB} - 1] \times 1000$$

Our sample values are calibrated to international and FSU laboratory standards ROY-CC (+0.67‰, −12.02‰), MB-CC (−10.5‰, −3.5‰) NBS-19 (1.95‰, −2.2‰) and PDA (−1.3‰, −5.34‰) with standard deviations for $\delta^{13}C$ and $\delta^{18}O$ of ±0.05‰ and ±0.1‰, respectively (1σ).

Results

The full data set produced from the sample analyses is available in Table 1. The $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values are plotted in Fig. 3, against the local stratigraphy and the inferred sea level changes recognized by Lindskog & Eriksson (2017; see detailed discussion therein). In order to facilitate descriptions of the changes in isotopic values below, some intervals with distinct trends in the data are labelled C and O followed by a number. With the exception of the well-documented Middle Darriwilian Isotopic Carbon Excursion (MDICE; see Ainsaar *et al.* 2010), we

Fig. 2. Field photograph from the Hällekis quarry, northwestern Kinnekulle, Västergötland, south-central Sweden. Visible rock units referred to in the main text are indicated. The thickness of the figured succession is approximately 30 m. [Colour figure can be viewed at wileyonlinelibrary.com]

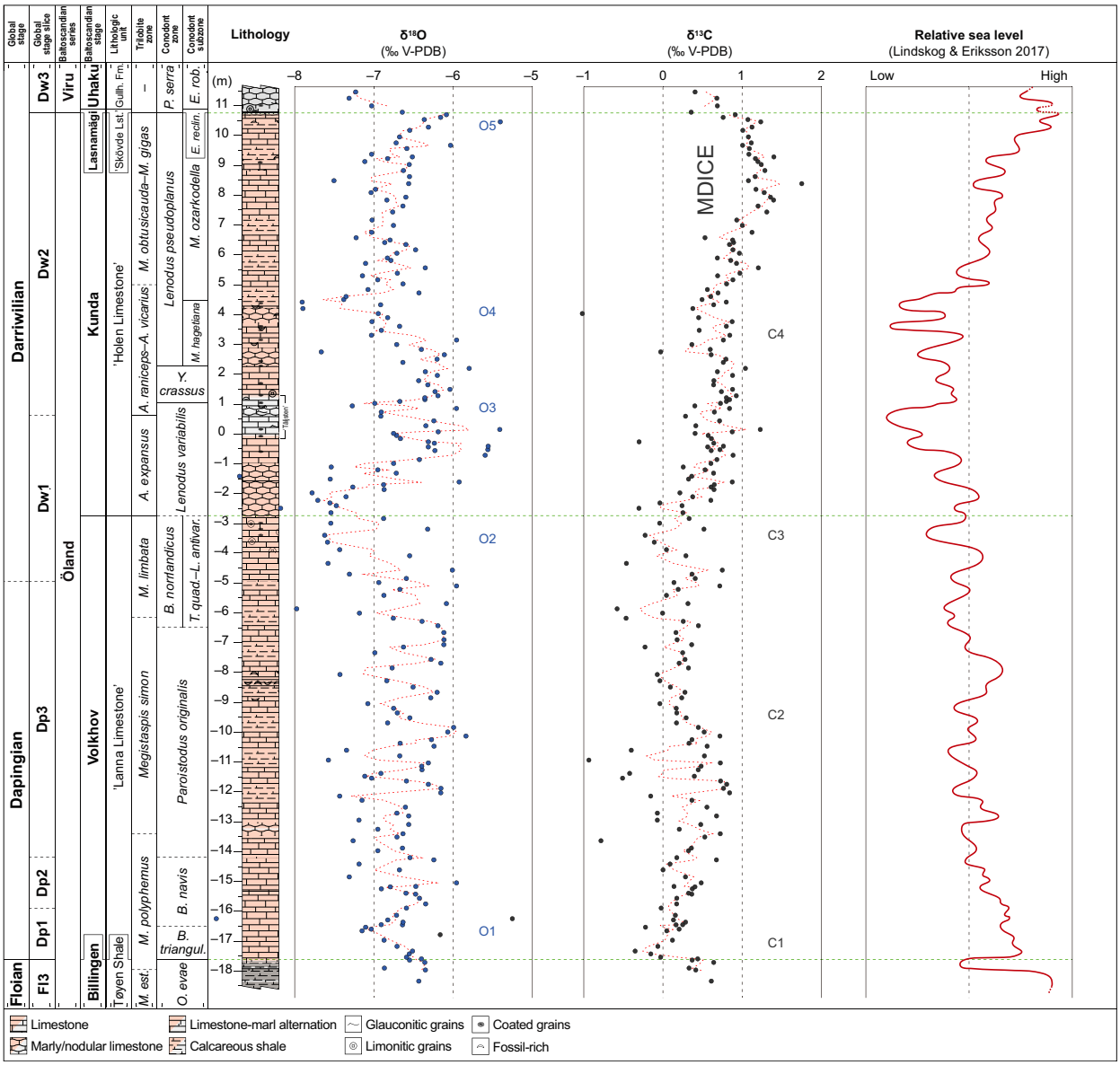

Fig. 3. Sedimentary profile of the 'orthoceratite limestone' and enclosing strata at the Hällekis quarry (modified from Lindskog & Eriksson 2017). Carbon ($\delta^{13}C_{carb}$) and oxygen ($\delta^{18}O_{carb}$) isotope curves are plotted together with an interpretation of relative sea level *sensu* Lindskog & Eriksson (2017). Two-point moving averages (stippled line; calculated from base) are indicated. Biostratigraphy after Tjernvik (1956), Zhang (1998a,b), Bergström & Löfgren (2009), Mellgren & Eriksson (2010), Schmitz *et al.* (2010) and Lindskog *et al.* (2014), with the middle part of the 'Lanna Limestone' extrapolated from Nielsen (1995) and Löfgren (1995). [Colour figure can be viewed at wileyonlinelibrary.com]

refrain from identifying these labelled shifts as named 'excursions' in order not to add to the already inflated number of minor isotopic kinks, or slight shifts from 'baseline' values, identified as such in the Palaeozoic despite being of questionable correlative value and event stratigraphical applicability. Hence, the letter-number scheme employed here for our observed trends in $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ may only have local to regional significance at this time due to many factors that affect these stable isotope records in shallow water carbonate platform settings: water mass connectivity/restriction, pH, temperature, salinity, organic matter decomposition, $CO_{2(aq)}$, diagenesis and carbonate mineralogy (e.g. Holmden *et al.* 1998; Immenhauser *et al.* 2003; Panchuk *et al.* 2005; Swart 2008).

A carbon isotope profile at Hällekis quarry was previously documented by Schmitz *et al.* (2010), but their study only covered part of the 'orthoceratite limestone' succession and at relatively low stratigraphical resolution. Nonetheless, a comparison between the results of Schmitz *et al.* (2010) and the

Table 1. List of samples, in stratigraphical order, and their $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values, expressed as ‰ relative to the Vienna Pee Dee Belemnite (V-PDB). The stratigraphical position of the respective samples can be found in Lindskog & Eriksson (2017, Figs 6, 7).

Stratigraphical unit	Sample	$\delta^{13}C_{carb}$	$\delta^{18}O_{carb}$
Gullhögen Formation	HÄ12-ÖP4	0.42	−7.21
Gullhögen Formation	HÄ12-ÖP3	0.69	−7.30
Gullhögen Formation	HÄ12-ÖP2	0.70	−7.02
'Skövde Limestone'	HÄ12-ÖP1	0.37	−6.60
'Holen Limestone'	HÄ12-ÖP5	0.92	−6.05
'Holen Limestone'	HÄ12-ÖP6	0.75	−6.15
'Holen Limestone'	HÄ12 ÖP7	1.10	−6.32
'Holen Limestone'	HÄ12 ÖP8	1.25	−5.35
'Holen Limestone'	HÄ12 ÖP9	1.11	−6.27
'Holen Limestone'	HÄ15-1	1.01	−6.52
'Holen Limestone'	HÄ15-2	1.08	−6.66
'Holen Limestone'	HÄ15-3	1.13	−6.69
'Holen Limestone'	HÄ12 ÖP10	1.02	−6.01
'Holen Limestone'	HÄ15-4	1.07	−6.52
'Holen Limestone'	HÄ15-5	1.10	−7.00
'Holen Limestone'	HÄ12 ÖP11	1.40	−6.49
'Holen Limestone'	HÄ15-6	1.18	−6.80
'Holen Limestone'	HÄ15-7	1.19	−7.08
'Holen Limestone'	HÄ15-8	1.24	−6.55
'Holen Limestone'	HÄ15-9	1.29	−6.62
'Holen Limestone'	HÄ15-10	1.15	−6.52
'Holen Limestone'	HÄ12-ÖP12	1.10	−7.48
'Holen Limestone'	HÄ15-11	1.76	−6.53
'Holen Limestone'	HÄ15-12	1.16	−6.97
'Holen Limestone'	HÄ15-13	1.30	−7.03
'Holen Limestone'	HÄ12 ÖP13	1.38	−6.59
'Holen Limestone'	HÄ15-14	1.39	−6.80
'Holen Limestone'	HÄ15-15	1.19	−6.60
'Holen Limestone'	HÄ15-16	1.33	−6.74
'Holen Limestone'	HÄ15-17	0.93	−7.03
'Holen Limestone'	HÄ15-18	1.01	−6.74
'Holen Limestone'	HÄ15-19	1.11	−7.00
'Holen Limestone'	HÄ12-38	0.54	−7.22
'Holen Limestone'	HÄ15-20	0.87	−6.76
'Holen Limestone'	HÄ12-37	0.87	−6.84
'Holen Limestone'	HÄ15-21	0.84	−6.57
'Holen Limestone'	HÄ12-36	0.88	−6.45
'Holen Limestone'	HÄ15-22	0.95	−6.69
'Holen Limestone'	HÄ15-23	0.71	−6.82
'Holen Limestone'	HÄ12-35	0.85	−6.77
'Holen Limestone'	HÄ15-24	0.93	−7.09
'Holen Limestone'	HÄ15-25	1.19	−6.32
'Holen Limestone'	HÄ15-26	0.98	−6.67
'Holen Limestone'	HÄ12-34	0.67	−7.13
'Holen Limestone'	HÄ15-27	0.90	−6.94
'Holen Limestone'	HÄ15-28	0.80	−6.61
'Holen Limestone'	HÄ12-33	0.57	−7.03
'Holen Limestone'	HÄ15-29	0.68	−6.43
'Holen Limestone'	HÄ12-32	0.60	−7.33
'Holen Limestone'	HÄ12-31	0.48	−7.35
'Holen Limestone'	HÄ12-30	0.80	−7.90
'Holen Limestone'	HÄ12-29	0.66	−6.87
'Holen Limestone'	HÄ12-28	0.38	−7.90
'Holen Limestone'	HÄ12-27	−1.03	−6.93
'Holen Limestone'	HÄ12-26	0.46	−6.81
'Holen Limestone'	HÄ12-25	0.89	−6.99
'Holen Limestone'	HÄ12-24	0.81	−6.64
'Holen Limestone'	HÄ12-23	0.46	−6.88
'Holen Limestone'	HÄ12-22	0.84	−7.00
'Holen Limestone'	HÄ12-21	0.78	−5.94
'Holen Limestone'	HÄ12-20	0.38	−6.70
'Holen Limestone'	HÄ15-30	0.63	−6.38
'Holen Limestone'	HÄ12-19	−0.02	−7.64
'Holen Limestone'	HÄ15-31	0.60	−6.09
'Holen Limestone'	HÄ15-32	0.80	−6.16
'Holen Limestone'	HÄ12-18	0.75	−6.60
'Holen Limestone'	HÄ12-17	1.05	−5.77
'Holen Limestone'	HÄ12-16	0.69	−6.31
'Holen Limestone'	HÄ12-15	0.90	−6.18
'Holen Limestone'	HÄ12-14	0.63	−6.42
'Holen Limestone'	HÄ12-13	0.65	−6.31
'Holen Limestone'	HÄ12-12	0.87	−6.01
'Holen Limestone'	HÄ15-33	0.72	−6.19
'Holen Limestone'	HÄ15-34	0.93	−6.17
'Holen Limestone'	HÄ12-11	0.82	−6.34
'Holen Limestone'	HÄ15-35	0.85	−6.34
'Holen Limestone'	HÄ15-36	0.81	−6.66
'Holen Limestone'	HÄ12-10	0.72	−6.95
'Holen Limestone'	HÄ12-9	0.39	−7.26
'Holen Limestone'	HÄ12-8	0.83	−5.91
'Holen Limestone'	HÄ12-7	0.65	−6.87
'Holen Limestone'	HÄ12-6	0.30	−6.90
'Holen Limestone'	HÄ12-5	0.75	−6.20
'Holen Limestone'	HÄ12-4	0.41	−6.33
'Holen Limestone'	HÄ12-3	1.24	−5.39
'Holen Limestone'	HÄ12-2	0.88	−6.18
'Holen Limestone'	HÄ12-1	0.40	−6.72
'Holen Limestone'	HÄ12-39	0.57	−6.69
'Holen Limestone'	HÄ12-40	0.61	−6.63
'Holen Limestone'	HÄ12-41	−0.30	−6.30
'Holen Limestone'	HÄ15-37	0.64	−6.19
'Holen Limestone'	HÄ12-42	0.79	−5.52
'Holen Limestone'	HÄ15-38	0.56	−6.27
'Holen Limestone'	HÄ12-43	0.72	−5.54
'Holen Limestone'	HÄ15-39	0.59	−6.20
'Holen Limestone'	HÄ15-40	0.90	−5.57
'Holen Limestone'	HÄ12-44	0.70	−6.41
'Holen Limestone'	HÄ12-45	0.62	−6.72
'Holen Limestone'	HÄ12-46	0.24	−7.52
'Holen Limestone'	HÄ12-47	0.52	−6.92
'Holen Limestone'	HÄ12-48	0.65	−6.69
'Holen Limestone'	HÄ12-49	0.35	−8.68
'Holen Limestone'	HÄ12-50	0.34	−7.52
'Holen Limestone'	HÄ12-51	0.90	−5.90
'Holen Limestone'	HÄ12-52	0.65	−6.84
'Holen Limestone'	HÄ12-53	0.59	−7.24
'Holen Limestone'	HÄ12-54	0.66	−6.84
'Holen Limestone'	HÄ12-55	0.23	−7.77
'Holen Limestone'	HÄ12-56	0.38	−7.34
'Holen Limestone'	HÄ12-57	0.59	−7.69
'Holen Limestone'	HÄ12-58	−0.02	−7.55
'Holen Limestone'	HÄ12-59	0.23	−7.43
'Holen Limestone'	HÄ12-60	−0.30	−8.16
'Holen Limestone'	HÄ12-61	0.23	−7.52
'Lanna Limestone'	HÄ12-62	0.34	−6.87
'Lanna Limestone'	HÄ12-63	−0.02	−7.54
'Lanna Limestone'	HÄ12-64	0.53	−6.27
'Lanna Limestone'	HÄ12-65	−0.21	−7.60
'Lanna Limestone'	HÄ12-66	−0.10	−7.57
'Lanna Limestone'	HÄ12-67	0.03	−7.42
'Lanna Limestone'	HÄ12-68	0.27	−6.52
'Lanna Limestone'	HÄ12-69	−0.46	−7.56
'Lanna Limestone'	HÄ12-70	0.76	−5.96
'Lanna Limestone'	HÄ12-71	0.36	−7.28
'Lanna Limestone'	HÄ12-72	0.42	−6.58
'Lanna Limestone'	HÄ12-73	0.11	−6.92
'Lanna Limestone'	HÄ12-74	0.71	−5.95
'Lanna Limestone'	HÄ12-75	0.22	−6.64

(continued)

Table 1. (continued)

Stratigraphical unit	Sample	$\delta^{13}C_{carb}$	$\delta^{18}O_{carb}$
'Lanna Limestone'	HÄ12-76	0.04	−6.86
'Lanna Limestone'	HÄ12-77	0.33	−6.05
'Lanna Limestone'	HÄ12-78	−0.59	−6.63
'Lanna Limestone'	HÄ12-79	0.02	−7.18
'Lanna Limestone'	HÄ12-80	−0.45	−6.75
'Lanna Limestone'	HÄ12-81	0.26	−6.38
'Lanna Limestone'	HÄ12-82	0.44	−6.19
'Lanna Limestone'	HÄ12-83	0.18	−6.08
'Lanna Limestone'	HÄ12-84	0.17	−6.08
'Lanna Limestone'	HÄ12-85	0.35	−6.09
'Lanna Limestone'	HÄ12-86	−0.23	−6.62
'Lanna Limestone'	HÄ12-87	0.25	−6.98
'Lanna Limestone'	HÄ12-88	0.28	−6.26
'Lanna Limestone'	HÄ12-89	0.22	−6.12
'Lanna Limestone'	HÄ12-90	0.34	−6.78
'Lanna Limestone'	HÄ12-91	−0.08	−7.40
'Lanna Limestone'	HÄ12-92	−0.05	−6.79
'Lanna Limestone'	HÄ12-93	0.10	−6.51
'Lanna Limestone'	HÄ12-94	0.27	−6.18
'Lanna Limestone'	HÄ12-95	0.27	−6.24
'Lanna Limestone'	HÄ12-96	−0.03	−7.04
'Lanna Limestone'	HÄ12-97	0.15	−6.72
'Lanna Limestone'	HÄ12-98	0.19	−6.67
'Lanna Limestone'	HÄ12-99	0.29	−6.53
'Lanna Limestone'	HÄ12-100	0.18	−6.80
'Lanna Limestone'	HÄ12-101	0.46	−5.99
'Lanna Limestone'	HÄ12-102	0.52	−6.03
'Lanna Limestone'	HÄ12-103	0.71	−5.81
'Lanna Limestone'	HÄ12-104	0.37	−6.26
'Lanna Limestone'	HÄ12-105	0.32	−6.64
'Lanna Limestone'	HÄ12-106	0.58	−6.20
'Lanna Limestone'	HÄ12-107	−0.40	−7.34
'Lanna Limestone'	HÄ12-108	0.53	−6.64
'Lanna Limestone'	HÄ12-109	−0.97	−7.55
'Lanna Limestone'	HÄ12-110	0.73	−6.28
'Lanna Limestone'	HÄ12-111	0.49	−6.36
'Lanna Limestone'	HÄ12-112	0.46	−6.36
'Lanna Limestone'	HÄ12-113	−0.44	−6.90
'Lanna Limestone'	HÄ12-114	0.43	−7.09
'Lanna Limestone'	HÄ12-115	−0.52	−7.02
'Lanna Limestone'	HÄ12-116	0.71	−6.57
'Lanna Limestone'	HÄ12-117	0.79	−6.29
'Lanna Limestone'	HÄ12-118	0.76	−6.13
'Lanna Limestone'	HÄ12-119	0.85	−6.15
'Lanna Limestone'	HÄ12-120	−0.14	−7.42
'Lanna Limestone'	HÄ12-121	0.36	−7.14
'Lanna Limestone'	HÄ12-122	0.57	−6.59
'Lanna Limestone'	HÄ12-123	−0.07	−6.69
'Lanna Limestone'	HÄ12-124	0.68	−6.54
'Lanna Limestone'	HÄ12-125	−0.05	−7.19
'Lanna Limestone'	HÄ15-41	0.50	−6.53
'Lanna Limestone'	HÄ12-126	0.20	−6.92
'Lanna Limestone'	HÄ15-42	0.72	−6.60
'Lanna Limestone'	HÄ12-127	0.53	−6.69
'Lanna Limestone'	HÄ12-128	−0.80	−7.24
'Lanna Limestone'	HÄ15-43	0.35	−6.59
'Lanna Limestone'	HÄ12-129	0.34	−6.94
'Lanna Limestone'	HÄ15-44	0.18	−6.52
'Lanna Limestone'	HÄ12-130	0.67	−6.22
'Lanna Limestone'	HÄ12-131	0.10	−7.19
'Lanna Limestone'	HÄ12-132	0.02	−6.66
'Lanna Limestone'	HÄ12-133	0.27	−7.27
'Lanna Limestone'	HÄ12-134	0.47	−5.94
'Lanna Limestone'	HÄ12-135	0.11	−6.45
'Lanna Limestone'	HÄ15-C6	0.42	−6.76
'Lanna Limestone'	HÄ15-C5	0.38	−6.89
'Lanna Limestone'	HÄ15-C1	0.34	−6.58

(continued)

Table 1. (continued)

Stratigraphical unit	Sample	$\delta^{13}C_{carb}$	$\delta^{18}O_{carb}$
'Lanna Limestone'	HÄ15-45	0.35	−6.46
'Lanna Limestone'	HÄ15-B1	0.17	−6.40
'Lanna Limestone'	HÄ15-B2	0.19	−6.34
'Lanna Limestone'	HÄ15-B3	−0.04	−6.58
'Lanna Limestone'	HÄ15-B4	0.19	−6.70
'Lanna Limestone'	HÄ15-C2	−1.89	−9.02
'Lanna Limestone'	HÄ15-D12	0.13	−6.80
'Lanna Limestone'	HÄ15-B5	0.31	−6.60
'Lanna Limestone'	HÄ15-C3	0.25	−6.88
'Lanna Limestone'	HÄ15-D11	0.16	−6.63
'Lanna Limestone'	HÄ15-D10	−0.21	−7.10
'Lanna Limestone'	HÄ15-C4	0.22	−7.02
'Lanna Limestone'	HÄ15-D9	0.04	−7.12
'Lanna Limestone'	HÄ15-D8	−2.83	−17.07
'Lanna Limestone'	HÄ15-D7	0.13	−6.85
'Lanna Limestone'	HÄ15-D6	−0.06	−6.70
'Lanna Limestone'	HÄ15-D5	−0.37	−6.49
'Lanna Limestone'	HÄ15-D4	−0.13	−6.52
'Lanna Limestone'	HÄ15-D3	−0.05	−6.58
'Lanna Limestone'	HÄ15-D2	0.45	−6.38
'Lanna Limestone'	HÄ15-D1	0.36	−6.55
Tøyen Shale	HÄ15-D13	0.65	−6.33
Tøyen Shale	HÄ15-D14	0.33	−6.83
Tøyen Shale	HÄ15-D15	0.41	−6.34
Tøyen Shale	HÄ15-D16	0.62	−6.40

present study shows that the data sets match up well and that the $\delta^{13}C_{carb}$ curves coincide in terms of both stratigraphical pattern and value ranges. This attests to the reliability of the overall approach and data used whilst simultaneously comparing two different carbon isotope laboratories.

Carbon isotopes ($\delta^{13}C_{carb}$)

The $\delta^{13}C_{carb}$ values recorded are relatively uniform, mainly varying within the range of 0‰ to +1‰ throughout the study succession, although a long-term trend towards successively more positive values occurs up-section (Fig. 3). Beginning at the base of the studied succession, there is a marked albeit minor (~0.5‰) negative shift in values across the Billingen–Volkhov (~Floian–Dapingian) boundary interval, marked C1 in Fig. 3. This part of the local succession is characterized by abundant limonitic discontinuity surfaces (Lindskog & Eriksson 2017). A gradual trend towards more positive values (~1‰ magnitude shift) is seen throughout the lower Volkhovian. Within this general positive trend in $\delta^{13}C_{carb}$ values, there is a relatively large scattering of data points that coincides with a distinct limestone–marl alternation interval in the 'Lanna Limestone' at Kinnekulle (between C1 and C2 in Fig. 3). A ~0.5‰ drop to lower $\delta^{13}C_{carb}$ values occurs in the middle-most Volkhovian (C2). This negative shift is not related to any obvious lithological change, although detailed biotic and sedimentary facies analyses

indicate a rise in the relative sea level (see Lindskog & Eriksson 2017). Succeeding $\delta^{13}C_{carb}$ values increase gradually up-section until a minor but marked negative shift occurs again close to the Volkhov–Kunda boundary (C3). These beds contain numerous limonitic discontinuity surfaces and limonite-stained grains, and locally derived intraclasts. The basal Kunda is associated with a rapid rise in $\delta^{13}C$ ‰ values. A slight but distinguishable decline to slightly lighter (~0.25‰ magnitude) values occurs in the middle Kunda interval (C4). This interval of lowered $\delta^{13}C_{carb}$ values coincides closely with the *Microzarkodina hagetiana* Subzone of the *Lenodus pseudoplanus* conodont Zone (Zhang 1998a). It is followed up-section by increasingly higher values (up to +1.40‰) – with minor but pronounced rhythmic/cyclic fluctuations – leading into the MDICE interval. The onset of the MDICE is close to the base of the *Microzarkodina ozarkodella* Subzone of the *L. pseudoplanus* conodont Zone (ibid.) and is associated with a change into rocks with a clearly different lithology compared to those in the underlying strata, and the establishment of a pronounced alternation between limestone and marly/shaly interbeds (Lindskog & Eriksson 2017). The limestone of these strata is of exceptionally fine carbonate texture compared to that of the lower and middle parts of the 'Holen Limestone', and the fossil fauna is poor. The disconformable contact between the 'Holen Limestone' and the 'Skövde Limestone' is associated with a negative shift (~0.75‰ magnitude) in $\delta^{13}C_{carb}$ values, although the values increase again somewhat in the basal Gullhögen Formation (see also Schmitz *et al.* 2010). This part of the study succession is characterized by irregular beds and lenses of greyish limestone (~mudstone) and calcareous shale containing ferruginous and phosphatic ooids and skeletal debris (see Jaanusson 1964; Lindskog & Eriksson 2017).

Oxygen isotopes ($\delta^{18}O_{carb}$)

Most $\delta^{18}O_{carb}$ values vary between −7‰ and −6‰ throughout the succession, although there are some notable fluctuations (Fig. 3). Scatter within the $\delta^{18}O_{carb}$ data is typically quite pronounced, but more so in the Volkhovian than in the upper part of the succession. The transition between the Billingen and Volkhov Baltoscandian stages (~Tøyen Shale–'Lanna Limestone') is associated with a distinct, ~0.5‰ magnitude shift into lower $\delta^{18}O_{carb}$ values, marked O1 in Fig. 3. The Volkhov Stage is thereafter characterized by $\delta^{18}O_{carb}$ values scattering across a similar long-term mean (~−6.5‰). A distinct ~1‰ negative shift (from −6.5‰ to −7.5‰) in $\delta^{18}O_{carb}$ values is then seen across the Volkhov–Kunda boundary interval (O2). This is followed by a rapid rise in values in the lower Kunda interval, although relatively low values (−7.0‰) occur in the upper 'Täljsten' beds (O3), in the *Asaphus expansus–Asaphus raniceps* trilobite Zone and *Lenodus variabilis–Yangtzeplacognathus crassus* conodont Zone boundary interval. After a sharp rise in the overlying strata to $\delta^{18}O_{carb}$ values near −6.0‰, a marked drop in values to ~−7.0‰ is then observed in the middle Kunda interval (O4), within the *M. hagetiana* Subzone of the *L. pseudoplanus* conodont Zone. This is associated with a gradual change in lithology, with the rocks becoming increasingly marly until the abovementioned limestone–marl/limestone–shale alternation is fully developed (see Lindskog & Eriksson 2017). The $\delta^{18}O_{carb}$ values are relatively stable, near −7.0‰, up-section, but a consistent increase is observed in the uppermost Kunda beds (O5). The transition between the 'Holen Limestone' and the 'Skövde Limestone'–Gullhögen Formation is marked by a ~1.0‰ magnitude negative shift in $\delta^{18}O_{carb}$ values.

Discussion

The thermally and tectonically undisturbed strata of the 'orthoceratite limestone' at the Hällekis quarry appear to preserve a relatively good record of regional to global changes in marine $\delta^{13}C_{carb}$ and in part $\delta^{18}O_{carb}$. Several recently published papers deal with stratigraphical variations in $\delta^{13}C$ in successions coeval with the Hällekis study interval, but few have discussed the corresponding $\delta^{18}O$ records (although many studies include $\delta^{18}O$ data sets). In large part, this is likely due to the observation that $\delta^{18}O$ is relatively sensitive to lithological variations and diagenetic alterations compared to $\delta^{13}C$ (e.g. Kah 2000; Melim *et al.* 2001; and references therein). The broad scattering of the Hällekis data points indicates that $\delta^{18}O_{carb}$ values are indeed strongly influenced by both lithological characteristics and diagenetic processes, and thus less reliable for palaeoenvironmental interpretations and stratigraphical correlation than the corresponding $\delta^{13}C_{carb}$ data. Still, the stratigraphical patterns in the $\delta^{18}O_{carb}$ data show some clear similarities to those of other regional localities and different proxies that deserve closer inspection and discussion (see below).

As discussed recently in some detail by Saltzman & Edwards (2017), much information about the chemical and physical characteristics of the atmosphere and the oceans during the Ordovician remains to be unravelled. Carbon and oxygen

isotopes, together with some other geochemical proxies, show promise to help in elucidating such information. However, in order to better understand the dynamics of long-term carbon and oxygen cycles of the Palaeozoic, great care must be exercised in assessing similarities and differences in stable isotope data sets. With increasing distance from open oceanic settings, within the epeiric basins, comparisons at the global scale are likely to become more difficult due to increasingly restricted oceanographical circulation.

Here, we first evaluate our new data within the context of factors that may affect the preservation of primary stable isotopic trends (i.e. diagenesis, carbonate mineralogy). Once established that our records reflect mostly primary Ordovician marine chemistry, we go on to interpret these records in the context of factors that influence these stable isotopic data within shallow water carbonate platform settings: water mass restriction/connectivity, organic matter decomposition, temperature, salinity and $CO_{2(aq)}$ (e.g. Holmden *et al.* 1998; Immenhauser *et al.* 2003; Panchuk *et al.* 2005; Swart 2008). With the aforementioned in mind, we explore possible changes to the regional to global environment (i.e. temperature, salinity, $CO_{2(aq)}$, burial flux of organic carbon) to produce these small but distinct changes in the $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ records from the Baltoscandian palaeobasin.

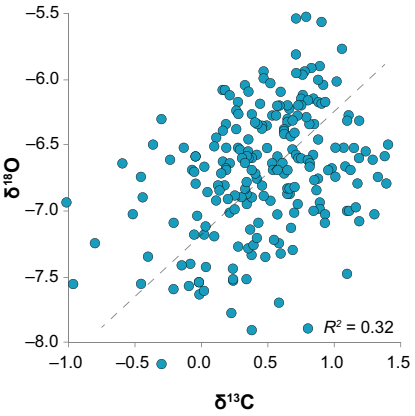

Fig. 4. Cross-plot of the $\delta^{13}C$ and $\delta^{18}O$ data with linear (least-squares regression) trend line indicated. [Colour figure can be viewed at wileyonlinelibrary.com]

Diagenetic influence on the studied rocks

The 'orthoceratite limestone' at Kinnekulle typically shows evidence of early lithification and only very limited (late) diagenetic alteration (see Lindskog & Eriksson 2017; and references therein). The early lithification, together with a generally fine-grained composition and negligible primary and secondary porosity, has ensured that the rocks have retained many of their original chemical and physical characteristics throughout time; few beds show any significant recrystallization effects and little interchange of materials between beds can be detected (e.g. Lindskog *et al.* 2017). The Kinnekulle area has largely been spared from tectonic disturbances throughout the Phanerozoic, and the colour alteration index (~1) of conodonts from the 'orthoceratite limestone' reveals negligible heating of the rocks (e.g. Mellgren & Eriksson 2010). Cross-plotting of the $\delta^{13}C$ and $\delta^{18}O$ data (Fig. 4) shows only limited correlation between numbers ($R^2 = 0.32$), indicating a general lack of influence from meteoric diagenesis (e.g. Allan & Matthews 1982; Jones *et al.* 2015). Possible meteoric influence is especially apparent close to some biostratigraphical boundaries, which likely coincide with slowed and/or temporarily halted deposition, although there are no obvious signs of subaerial exposure at Kinnekulle (Fig. 3). Even without meteoric influence, however, some correlation (positive or negative) can be expected if the two isotope systems were influenced by the same ambient environmental conditions and processes (see below). Original lithological properties may also contribute. Collectively, the conditions suggest that the studied rocks retain relatively pristine records of carbon and oxygen isotopes.

Geographical comparisons and correlations

Coeval carbon isotope curves from Baltoscandian localities (outcrops and drill cores) often show consistent general patterns and trends, both in shorter and longer time perspectives (Fig. 5). The prominent negative shift (C1) in the lowermost Volkhov interval appears to be well reflected in successions that, like Kinnekulle, show a change in facies from argillaceous rocks to relatively pure limestone in this interval, but this shift is absent or poorly developed in successions completely dominated by limestone. As at Kinnekulle, many localities show a two-step (cyclic?) $\delta^{13}C$ development in the Volkhov Baltoscandian Stage. This can be observed as gradually increasing $\delta^{13}C$ values upward from the base to about the middle Volkhov, where there is a slight but marked temporary dip in values (C2) in the *Baltoniodus norrlandicus* conodont Zone (*sensu* Wu *et al.* 2017). Except in the most condensed successions, where this interval is characterized by a marked hiatus, a small magnitude (~0.5‰) negative shift in $\delta^{13}C_{carb}$ values (C3) is evident also near the Volkhov–Kunda boundary (the *Asaphus lepidurus*/*Megistaspis limbata*–*A. expansus* trilobite Zone

boundary). The succeeding Kunda Stage is consistently characterized by a gradual but fluctuating rise in $\delta^{13}C$ values, with another small magnitude negative shift of ~0.25–0.5‰ (C4) in the lower *L. pseudoplanus* conodont Zone. This is followed by the geographically widespread and globally documented MDICE. A slight negative shift in $\delta^{13}C$ values is typically seen also in the topmost Kunda Stage, within the MDICE interval (the uppermost *L. pseudoplanus* Zone/lowermost *Eoplacognathus suecicus* conodont Zone; see also Lehnert *et al.* 2014). It appears that different 'phases' of the overall stratigraphical $\delta^{13}C$ development are of varying relative thickness throughout Baltoscandia, revealing important information on the spatiotemporal depositional dynamics in the regional palaeobasin – that is, that different areas were characterized by deposition and/or erosion at different times. At Kinnekulle, the conspicuous hiatus below the 'Skövde Limestone' cuts out most of the MDICE (see also Schmitz *et al.* 2010). A corresponding stratigraphical gap can be traced along a belt southwards and into central Eastern Europe (and a similar gap also occurs elsewhere; e.g. Schmitz *et al.* 2010), but much of Baltoscandia experienced significant and essentially continuous deposition during this time (Fig. 5; Lindskog & Eriksson 2017).

Whilst most successions in Baltoscandia show similar overall $\delta^{13}C$ patterns through the Volkhov–Kunda interval, significant variations are observable between different areas and localities. For example, the Siljan area in Dalarna, central Sweden, deviates from other areas as it shows a long-term trend of strongly declining $\delta^{13}C$ values throughout the Volkhov Stage that culminates in a pronounced low, or 'negative excursion', in values across the Volkhov–Kunda boundary (LDNICE of Lehnert *et al.* 2014; possibly a condensation-induced amalgamation of

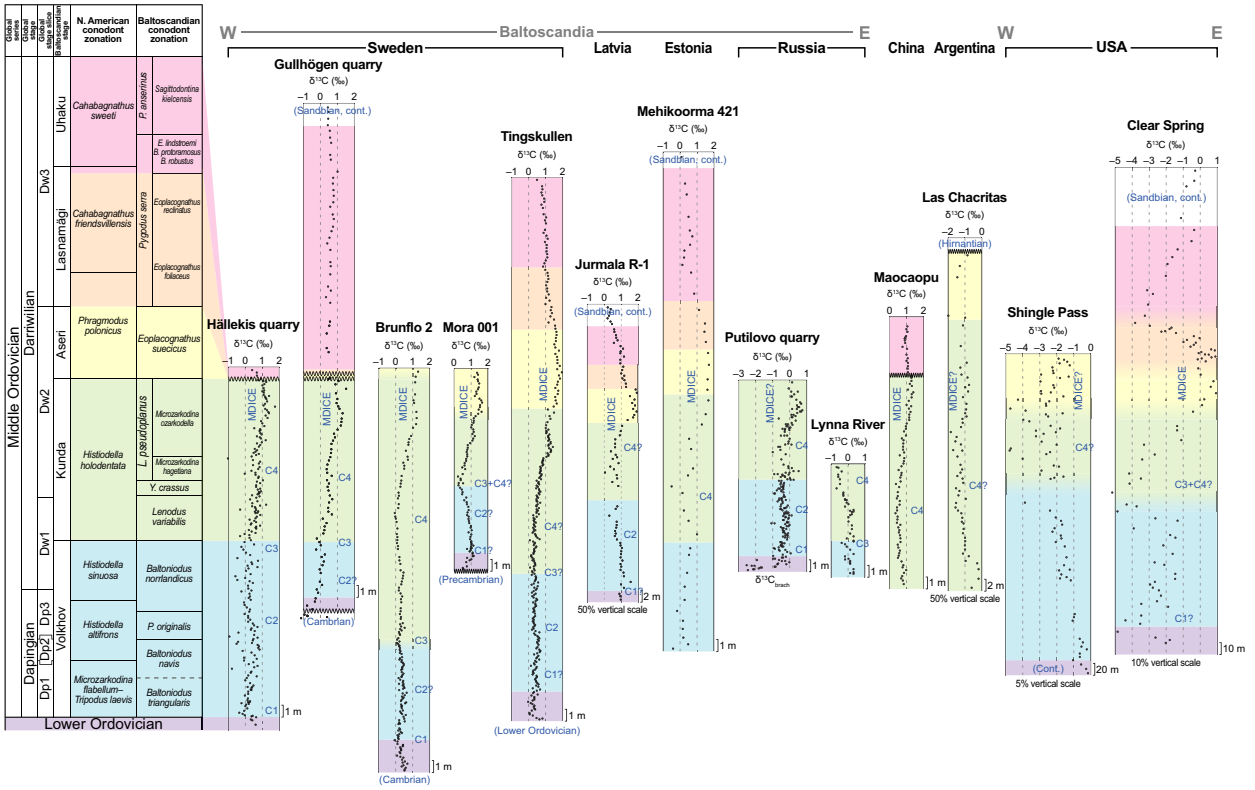

Fig. 5. Comparison between the carbon isotope ($\delta^{13}C_{carb}$) curve at Hällekis with (portions of) select $\delta^{13}C_{carb}$ curves from other localities (Gullhögen quarry, outcrop: Meidla *et al.* 2004; Brunflo 2, drill core: Wu *et al.* 2017; Mora 001, drill core; Lehnert *et al.* 2014; Tingskullen, drill core: Wu *et al.* 2017; Jurmala R-1, drill core: Ainsaar *et al.* 2004; Mehikoorma 421, drill core: Kaljo *et al.* 2007; Putilovo quarry, outcrop: Rasmussen *et al.* 2016; Lynna River, outcrop: Zaitsev & Pokrovsky 2014; Maocaopu, outcrop: Schmitz *et al.* 2010; Las Chacritas, outcrop: Albanesi *et al.* 2013; Shingle Pass, outcrop: Edwards & Saltzman 2014; Clear Spring, outcrop: Saltzman & Edwards 2017). All sections are shown to the same vertical scale (i.e. their relative thicknesses are represented correctly), except where noted. Colour coding indicates biostratigraphical correlation as tied to the Baltoscandian stratigraphical framework (modified from Lindskog *et al.* 2017); graded transitions indicate uncertain boundaries (biostratigraphical data from Jaanusson & Mutvei 1951; Jaanusson 1964; Karis 1998; Zhang 1998a; Löfgren 2003; Ainsaar *et al.* 2004; Meidla *et al.* 2004; Bergström 2007; Kaljo *et al.* 2007; Schmitz *et al.* 2010; Albanesi *et al.* 2013; Edwards & Saltzman 2014; Rasmussen *et al.* 2016; Lindskog & Eriksson 2017; Saltzman & Edwards 2017; Wu *et al.* 2017; see further references therein). [Colour figure can be viewed at wileyonlinelibrary.com]

C3 and C4). Thereafter, the Kunda interval is characterized by a steady increase from 'baseline' values leading directly into the MDICE (Ainsaar *et al.* 2010; Lehnert *et al.* 2014). In the Tingskullen drill core from northern Öland in southeastern Sweden, the negative shift in $\delta^{13}C$ values that is characteristic of the middle Kunda (C4) is essentially not developed, whereas this shift is especially well pronounced in the Jämtland region, central Sweden, and in the easternmost Baltic successions (Ainsaar *et al.* 2010; Zaitsev & Pokrovsky 2014; Wu *et al.* 2015, 2017; Rasmussen *et al.* 2016). The $\delta^{13}C$ values are relatively low in western Russia compared to those of other areas in the Baltoscandian palaeobasin (cf. Immenhauser *et al.* 2003). Variations in stratigraphical $\delta^{13}C$ patterns and values between different areas possibly reflect varying weathering sources and intensity of sediment input and that all parts of the Baltoscandian palaeobasin were not always well connected in terms of oceanographical circulation and exchange with the open ocean (especially during lowstands; see Saltzman & Edwards 2017 and references therein). The timing and tempo of traceable changes also appears to differ somewhat (cf. Fanton & Holmden 2007; Schiffbauer *et al.* 2017; Rasmussen & Stouge 2018). These differences stress the need for caution when interpreting $\delta^{13}C$ curves in extreme high-resolution details and highlight the importance of biostratigraphical control when correlating sections. Biostratigraphy in itself is obviously not without problems, however, as the timing of biostratigraphical boundaries appears to vary somewhat between well-studied sections (i.e. they are arguably asynchronous; e.g. Lindskog & Eriksson 2017).

Detailed comparisons between $\delta^{13}C$ curves become more problematic when expanding the scope to the global scale, although some general patterns appear to be recognizable (Fig. 5). The study interval is characterized by a long-term increase in $\delta^{13}C$ values in most successions, although there are exceptions and the amplitude of changes differs drastically between regions. Most significantly, data sets from the Great Basin in western USA and the Precordillera of Argentina show long-term standstill or even decrease in $\delta^{13}C$ values, and the MDICE is not clearly discernible (cf. Albanesi *et al.* 2013; Edwards & Saltzman, 2014; Saltzman & Edwards 2017; Henderson *et al.* 2018). Moreover, the stratigraphical patterns of $\delta^{13}C$ curves differ significantly between western USA, as represented by the Shingle Pass section in Nevada, and eastern USA, as represented by the Clear Spring section in Maryland; the long-term trends throughout the Dapingian–Darriwilian interval are essentially mirrored. Regardless of differences in stratigraphical patterns, both Argentinian and North American successions typically show distinctly lower $\delta^{13}C$ values compared to those from Baltoscandia. Whereas a minor part of this may be explained by temperature-dependent fractionation (e.g. Goericke & Fry 1994; Hayes *et al.* 1999), and other physical processes, most of the differences are likely due to global/regional differences in carbon stocks (e.g. Saltzman & Edwards 2017, and references therein). Furthermore, different sections appear to have been strongly influenced by local physical and chemical syn- and post-depositional conditions.

Although the data scatter conspicuously, most sections in Baltoscandia appear to record the same overall trend in $\delta^{18}O$ values (Fig. 6). The study interval is characterized by a long-term rise in $\delta^{18}O$ values with recurring (cyclic?) negative shifts that appear traceable between sections. The amplitude of variations is much less distinct in Jämtland than in other parts of Baltoscandia. This likely reflects that the Jämtland area was a relatively deep-water setting, but the relatively moderate scattering in the Brunflo 2 drill core data may also indicate homogenization during diagenesis (cf. Wu *et al.* 2015). The data from the Mehikoorma 421 drill core in Estonia differ drastically from the other Baltoscandian data sets in that the post-Kunda interval is characterized by relatively low $\delta^{18}O$ values. The data from the United States also show a long-term trend of increasing $\delta^{18}O$ values, although with distinct differences in stratigraphical range/position and amplitude of changes between sections. Curiously, Clear Spring shows a similar small magnitude drop in $\delta^{18}O$ values approximately through the same stratigraphical interval as in Mehikoorma 421. Despite the susceptibility of oxygen isotopes to alteration, data from bulk carbonates show remarkably similar patterns to those produced from conodont elements and brachiopod shells (Trotter *et al.* 2008; Zaitsev & Pokrovsky 2014; Rasmussen *et al.* 2016). There is a tendency for bimodality in the $\delta^{18}O$ values in some parts of the Hällekis succession, with a marked and potentially systematic difference between minimum and maximum values in the data scatter, as can also be observed in the Tingskullen drill core (Fig. 6). This may reflect high-frequency palaeoenvironmental and/or diagenetic variations.

A distinct trend is observed in the $\delta^{18}O$ data across Baltoscandia, wherein values become lower roughly from east to west; values typically vary between $\sim -4‰$ and $-5‰$ in the easternmost part of the region (western Russia), and between $\sim -8‰$ and $-10‰$ in its western parts (Fig. 6; cf. Immenhauser *et al.* 2003). Assuming that $\delta^{18}O$ reflects water temperature in the depositional environment, the data

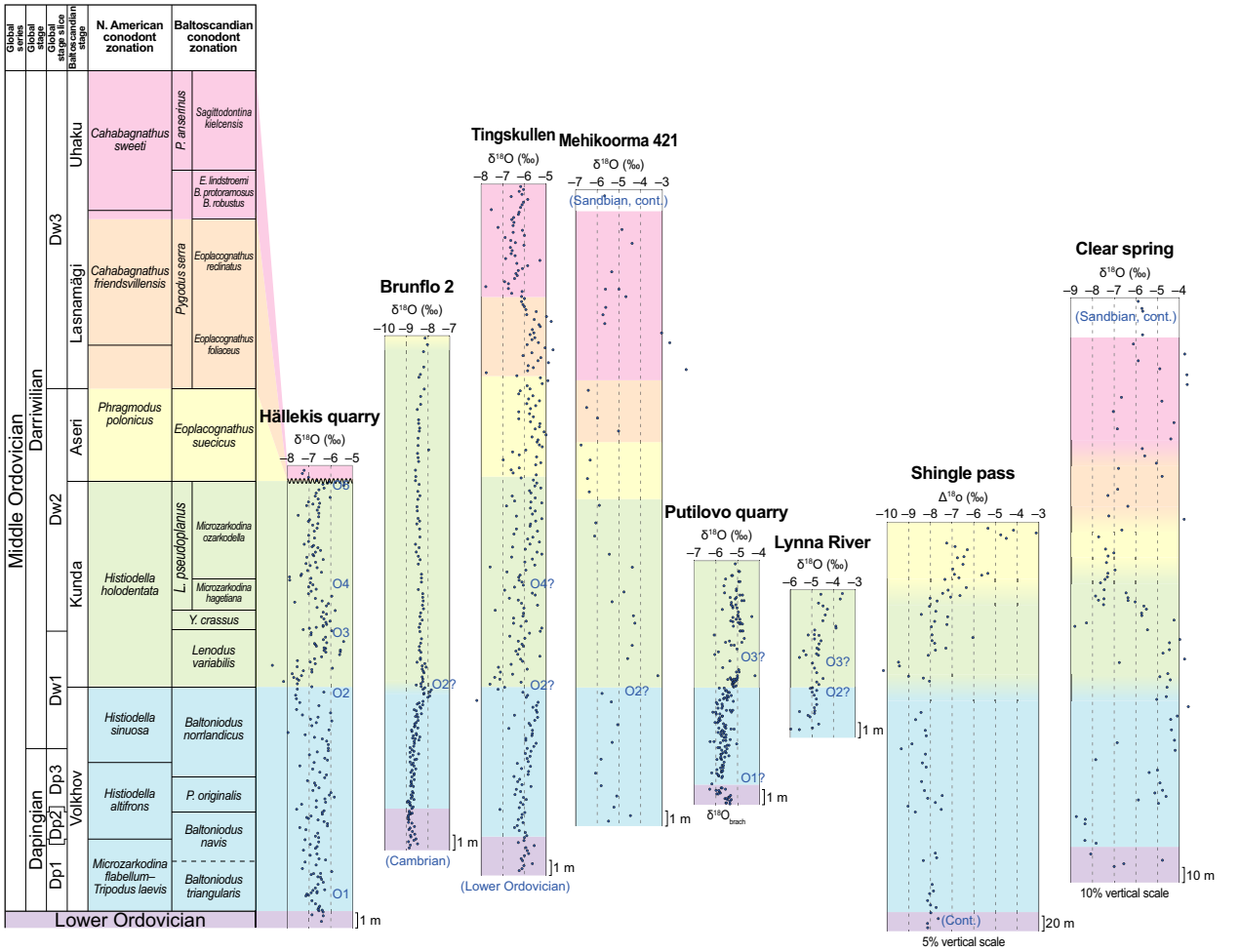

Fig. 6. Comparison between the oxygen ($\delta^{18}O_{carb}$) isotope curve at Hällekis with select $\delta^{18}O_{carb}$ curves from other localities (from works providing both $\delta^{13}C$ and $\delta^{18}O$ data sets). All sections are shown to the same vertical scale, except where noted. Colour coding indicates biostratigraphical correlation as tied to the Baltoscandian stratigraphical framework; graded transitions indicate uncertain boundaries. For references, see Fig. 5. [Colour figure can be viewed at wileyonlinelibrary.com]

suggest a gradient of ~15–20°C (Epstein *et al.* 1953) with warmer water to the west. This may seem to be in contrast with what can be intuitively expected as water generally became deeper and more oceanic in western direction, and thus likely colder, especially when considering potential upwelling areas (e.g. Lindström & Vortisch 1983; Rasmussen *et al.* 2016; cf. Lindström 1984). However, palaeogeographical reconstructions show that the present-day western edge of Baltica was oriented northwards in the Middle Ordovician (e.g. Torsvik & Cocks 2017). Thus, this part of the palaeocontinent was facing the Iapetus Ocean, and the regional trend in $\delta^{18}O$ could reflect increasing exposure to warmer waters coming in from lower latitudes. Conodont biofacies analyses support this interpretation (Rasmussen & Stouge 2018), although further studies are needed in order to elucidate what (and how) other factors and processes may have influenced the observed $\delta^{18}O$ trends. In the United States, $\delta^{18}O$ values span the entire range of those seen in Baltoscandia, and stratigraphical variations are somewhat contradictory (Fig. 6). This suggests that specific values do not translate directly into temperature, as Laurentia was consistently situated in distinctly warmer latitudes as compared to most of Baltica (e.g. Torsvik & Cocks 2017), and the amplitude of variations in $\delta^{18}O$ indicates relatively complex depositional and/or diagenetic histories compared to sections elsewhere. Still, the similarity between different $\delta^{18}O$ data sets and trends indicates that bulk rock oxygen isotopes commonly retain primary signals and, thus, may provide valuable palaeoenvironmental information that is rarely acknowledged or even discussed in the recent literature (cf. Lindström 1984).

Carbon and oxygen isotopes vs. palaeoenvironmental development

Comparison between isotopic, palaeontological and sedimentological data (see Lindskog & Eriksson

2017; and references therein) from the Hällekis quarry indicates a near inverse relationship between carbon isotope values and estimated relative sea level through most of the study interval, although the labelled shifts (C1–C4) deviate from this general pattern (Fig. 3). This probably reflects that times of higher sea level were associated with influx of more open marine waters into the Baltoscandian palaeobasin and that lowstands (and shallower water areas in general) were associated with a more 'regional' and variable $\delta^{13}C$ signal (e.g. Immenhauser *et al.* 2002; Immenhauser *et al.* 2003; Bergström *et al.* 2011; Edwards & Saltzman 2014). Oxygen isotopes also appear to have covaried with relative sea level, although scattering and variability in the data preclude confident interpretations. It is noteworthy, however, that the coincident patterns suggest that temperature (and its influence on water and ice volumes) was an important driver of sea level change, and the global data support interpretations of a cooling trend through the studied time interval (see below; e.g. Trotter *et al.* 2008; Rasmussen *et al.* 2016).

The apparent pattern of inverse covariation between $\delta^{13}C$ values and sea level is decoupled in the uppermost part of the study succession, when entering into the MDICE interval. This occurs in association with a marked lithological change at Kinnekulle, as is also the case in many other areas of Sweden. The collective results and observations suggest migration of the regional depocentre(s) into the interior parts of the Baltoscandian craton, likely as a result of rising sea level (see detailed discussion in Lindskog & Eriksson 2017). This may have changed the conditions for the circulation and transport of water across the palaeobasin, here reflected in a shift in the relationship between $\delta^{13}C$ and sea level. The widespread development of the MDICE is consistent with a global rise in sea level, as continents were extensively flooded and epeiric seas brought into better contact with isotopically heavier oceanic water masses (e.g. Allan & Matthews 1982; Patterson & Walter 1994; Immenhauser *et al.* 2003; Fanton & Holmden 2007). A net trend of rising sea level through the MDICE interval has been inferred globally, although exceptions occur locally and it remains difficult to disentangle epeirogenic/isostatic signals from eustatic ones at higher resolutions (e.g. Barnes 1984; Haq & Schutter 2008; Agematsu & Sashida 2009; Dronov *et al.* 2009; Saltzman & Edwards 2017; Henderson *et al.* 2018). The expression of the MDICE may have been enhanced by changes in bioproductivity and carbon burial due to increasing shelf areas (e.g. Fanton & Holmden 2007). The predominantly small magnitude shifts in Middle Ordovician stable carbon isotopes suggest that changes in the global burial fluxes of carbon were likely small in magnitude as compared to, for example, the Late Ordovician–Silurian interval, which is characterized by multiple large-magnitude positive excursions (e.g. Saltzman 2005).

The most distinct deviations in both carbon and oxygen isotopic values (C1–C4, O1–O5; Figs 5, 6) coincide with notable palaeontological and/or sedimentological changes in the Baltoscandian palaeobasin, reasonably related to changes in sea level (e.g. Bergström *et al.* 2013; Lindskog & Eriksson 2017; Rasmussen & Stouge 2018; and references therein). As such, the typically negative shifts appear to reflect times of increased riverine input into the palaeobasin during lowstand conditions. The amplitude of changes tends to increase into the shallower (and/or proximal) parts of the palaeobasin (Fig. 1), likely as a combined result of more biotic and hydrodynamic activity and a general decrease in stratigraphical completeness due to increasing condensation and the presence of more stratigraphical gaps.

The long-term trends in ^{18}O at the global scale suggest a cooling climate through the studied time interval, theoretically of some 5–10°C in average temperature (Epstein *et al.* 1953), although these numbers unrealistically assume a lack of other influences (e.g. variations in evaporation, input of weathering products, differential diagenesis) on $\delta^{18}O$ in all concerned rock sections. Nevertheless, the notion of a cooling climate during the Middle Ordovician is consistent with interpretations by, for example, Trotter *et al.* (2008), Pohl *et al.* (2016), Rasmussen *et al.* (2016) and Rasmussen & Stouge (2018).

However interpreted, disentangling one palaeoenvironmental signature from another and/or from diagenetic ones remains a complex and problematic task that is often impossible with $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ data sets alone.

Conclusions

A high-resolution chemostratigraphical (coupled $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) study of the topmost Floian through the middle Darriwilian succession at the Hällekis quarry, Kinnekulle, southern Sweden, shows relatively steady carbon and oxygen isotope values, although some notable short-term and long-term shifts are discernible. The only confidently identified shift from 'baseline' values, currently regarded as a true (named) global isotopic excursion, is the positive MDICE.

Comparison between Middle Ordovician data sets from various localities worldwide suggests that smaller magnitude (~0.25–0.5‰) and shorter duration (10–>100 kyr) shifts in different carbon and oxygen isotope curves cannot be confidently correlated in most cases, although longer-term (≥Myr time-scales) and broader patterns appear quite consistent, especially at the regional scale. The differences in stratigraphical patterns of both carbon and oxygen isotopes between localities suggest strong secular development at several spatiotemporal scales, and global perturbations of the carbon cycle that resulted in isotopic shifts <1% in magnitude are often masked/subdued by local and regional water mass and diagenetic effects. Efforts to construct composite 'reference standards' for chemostratigraphical purposes must take into account geographical variability. Still, the stable isotope data sets support the growing body of proxy evidence suggesting progressive cooling of the Middle Ordovician oceans (e.g. Trotter *et al.* 2008; Rasmussen *et al.* 2016) and that redox conditions oscillated from oxic to dysoxic/anoxic in parts of the ocean (e.g. Young *et al.* 2016). Moreover, our results suggest that isotope data, in addition to forming a powerful correlation tool, can help in elucidating palaeogeographical scenarios.

Robust interpretation of the climatic development and its specific parameters in terms of more absolute numbers remains hampered by different tectonic histories of the palaeocontinents, and the restriction and poor circulation across intracratonic basins cause problems in disentangling global carbon and oxygen conditions from the often conflicting local, regional and global patterns. Moreover, due to the marked faunal provincialism and associated lack of common key index fossils during the Ordovician, precise biostratigraphical correlations of geographically widely separated successions are often difficult. In the absence of good biostratigraphical control of study successions, detailed comparison of isotope curves from different localities is unlikely to produce very reliable results.

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