Subaerial speleothems and deep karst in central Sweden linked to Hirnantian glaciations

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Abstract: The limestones of the upper Katian Boda mud mounds (Ordovician) of the Siljan district in central Sweden are deeply fractured. The fissures were partly synsedimentary and are often lined with stromatolite-like crusts. These crusts thus far are the only known subaerial Ordovician speleothems. They reach depths of up to 30 m below the former mound top. Macroscopically the crusts form decimetre-sized, cone-shaped domal aggregates, stalactites and stalagmites. Microfabric and morphology identify them as microbially mediated speleothems in a dark environment. Combined Sr and C isotope values indicate a formation of the speleothems from meteoric waters without influence of a significant soil horizon.

For the first time the age of the speleothems can be precisely constrained by δ13C whole-rock and brachiopod shell isotope data to the mid-Hirnantian. Repeated and/or prolonged subaerial exposure of the Boda mud mounds during the Hirnantian is evident from karst surfaces and early cements in the mound capping carbonates. The speleothems and the karst surfaces record an estimated sea-level fall in the range of 80–130 m within the time window of the Hirnantian Isotopic Carbon Excursion. This massive regression coincides with maximum ice sheet extent inferred from sections in West Gondwana.

Supplementary material: 87Sr/86Sr isotope ratio of selected brachiopod shells and results of Energy-dispersive-X-ray spectroscopy are available from http://www.geolsoc.org.uk/SUP18809

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Orдовицкий палеокарст становится широко известным на карбонатных отложениях Авалонии, Лавентании, Гондваны, Новый Китай, платформа и Балтика (например, Desrochers & James 1988; Brenchley et al. 2006; Calner et al. 2010b; Brasier 2011; Semeniuk 2011; Lehner et al. 2012; Dahlqvist et al. 2013). Субаэриальный Ордовикский карст распространяется на значительную часть области в кратонных интерьерах Северной Америки и вертикально влияет на прилегающие слои до 150 м и более (например, Keller & Lehner 2010). Субаэриальный карст является макрофациальным карстовым процессом (например, Calner et al. 2010b; Keller & Lehner 2010), который характеризуется образованием карста, включающего палеоосоли и кальцефиты (Semeniuk 2011). Однако, Ордовикский палеокарст, как правило, не описан в более ранних публикациях, и его возникновение осталось невыясненным.

Доказательства для существования подземного карста были получены в период формирования последних слоев карста Боды, представленных ранее (Jux 1966; Jaanunus 1982; Marshall & Middleton 1990; Brenchley et al. 1994). Доказательства существования подземного карста не были найдены в более ранних публикациях, что свидетельствует о том, что эта проблема является не решенной.

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δ13C analysis of whole-rock samples

The δ13C analysis of whole-rock samples was performed in the stable isotope laboratory of Michael Joachimski (GeoZentrum Nordbayern, Erlangen, Germany). The carbonate powders from
the whole-rock samples were recovered by a dental drill and reacted with 103% phosphoric acid at 70°C using a Gasbench II connected to a ThermoFinnigan Five Plus mass spectrometer. All values are reported in per mil relative to V-PDB by assigning a $\delta^{13}C +1.95‰$ to NBS19. Reproducibility and accuracy were monitored by replicate analysis of laboratory standards calibrated to NBS19 and are better than ±0.05‰ (1σ).

Brachiopod shells from key horizons were chosen for further carbonate geochemical analysis to support the bulk-rock analysis. For both the stable isotope ratios of carbon and oxygen, as well as the strontium analyses, calcite was carefully obtained from the fibrous secondary layer of the brachiopod shells. For the C + O analysis roughly 0.5 mg calcite was obtained per sample. When possible, two samples were prepared from each specimen to account for uncertainties related to growth and seasonally changing environmental conditions during shell accretion. In addition, material was saved for further SEM analysis to check for visible signs of alterations in the ultrastructure, which could have been caused by secondary diagenetic overprint. For the strontium analysis on average 2 mg was obtained from the secondary shell layer.

Geochemical analyses at the University of Copenhagen were carried out according to method described by Ullmann et al. (2013). Carbon and oxygen isotope samples were crushed and the powders were placed in glass vials, and subsequently reacted with c. 0.05 ml of >100% H$_3$PO$_4$. The samples were then equilibrated for >100 min at 70.0°C and the resultant carbon dioxide was analysed for $\delta^{13}C$ and $\delta^{18}O$ by use of an IsoPrime gas source isotope ratio mass spectrometer using the continuous flow technique (Spötl & Vennemann 2003). Correction for weight-dependent effects on the raw data was conducted by measuring a set of samples of the Copenhagen laboratory reference material (Carrara Marble: ‘LEO’), covering the weight range of the samples. The reproducibility (2 SD) of the analyses was 0.08‰ for C and 0.18‰ of O isotope ratios.

C and O isotope samples were also analysed for trace elements (Sr/Ca and Mn/Ca ratios) to assess whether any secondary diagenetic overprint had affected the samples. The trace elemental ratios were quantified by inductively coupled plasma optical emission spectrometry (ICP-OES) using a Perkin Elmer Optima 7000 DV system on the reacted carbonate aliquots, which remained from the H$_3$PO$_4$ treatment (Coleman et al. 1989). Samples were diluted with 2% HNO$_3$ to a nominal Ca concentration of 25 µg g$^{-1}$ and analyses were conducted against a set of three matrix matched synthetic
(2 RSD) (Ullmann et al. 2013). Measured average Mn/Ca and Sr/Ca ratios agree within 0.5% with values published by Imai et al. (1996), but Mg/Ca ratios are measured 8% lower than ratios computed from Imai et al. (1996).

$^{87}$Sr/$^{86}$Sr ratios were obtained by thermal ionization mass spectrometry (TIMS) using a Sector VG 54 instrument. Material was dissolved in 0.5M HCl, dried on a hotplate at 80°C, then redissolved in 0.2ml of 3M nitric acid. The strontium was then purified using Sr-Spec resin (Horwitz et al. 1992), eluted with de-ionized water, and after additions of 25µl of 0.1M HPO$_4$ was dried on a hotplate at 80°C. The material was then loaded on single rhenium filaments in 5µl of Ta$_2$O$_5$–H$_3$PO$_4$–HF matrix. $^{87}$Sr/$^{86}$Sr ratios were then measured on the TIMS system with Faraday cups in dynamic multi-collection mode at 1250–1400°C. Accuracy and reproducibility were checked with NISTSRM-987, which gave an average result of 0.710243±0.000022 (2 SD, n=15). The Sr isotope values obtained from the brachiopod shells were consistently higher than expected and are probably diagenetically overprinted.

**Geological setting and stratigraphical framework**

The Boda Limestone Formation of Dalarna, central Sweden, is part of the Palaeozoic succession that is preserved within the Siljan Ring impact structure (Fig. 1). The crater forms a ring-like depression with a present-day diameter of c. 52 km (Juhlin et al. 2012), and is dated as late Devonian (Frasnian, c. 380 Ma; Jourdan et al. 2012). During the Ordovician this region constituted the western, ocean-facing, part of an extensive epeiric sea (Jaanusson 1976).

The Boda Limestone Formation is exposed within the ring-graben, where it forms lenses of carbonate buildups and associated sediments (Ebbestad & Högström 2007; Fig. 1). The buildups reach dimensions of up to 140 m in thickness and 1000 m in diameter (Jaanusson 1982) and consist of massive, pure limestones. The general absence of frame builders and the ubiquity of large stromatolites structures in the micrite core facies make the buildups textbook examples of carbonate mud mounds (Riding 2002). Within the mounds deep horizontal to vertical syndepositional features are common. These are filled mainly by sediments of late Katian–Hirnantian age, but also contain mid-Silurian fillings (Suzuki & Bergström 1999; Suzuki 2002; Ebbestad & Högström 2007).

The age of the Boda mud mound facies is Katian whereas a terminal carbonatic facies development has been shown to be Hirnantian (Schmitz & Bergström 2007; Rasmussen et al. 2010; Ebbestad et al. 2015). These are the Boda Core Member and the Upper Boda Member respectively (Suzuki et al. 2009; Fig. 2). The strata of the Upper Boda Member record the Hirnantian Isotopic Carbon Excursion (HICE; Schmitz & Bergström 2007; Ebbestad et al. 2015). The onset of the HICE is globally recognized as nearly time equivalent to the lower boundary of the Hirnantian Stage (see, e.g. Melchin et al. 2013).

Ghienne et al. (2014) demonstrated that the HICE by high probability in many sections combines several δ13C excursions, which are locally obscured by stratigraphic gaps. This probably is also the case in the Siljan district, where the succession is heavily affected by erosional unconformities and hiatuses (see below). Combined chemo- and biostratigraphy allows for relatively precise correlation of these horizons and, although problems remain, here the scheme of Ebbestad et al. (2015) is followed. The Ordovician succession in the Siljan district is terminated by the Glisstjärn Formation, which unconformably overlies the Upper Boda Member. Chemo- and biostratigraphy indicate a post-HICE Hirnantian age of the Glisstjärn Formation (Bergström et al. 2014).
Table 1. δ¹³C samples from brachiopod shells within the Boda Limestone and the overlying Glisstjärn Formation

| Sample | Taxon             | Locality           | δ¹⁸O (‰ PDB) | δ¹³C (‰ PDB) | Stratigraphy         |
|--------|------------------|--------------------|--------------|--------------|----------------------|
| 221    | *B. umbosulcata* | Solberga, south wall | −4.70        | 1.03         | Post-HICE, Hirnantian |
| 222    | *B. kjerulfi*    | Osmundsberget 1    | −3.70        | 4.01         | UB Mbr, Hirnantian   |
| 223a   | Clorilamnulella  | Osmundsberget 1    | −3.23        | 4.64         | Glisstjärn Fn, Hirnantian |
| 223b   | Clorilamnulella  | Osmundsberget 1    | −5.17        | 2.73         | Glisstjärn Fn, Hirnantian |
| 224a   | *E. rombica*     | Osmundsberget 1    | −4.17        | 1.59         | BC Mbr, flank, Katian |
| 224b   | *E. rombica*     | Osmundsberget 1    | −4.60        | 1.40         | BC Mbr, flank, Katian |
| 225a   | *B. kjerulfi*    | Osmundsberget 1    | −5.49        | 3.29         | UB Mbr, Hirnantian   |
| 225b   | *B. kjerulfi*    | Osmundsberget 1    | −4.98        | 3.10         | UB Mbr, Hirnantian   |
| 226a   | *H. terebratulina* | Osmundsberget 4 | −4.67        | 5.31         | UB Mbr, Hirnantian   |
| 226b   | *H. terebratulina* | Osmundsberget 4 | −2.60        | 5.52         | UB Mbr, Hirnantian   |
| 227a   | *C. aff. psittacina* | Osmundsberget 4 | −3.49        | 1.42         | Glisstjärn Fn, Hirnantian |
| 227b   | *C. aff. psittacina* | Osmundsberget 4 | −3.27        | 1.49         | Glisstjärn Fn, Hirnantian |

UB Mbr, Upper Boda Member; BC Mbr, Boda Core Member; B., Brevilamnulella; C., Cliftonia; E., Eoplectodonta; H., Hindella.

Fig. 4. Diagram comparing δ¹³C and δ¹⁸O data from brachiopod shells (see Table 1) with δ¹³C bulk-rock data from Ebbestad et al. (2015) and δ¹⁸O brachiopod shell data from Marshall & Middleton (1990). The Marshall & Middleton trend is shown by the narrowly dotted line; the Ebbestad et al. trend is depicted by the continuous line. Circles with symbols show the stratigraphical position of the brachiopod samples analysed for this study tied to the δ¹³C bulk-rock curve from Osmundsberget (see legend for details). Error bars show the stratigraphical thickness of the Hindella and *B. kjerulfi* coquinas. It should be noted that the current dataset extends into the persculptus Zone, and that brachiopod shell data roughly reproduce bulk-rock data. G.F., Glisstjärn Formation; U. Boda Mbr., Upper Boda Member; M. extraord., *Metabolograptus extraordinarius*; M. persc., *Metabolograptus persculptus*; B., Brevilamnulella; C., Cliftonia; E., Eoplectodonta; H., Hindella.

Fig. 5. Morphology of Hirnantian speleothems and karstic voids. (a) Stalactite- and stalagmite-like speleothems. (b) Flowstone covered by greenish, calcareous siltstone. (a) and (b) are in syndepositional cracks within the Boda Core Member at Solberga quarry. (c) Horizontal dissolution void within Hindella beds of the Upper Boda Member, covered with heavy botryoidal cements and greenish, calcareous siltstone. Osmundsberget quarry.
Stratigraphic correlation of the Upper Boda Member

Herein we distinguish two successions of coquinas within the Upper Boda Member: a lower succession dominated by Hindella terebratulina and an upper succession dominated by Brevillamnula kjerulfi (Fig. 3). Each succession is topped by an erosional unconformity with evidence of subaerial exposure. The lower succession roughly corresponds to Unit B of Suzuki et al. (2009) whereas the upper corresponds to the overlying units C and D. The coquinas can also be related to δ^{13}C and δ^{18}O, chemostratigraphy and to the expression of the HICE. At the localities Osmundberget 4 and 5 of Ebbestad & Högström (2007) the Hindella coquinas represent the peak of the HICE and the B. kjerulfi coquinas the falling limb of the HICE, both based on bulk and brachiopod shell values (Ebbestad et al. 2015; Table 1, Fig. 4).

Hindella terebratulina from the Boda Limestone has previously been shown to record extraordinarius interval δ^{13}C and δ^{18}O values (Marshall & Middleton 1990; Brenchley et al. 1994). Rasmussen et al. (2010) interpreted the age of the B. kjerulfi coquinas of the Boda Limestone, reported from the locality Osmundberget 1 of Ebbestad & Högström (2007), to be latest Katian. This interpretation is modified herein. Newly collected material places B. kjerulfi from within an interval of up to c. 1 m below the top of the Upper Boda Member at Osmundberget 5 (corresponding to the rhychnonellid coquina of Unit C of Suzuki et al. 2009) and from the uppermost 0.3 m of that member at Kallholn (Unit D of Suzuki et al. 2009). This strongly suggests that the range of B. kjerulfi is restricted to the youngest interval of the Upper Boda Member at Osmundberget 5 and at Kallholn (upper Unit C and Unit D of Suzuki et al. 2009). B. kjerulfi was described by Cocks (1982) from the upper Katian Langåra Formation in the Oslo region, Norway. Brenchley & Cocks (1982), in their thorough investigation of the depositional environment and stratigraphy of the Oslo region, instead stated that the range of B. kjerulfi could be confined to the overlying Langøyene Formation, which is Hirnantian in age. This has led to some confusion as to what the actual range of this species is, but L. R. M. Cocks (pers. comm. 2013) has confirmed that B. kjerulfi is solely known from the Hirnantian Langøyene Formation in the Oslo region. Low δ^{13}C isotope values were recorded by Ebbestad et al. (2015) from whole-rock samples in the B. kjerulfi levels at Osmundberget 1 and 5, and new δ^{13}C values obtained from the shells of B. kjerulfi from Osmundberget 1 and from bulk-rock samples of Osmundberget 5, presented herein (Table 1, Fig. 4), reproduce these and confirm a position of the B. kjerulfi coquinas on the falling limb of the HICE.

Evidence for subaerial exposure in the Boda Limestone

The Upper Boda Member locally is strongly affected by fissures. Secondary dissolution and subsequent cementation of the sediment interspaces around these fissures, and seemingly independent from them, is widespread and locally variable. Dissolution and subsequent void filling took place at multiple scales. Dissolution voids range from decimetre to millimetre size (Figs 5c, 6b and 7c). The macroscopic shape of the system of voids and their fillings is similar to features known from other stratigraphical levels in the Siljan district (Calner et al. 2010a). Fissures, and to a lesser extent voids, also occur throughout the underlying Boda Core Member, but no such structures are known from the overlying Glisstjärn Formation, or from Silurian strata.

In addition to these secondary effects of dissolution and recrystalization, primary gravitational, bladed cements indicate an early diagenesis under the influence of vadose and phreatic meteoric waters. These structures have been detected in two distinctive horizons within the Upper Boda Member: at the top of the beds with H. terebratulina, and at the top of those with B. kjerulfi (Figs 6a and 7c). Both horizons are truncated by major unconformities.
In many mounds stromatolite-like crusts occur on the rugged surfaces of the fissures (Fig. 5a and b). In the Jutjärn quarry these linings can be found in fissure depths down to 30 m below the top of the mounds. The crusts are 10–100 mm thick and form areas with a texture that is characteristic of flowstones. In some places dripstones have formed massive stalagmites and stalactites (Fig. 5b). The crusts consist of a fine-grained layered peloidal grainstone with peloids in the silt size range (30–70 µm). The c. 100–300 µm thick layers reflect differences in packing density and peloid size, and in density of microsparitic interlayers (Fig. 7a and b). Often, widely spaced fenestrae occur within the grainstone, reaching sizes in some places more than 100 µm in diameter. Locally the layers are less regular and form clusters of clotted peloidal–microsparitic areas. Energy-dispersive X-ray spectroscopy (EDS) indicates low Mg-calcite for both peloids and microspar.

**Age of the speleothems**

Three lines of evidence support the hypothesis of a mid-Hirnantian age for the formation of speleothems, as follows.

1. The δ13C isotope values of whole-rock samples of the stromatolite-like crusts range between 4.4 and 6.3‰ (Table 2). This record is similar to peak HICE values measured elsewhere in the Boda Limestone Formation (Schmitz & Bergström 2007; Ebbestad et al. 2015; see above).

2. Often the speleothem crusts are covered by a platy, greenish calcareous siltstone (Fig. 3b). Lithological comparison led to correlations with a silty–sandy inter-reef limestone yielding a Hirnantian fauna found elsewhere in the Siljan district (Thorslund 1935; Jux 1966). The siltstone cover is devoid of macrofossils, but δ13C bulk-rock values between 2.6 and 4.4‰ make a late HICE Hirnantian interval likely for its deposition (Table 2).

3. At the southern wall of Solberga quarry, the speleothems on fissures in the Boda Core Member are directly covered by a coquina, containing masses of *B. umbosulcata*. The age of this brachiopod species has been interpreted as latest Katian by Rasmussen et al. (2010). However, new collections and δ13C and δ18O isotope values of the shells show the range of the species to be constrained within the post-HICE interval (see above).

The age of the speleothems at Solberga thus can be constrained to be time equivalent to the Upper Boda Member of the Boda Limestone Formation, representing two major unconformities, situated early and late within the HICE interval (Fig. 8).

**Discussion**

The Boda speleothems formed in subaerial caves. This is clearly indicated by the presence of stalactites and stalagmites, which are here documented for the first time. To date these are the only known Ordovician subaerial speleothems. However, they are not regular dripstones as they do not consist of layered sparitic calcite crystals, like most of modern cave sinters. Instead, they consist of stromatolite-like, layered–clotted fine peloidal grainstone, which is known from the fossil record as spongiostrome (Riding 2011). Experimental results of freshwater calcite precipitation suggest that spongiostromes are produced as early diagenetic products of decaying microbial mats (Pedley et al. 2009). Microbially mediated stromatolites are not confined to cyanobacteria and daylight (Jones 2001; Melim 2009).

Indirect evidence for a vadose precipitation and microbial origin of the speleothems comes also from the isotopic signature. The Sr isotope values of the Hirnantian brachiopod shells of the Upper Boda Member are consistently higher than expected under marine equilibrium conditions, which is probably a result of an early diagenetic overprint and the influence of meteoric water (compare Brand 1991). In contrast, no significant depletion in 13C can be detected in the speleothems and brachiopod shells. Modern meteoric carbonates are often strongly depleted relative to 13C because...
Table 2. Whole-rock $\delta^{13}$C data from syndepositional fissure fillings within the Boda Limestone

| Sample | Lithology                        | Locality                          | $\delta^{13}$C (‰ PDB) | Stratigraphy |
|--------|----------------------------------|-----------------------------------|-------------------------|--------------|
| J 1    | Greenish calcareous siltstone    | Jutjärn quarry, fissure filling    | 2.79                    | K2, Hirnantian |
| OS 4   | Greenish calcareous siltstone    | Osmundsberget quarry, void filling | 2.55                    | K2, Hirnantian |
| OS 5   | Greenish calcareous siltstone    | Osmundsberget quarry, void filling | 3.07                    | K2, Hirnantian |
| OS 16  | Greenish calcareous siltstone    | Osmundsberget quarry, void filling | 2.7                     | K2, Hirnantian |
| OB 19  | Greenish calcareous siltstone    | Östbjörka quarry, fissure filling  | 3.21                    | K2, Hirnantian |
| SO2    | Greenish calcareous siltstone    | Solberga quarry, south wall fissure filling | 4.35 | K1, Hirnantian |
| S1A    | Brachiopod coquina with *B. umbosulcata*, cover of S1B | Solberga quarry, south wall fissure margin | 2.16 | K2, Hirnantian |
| S2A    | Laminated, peloidal grainstone, stromatolite-like speleothem | Solberga quarry, east wall fissure margin | 2.39 | latest Katian |
| S2B    | Laminated, peloidal grainstone, stromatolite-like speleothem, crust on S2A | Solberga quarry, south wall fissure crust | 6.28 | K1, Hirnantian |

K1 and K2 refer to the two distinctive Hirnantian karst intervals (see Fig. 3).

Fig. 8. Scheme of the timing of the Hirnantian subaerial exposure and karst development of the Boda Limestone Formation. *D. Dicellograptus*; *M. Metabolograptus*; HICE, Hirnantian Carbon Isotope Excursion; K1 and K2 refer to two distinctive Hirnantian karst intervals (compare Fig. 3). Stage slices after Bergström et al. (2009), Baltic isotope zones after Ainsaar et al. 2010; glaciation peaks after Holmden et al. (2013). of the strongly negative carbon isotopic composition of soil gas (Lohmann 1988). Speleothems in modern caves are formed as a result of CO$$_2$$ uptake of meteoric waters and related calcite dissolution in the soil zone, and subsequent precipitation at greater depths (e.g. Brasier 2011). A speleothem that is formed from meteoric waters without enhanced calcite dissolution in a soil zone would not be depleted relative to $^{13}$C and it would need an alternative process of precipitation such as microbial decay. In this context it is interesting to note that the $^{13}$C depletion in Ordovician meteoric carbon often is only minor, and that this fact was suggested to be a consequence of the lack of modern soils formed by vascular plants in the Ordovician world (Tobin et al. 1999, and references therein). Brasier (2010) assumed major differences in terrestrial calcite precipitation mechanisms during the earlier Palaeozoic, a time before the presence of an extensive plant cover. The Boda speleothems are among the oldest non-ambiguous speleothems and the fact that these are microbial mediated and stromatolite-like probably reflects these fundamental environmental differences.

The Boda speleothems are of Hirnantian age. The Hirnantian mound capping carbonates of the Upper Boda Member contain two distinctive horizons of subaerial exposure within and terminating the HICE. Most probably these horizons represent the interval of maximum sea-level drop and speleothem formation. The timing of these two horizons within the *M. extraordinarius* and *M. peresculptus* graptolite biozones is in accordance with the two inferred maximum ice sheet advances of the Hirnantian (Holmden et al. 2013; Melchin et al. 2013; roughly equivalent to LOGC 2 and 3 of Ghienne et al. 2014) (Fig. 9).

Given a maximum depth of the speleothem occurrences of c. 30 m below the mound top at Jutjärn and a presumed depositional depth of the mounds of between 50 and 100 m during the latest Katian (Riding 2002; Kröger & Ebbestad 2013), the sea-level fall was rapid and in the range of 80–130 m. A Hirnantian sea-level drop of >50 m and >80 m was inferred from sedimentary features in palaeotropical settings of eastern Canada (Desrochers et al. 2010) and from palaeokarst in the Welsh Basin (Brenchley et al. 2010), respectively. The Boda speleothems support these earlier estimations. Future analyses will have to identify more exposure surfaces related to the Late Ordovician glaciations and compare their exact timing with the pattern in glaciogenic successions of Gondwana.

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