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# Nd isotope composition and rare earth element distribution in early Paleozoic biogenic apatite from Baltoscandia: A signature of Iapetus ocean water

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## ABSTRACT

**Analyses of the Nd isotopic composition and REE distribution in biogenic apatite (organophosphatic brachiopods and conodont elements) from the Cambrian and Ordovician sequences of the Baltic plate give new insights into the development of the southeastern segment of the continental margin bounding the Iapetus ocean. The Nd isotope analyses show  $\epsilon_{Nd(t)}$  of  $\sim -8.0$  for the Cambrian, indicating that the main source of the sedimentary deposition came from weathered sedimentary rocks of Vendian and Cambrian age. The increase of  $\epsilon_{Nd(t)}$  to  $\sim -5.0$  for the Early Ordovician indicates the appearance of a new source of radiogenic Nd in the surrounding area—most likely a volcanic arc along the western borderland of Baltic plate from Arenigian time. Samples of Cambrian biogenic apatite show significantly lower total amounts of REE than do the Ordovician samples, and this is probably due to a shorter exposure to seawater before burial during Ordovician sedimentary accumulation in Baltoscandia. These preliminary results suggest that biogenic apatite from the Baltoscandian basin preserves geochemical signatures of the water masses that will be important for understanding the evolution of the Iapetus ocean during the early Paleozoic.**

## INTRODUCTION

Biogenic apatite is known to concentrate rare earth elements (REE) at the sediment-water interface and thus represents the chemical conditions at the sea floor. The enrichment occurs post mortem and so is not influenced by metabolism, which means that material from various fossil species can be used for comparison. As a result, old biogenic apatite (e.g., fish debris, teeth, thelodont and bones, conodont elements, phosphatic shells of brachiopods) is the carrier of readable geochemical signals concerning redox conditions (Wright et al., 1987), sedimentary environment (Laenen et al., 1997), and the Sr and Nd isotopic composition of contemporaneous seawater (Bertram et al., 1992; Bertram and Elderfield, 1993).

The neodymium isotopic composition of old biogenic apatite is commonly used for provenance analysis and reflects the proportion of radiogenic neodymium in the surrounding source area. Data are usually expressed as  $\epsilon_{Nd}$  representing deviations of  $^{143}\text{Nd}/^{144}\text{Nd}$ , in parts per 10000, from the chondritic uniform reservoir (CHUR). In small basins, like the Baltoscandian epicontinental sea, for which there are only a few possible source areas, the Nd isotopic characteristics should be a good indicator of REE provenance. Late diagenetic alteration could affect the acquired REE patterns, but a series of publications (see Holser, 1997) support the hypothesis that the REE signature in fossil apatite has an early diagenetic origin.

During the Cambrian and Early Ordovician, Baltica (the East European plate) was located at high or temperate southern latitudes and formed the southeastern margin of the Iapetus ocean (Mac Niocaill et al., 1997). From the beginning of the Cambrian, the plate was mainly covered by an epeiric sea (Baltoscandian and Moscow basins) that persisted until near the end of the Silurian. During this time, condensed sequences of mature siliciclastic sediments and carbonates were formed, containing perfectly preserved biogenic phosphates. The current study of the REE content and Nd isotope compositions in the Late Cambrian to Early Ordovician organophosphatic brachiopods and conodont elements was undertaken to investigate variations in depositional environments in Cambrian–Early Ordovician Iapetus seawater, as reflected in the preserved geochemical signature in biogenic apatite from Baltoscandia.

Material selected for our study includes samples of Late Cambrian–Early Ordovician organophosphatic brachiopods and Early Ordovician conodont elements. Lingulids of the genera *Ungula*, *Obolus*, and *Schmidtitites*, as well as the siphonotretide *Helmerseniania ladogensis*, represent Late Cambrian brachiopods. Early and Middle Ordovician brachiopods include lingulid genera *Thysanotus*, *Aulonotreta*, and *Lingulasma* as well as a number of taxa of microbrachiopods, mostly of acrotretides and siphonotretides. Conodont samples constitute a mixture of various taxa of euconodonts.

Upper Cambrian and some Ordovician samples used in the analysis were separated from the clastic host rock by deionized water, and for the limestone rocks weak acetic acid was used (for details see Methods in Data Repository).<sup>1</sup> All the conodonts are light brown or yellowish and have conodont alteration indices (CAI) within the range of 1–1.5, indicating minor thermal alteration. Phosphatic brachiopods from the samples also show well-preserved original structures (Holmer, 1989). Each sample was carefully examined under the microscope, and no sample with impurities of pyrite, clay, etc., was used. Scanning electron microscope with energy dispersive spectrometer (EDS), instrumental neutron activation analysis (INAA), and X-ray diffraction analysis were used to reveal any degree of calcification or silicification. However, most of the Late Cambrian and Early Ordovician lingulid shells contain internal secondary apatite deposited as a result of post mortem recrystallization and bacterial degradation of the organics within the shells (Nemliher and Puura, 1997).

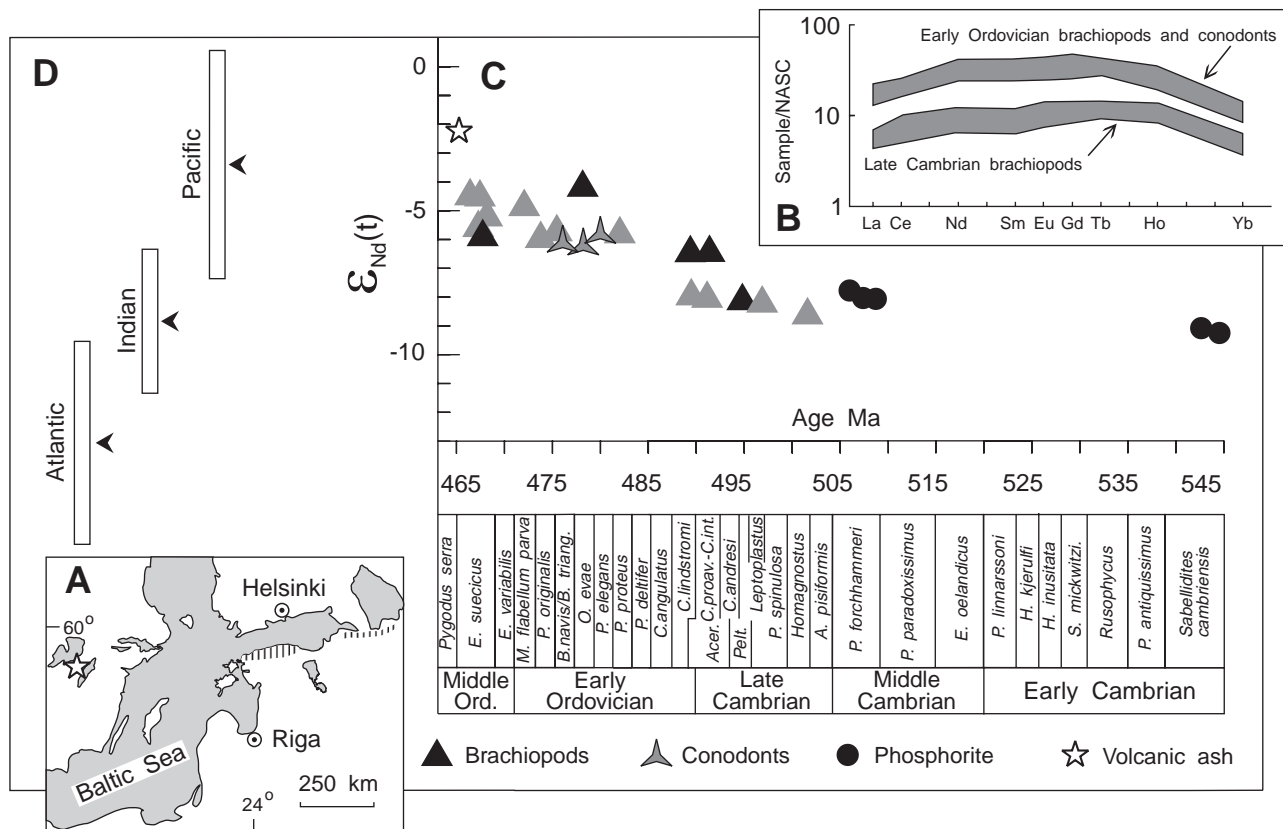
The REE concentration in selected samples was determined with isotope dilution–mass spectrometry (ID-MS) and instrumental neutron activation analysis (INAA).

## REE PATTERNS AND Nd ISOTOPIC SIGNATURE IN ORGANOPHOSPHATIC BRACHIOPODS AND CONODONTS

Shale-normalized REE patterns for early Paleozoic biogenic apatite are shown in Figure 1B. The shape of the normalized patterns is rather uniform for the Late Cambrian and Early Ordovician organophosphatic brachiopod shells and conodont elements: slight enrichment of middle REE and similar values of La/Sm and Sm/Yb for both Late Cambrian and Early Ordovician phosphatic fossils. There are two distinct groups of REE concentrations. All the analyzed Late Cambrian brachiopods display a REE enrichment

<sup>1</sup>GSA Data Repository item 9895 (methods used, Sm–Nd isotope data, and description of analyzed material [Table A]; rare earth element data [Table B]), is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301. E-mail: editing@geosociety.org.

Data Repository item 9895 contains additional material related to this article.



**Figure 1.** A: Schematic map of Baltoscandia. Striped areas show sampling sites for biogenic phosphate in North Estonia (west) and St. Petersburg–Ladoga Lake region (east). Star shows sampling site for volcanic ash in Sweden. Middle Cambrian phosphorite samples were taken from North Greenland, ~300 km northwest, and Early Cambrian and Vendian phosphorite and shale samples were taken in the Moscow syncline, ~600 km southeast of mapped area. B: Rare earth element (REE) patterns of Late Cambrian–Early Ordovician organophosphatic brachiopods and conodont elements from East Baltic, North American shale composite (NASC)–normalized, mean value  $\pm$  one standard deviation. Samples from Estonia and Sweden are filled-in gray, from St. Petersburg–Ladoga Lake region are filled-in black. C: Variation of  $\epsilon_{Nd}(t)$  values in Cambrian–Early Ordovician biogenic apatite.  $\epsilon_{Nd}(t)$  scale is common to C and D. Our project is focused on faunistic changes and their causes, from the Cambrian–Ordovician transition to Middle Ordovician, which explains concentration of samples in this time span. We have so far no data for Early–Middle Cambrian part of scale. D: Nd isotopic composition of modern-day oceans as  $\epsilon_{Nd(0)}$ , (95% confidence level; mean values are indicated with arrows), modified from Bertram and Elderfield (1993). Conodont and trilobite zones of Baltoscandia are after Mens et al. (1984) and Dronov et al. (1996); age scale used for calculation of  $\epsilon_{Nd}(t)$  is after Shergold (1995). Data for B and C are in the GSA Data Repository (see footnote 1).

within a factor of 5 to 10 of the standard shale values (North American shale composite, NASC); for Early Ordovician fossils this factor ranges from 8 to 50. Taking Nd concentrations as a proxy of bulk REE contents gives Nd = 251  $\pm$  49 ppm ( $n$  = 16) for the Late Cambrian and Nd = 883  $\pm$  358 ppm ( $n$  = 18) for the Early Ordovician. The samples are taken from a large area (Fig. 1A), so the higher REE contents in Early Ordovician biogenic apatite are to be regarded as a regional geochemical signature of the Early Ordovician organophosphatic brachiopods and conodonts from the East Baltic area.

Nd isotopic composition ( $\epsilon_{Nd(t)}$  as a proxy for evaluation) in the fossil apatite of Early Ordovician (Fig. 1C) varies from  $-6.2$  to  $-4.4$ ; mean  $\epsilon_{Nd(t)}$  =  $-5.3 \pm 0.7$ ,  $n$  = 12, (mean value  $\pm$  one standard deviation), which is close to the  $-5.4$  value estimated for Early Ordovician Iapetus seawater on the basis of  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios of the metalliferous sediments from Southern Uplands

of Scotland (Hooker et al., 1981). Late Cambrian brachiopods from the East Baltic area show significantly lower values of  $\epsilon_{Nd(t)}$  =  $-7.9 \pm 0.9$  ( $n$  = 7) and a pronounced increase near the Cambrian–Ordovician boundary. Early Cambrian phosphorites from the central part of the East European platform (Gavrilov–Yam borehole, ~1000 km southeast of the Baltic Sea) have  $\epsilon_{Nd(t)}$  ranging from  $-8.6$  to  $-9.0$ , whereas Middle Cambrian phosphorites from the Holm Dal Formation of North Greenland have  $\epsilon_{Nd(t)}$  of  $-8.0$  (three samples).

Notwithstanding the commonly observed wide scatter of  $\epsilon_{Nd(t)}$  values in biogenic phosphate and fossil apatite (Holser, 1997), the difference in Nd isotopic signatures between the Late Cambrian and Early Ordovician of the East Baltic area is statistically significant and the  $\epsilon_{Nd(t)}$  values vary within remarkably narrow limits.

Similar Nd isotope compositions between late Middle Cambrian (*Lejopyge laevigata* zone) sedi-

mentary phosphorites from North Greenland and early Late Cambrian (*Agnostus pisiformis*–*Homagnostus* zones) obolids from the East Baltic area, which are only slightly younger and located on the opposite side of Iapetus ocean, suggest that relatively low  $\epsilon_{Nd}$  values within the range  $-8.0$  to  $-9.0$  represent the general characteristics of water masses of the Iapetus ocean at the beginning of the Late Cambrian.

#### DEPOSITIONAL ENVIRONMENTS

The Upper Cambrian and lowermost Ordovician deposits in the East Baltic area are represented by condensed, unconsolidated quartzose sands and silts, which form several lithostratigraphic units separated by discontinuity surfaces of highly varying morphology, that locally have traces of subaerial exposure. There are frequent occurrences of bidirectional and multidirectional cross-bedding, wave ripple marks, *Skolithos* trace fossils, and beds of obolid coquina; these charac-

teristics suggest that the depositional environment was above storm wave base for most of the period, and mostly within the peritidal zone of the shallow shelf (Popov et al., 1989). The mature character of the sands, the condensed character of the sequences in the East Baltic area, and the absence of evidence of any significant erosion of the Precambrian basement in the adjacent areas all suggest that weathering of Vendian and Lower Cambrian siliciclastic sedimentary rocks was the major sediment source.

Studies of the taphonomy of Holocene lingulids demonstrate that under nearshore conditions, the chance of preservation is very low, and the rapid burial of lingulid shells during storm events is an important factor in their fossilization (Emig, 1997). It is also probable that the layers and lenses of coquina, which are the main supply of the specimens of the Late Cambrian organophosphatic brachiopods in this study, were formed mainly during storms within a beach and bar system, which was sometimes subaerially exposed.

During the Early Ordovician (*Paltodus deltifera*–*Paroistodus proteus* zones) an accumulation of glauconitic sands and clays began, which was later replaced by carbonate sedimentation. The net rate of deposition of carbonates in Baltoscandia was very low and did not exceed 1–5 mm/1000 yr through the Ordovician (Jaanusson, 1972), which is comparable to the average rate of deposition of radiolarian ooze in recent oceans. On the basis of conodont studies, Schmitz et al. (1996) estimated the net sedimentation rate to  $2 \pm 1$  mm/k.y. during Lower–Middle Ordovician in Sweden. The siliciclastic influx into the epicontinental sea was low and very fine grained. Numerous discontinuity surfaces that have traces of submarine erosion are also characteristic. There is no evidence, however, of subaerial exposure and erosion of carbonate sediments (Lindström, 1979). These deposits were formed within a shallow shelf environment under stable marine conditions, mostly below the seasonal storm wave base.

The biogenic apatite of living organisms (thelodonts, fish teeth, and bones) is not enriched in REE. Typical values do not exceed 10–20 ppm of bulk REE content (Wright et al., 1987) owing to very low REE concentrations in surface, bottom, and pore waters ( $10^{-7}$ – $10^{-9}$  of standard shale values; Möller et al., 1994). An exposure time providing water/fossil mass ratios to  $10^8$  leads to the observed REE enrichment in old biogenic apatite of as much as thousands of parts per million. The sedimentation rate and taphonomic conditions are the most important factors controlling the incorporation of REE into brachiopod shells and conodont elements at the sediment-water interface. As a result, recent and old biogenic apatite from pelagic deep-water sediments have REE concentrations of one order of magnitude and higher in comparison to apatite from

shallower water sediments on continental shelves and epicontinental basins (Elderfield and Pagett, 1986; Wright et al., 1987).

The enhanced REE contents in Early Ordovician versus Late Cambrian organophosphatic brachiopods and conodont elements are consistent with different paleogeographical and taphonomical conditions: Late Cambrian brachiopods were preserved by rapid burial, whereas most of the Early and Middle Ordovician brachiopods were exposed considerably longer to seawater.

Disarticulated valves and shell fragments of the organophosphatic brachiopods, mostly obolids, varying in size from 2 to 20 mm, represent the single source of bioclasts in the Upper Cambrian deposits, whereas the biogenic phosphates in the Ordovician carbonates are mostly represented by 0.5–2.5 mm microbrachiopods and conodonts. The content of biogenic phosphates in the host rock for the Lower and Middle Ordovician is 10–100 times lower than in the Upper Cambrian. As the REE concentration in biogenic apatite can reach up to  $10^{12}$  of normal marine water concentration, phosphatic bioclasts must be considered an important depository of REE removed from the water during fossilization, particularly in the shallow shelf environments typical for the East Baltic area during the early Paleozoic. The effect of a simple geochemical dilution may also be responsible for the observed enrichment in REE in the Early and Middle Ordovician biogenic apatite. Assuming a fairly stable input of REE in the basin, confirmed by a stable tectonic source area (peneplain), a smaller amount of REE-scavenging biogenic apatite on the seafloor should concentrate more REE per weight unit.

#### **Nd ISOTOPE COMPOSITION OF THE BALTOSCANDIAN SEGMENT OF IAPETUS OCEAN IN THE EARLY PALEOZOIC**

The paleogeography and mode of sedimentary accumulation of the East Baltic area in the Late Cambrian–Early Ordovician are important controls on  $\epsilon_{Nd}$  variation in biogenic phosphate. The  $\epsilon_{Nd}$  in recent seawater is controlled by the proportion of radiogenic Nd in the source terrane surrounding the basin (Goldstein and Jacobsen, 1987). The short residence time of Nd in a marine basin (only a few hundred years) in relation to the  $\sim 10^3$  yr of mechanical turnover of the deep ocean results in the wide regional and vertical scatter of  $\epsilon_{Nd}$  values in recent and ancient oceans (Holser, 1997). Nevertheless, the different Nd isotopic compositions of modern-day oceanic water masses are well documented and  $\epsilon_{Nd(0)}$  of seawater varies from  $-15.1 \pm 3.4$  (mean value  $\pm$  one standard deviation) in the northern Atlantic Ocean, through  $-8.3 \pm 1.2$  for the Indian Ocean, to  $-3.5 \pm 1.7$  in the Pacific, reflecting the prevalent composition of surrounding source areas and large-scale circulation patterns (Bertram

and Elderfield, 1993). Circum-Pacific volcanic arcs provide more radiogenic Nd in the ocean in comparison to the relatively old oceanic crust and crystalline shields bordering the northern Atlantic Ocean.

Only siliciclastic sedimentation took place within the northwestern part of the East European platform during the Vendian. Fine-grained shale has Nd isotope compositions from  $^{143}\text{Nd}/^{144}\text{Nd}_{\text{measured}} = 0.511505 \pm 0.000013$  ( $\epsilon_{Nd(t)} = -15.7$ ) in the basal Redkino Stage through  $0.511520 \pm 0.000009$  ( $\epsilon_{Nd(t)} = -10.5$ ) in the Kotlin Stage to  $0.511890 \pm 0.000006$  ( $\epsilon_{Nd(t)} = -9.1$ ) in the basal Cambrian Rovno Stage. The phosphorite pebbles collected from the Lower Cambrian Lezha Formation (Gavrilov-Yam borehole), central part of the East European platform show  $\epsilon_{Nd(t)}$  from  $-8.6$  to  $-9.0$ , probably illustrating the evolution of the Nd isotopic composition of the epicontinental marine basin in the Early Cambrian. There is no evidence of volcanic activity in the proximity of Baltoscandia in the Late Cambrian, and  $\epsilon_{Nd(t)}$  in the shells of organophosphatic brachiopods clearly indicates the predominance of rocks with low proportions of radiogenic Nd as a source area for respective parts of the Iapetus ocean.

A substantially different situation was developed in the Early and Middle Ordovician in Baltoscandia. Beds of tephra are reported from the Billingen (lower Arenig) deposits of south-central Sweden (Lindström, 1979). High aluminum basalt-andesite-dacite volcanic material and tuff are characteristic of the Arenig–early Llanvirn of the Smöla, Trondheim area, of Norway (Roberts, 1982). Numerous K-bentonite layers are characteristic of the Ordovician (Caradoc–Ashgill) and Silurian deposits of Baltoscandia (Snäll, 1977; Huff et al., 1992). The trace element geochemistry of these volcanic strata indicates that the ash is derived from a continental-crust–based, mature island arc on a destructive Andean-type plate margin setting (Roberts, 1982; Huff et al., 1992).

In the Arenig, a subduction zone was located several hundred kilometers from the epicontinental basin of Baltoscandia (Prigmore et al., 1997). An air-fall magnitude of the Ordovician eruptions of 100–1000 km<sup>3</sup> for every discrete event indicates large-scale explosive volcanism close to the margin of the Baltic plate. The Nd signature of the late Llanvirnian Våmb volcanic ash in south Sweden (Stuesson, 1992) ( $^{143}\text{Nd}/^{144}\text{Nd} = 0.512244 \pm 0.000005_{\text{measured}}$  and  $\epsilon_{Nd(t)} = -2.4$ ) shows that juvenile crust was also a source of tephra covering most of Baltoscandia. As a result of Nd input, this part of the Iapetus ocean became increasingly radiogenic from the Late Cambrian to the Early and Middle Ordovician. The organophosphatic brachiopods and conodonts of that age have an  $\epsilon_{Nd(t)}$  signature of about  $-5.0$  reflecting the change in Nd isotope composition of the ancient water masses.

On the basis of the Nd isotope data presented here, paleotectonic reconstructions, and  $\epsilon_{\text{Nd}(t)}$  values of different recent oceans (Fig. 1D), the Pacific Nd isotopic signature seems to be a more suitable model for this part of Iapetus ocean in the Early and Middle Ordovician, not the modern-day Atlantic Ocean (Arctic or circumpolar) as proposed by Hooker et al. (1981). The  $\epsilon_{\text{Nd}(t)}$  values are characteristic signatures of single-reservoir water mass (Holmden et al., 1996), and the variations of  $\epsilon_{\text{Nd}(t)}$  values in fossil apatite from the early Paleozoic of Baltoscandia can be regarded as genuine signatures of the part of Iapetus ocean close to the Baltic paleoplate. This interpretation is supported by the  $\epsilon_{\text{Nd}}$  isotope composition of the Mohawkian sea between the 454 Ma Millbrig and Deicke bentonites in eastern Laurentia, as recorded in conodonts by Holmden et al. (1998). They identified three “aqua facies”: a shallow-water carbonate platform with very negative Nd values ( $\epsilon_{\text{Nd}} \approx -15$ ), a foreland basin ( $\epsilon_{\text{Nd}} \approx -7.5$ ), and unmodified Iapetus ocean water ( $\epsilon_{\text{Nd}}$  from  $-0.6$  to  $-5$ ). In their terms, our data correspond to an “aqua facies” of a foreland basin with a possible influence of deeper Iapetus ocean water.

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