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Paleoenvironmental changes in the Silurian indicated by stable isotopes in brachiopod shells from Gotland, Sweden

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Abstract—Ratios of stable carbon and oxygen isotopes in brachiopod shells (more than 370 specimens, esp. *Atrypa reticularis*) from the Silurian of Gotland, Sweden, have been analysed. Preservation of biological skeletal ultrastructures, observed in SEM-micrographs, and cathodoluminescence analyses indicate that usually no diagenetical alteration occurs.

The Silurian of Gotland consists of 440 m carbonate deposits, spanning the late Llandovery to late Ludlow epochs (431–411 m.y.). Repeatedly, uniform sequences of micritic limestones and marls are interrupted by complex-structured reefs and adjacent platform sediments. Previously, the alternation of facies is interpreted as the result of sea level fluctuations caused by a gradual regression with superimposed minor transgressive pulses. The Silurian sequence of Gotland exhibits principally parallel carbon and oxygen isotope records corresponding closely to the topostratigraphic units. Lower values occur in periods dominated by deposition of marly sequences. Higher values are observed in periods dominated by reefs and extended carbonate platforms.

The isotope ratios are influenced by local as well as global factors. The oxygen isotope ratios are interpreted to reflect paleosalinity changes due to varying freshwater input, rather than changes in paleotemperature. Consequently, the facies distribution of Gotland is interpreted as resulting from changes in terrigenous input caused by different rates of continental weathering and freshwater runoff rather than by sea level fluctuations. Periods of arid climate and, therefore, anti-estuarine downwelling of oxygenated surface water appear as short episodes of reef growing ($\leq$1.5 m.y.) in an epoche characterized by a tropic humid climate, which causes an estuarine circulation and the upwelling of CO_2-rich deep water. Carbon isotope ratios are obviously connected to these changes in circulation by the advection of ^{13}C-rich surface water (arid episodes) or upwelling of ^{13}C-depleted deep water (humid episodes) of a Silurian ocean which itself reveals generally euxinic deep water conditions due to the burial of organic carbon in black shales. *Copyright © 1997 Elsevier Science Ltd*

1. INTRODUCTION

The Silurian Period, with its relatively short duration of about 30 million years, is generally thought to be an epoch of stable environmental conditions and warm climates. Vast platform seas extended across the low relief of many paleocontinents (Scotese and McKerrow, 1990), the distribution of carbonate sediments shows an extraordinary expansion (Boucot et al., 1968; Wilde et al., 1991; Frakes et al., 1992), and continental ice sheets are only reported for early Silurian times in South America (Grahn and Caputto, 1992). Fischer (1983) assigned the Silurian earth to be in a greenhouse mode. In the history of Silurian biota as a whole no bigger catastrophes occurred like the first order bio-event at the Ordovician-Silurian boundary.

Nevertheless, several paleontological studies show that the Silurian was not a quiet period. Kaljo et al. (1995) summarized fifteen remarkable bio-events, among others the most severe extinction of conodonts and acritarchs at the Llandovery-Wenlock (Ireviken Event) or the middle Ludfordian Event comprising many lineages of vertebrates, graptolites, conodonts, and corals. Furthermore, recent studies in stable isotopes, measured in brachiopod shells as well as in carbonate sediments, showed several distinct shifts in both carbon and oxygen isotopes, which mostly coincides with changes in lithology of the surrounding environment (Corfield et al., 1992; Jux and Steuber, 1992; Samtleben et al., 1996; Talent et al., 1993; Wenzel and Joachimski, 1996). These isotope shifts are in the order of the variability observed for the entire Paleozoic, which include data from fossils (Popp et al., 1986; Veizer et al., 1986; Hudson and Anderson, 1989; Railsback, 1990; Wadleigh and Veizer, 1992) and abiotic marine calcites (Lohmann and Walker, 1989; Carpenter et al., 1991). Some of these shifts, especially in $\delta^{13}C$, exceed by far the variability in isotopes observed in Neogene and Quaternary times which are explained by changes in earth climate systems like ice volume, ocean circulation, and surface water productivity.

There is an ongoing debate whether the isotopic changes in Palezoic sediments and invertebrate shells are influenced or even controlled by effects of diagenetic alteration (Qing and Veizer, 1994; Land, 1995; Veizer, 1995). On the other hand, modern brachiopods secrete shell carbonate at or near isotopic equilibrium with the surrounding seawater. The same relation is assumed for ancient species. A detailed study by Carpenter and Lohmann (1995) showed a stronger deviation by vital effects only for the primary layer calcite and specialized portions of the skeleton, whereas the secondary layer calcite approximates equilibrium values. Shells of articulate brachiopods consist of low-Mg calcite and are,

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therefore, rather resistant to diagenetic alteration (Lowenstam, 1961; Popp et al., 1986). If the assumption is correct that they mostly exhibit their primary isotopic composition, then the following questions arise: (1) What processes control the variability in isotopes, especially the rapidness of shifts? (2) What are the causes for the observed extreme isotopic fractionations? (3) Are these processes global effects or changes in regional environmental parameters?

In this paper we present a fairly continous record of stable carbon and oxygen isotopes in brachiopods from the Silurian of Gotland. We try to give plausible explanations for the fractionation processes of both carbon and oxygen isotopes, which are consistent with the interpretation of the facies succession and the paleontological record of Gotland, and discuss their paleoceanographic implications.

2. GEOLOGIC SETTING

The Silurian of Gotland, Sweden, consists of approximately 440 m of limestones and marls deposited in an epicontinental sea near the Silurian equator. Along the edge of this cratonic basin, a wide belt of shallow-water carbonate sediments with sporadic reefs was developed, spanning from Gotland, north and central Estonia to eastern Lithunia (Nestor, 1995). This belt was to the east surrounded with land areas and deepened westwards into the Iapetus Ocean and southwards into the Rheic Ocean (Scotese and McKerrow, 1990; McKerrow et al., 1991). During Ordovician and Silurian times, the Baltic Contintent drifted from about 50° south with a counterclockwise rotation towards the northeastern edge of Laurentia in a low latitudinal position (Torsvik et al., 1992). This plate movement resulted in the closure of the Iapetus Ocean, followed by the Caledonian orogeny at the end of the Silurian. However, deposited on the stable Baltic Shield, the strata of Gotland never underwent tectonic deformation or deep burial (Jeppsson, 1983; Frykman, 1986). For this reason the sediments show rather low grades of diagenetic alteration with an excellent preservation of microfacies character and fossils, particularly those with originally calcitic skeletons.

Due to the gentle SE-dipping of the strata the stratigraphic succession is laterally exposed with the oldest beds located in the NW, the youngest in the SE of the island (Fig. 1). Based on lithological and paleontological observations, Hede (1921, 1960) defined thirteen topostratigraphic units. While uniform sequences of micritic limestones and marls are developed on the western side of Gotland, the eastern part of the island shows extensive reef complexes and shallow marine to lagoonal deposits with frequent discontinuities. The distribution of these facies shifted several times in an east-west direction during the Silurian. In the central part of the island, the sequence of strata shows repeated changes in facies formation: Three times (Högklint Beds, Klinteberg Beds, and Hamra-Sundre Beds) carbonate platforms have been formed by massive bioherms surrounded by shallow shoal areas with biodetritic deposits. These alternate with sequences of micritic limestones and marls (Lower Visby Beds, Slite Beds, and Hemse Beds) which were deposited in shelf basin areas.

The biostratigraphic subdivision of the Silurian sequence on Gotland and its correlation with the standard (British) biostratigraphic classification is relatively certain in spite of the absence of a continuous graptolitic succession. The boundary between Llandovery and Wenlock is situated 1.5–5 m (depending on locality) below the top of the Lower Visby Beds (Jeppsson, 1983; Aldridge et al., 1993), and the Wenlock/Ludlow boundary is within the Klinteberg Beds (Martinsson, 1967). The youngest part of the Sundre Beds belongs to the upper Ludlow (Ludfordian) (Jeppsson et al., 1994). In total, the succession of the Silurian on Gotland comprises the period from 431 to about 411 m.y. B.P. (Harland et al., 1990).

3. PREVIOUS STUDIES

Earlier reconstructions of the paleogeography and sedimentological conditions are based on the lithological and paleontological record. Generally, the formation of the various facies complexes is interpreted as the result of changing water depths connected with a gradual regression and a migration of the facies belts to the southeast (e.g., Manten, 1971; Laufeld and Basset, 1981). The interruption of reef formation at times along with the deposition of micritic limestones appears to represent intermittent episodes of transgression superimposed on a general regression trend (Laufeld and Bassett, 1981; Riding, 1981). At the base of the Wenlock, the shoreline is reconstructed to be in a northeast-southwest direction north of Gotland and to migrate to the south during the Silurian. At the end of the Ludlow the major part of Gotland is supposed to have been rested above sea level (Brood, 1976; Stel and de Coo, 1977; Bassett et al., 1989).

Jux and Steuber (1992) analysed the isotopic composition (C_{org}, C_{carb}, O_{carb}) of rocks and reef building organisms on Gotland. Shifts in isotope values are interpreted as the result of worldwide sea level changes and interacting crustal movements. The ratios of stable carbon isotopes represent fluxes in the global carbon cycle, but decisive paleoclimatic changes are not inferred. Similar conclusions are drawn by Wenzel and Joachimski (1996), who measured oxygen and carbon isotopes in brachiopod shells from the Silurian of Gotland. The variations in the isotope signals correlate with changes in sea level. Sea level highstands correspond to lower oxygen isotope values indicating formation of warm saline waters on subtropical shelves leaving shallow waters depleted with respect to ^{18}O due to a storage of ^{18}O in the deep ocean, as proposed by Railsback (1990) for the Ordovician ocean. Low carbon isotope values indicate a low surface water productivity. During periods of sea level lowstands, colder waters were formed in high latitudes, resulting in a better ventilation of the basins and in high ^{18}O values. The corresponding high ^{13}C values are attributed to an enhanced primary productivity.

Jeppsson (1990) observed a close correlation between the facies development on Gotland, alternating between shelf basin facies and enhanced carbonate platform formation with cycle lengths in the order of a few million years, and a distinct conodont faunal cyclicity. During periods of shelf basin character the conodont assemblages exhibited high abundances and diversities. In contrast, during periods of laterally expanding reef and platform facies an impoverished and low diverse conodont fauna occurred. The change of

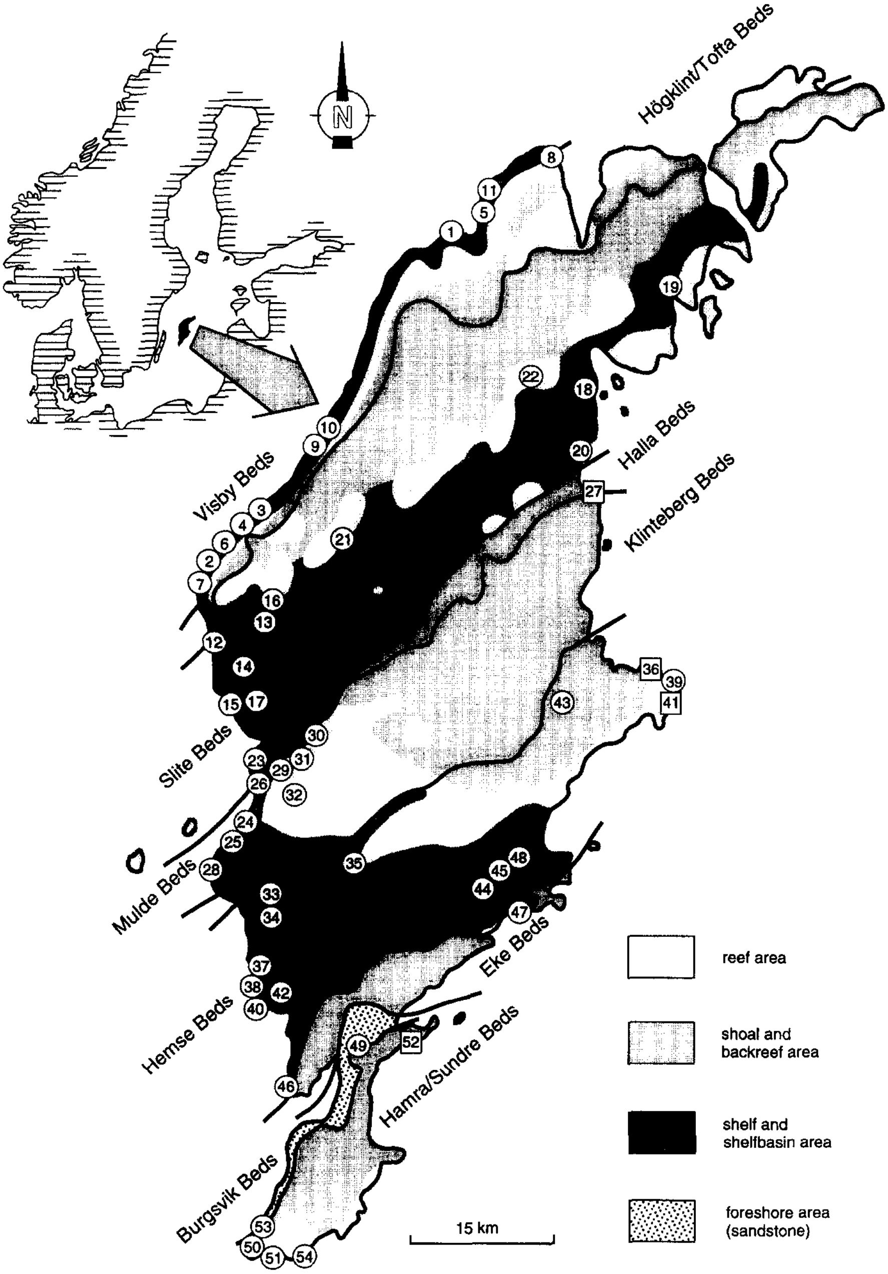
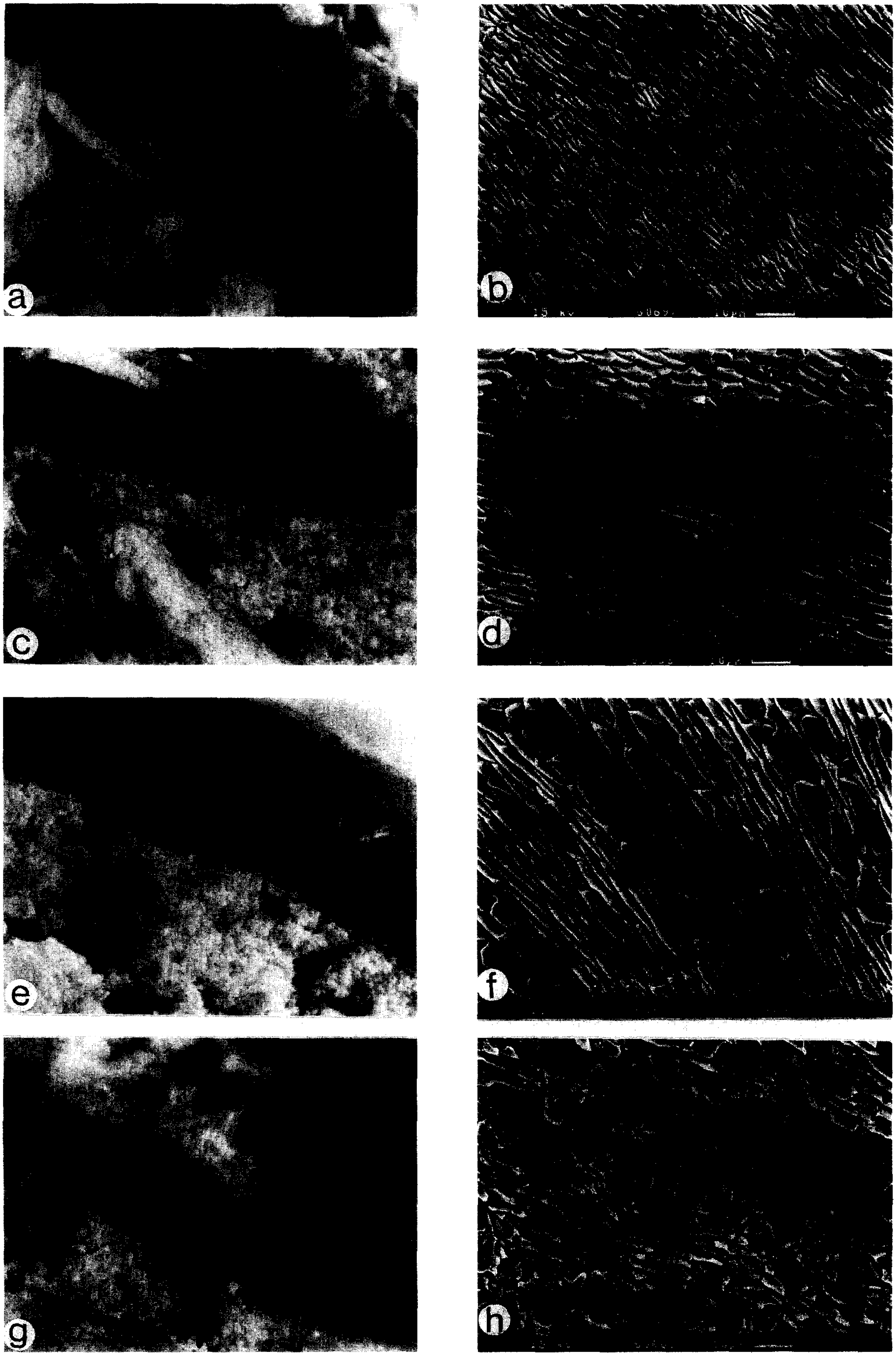


Fig. 1. Map of Gotland including topostratigraphy (Hede, 1921, 1960), facies interpretation, and sample localities (circles = mostly *Atrypa reticularis*, squares = exclusively other brachiopod species).

conodont assemblages preceded the facies development. This conodont cyclicity is observed world widely (except for the malvinokaffric realm) and is elsewhere connected with lithological changes (Jeppsson 1989, 1990). Changes from high to low diverse faunas are abrupt and connected with global general extinction events (Kaljo et al., 1995). A similar, although less regular development shows the record of acritarchs in the Silurian of Gotland (Kaljo et al., 1995).

Jeppsson (1990) introduced a model for global oceanic cycles, in which a humid low latitude and cooler high latitude climate (P-episode) alternate with a dryer low latitude and a slightly warmer high latitude climate (S-episode). Sedimentary changes involve fluctuations in the rate of clay deposition and carbonate production and the advance or retreat of carbonate deposition on shelves. As a driving mechanism for these cycles the intensity of deep water formation in high latitudes is proposed. During P-episodes, cold and dense bottom water which was rich in oxygen was formed in high latitudes. During S-episodes, warmer surface temperatures in high latitudes reduced the ventilation of the deep ocean, consequently euxinic conditions developed.

The present authors (Samtleben et al., 1996) re-interpreted the facies succession of Gotland, based on both lithofacies analysis and stable isotope measurements in brachiopods, to result from global paleoclimatic conditions (alternation of humid and arid stages), applying the model of Jeppsson (1990; see above). The oxygen isotope ratios are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interpreted to reflect paleosalinity changes due to varying freshwater input, rather than to paleotemperature. Carbon isotope ratios are believed to have been connected to global changes in the burial of organic carbon in black shales during periods of euxinic deep water conditions. Consequently, the carbonate formation on Gotland is not controlled by different water depths due to sea level fluctuations but by changes in terrigenous input due to different rates of weathering and freshwater runoff. At the end of the Silurian sequence no regression or even emersion is indicated by the facies development on Gotland.

4. MATERIAL AND METHODS

About 370 brachiopods from eighty-nine sites at fifty-four localities, mainly from the western part of Gotland (Fig. 1) were analyzed. The stratigraphic positioning of the samples follows the topostratigraphic classification of Hede (1921, 1960) and is presented as normative vertical distance to the top of the Lower Visby Beds (*Phaulactis* layer, Samtleben et al., 1996), which coincides roughly with the Llandovery/Wenlock boundary.

For the investigation of the Silurian of Gotland, the spiriferid brachiopod *Atrypa reticularis* has been chosen because of its occurrence throughout the entire stratigraphic succession and its widespread distribution in most facies areas. By consistent restriction to one species, possible different reactions of various species to environmental conditions can be avoided. However, *A. reticularis* is generally rare in biohermal and biostromal deposits and absent in marginal marine and some backreef areas. In these cases other brachiopod species have been sampled and analysed. Where possible, species have been investigated which are systematically related to *A. reticularis* and show the same skeletal ultrastructure: *Microsphaeridiorhynchus nucula* (Rhynchonellida), *Ferganella borealis* (Rhynchonellida), *Howellella elegans* (Spiriferida), *Delthyris elevata* (Spiriferida), *Ivanothyris* cf. *gibbosa* (Spiriferida), and in reef complexes *Didymothyris didyma* (Spiriferida). In order to determine the degree of correspondence with the isotope values from *A. reticularis* several parallel analyses were made from locations where both *A. reticularis* and other species occurred. In general, the interspecies differences were smaller than ±0.2‰ and, therefore, well within the variance of *A. reticularis* of the same collected sample (Samtleben et al., 1996).

The original shell ultrastructure of brachiopods consists of a primary layer calcite, which forms the thin outer layer of the shell, and a secondary layer below, build up by long, intracellularly precipitated and closely packed calcite prisms. If diagenesis occurs, this structure is progressively altered, resulting in a fusion of the prisms and an aggrading recrystallization. The state of preservation of the shell ultrastructure was determined using a scanning electron microscope (SEM). In addition, cathodoluminescence (CL) investigations were carried out. These investigations showed a strict correlation between CL and diagenetic alteration of the shell ultrastructure (Fig. 2). Most of the investigated samples (ca. 90%) showed original shell structures without recrystallization. Indications of diagenesis were observed in some samples from reefs and bioclastic limestones (25 m, 35 m, 120 m, 180 m, 320 m, 375 m, and 383 m above the *Phaulactis* layer). However, other specimens from these samples showed no diagenetical alteration or CL and were, therefore, used for determination of isotopic composition.

The brachiopods were cleaned mechanically and in an ultrasonic bath. Samples were taken from the shell surface by abrasion with a scalpel under the binocular microscope. To avoid influences on isotopic values due to adherent micritic material or sparitic cements especially close to the beak and the edge of a shell, only the central area of the exterior part of the shells has been sampled. To avoid vital effects on isotope composition (see Carpenter and Lohmann, 1995) and because the outer (primary) layer of the shells always showed CL, samples were taken exclusively from the prismatic (secondary) layer. Due to the fact that sometimes a gradual transition between shell and a sparitic filling of the brachiopods was observed, samples were taken mainly from the exterior part of the prismatic layer.

60 to 80 μg of the homogenized carbonate material from each sample were analyzed using a Finnigan MAT 251 micromass-spectrometer coupled with a Finnigan automated carbonate preparation device at the University of Bremen. The carbonate was reacted with orthophosphoric acid at 75°C. The reproducibility of the measurements, as referred to an internal carbonate standard (Solnhofen limestone), is ± 0.07‰ and ± 0.05‰ (1σ over a one year period) for oxygen and carbon isotopes, respectively. The conversion to the PDB scale was performed using the international standard NBS 19. The isotope data, used in this study, are available from the first author via internet upon request.

5. RESULTS

In Fig. 3 stable isotope measurements in brachiopod shells, mainly *A. reticularis*, are plotted against the height of each sample in the normative profile of the Silurian of Gotland. The oxygen and carbon isotope records generally exhibit parallel variations. The values range from −7 to −3‰ in oxygen and −1 to +8‰ in carbon isotopes. The variability of various individual shells at a given site depends on the kind of brachiopod assemblage which has been sampled. In groups of shells, found in obviously autochthonous or even life position, the internal variability is ±0.4‰ for oxygen and 0.3‰ for carbon isotopes. A somewhat larger range (±1.0‰ and ±0.4‰, respectively) is observed in assemblages of allochthonous specimens (for further discussion see Samtleben et al., 1996). However, the range of the variability in autochthonous samples is clearly lower than the shifts observed in the isotope records and, therefore, does not prohibit their interpretation.

Both carbon and oxygen isotope records are closely related to the stratigraphic succession and the general facies development (Fig. 3). Compared to the lithostratigraphic sequence, high carbon and oxygen values occur in periods characterized by reef complexes and extensive carbonate platforms (Högklint Beds, Klinteberg Beds, Hamra-Sundre Beds). Low values occur in sequences dominated by micritic limestones and marls (Lower Visby Beds, Slite Beds, Hemse Beds). In general, marly series show less internal variability indicating more stable environmental conditions during their deposition compared to series of increased reef growth. However, on closer examination, it is remarkable that changes in isotope composition clearly precede changes in carbonate facies development. That is, increases in both δ^{13}C and δ^{18}O occur 12–15 m below facies transitions from marl-

Fig. 2. Cathodoluminescence (CL) of *Atrypa reticularis* shells in polished sections (×40; on the left) in comparison with their skeletal ultrastructure in SEM micrographs (×700; on the right). Black arrows indicate the position of the SEM section on CL photomicrographs. (a–b) Nonluminescent shell with original ultrastructure (Stavsklint, Högklint Beds, 35 m); (c–d) Small patch of CL (width 0.1 mm) in an nonluminescent shell showing initial recrystallization within unaltered shell structure (Uddvide, Hamra Beds, 375 m); (e–f) Alternation of luminescent and nonluminescent layers within a diagenetically affected shell (Irevik/Snipan, Högklint Beds, 25 m); (g–h) Shell with distinct CL and strongly recrystallized ultrastructure (Stavsklint, Högklint Beds, 35 m).

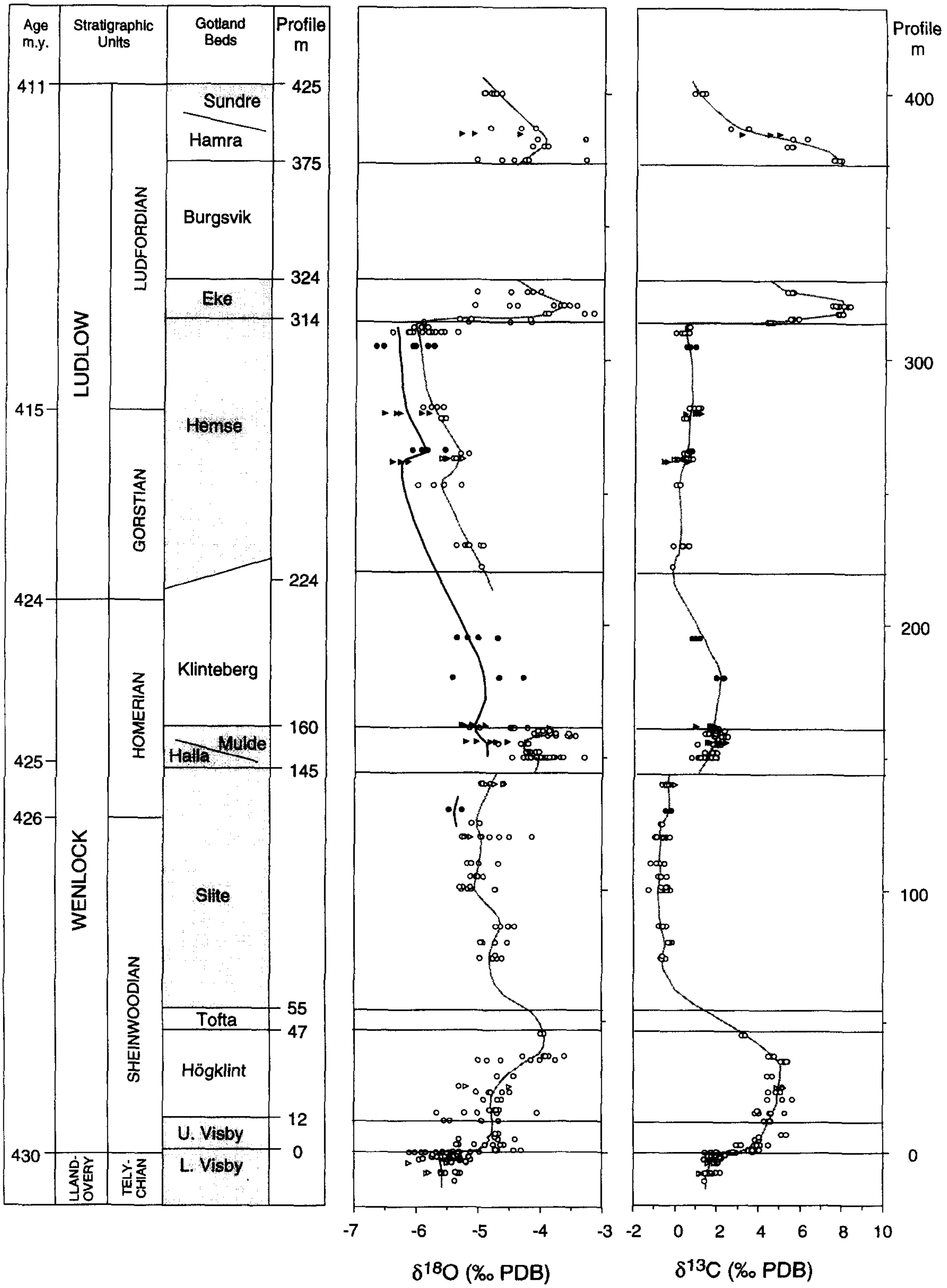

Fig. 3. Synoptic presentation of the stratigraphic frame (Harland et al., 1990), topostratigraphy of Gotland (Hede, 1921, 1960) and the oxygen and carbon isotopes, measured in brachiopod shells, vs. vertical distance to *Phaulactis* layer (open symbols = western side facies, black symbols = eastern side facies, circles = *Atrypa reticularis*, triangles = other brachiopod species). Intervals according to marl-dominated stratigraphic units are shaded.

dominated facies to periods characterized by reef carbonates, and also the decreases in isotope values start well below the top of each reef-dominated unit (Fig. 3).

Although carbon and oxygen isotopes seem to correspond closely one to each other, it is surprising that the correlation between both isotopes, checked by means of a linear regres-

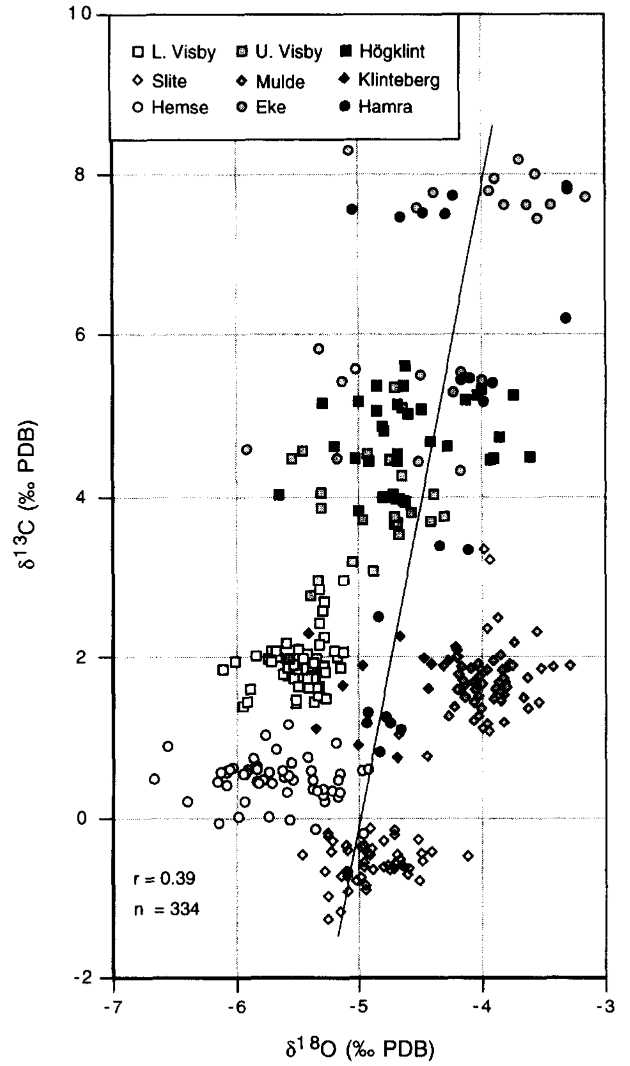

Fig. 4. Oxygen vs. carbon isotopes, measured in *Atrypa reticularis* in different topostratigraphic units, show a positive though weak correlation.

sion, is poor ($r = 0.39$; Fig. 4). This is caused by long-term trends (10^7 y) of the isotope records, which are superimposed by the secular changes (10^6 y) described above. If solely the $\delta^{13}C$ and $\delta^{18}O$ values of brachiopods sampled in stratigraphic units corresponding to P-states (Jeppsson, 1990; Lower Visby Beds, Slite Beds, Hemse Beds, uppermost Hamra-Sundre Beds) are considered (because they are assumed to represent the normal conditions during the Silurian; see the discussion below), linear regression gives a much better coefficient ($r = 0.59$; Fig. 5a) but with a negative sign due to the contrary course of the long-term trends of both isotope records (Fig. 5b). If these trends are approximated each by a third-order polynom and subtracted from the isotope values of each site, an excellent correlation between the detrended $\delta^{13}C$ and $\delta^{18}O$ values arises ($r = 0.79$; Fig. 5c), despite the fact that the $\delta^{13}C$ variability exceeds that of $\delta^{18}O$ by a factor of 2.5 (Fig. 5d). In summary, there is a covariation of carbon and oxygen isotopes related to the shorter secular changes in the facies succession of Gotland, but this covariation is masked by the opposite direction of isotope changes on longer timescales.

Lateral facies changes in synchronous strata, predominantly in an northeast-southwest direction, have minor, though significant influences on oxygen isotope compositions. Samples from reef areas and extremely shallow marine deposits on the eastern side of Gotland show generally lower values of oxygen isotopes (about 0.5‰) compared to samples from micritic shelf basin deposits on the western side. In parts of the stratigraphic sequence (upper Slite Beds, Klinteberg Beds, Hemse Beds) a second oxygen isotope curve marks the deviation of the isotope compositions in these extremly shallow marine deposits (Fig. 3). Due to local influences this curve is less regular than the one from the western side, although it follows the same general trend. In carbon isotope compositions no differences are observed between synchronous facies.

6. DISCUSSION

6.1. Secular Changes in Oxygen Isotopes

The oxygen isotope ratios in carbonate shells are a function of both temperature and the $\delta^{18}O_w$ of the surrounding seawater. Among all the paleotemperature equations which have been developed since the first one of Epstein et al. (1953), but which are not much different in their results (Wefer and Berger, 1991), we chose the equation of Erez and Luz (1983), which is based on laboratory experiments with planktonic foraminifera.

$$T = 17.0 - 4.52 * (\delta c - \delta w) + 0.03 * (\delta c - \delta w)^2$$

T	in-situ temperature during shell precipitation (°C)
δc	$\delta^{18}O$ value of the calcite (‰ PDB)
δw	$\delta^{18}O$ value of the seawater (‰ PDB)

The $\delta^{18}O_w$ of seawater itself is controlled by fractionation effects due to evaporation and precipitation at the sea surface, freezing of ice in polar regions, the admixing of water masses containing different $^{18}O/^{16}O$ ratios such as melt water, river runoff, etc., and the global isotope content of the oceans. Since the salinity of seawater is similarly affected by these processes, Craig and Gordon (1965) and later Fairbanks et al. (1992) defined a set of regression relationships between salinity and $\delta^{18}O_w$ with different slopes for several modern water masses, varying between 0.1 for humid tropical and 1.0 for arid polar surface water masses. Although Gotland was located close to the Silurian equator, we choose the global mean of 0.49 instead of the tropical value for the following calculations, because the relationships between $\delta^{18}O$ and salinity for the Silurian are still unknown.

Interpreting the oxygen isotope record of the Silurian of Gotland as to exhibit only paleotemperature variations and assuming a marine isotopic composition close to the modern global mean ($\delta^{18}O_w = 0$‰), the temperatures would range from 33° to 46°C (Fig. 6, Variant 1) with a mean of 39.5°C and an error of ±1.8°C (due to a range of ±0.4‰ in $\delta^{18}O$ in autochthonous brachiopod groups, see above). These temperatures seem unrealistic, because they exceed the thermal lethal limits for marine invertebrates, e.g., 31°C for corals (Kinne, 1970; Brock, 1985). Based on the paleogeography of the continents and a pCO_2 level four times higher than the present-day level of the atmosphere (i.e., preindustrial content), Moore et al. (1994) simulated the Silurian ocean

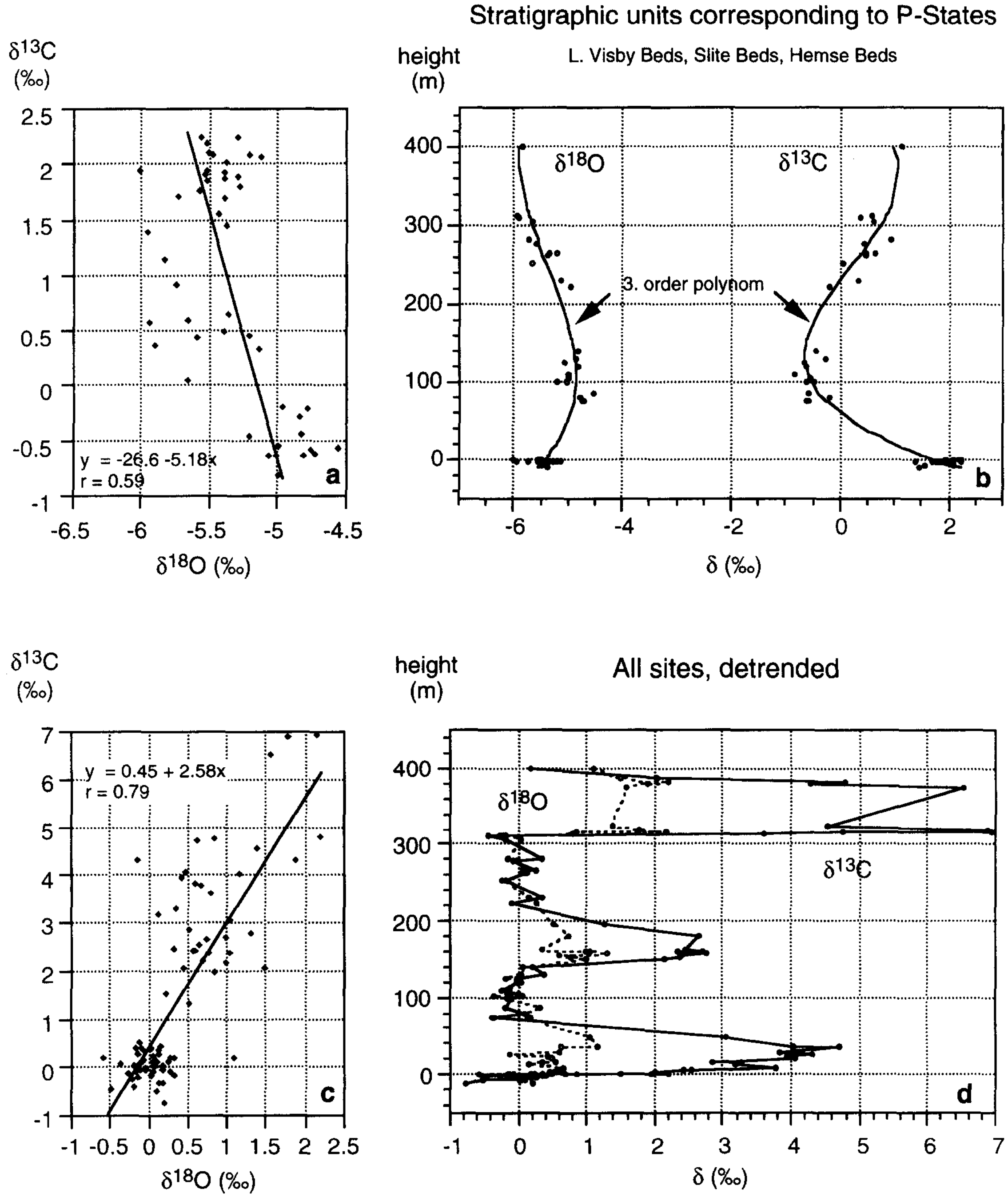

Fig. 5. Oxygen and carbon isotopes, measured in *Atrypa reticularis,* show a negative correlation in stratigraphic units corresponding to P-states (L. Visby, Slite, Hemse Beds) (a), due to their opposite longterm trends (b). If corrected for this trends by subtraction of 3. order polynoms, $\delta^{18}O$ and $\delta^{13}C$ of all sites show a much better correlation (c), despite the about 2.5 times higher amplitudes in the $\delta^{13}C$ records (d).

environmental conditions. The best fit with climatically sensitive biofacies and lithofacies data resulted in sea surface temperatures of about 30°C for tropical seas, well below the temperatures calculated from Gotland oxygen isotopes above.

Several authors have suggested a lower global isotopic composition for the Paleozoic oceans on the order of −1‰ to −3.5‰ (Hudson and Anderson, 1989; Bates and Brand, 1991; Veizer et al., 1986; Qing and Veizer, 1994). This depletion in ^{18}O is explained by enhanced interactions of seawater with fresh, silicate rocks at lower temperatures during the Paleozoic compared to younger epochs (Carpenter et al., 1991; Walker and Lohmann, 1989). This explanation has been questioned because studies of the dynamics of mid-ocean-ridge/seawater interactions suggest that the oxygen isotopic composition of seawater should have been buffered at values close to the modern ocean composition (Hoffman et al., 1986; Muehlenbachs, 1986). On the other hand, Bar-

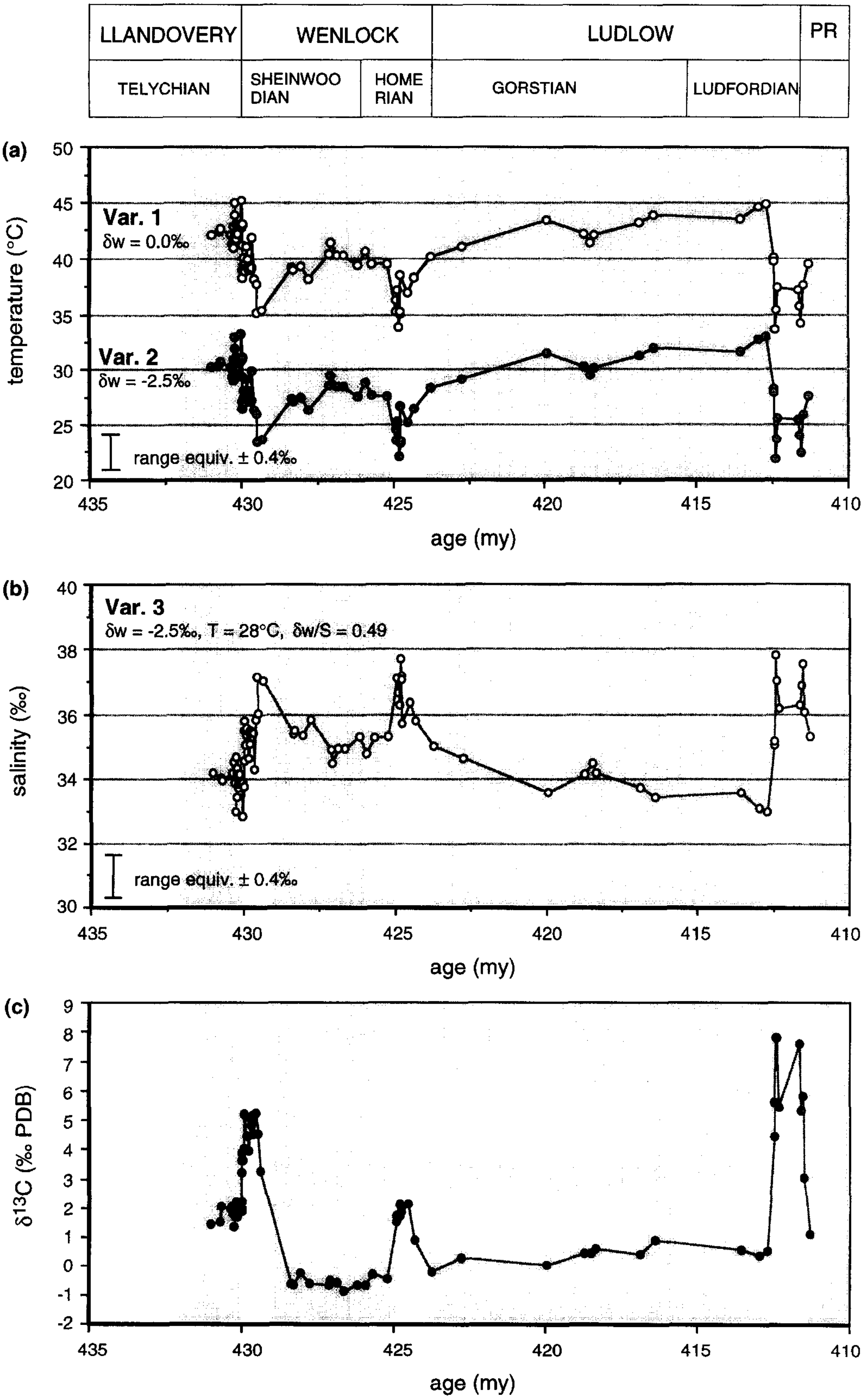

Fig. 6. Paleotemperature (a) and paleosalinity (b), discussed as three variants for interpreting oxygen isotope measurements in *A. reticularis*, using the paleotemperature equation of Erez and Luz (1983) and the global salinity-$\delta^{18}O$ gradient of 0.49 (Fairbanks et al., 1992). $\delta^{18}O$ value of seawater is assumed to be 0.0‰ (Var. 1) or −2.5‰ (Var. 2 and 3) according to Railsback (1990). For multiple $\delta^{18}O$ values of a horizon only the average value is presented, the error for a deviation of ±0.4‰ (autochthonous groups; see text) is given as a vertical bar. Carbon isotopes (c) are presented vs. age for comparison. Intervals corresponding to P-episodes (Jeppsson, 1990) are shaded.

rett and Friedrichsen (1989) showed that low temperature exchange reactions may dominate in some ophiolite sequences, implying a possible imbalance between reactions controlling the $\delta^{18}O$ of seawater. Walker and Lohmann (1991) suggested that possible rates of change appear to be quite slow (1‰ in 10^8 years) because of the large size of the oceanic reservoir. Therefore, such considerations make it seem especially unlikely that ocean compositions could undergo rapid changes. Railsback (1990) approximated the rising of $\delta^{18}O_w$ during the Paleozoic by a third-order relation to simulate a slow and nonreversible changing ocean, giving an average oxygen isotopic composition of −1.5‰ for the Silurian. Due to the nearly ice-free conditions at that time (Grahn and Caputo, 1992), an additional −0.8‰ has to be considered for the Silurian ocean. Thus, applying a $\delta^{18}O_w$ of −2.5‰ to the data of Gotland, the temperatures would range from 22°C to 34°C (Fig. 6, Variant 2) with a mean of 27.7°C and an error of ±1.8°C (see above). Although these temperatures are more realistic than the previously calculated data, their range of 12°C is still too large, compared to the relatively stable conditions generally assumed for tropical sea surface waters (e.g., 1–2°C for the last glacial/interglacial transition in the late Quaternary; CLIMAP, 1981).

In this study, we propose a third explanation for the observed variation in oxygen isotopes measured in brachiopods from the Silurian of Gotland. From the discussion above it seems reasonable that the short-term secular changes in $\delta^{18}O$ may not be interpreted as a paleotemperature record, but as a paleosalinity record. Assuming again the oxygen isotopic composition of seawater to be −2.5‰ (Railsback, 1990), a slope between salinity and $\delta^{18}O_w$ of 0.49 (the modern global average), and the temperature to be constant at 28°C, the resulting salinities range between 32.5‰ and 38.0‰ with a mean value of 35.3‰, close to the modern ocean salinity (Variant 3, Fig. 6). Investigations on fluid inclusions of Cambrian and Ordovician marine cements suggest that ocean salinity has not changed significantly during the Phanerozoic (Johnson and Goldstein, 1993). The range of salinities, we calculated, is also observed in modern ocean surface waters (the Gulf of Bengal as an example for humid, and the Red Sea for arid conditions; Levitus, 1982) and does not violate the stenohaline conditions preferred by modern brachiopods (≥32‰, Lowenstam, 1961; Lepzelter et al., 1983).

Silurian paleogeographic reconstructions place Gotland near the equator (Scotese and McKerrow, 1990; Torsvik et al., 1992). The salinity variations are, therefore, presumed to result from changes in the tropical climate. Lower salinities result from high rates of precipitation during humid climatic periods, admixing freshwater to near surface water masses. Humid precipitation would also lead to high rates of weathering, erosion, and input of clay minerals resulting in turbid waters which decrease the benthic carbonate production and inhibit reef formation. And indeed, periods of low salinities coincide with lithofacies units which are characterized by marly deposits with minor or no reef formation (Lower Visby Beds, Slite Beds, Hemse Beds). During periods of arid climate, higher salinities occur, and decreased rates of terrigenous input permit widespread reef growth (Högklint Beds, Klinteberg Beds, Hamra/Sundre Beds). This salinity hypothesis is confirmed by the fact that changes in facies on Gotland slightly lag behind changes in oxygen isotopes. This phase relationship constrains that facies changes are caused by the mechanism which control the variability in oxygen isotopes, i.e., the variability of freshwater run off.

Wenzel and Joachimski (1996) attributed the variability in oxygen isotopes on Gotland to sea level changes (Johnson et al., 1991). Periods with high isotope values and increased reef formation are connected with nutrient rich surface waters due to strong terrigenous influx and vice versa. This explanation is rejected for the following reasons: (1) Due to the lack of continental ice sheets at least during Wenlock and Ludlow times (Grahn and Caputto, 1992), rapid glacio-eustatic sea level changes, as known from late Pleistocene climates, are unlikely to occur in the studied Silurian interval. On the other hand, tectono-eustatic sea level movements would be too slow to produce as abrupt changes in environmental conditions as recorded in the isotope records of Gotland. If an age model for the isotope records of Gotland is defined by interpolating between the datings of the Silurian stratigraphic units (Fig. 3), the isotope shifts connected to the transitions from marl- to overlying limestone-dominated units lasted less than 500 ky (Slite/Mulde Beds, Hemse/Eke Beds) or even less than 100 ky (Lower/Upper Visby Beds). Such rapid changes can hardly be attributed to tectono-eustatic sealevel changes. (2) The sea level reconstruction for the Silurian by Johnson et al. (1991) is questionable, because it is based mainly on changes in faunal assemblages. On Gotland, field observations constrain that the regional development of water depth is not the cause, but the result of carbonate production and accumulation (Samtleben et al., 1996). That is, under climatic conditions which caused high carbonate production (arid states) the carbonate platforms enlarged laterally and prograded into the adjacent shelf basins causing shallow water habitats. In periods of reduced carbonate formation (humid states) the platforms retreated while the marly sequences of deeper shelf areas expanded. (3) The paleonvironmental reconstruction of Wenzel and Joachimski (1996) is also inconsistent with the facies succession, because it assumes high terrigenous influx in times of intense reef growth. This contradicts the general (fossil and recent) ecologic conditions for reef formation (e.g., James, 1983; Schuhmacher, 1982).

Abrupt changes in environmental conditions as recorded in the isotope records of Gotland are easily explained by changes in tropical climate like the interactions between monsoon and trade wind realms as known from Plio-Pleistocene sediments from the Mediterranean Sea. Episodic occurrence of sapropels, intercalated as thin layers in generally oxic sediments, is attributed to short periods (≈20 ky) of intense freshwater influx, which prohibits the formation of warm, saline deep water, forming under usually arid climatic conditions, and leads to anaerobism in the normally deeply ventilated ocean basin (e.g., Rohling, 1994). Such a mechanism is also confirmed by the interpretation of the carbon isotope records and the lithological observations, as will be discussed in the following chapter.

As argued above, the development of the oxygen isotope values on Gotland was dominated by changes in the salinity due to varying freshwater influx. However, the lateral variability in $\delta^{18}O$ of contemporaneous strata gives evidence that these differences are influenced by temperature. The about 0.5‰ lower $\delta^{18}O$ values of the eastern side samples compared to those of the western side indicate local environmental effects on oxygen isotopes superimposed on the paleosalinity changes (Fig. 3). From the lithofacies interpretation given above it is evident that the eastern side strata represent shallower water conditions than those of the western island. Thus brachiopods of this facies reflect higher temperature and/or different salinity conditions in their isotope values while living closer to the sea surface.

6.2. Secular Changes in Carbon Isotopes

The carbon isotopic composition of ΣCO_2 in seawater is controlled mainly by two processes, biochemical fractionation due to the formation and decay of organic matter, and the physical fractionation during gas exchange at the air-sea boundary (Broecker and Maier-Reimer, 1992). Surface water is enriched in ^{13}C, because photosynthesis preferentially removes ^{12}C from the ΣCO_2. Deeper water masses have lower $\delta^{13}C$ values, since nearly all of the organic matter that is produced by photosynthesis is subsequently remineralized in the water column. On the other hand, cold surface water is enriched in ^{13}C compared to warm water due to the larger fractionation effect during the air-sea gas exchange.

Generally, three explanations are given for changes of $\delta^{13}C$ distribution in the ocean: (1) changes in the surface-ocean productivity which cause variable fractionation between surface and deep water carbon isotopic composition, (2) changes in the gas exchange rates between ocean and atmosphere due to changes in surface temperatures and ocean circulation, and (3) changes in the marine carbon budget by variations in the reservoirs of the atmosphere, the ocean, or the lithosphere.

Wenzel and Joachimski (1996) explained the periods of high $\delta^{13}C$ values on Gotland by a high productivity due to an intense supply of nutrients caused by continental runoff in times of sea level lowstand. Beside the fact that periods associated with high $\delta^{13}C$ values contain sparse and low diverse conodont faunas, which indicate periods of low surface water productivity (S-states; Jeppsson, 1990), it is questionable whether productivity changes alone could explain the large shifts in carbon isotopes observed in the Silurian of Gotland. Due to the fact that more than 90% of the organic material is recycled within the water column (Berger et al., 1989), it is unlikely to develop such an extreme fractionation necessary to produce the observed ^{13}C enrichment under aerobic conditions. Furthermore, modelling of modern ocean conditions reveals that the difference between a maximally acting biological pump and a strangelove ocean state is only on the order of 2–3‰ (Berger and Vincent, 1986). This would explain only 25–35% of the maximal shift observed at the transition between Hemse Beds and Eke Beds (Fig. 3).

Also the second explanation (changes in the air/sea fractionation) seems unlikely because of the stable climatic conditions during Silurian times. Fischer (1983) assigned the Wenlock and Ludlow series to a greenhouse mode with only little or no glaciation on the continents. There is no cause for greater changes in the surface temperature pattern which would be large enough to result in the carbon isotopic shifts observed in the isotope records of Gotland.

Therefore, we favor reservoir changes due to changes in ocean circulation as the explanation for the variation in carbon isotopes measured in brachiopods from the Silurian of Gotland. Over geologic time, the influx and burial of sedimentary inorganic and organic carbon exert a primary control on the $\delta^{13}C$ of the ocean and the atmosphere. Main sources for carbon comprise the erosional flux of sedimentary carbon (as C_{org} and carbonate) and the degassing of volcanic CO_2, main sinks are the burial of organic matter and the deposition of carbonate. Because of the large kinetic isotope effect associated with photosynthetic fixation of carbon, the carbon isotopic composition of marine sediments permits an estimate of the relative importance of carbon burial in its reduced or oxidized form (Derry and France-Lanord, 1996).

The deep Silurian ocean is assumed to exhibit permanent anoxic conditions (Wilde et al., 1991). This is concluded from lower oxygen contents, which are estimated to have been about 65% of present atmospheric level (PAL) at the beginning of the Silurian and declining to about 35% PAL at the beginning of the Devonian (Budyko et al., 1987). Assuming an atmospheric value of 50% PAL, the oxygen content in tropical waters would decline to zero at about 100 m depth (Wilde et al., 1991), and reducing conditions and anoxic waters would extend downward into the deep ocean. Such conditions encourage intense carbon burial in deep sea sediments, what is documented by the widespread black shale deposits during that time (Jaeger, 1976; Kemp, 1991), and lead to extreme carbon isotope fractionation between surface and deep water. Applying a simple equation for carbon isotope mass balance, given by Derry and France-Lanord (1996), a +8‰ increase (the maximum shift observed on Gotland, at the Hemse-Eke transition) in surface water $\delta^{13}C$ would be produced, if about 50% of carbon input in the ocean is buried as organic matter. In the modern ocean, an 8‰ difference between surface and deep water $\delta^{13}C_{DIC}$ is observed in the Black Sea, where freshwater influx to the surface ocean causes stagnant and anoxic conditions below 250 m water depth (Fry et al., 1991).

For the secular changes in carbon isotopes measured in brachiopods of Gotland we, therefore, propose that changes in the circulation of epicontinental seas are responsible for rapid shifts observed in the Silurian record. In periods characterized by humid tropical climates, intense continental runoff and, therefore, marl-dominated deposits (HUMID states in Fig. 7a), the freshwater influx leads to an estuarine circulation, comparable to modern hydrographic pattern in the Black Sea, and allows anoxic deep ocean waters to invade onto deeper shelf areas. The consequent admixture of ^{12}C-enriched deep waters lowers the $\delta^{13}C$ values of the shallow ocean water. On the other hand, in periods characterized by arid tropical climates and reef growth (ARID states in Fig. 7b), evaporation leads to downwelling of warm, saline, and well oxygenated surface water, which is balanced by lateral advection of ^{13}C-enriched open ocean surface water. Such an anti-estuarine circulation, known today from the Mediterranean Sea, pushes the top of the anoxic layer down to greater water depths. Nevertheless, it is important to note that the deep Silurian ocean remained anoxic.

Several arguments support the proposed paleoceanographic scenario: First, the strong correlation between oxygen and carbon isotopes on short-term changes (see Fig.

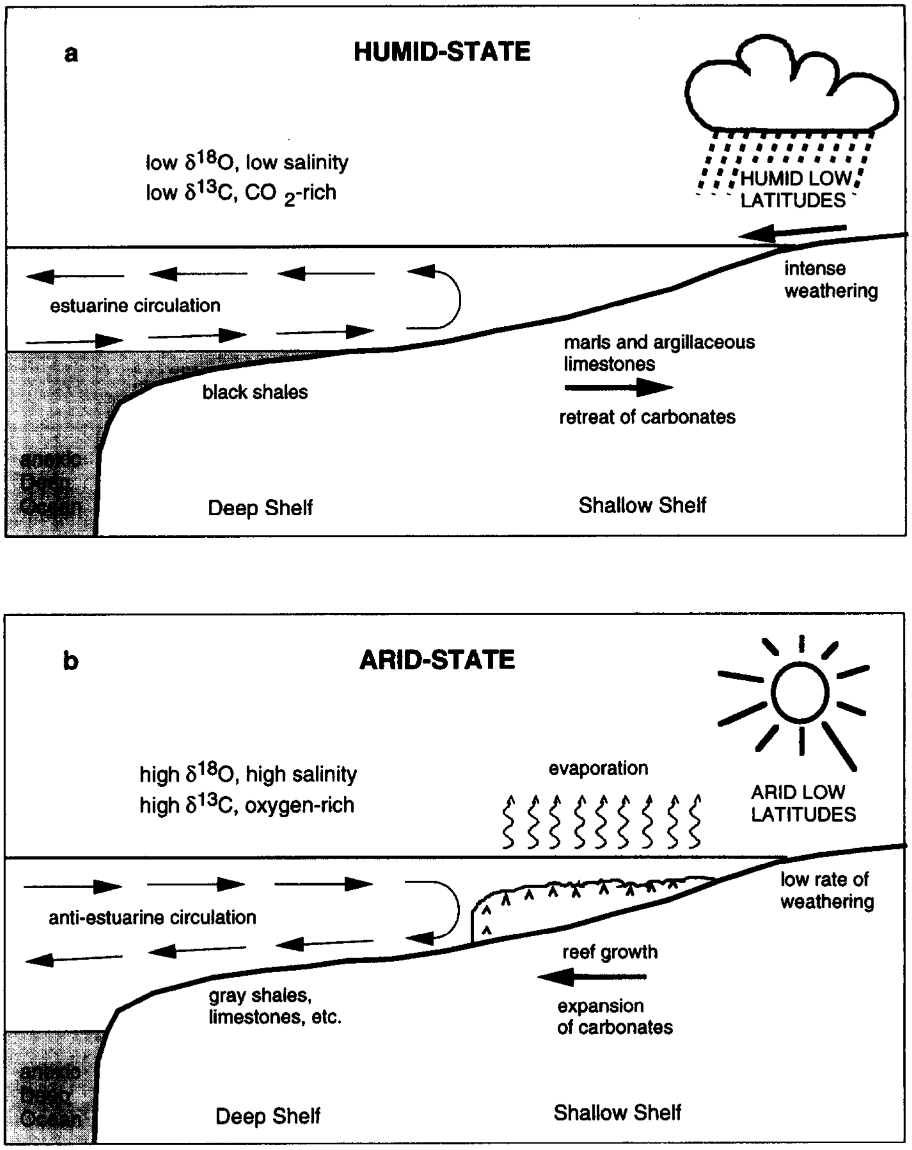

Fig. 7. Schematic view of the Silurian paleoceanography during (a) HUMID-episodes and (b) ARID-episodes, modified after Jeppsson (1990).

5) strengthens that these changes are caused by the same mechanism, i.e., the circulation changes due to varying freshwater influx. Second, lithological records from deeper shelf areas show that the anoxic conditions assumed for HUMID states are documented by the deposition of black shales, while two short periods of limestone deposition adjacent to the Iapetus Ocean (lowest Sheinwoodian and upper Homerian; Kemp, 1991) and a period of gray shale deposition in the Paleotethys realm (Ludfordian; Jaeger, 1976) indicate episodes of better ventilation, which coincides roughly with the ARID states defined on Gotland. Third, the rapid changes in both oxygen and carbon isotopic composition coincide with the distinct cyclicity in conodont faunal development (Jeppsson, 1989, 1990).

The fact that the conodont faunal cyclicity is observed worldwide and is elsewhere connected with similar lithological changes constrains the global character of the forcing mechanism which is seen in the alternation of humid and arid climate systems (Jeppsson, 1990). A further confirmation for a global forcing is given by the fact that at least for one $\delta^{13}C$ shift to higher values close to a stratigraphic boundary (Llandovery/Wenlock) there is another example in Australia (Borenore Caves), although measured in whole rock samples instead of brachiopod shells (Talent et al., 1993). Therefore, although the paleoceanographic reconstruction in the Silurian of Gotland, proposed in this study, is based on environmental conditions of an epicontinental sea, it can be transferred to other tropical regions. The development of isotope values is expected to be principally coherent worldwide, whereas deviations in absolute values and amplitudes may occur due to regional differences in oceanographic conditions.

6.3. Long-Term Trends vs. Secular Variability of Oxygen and Carbon Isotopes

In order to determine the relevance of HUMID- and ARID-states for the Silurian paleoclimate, it is necessary to get an idea of the time frame of these changes, i.e., the duration of each climatic state. Due to the sparse number of absolute datings for Silurian stratigraphic units, it is difficult to define an age model with a sufficient time resolution.

Applying the timescale of Harland et al. (1990) and transferring the approximation of 1–1.5 m.y. for the Halla/Mulde-Klinteberg sequence to the two other reef-dominated facies units (Högklint/Tofta Beds and Hamra/Sundre Beds), periods of reef growth characterized by high salinities as well as by high $\delta^{13}C$ values seem to occur as rather short episodes ($\leq$1.5 m.y.) in between longer periods of marl-dominated sedimentation, low salinities, and low $\delta^{13}C$ values. This suggests that ARID states are extraordinary episodes in a usually HUMID paleoclimate epoch. If the small maximum in salinity within the Hemse Beds (at 418 m.y., Fig. 6) is assumed to represent a (suppressed) ARID state, then a periodicity of 6 ± 1 m.y. is obtained, which is a common cyclicity in earth history (see Frakes et al., 1992, for a review).

Although the development of $\delta^{13}C$ and $\delta^{18}O$ values have been shown to be parallel in the secular changes, the underlying long-term trends show antiparallel courses (Fig. 5b). This fact indicates that the factors controlling the long-term trends in isotopes are different from those assumed to control the secular changes. The long-term trends in the Silurian of Gotland interrupt the general increase of both carbon and oxygen isotope values during the Paleozoic. However, the causes responsible for these trends remain unknown.

7. CONCLUSIONS

CL and SEM-observations of ultrastructures reveal no evidence for diagenetic alteration in the brachiopod shells selected for this study. The shells are, therefore, assumed to reflect their primary isotopic composition.

The stratigraphic sequence of Gotland exhibit covarying alternations of stable carbon and oxygen isotopic compositions. These secular changes are underlain by long-term trends in both oxygen and carbon isotopes, which show a negative correlation.

Assuming a $\delta^{18}O$ value of $-2.5‰$ for Silurian seawater, short-term changes in oxygen isotope ratios are thought not to reveal a paleotemperature signal, exhibiting instead primarily changes in paleosalinities due to varying freshwater input (humid or arid tropical climate conditions).

Carbon isotope ratios are obviously connected to changes in oceanic circulation by the advection of ^{13}C-rich surface water (arid episodes) or upwelling of ^{13}C-depleted and CO_2-rich deep water (humid episodes) of a Silurian ocean which reveals generally euxinic deep water conditions due to the burial of organic carbon in black shales.

Dating of the Silurian sequence of Gotland shows that periods characterized by high salinities as well as by high $\delta^{13}C$ values, indicating the formation of warm saline deep water in low latitudes, seem to occur as rather short episodes of reef growth ($\leq$1.5 m.y.) in between longer periods of tropic humid conditions which cause an estuarine circulation.

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