

The SPICE carbon isotope excursion in Siberia: A combined study of the upper Middle Cambrian–lowermost Ordovician Kulyumbe River section, northwestern Siberian Platform

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Abstract

An integrated, high-resolution chemostratigraphic (C, O and Sr isotopes) and magnetostratigraphic study through the upper Middle Cambrian – lowermost Ordovician shallow-marine carbonates of the northwestern margin of the Siberian Platform is reported. The interval was analysed at the Kulyumbe section, which is exposed along the Kulyumbe River: an eastern tributary of the Enisej River. It comprises the upper Ust'-Brus, Labaz, Orakta, Kulyumbe, Ujgur, and lower Iltyk formations and includes the Steptoean positive carbon isotopic excursion (SPICE) studied here in detail from upper Cambrian carbonates of the Siberian Platform for the first time. The peak of the excursion, showing $\delta^{13}\text{C}$ positive values as high as +4.6‰ and least-altered $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.70909, is reported herein from the Yurakhian Horizon of the Kulyumbe Formation. The stratigraphic position of the SPICE excursion does not support traditional correlation of **the boundary between the Orakta and Labaz formations at Kulyumbe River with its supposedly equivalent level** in Australia, Laurentia, South China, and Kazakhstan, where the *Glyptagnostus stolidotus* and *G. reticulatus* biozones are known to immediately precede the SPICE excursion and span the Middle–Upper Cambrian boundary. The Cambrian–Ordovician boundary is probably situated in the middle Nyajan Horizon of the Iltyk Formation, in which carbon isotope values show a local maximum below a decrease in the upper part of the Nyajan Horizon, attributed herein to the Tremadocian. A refined magnetic polarity sequence shows that the geomagnetic reversal frequency was very high during the Middle Cambrian at 5–10 reversals per Ma, assuming a total duration of ~10 Ma and up to 100 magnetic intervals in the Middle Cambrian. By contrast, the sequence attributed herein to the Upper Cambrian on chemostratigraphic grounds contains only 10–11 magnetic intervals.

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1. Introduction

The Siberian Platform is one of the key areas in the world for research into the Cambrian-Ordovician interval. Continuous successions of sedimentary fossiliferous rocks little affected by metamorphism crop out there along river valleys. Three facies regions are traditionally recognized in the Cambrian of the Siberian Platform: Turukhansk-Irkutsk-Olyokma, Anabar-Sinsk, and Yudoma-Olenyok (Rozanov *et al.* 1992). In the upper Cambrian these regions correspond respectively to the inner shelf, transitional (with reefs, banks and bars), and slope/basin depositional environments (Sukhov, 1997; Fig. 1). At the northwestern margin of the platform the Cambrian deposits are often folded and well exposed in relatively compact sections. Among them biostratigraphy of the upper Middle – Upper Cambrian of the classical Kulyumbe River reference section has been most extensively studied in the second half of the the 20th century (e.g., Rozova, 1963, 1964, 1968; Rozova & Yadrenkina, 1967; Datsenko *et al.* 1968). This section, however, contains an endemic fauna that is a product of isolated environments at the margin of the inner shelf (Fig. 1). Without cosmopolitan genera and species of trilobites important for intercontinental correlation, faunal content and changes at Kulyumbe can not be directly translated to global biostratigraphic events.

For the purpose of precise correlation, carbon isotopic stratigraphy is the most suitable method in combination with biostratigraphy, but none of the areas on the Siberian Platform has been previously studied for carbon isotope chemostratigraphy within the upper Cambrian. This study provides the first high-resolution chemostratigraphic data from the upper Cambrian – lowermost Ordovician of Siberia. The Steptoean positive carbon isotopic excursion (SPICE) is an excellent global chemostratigraphic marker in the upper Cambrian usually rising far above the background noise (e.g., Saltzman *et al.* 2000, 2004). Documentation of the SPICE excursion from Siberia is valuable for correlation of richly fossiliferous Siberian sections with other well known Cambrian sections globally and, therefore, contributes much to understanding of biotic evolution, oceanography, climate, tectonics, and magnetic field at this time in Earth history.

2. Geological settings

The isotopic profile presented in this paper derives from several outcrops along the Kulyumbe River, located ~100 km E of the Enisej River (~68.0° N; 88.7–89° E), at the northwestern margin of the Siberian Platform (Rozova, 1968, 1984; Sokolov, 1982; Rozanov *et al.* 1992; Pavlov & Gallet 1998, 2001) (Fig. 1). The Middle Cambrian – Lower Ordovician sediments are exposed along ca 10 km of the Kulyumbe River, including in ascending order: the upper Ust'-Brus, Labaz, Orakta, Kulyumbe, Ujgur, and lower Iltyk formations (Fig. 2). These deposits are described in several published studies, such as Rozova, 1963, 1964, 1968, 1984; Rozova & Yadrenkina, 1967; Datsenko *et al.* 1968.

The Ust'-Brus Formation, sampled through the upper 250 m, is followed by 630 m of the Labaz Formation. They are characterised by gray, greenish-gray, and reddish clayey and silty dolomitic lime mud/wackestones with algal boundstones, lenses of intraclast flat-pebble conglomerates, local cross-bedding and ripple marks.

The Orakta Formation comprises 450 m of gray and greenish-gray clayey and silty dolomitic lime mud/wackestones with lenses of bio-grainstones, algal boundstones, intraclast flat-pebble conglomerates, local cross-bedding, ripple marks, and slump structures. Layers of breccia are developed in the lower part of the formation.

The Kulyumbe Formation is built of 645 m of gray, greenish-gray and reddish clayey and silty dolomitic and dolomitic lime mud/wackestones with gray algal boundstones, lenses of bio- and intraclast grainstones, intraclast flat-pebble conglomerates, local cross-bedding, ripple marks, and slump structures (in the lower and upper parts of the formation).

The overlying 210 m of the Ujgur Formation are represented by gray, greenish-gray and reddish clayey and silty dolomitic and dolomitic lime mud/wackestones with oolitic beds, algal boundstones and lenses of intraclast flat-pebble conglomerates. Slump structures and quartz-felspar sandstone beds are reported from the lower part of the formation.

The lower 125 m of the Iltyk Formation (Nyajan Horizon) was sampled in the uppermost part of the studied section. It is composed of gray clayey and silty dolomitic and dolomitic lime mud/wackestones, gray and rare reddish dolomitic/wackestones, as well as gray algal boundstones.

Folding and dislocations occur in the Kulyumbe section, where beds monoclinaly dip at 15–25° E–SE. They are locally intruded by sills reflecting Carboniferous–Triassic tectonic activity at the margin of the Siberian Platform (Pavlov & Gallet, 1998, 2001). These intrusions are not included in our stratigraphic column, but the positions of local marker horizons, such as the Madui and Mansi sills, are indicated by arrows (Figs 1 and 2). The Madui sill is ~20 m thick in the river outcrop and located 15–20 m above the base of the Kulyumbe Formation (within the lowermost part of the Entasian Horizon according to Rozova, 1984). The Mansi sill is ~60 m thick, and its base is situated c. 50 m above the base of the Ujgur Formation (base of the Mansian Horizon after Rozova, 1984).

3. Material and methods

Approximately 1100 hand-samples were collected in 1995 and 1998 along the Kulyumbe River from several outcrops of the Ust'-Brus, Labaz, Orakta, Kulyumbe, Ujgur, and Iltyk formations (Fig. 1) and studied for their remnant magnetization (Pavlov & Gallet, 1998, 2001, 2005). The remaining fragments of these samples have been analysed for carbon and oxygen isotopes (Table 1 of Supplementary material available online at <http://pangaea.de>). The samples were cut and the polished sections examined with a light microscope. A Dremel MiniMite micro-drill tool was used to extract rock powder from areas selected for their micritic composition. An amount of 200–400 µg was analysed from one or more spots (situated ca. 1 cm apart) from each sample. Carbon and oxygen isotopes from the carbonates of the samples were analysed with a Finnigan MAT 253 equipped with a ThermoFinnigan Gasbench II at the Department of Earth and Space Sciences, University of California, Los Angeles. The carbon and oxygen isotope composition is expressed in the conventional $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ notations relative to V-PDB.

Secondary standards used were NBS-19, IAEA-CO-1, IAEA-CO-8, and an internal laboratory standard, CARM-1.

The ThermoFinnigan Gasbench II enables a precision of the carbonate analyses that is better than $\pm 0.1\%$ for carbon and oxygen, which is comparable to dual-inlet techniques typically used for such a purpose. Powdered samples were loaded into septum-capped 12 mL glass vials (Labco exetainer) and placed into a heated block at 70°C (40 vials per run including 6 vials with the secondary and in-house standards). The vials were then successively flush-filled with helium (~6 min/vial), loaded with a few droplets of 100% orthophosphoric acid by a microliter pump, and analysed using an autosampler (~20 min/vial). The time interval between the beginning of sample digestion by orthophosphoric acid and measurements of isotope ratios in the resulting CO₂ gas was ~80 min.

Concentrations of major and trace elements (see Table 1 of Supplementary material) were measured with ICP-OES (Varian Vista Pro Ax) at the Department of Geology and Geochemistry, Stockholm University. Rock powder samples of 50 mg were dissolved in 2.5 ml ultrapure 7M HNO₃. After addition of distilled water the final volume of solution in each vial reached 50 mL. Mn/Sr and Mg/Ca ratios are used to estimate meteoric alteration and dolomite components of carbonates, respectively. These proxies were analysed from the Kulyumbe, Ujgur, and Iltyk formations, where dolostones abound and alternate with limestones.

Geochemical data from 70 additional samples collected in 1994 along the same Kulyumbe River section are also presented here (Table 2 of Supplementary material available online at <http://pangaea.de>). These samples were collected by Stefan Ebner and analysed for C, O and Sr isotopes as well as trace element concentrations at the Ruhr-

Universität Bochum between 1995 and 2001 (hereafter referred to as the Ottawa–Bochum dataset).

For Ca, Mg, Mn, Fe and Sr contents of the 1994 samples, 5–10 mg of powdered sample was dissolved in 10 mL of 2% HCl and analysed using a Varian AA 300 Atomic Absorption Spectrometer. The same powders were used for isotopic analyses. For stable isotopes, 1–5 mg of sample was reacted offline with 100% orthophosphoric acid at 50°C and analysed using a Finnigan 251 mass spectrometer. Analytical reproducibility for both $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ was within 0.1‰.

Sr isotope analyses were carried out on acetic acid leachates after an initial preleach with 0.01N HCl, which removed up to 10% of the sample. Leaching experiments were carried out to test the efficacy of this technique (see below). Standard techniques were used to concentrate Sr for analysis (Ebneht *et al.*, 2001; Shields *et al.*, 2003), and measurements were done on a Finnigan 262 thermal ionisation mass spectrometer under the supervision of D. Buhl. International standard NBS SRM 987 yielded 0.710231, which represents the mean value over 4.5 years and 550 measurements with a standard deviation (1 SD) of 38×10^{-6} and a standard error of 17×10^{-6} .

Five bulk limestone powders (K15, K17, K19, K20, and K21) were selected arbitrarily to test the efficacy of the strong acid preleach (Table 3 of Supplementary material available online at <http://pangaea.de>). They were all prepared in identical fashion. Initially, they were washed in a dilute solution of HCl (0.01 N), the supernate being retained for analysis. Thereafter, the residue was dissolved in 1 M acetic acid. The same powders were also prepared without any preleach using 1 M acetic acid and 2.5 N HCl in separate experiments. The preleach was more radiogenic than the subsequent acetic acid leach in all five cases, with the difference ranging from 9×10^{-6} to 86×10^{-6} . In

two out of five instances, the preleach was also significantly more radiogenic than the 2.5 N HCl bulk carbonate leach. In every case, the preleached 1 M acetic acid procedure yielded a less radiogenic ratio than the non-leached acetic acid procedure; however, the difference was only statistically significant in one case out of five, Ku 19 (39×10^{-6}).

4. Results of the geochemical analyses

The Kulyumbe profile exhibits $\delta^{13}\text{C}$ values which plateau around zero through the upper Ust'-Brus and Labaz formations, followed by a decreasing trend that includes minor oscillations with a peak-to-peak amplitude of 1–2‰ in the Orakta and lowermost Kulyumbe formations (Fig. 2). In the Yurakhian Horizon of the Kulyumbe Formation the $\delta^{13}\text{C}$ values rise as high as +4.6‰ and then drop, showing the characteristic features of the worldwide SPICE excursion (Fig. 2). The excursion in the Kulyumbe section is defined by >70 samples and extends through c. 200 m of carbonate rock. Previously, the excursion was recognised in the same section on the basis of five samples only (Stefan Ebner & Jàn Veizer, unpublished; and Table 2 of Supplementary material herein). A smaller positive $\delta^{13}\text{C}$ peak with amplitude of ca. 1.5‰ is observed immediately above the SPICE excursion, in the lower Khetian Horizon of the Kulyumbe Formation ('post-SPICE' in Fig. 2). The $\delta^{13}\text{C}$ values then decrease in an oscillatory fashion towards the lower boundary of the Ujgur Formation. At the boundary between the Kulyumbe and Ujgur formations, $\delta^{13}\text{C}$ values reach a minimum of ca. -1‰, before rising in the Ujgur Formation. They peak at ca. 1‰ in the upper Mansian Horizon and drop to ca. 0‰ in the upper Loparian Horizon (Fig. 2). In the uppermost Loparian and in the Nyajan Horizon of the Iltyk Formation the values oscillate near 0‰ and drop to ca. -1‰ in the upper Nyajan Horizon.

Trace element and oxygen isotope characteristics may help to assess the state of preservation of carbonate $\delta^{13}\text{C}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ values. Most of the samples show $\delta^{18}\text{O}$ values between -6‰ and -11‰. However, carbonates of the upper Entsian Horizon of the Kulyumbe Formation as well as several samples from other parts of the section yielded anomalously low $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values. In sedimentary carbonates, very low $\delta^{18}\text{O}$ values can result from a diagenetic resetting of $\delta^{18}\text{O}$ (Kaufman & Knoll, 1995). Although $\delta^{13}\text{C}$ is much less prone to diagenesis than $\delta^{18}\text{O}$, anomalously low oxygen isotope ratios are commonly associated with alteration of the carbon isotopic record as well. Therefore, outliers with $\delta^{18}\text{O} < -11‰$ are considered herein to be probably altered and are shown as void circles ($\delta^{13}\text{C}$ curve) and rectangles ($\delta^{18}\text{O}$ curve) in Fig. 2. All samples analysed show $\text{Mn}/\text{Sr} < 10$, except for sample K473 ($\text{Mn}/\text{Sr} = 12$) from the Yurakhian Horizon (Fig. 3). Although this sample might be excluded as probably altered (e.g. Kaufman & Knoll, 1995), its $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of respectively +3.16‰ and -7.15‰ are not anomalous.

Carbon isotope values from the earlier Ottawa–Bochum dataset (Table 2 of Supplementary material) are consistent with the higher resolution dataset herein, and with published data from Siberia (Brasier & Sukhov, 1998; Derry *et al.* 1994). Oxygen isotope values, however, are more internally consistent in the Ottawa–Bochum dataset, showing a clear trend towards more ^{18}O -depleted values upsection (Fig. 2; Tables 1 & 2 of Supplementary material). This difference would appear to reflect mineralogy, in that the smaller Ottawa–Bochum study used selected calcitic samples only, while the more variable $\delta^{18}\text{O}$ values of the upper Kulyumbe section studied herein seem likely to reflect variable degrees of dolomitisation in that part of the high-resolution profile (Fig. 2). This

interpretation is supported by the fact that Mg/Ca, Mn/Sr ratios and $\delta^{18}\text{O}$ (Fig. 3) show positive correlation (positive outliers in $\delta^{18}\text{O}$ are all partially dolomitised).

Sr isotope ratios range from 0.708732 to 0.709171 through the interval of the Middle Cambrian Mayan Stage to the Cambrian–Ordovician boundary (Table 2 of Supplementary material). In general, ratios rise through the Middle to Upper Cambrian. The generally negative correlation in older samples between Mn/Sr (and Mg/Ca, in Atdabanian samples only) and $^{87}\text{Sr}/^{86}\text{Sr}$ indicates that uppermost values, unusually, may best approximate contemporaneous seawater $^{87}\text{Sr}/^{86}\text{Sr}$ through the lower half of the section. By contrast, the positive correlation between Mn/Sr, Mg/Ca and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios as well as the generally high consistency of isotope values through the SPICE interval indicate a well-preserved $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.709095 for that level. Data scatter is reduced when only the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of samples below suitable diagenetic cut-off points are considered. If only samples with Mn/Sr < 0.5 and Mg/Ca < 0.04 are included, as being considered to be least altered (Fig. 2), then $^{87}\text{Sr}/^{86}\text{Sr}$ changed from 0.70885-0.70900 in the Middle Cambrian Labaz Formation to 0.70903-0.70905 in the Orakta Formation, and then to higher values of 0.70909-0.70911 in the Upper Cambrian Kulyumbe Formation.

5. The Middle-Upper Cambrian boundary in the Kulyumbe section

Traditionally, the Middle-Upper Cambrian boundary on the Siberian Platform has been placed at the top of the *Lejopyge laevigata*-*Aldanaspis truncata* Biozone of the Mayan Stage (Rozanov *et al.* 1992, Geyer & Shergold, 2000; Fig. 2). In the Kulyumbe section this level is usually traced to the boundary between the Labaz and Orakta formations (Rozova, 1963, 1964, 1968, 1984) and coincides with the base of the *Pedinocephalina*-*Toxotis* Biozone (Krasnov *et al.* 1983; Fig. 2). The ratification by the International Union

of Geological Sciences of the Middle–Upper Cambrian boundary between the *Glyptagnostus stolidotus* and *G. reticulatus* Biozones in the Paibi section on the South China Platform (Peng *et al.* 2004; Fig. 4) requires changes in position of this boundary on the Siberian Platform accordingly.

Khos-Nelege, situated on the northeastern Siberian Platform (Fig. 1), is the reference section through the Middle–Upper Cambrian boundary in Siberia (Lazarenko & Pegel', 2001). In this section, the boundary is now placed at the base of the *Glyptagnostus reticulatus*–*Eugonocare (Olenaspella) evansi* Biozone (Lazarenko & Pegel, 2001; Fig. 5). The Khos-Nelege section is associated with deeper-water **basin/slope facies (Fig. 1)** and cosmopolitan trilobites (for the purposes of this paper, the term 'trilobites' includes also agnostids). By contrast, the Middle–Upper Cambrian transitional strata of the Kulyumbe section were deposited on the carbonate platform, **in the inner shelf environments (Fig. 1)**, where almost all the trilobites are endemic (Rozova, 1963, 1964, 1968, 1984; Rozanov *et al.* 1992; Sukhov, 1997; Varlamov, Pak & Rozova, 2006). Therefore, biostratigraphic correlation between these two sections is difficult, and alternative methods, such as chemostratigraphy, are needed.

No major $\delta^{13}\text{C}$ oscillations are revealed from the upper Ust'-Brus, Labaz, and Orakta formations, correlated herein with the upper part of the Mayan Stage (Fig. 2). This is in agreement with data from Brasier & Sukhov (1998) showing a plateau during this interval from the Siberian Platform and with the values oscillating near zero in Laurentia reported by Saltzman (2005) and China (Zhu *et al.* 2004; Peng *et al.* 2006). Mild oscillations in the Ust'-Brus and Labaz formations (Fig. 2 herein) are similar to those shown by Peng *et al.* (2006, figs 6 & 7) from the *Lejopyge laevigata* Zone and upper Drumian Stage of China. The upper Stage 4 – lowermost Drumian Stage interval in

China (Zhu *et al.* 2004, figs 2-3, 6; Peng *et al.* 2006, fig. 7; Zhu, Babcock & Peng, 2006), as well as the traditional Middle Cambrian of Laurentia (Montañez *et al.* 2000) show pronounced oscillations, probably present in the lower Mayan and Amgan Stages of the Middle Cambrian of Siberian Platform (Brasier & Sukhov, 1998; A. Kouchinsky, unpublished observations). According to biostratigraphic correlation (Geyer & Shergold, 2000; Babcock *et al.* 2005; Peng *et al.* 2006) these excursions belong, however, to an interval below the profile sampled at Kulyumbe.

In addition to Siberia, SPICE excursion has been documented from Laurentia (Saltzman, Runnegar & Lohmann, 1998; Saltzman *et al.* 1995, 2000, 2004; Glumac & Walker, 1998; Buggisch (in press)), Australia (Saltzman *et al.* 2000), Kazakhstan (Saltzman *et al.* 2000), China (Saltzman *et al.* 2000; Peng *et al.* 2004; Zhu *et al.* 2004), Antarctica (Buggisch, 2006), and Scandinavia (Ahlberg *et al.* 2006) (Fig. 4). There is some uncertainty, however, in the position of the basal *G. reticulatus* Biozone with respect to the rising trend of the SPICE excursion (Saltzman *et al.* 2000). Although the Middle–Upper Cambrian boundary is considered to correspond to the onset of the SPICE excursion, it is situated almost in the middle of the rising trend in its stratotype at Paibi (see Peng *et al.* 2004, fig. 6; Fig. 4 herein). In Kazakhstan, Australia and Laurentia, the *G. stolidotus*–*G. reticulatus* boundary is associated with carbon isotope values of ca. 1‰ at the beginning of the rising trend (Saltzman *et al.* 2000 and Fig. 4 herein).

The two biozones constraining the Middle–Upper Cambrian boundary are now regarded as phylozones, because *G. stolidotus* is believed to be ancestral to *G. reticulatus* (Peng *et al.* 2004). Non-evolutionary changes in trilobite complexes, such as migrations, have been suggested, however, to be common especially at the beginning of the *G. reticulatus* Biozone (Rozanov *et al.* 1992). Palmer (1965, 1979) also noted that

diachronous boundaries of biostratigraphic units occurred in the Middle and Upper Cambrian, but he later suggested that the difference was not significant for biostratigraphic correlation (Palmer, 1984).

A combined carbon isotopic curve through the Middle–Upper Cambrian strata at Paibi and Wa’ergang sections in South China is provided by Saltzman *et al.* (2000) (Fig. 4 herein; a vertical arrow in this combined profile indicates a junction between $\delta^{13}\text{C}$ profiles of the Paibi section below and the Wa’ergang section above). Sampling intervals are rather large (several or more than ten meters), but a more detailed profile comprising 50 samples through the transitional 12 m at the *G. stolidotus*–*G. reticulatus* boundary in the Paibi section has been published (Peng *et al.* 2004). The combined data from South China suggest that the Middle–Upper Cambrian boundary in the stratotype is situated at the beginning of the rising trend of the SPICE excursion. Carbon isotopic values through the transitional strata in Paibi oscillate between 1‰ and 2.5‰, with the boundary lying at 1.5–2.0‰ (Peng *et al.* 2004, fig. 6) and at ca. 3‰ in Wa’ergang (Saltzman *et al.* 2000). Values of ca. 3‰ at the level of first appearance of *G. reticulatus* are also reported from the Wangcun section in South China (Zhu *et al.* 2004).

Based on the position of the SPICE excursion in the Kulyumbe section, the Middle–Upper Cambrian boundary is situated between the base of the Yurakhian Horizon of the Kulyumbe Formation, where carbon isotope values begin to rise, and a level showing values around ca. 2‰ at the top of section 6 (Figs 1 & 2). The position of the Middle–Upper Cambrian boundary according to the local trilobite biostratigraphy, between the Labaz and Orakta formations, is thus at least 700 m lower than that indicated by chemostratigraphy. Therefore, biostratigraphic correlation of the basal Orakta Formation with the *G. reticulatus* Biozone at Khos-Nelege is unlikely correct.

A worldwide mass extinction of trilobites at the base of the *Glyptagnostus reticulatus* Biozone marks the beginning of the SPICE excursion (Saltzman *et al.* 2000). Eighty species from the *Glyptagnostus stolidotus* zone do not proceed into the *G. reticulatus* Biozone in Laurentia. The diversity of trilobites increases later, during the rising trend of SPICE. In the Kulyumbe section the diversity of trilobites declines from the Labaz Formation (upper Middle Cambrian) towards the Cambrian-Ordovician boundary, with the most dramatic decline in number of species taking place in the upper Orakta Formation (Fig. 2; Table 4 of Supplementary material available online at <http://pangaea.de>). Trilobite complexes show significant renewal through each boundary between biozones and horizons in the section, including the Middle–Upper Cambrian boundary transition identified herein by chemostratigraphy at the base of the Yurakhian Horizon. The trilobite fauna of the Yurakhian Horizon is completely renewed with respect to that of the Entsian Horizon (Fig. 2), but in contrast with data from Saltzman *et al.* (2000), there is no evidence for increase in diversity during the rising trend of the SPICE excursion in the Kulyumbe section. Although new species and genera of trilobites continue to appear throughout the Kulyumbe Formation (Datsenko *et al.* 1968; Rozova, 1968, 1984), the diversity generally declines (Fig. 2; Table 4 of Supplementary material). The trilobites from the Kulyumbe section are endemics (Rozova, 1963, 1964, 1968, 1984; Rozanov *et al.* 1992; Sukhov, 1997; Varlamov, Pak & Rozova, 2006), and therefore their evolutionary pattern and diversity changes differ from those of forms with global distribution. Hence, the observed decline in diversity at Kulyumbe could be related to local palaeogeography and environments.

6. The Cambrian-Ordovician boundary in the Kulyumbe section

There are two distinct proposals for how to correlate the Kulyumbe section with the base of the Ordovician System. The first one places the Cambrian–Ordovician boundary on the Siberian Platform at the base of the Mansian Horizon (Krasnov *et al.* 1983; Fig. 2). In the second one (Rozova, 1968, 1984), the boundary is positioned at the base of the Nyajan Horizon. The latter option was supported by the Cambrian–Ordovician boundary International Working Group in 1986 (Fig. 2).

A lower boundary of the Ordovician System was finally ratified by the Cambrian–Ordovician boundary International Working Group at the base of the Tremadocian Stage at Green Point in western Newfoundland, where it was defined by the first appearance of the conodont *Iapetognathus fluctivagus* (Cooper & Nowlan, 1999). Thus, the occurrence of *Cordylodus proavus* can now be used to assign the Loparian and Mansian horizons to the Upper Cambrian (Dubinina, 2000; Tesakov *et al.* 2003). **The Ordovician *C. angulatus* was found in the Nyajan Horizon (Tesakov *et al.* 2003)**, but taxonomic revision of Siberian *Cordylodus* species is still needed (Tatiana Tolmacheva, pers. comm. 2006).

Although the carbon isotopic record from the Cambrian–Ordovician boundary stratotype at Green Point seems to be distorted by diagenesis (Cooper, Nowlan & Williams, 2001), a combination of data from other sections, such as Dayangcha in China (Ripperdan *et al.* 1993), Black Mountain in Australia (Ripperdan *et al.* 1992; **Miller *et al.* 2006**), and Lawson Cove in Utah (Ripperdan & Miller, 1995; **Miller *et al.* 2006**) enabled a generalized (global) curve to be compiled (Cooper, Nowlan & Williams, 2001, fig. 4; Fig. 4 herein). The Cambrian-Ordovician boundary is marked by the highest $\delta^{13}\text{C}$ value of the transitional interval, in the *Iapetognathus* Biozone, before the value decreases

through the *C. angulatus* Biozone (Cooper, Nowlan & Williams, 2001). There are also two main positive cycles below this excursion, between the base of the *Cordylodus proavus* and the base of the *C. lindstromi* Biozone (Ripperdan & Miller, 1995; Cooper, Nowlan & Williams, 2001, fig. 4; **Miller *et al.* 2006**).

The Lower Ordovician isotopic record is still in its inception, and conclusions on the exact number and position of excursions are premature. A Lower–Middle Ordovician carbon isotopic curve of high resolution and biostratigraphically constrained by Laurentian conodont biozones was published by Buggisch, Keller & Lehnert (2003) from an isolated terrane of a probable Laurentian provenance in Argentina. However, there are no species of *Iapetognatus* and *Cordylodus* known from that section, probably because of unfavourable facies. The SPICE excursion is also missing from that profile, because of a stratigraphic hiatus. The lower and upper boundaries of the Tremadocian Stage are marked by falling values from local maxima at the Cambrian–Ordovician boundary and in the upper Tremadocian (see Buggisch, Keller & Lehnert, 2003, fig. 6). A single positive excursion of 2‰ amplitude (from -1‰ to 1‰) in the *R. manitouensis* Biozone, is unknown beyond Argentina owing to lack of data from this stratigraphic interval (Buggisch, Keller & Lehnert, 2003). Otherwise, the Upper Cambrian–Tremadocian portion of this curve can be satisfactorily correlated with other transitional profiles (Buggisch, Keller & Lehnert, 2003; Fig. 4 herein).

There are apparently three positive excursions and four troughs above SPICE in the Kulyumbe section (Fig. 2). The lower maximum is situated in the lower Khetian Horizon of the Kulyumbe Formation (‘post-SPICE’ excursion in Fig. 2). It is similar in position and magnitude to a reported positive $\delta^{13}\text{C}$ feature with an amplitude up to 2‰ observed immediately above the SPICE excursion in Laurentia, above the last occurrence

of *Irvingella* (Saltzman, 2001), and above the *Ivshinagnostus ivshini* Biozone in Kazakhstan (Saltzman *et al.* 2000; Fig. 4). The subsequent decrease in values coupled with the SPICE excursion is recognized as the HERB event, a Sunwaptan-aged negative $\delta^{13}\text{C}$ anomaly identified in Australia and Laurentia (Ripperdan *et al.* 1992; Ripperdan, 2002; **Miller *et al.* 2006**). It is represented by minimum values of -3‰ to -4‰ in Australia (Ripperdan *et al.* 1992; **Miller *et al.* 2006**; Fig. 4) or a less pronounced trough in Laurentia (Buggisch, Keller & Lehnert, 2003; Saltzman *et al.* 2004; **Miller *et al.* 2006**; Fig. 4). Buggisch, Keller & Lehnert (2003) suggested, however, that negative anomalies, such as HERB, could have been produced or enhanced by diagenesis at sequence boundaries rather than representing the true magnitudes of primary oceanic carbon reservoir changes. Although more pronounced, the HERB minimum can be tentatively correlated to an isotopic minimum of -1‰ at the boundary between the Kulyumbe and Ujgur formations (HERB? in Figs 2 & 4). The location of these features in the Khetian Horizon is consistent with its terminal Upper Cambrian affinity.

At least two more peaks appear in the Kulyumbe section, within the Ujgur and Iltyk formations. Through a combination of chemostratigraphy and biostratigraphy, a lowermost possible and a most probable position of the Cambrian–Ordovician boundary in Kulyumbe may be indicated. The upper portion of the Iltyk Formation is referred to as the Ugorian Horizon (not shown here) and tentatively attributed to the ‘Arenig Stage’ of the Lower Ordovician (Rožanov *et al.* 1992). According to biostratigraphy, the lower Tremadocian boundary is situated below the Ugorian Horizon. On the other hand, based on the available chemostratigraphic information, the Cambrian–Ordovician boundary is unlikely to be situated at the base of the Mansian Horizon because of a well-defined isotopic trough there (HERB? in Figs 2 & 4). Two intervals in the Kulyumbe profile may

therefore be suggested for potential placement of the Cambrian–Ordovician boundary: the upper Mansian and the upper Loparian – middle Nyajan horizons. The upper Mansian isotopic maximum represents the first local maximum after the post-SPICE and HERB? excursions, and its position is therefore regarded to be too low for the Cambrian–Ordovician boundary (Figs 2 & 4). A more probable location for this boundary lies within the upper Loparian – middle Nyajan interval, but the positive excursion there may be composite, since values oscillate throughout these beds, and the isotopic signature may be insufficiently resolved owing to larger sampling intervals (Figs 2 & 4). Nevertheless, a fall in carbon isotope values in the upper Nyajan Horizon and its biostratigraphic correlation with the lowermost Ordovician (Dubinina, 2000; Tesakov *et al.* 2003) is consistent with our proposition that the most probable position of the Cambrian–Ordovician boundary at Kulyumbe is in the Nyajan Horizon, possibly in its middle part, in outcrop 7322 (Figs 1, 2 & 4).

7. Strontium isotopic interpretation

Our data imply a subtle rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ from the Middle Cambrian to the middle of the Late Cambrian. This rise might continue to all-time high values for the Phanerozoic, possibly as high as 0.70920, as recorded in slightly younger upper Pterocephaliid biomere in the USA (Saltzman *et al.* 1995). Published micrite and conodont $^{87}\text{Sr}/^{86}\text{Sr}$ data show a subsequent decrease across the Cambrian–Ordovician boundary worldwide, from 0.7091 to 0.7090 (Saltzman *et al.* 1995; Ebner *et al.* 2001). Strontium isotope ratios were as high during the Middle–Late Cambrian as they are today, and possibly even higher (Saltzman *et al.* 1995; Montañez *et al.* 2000), which indicates a unique importance during that time of chemical weathering on ocean

composition relative to that of submarine hydrothermal exchange with juvenile ocean crust (Ebner *et al.* 2001). The unique situation during the late Cambrian is underscored by the fact that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of carbonate rocks undergoing weathering and that of the depleted mantle would have been considerably lower than today, thus pushing seawater $^{87}\text{Sr}/^{86}\text{Sr}$ to lower, rather than higher values. The high $^{87}\text{Sr}/^{86}\text{Sr}$ of the late Cambrian ocean has been attributed to elevated chemical weathering rates of crustal silicate minerals uplifted during the Pan-African (Montañez *et al.* 2000) and Transgondwanan orogenies (Squire *et al.* 2006).

8. Paleomagnetism

The detailed magnetostratigraphic log of the Kulyumbe section presented herein combines published magnetostratigraphic columns for the Middle Cambrian–Lower Ordovician interval (Pavlov & Gallet, 1998, 2001), corrected, refined and complemented with additional data from the Orakta Formation (Table 1 of Supplementary material available online at <http://pangaea.de>).

Additional sampling in 1998 from the lower part of the Entsian horizon revealed three intervals of normal polarity instead of the two reported by Pavlov & Gallet (1998) (see Pavlov & Gallet, 2005, fig. 4; Fig. 2 herein). There are also two new very narrow reversed-polarity zones confirmed only by single samples from the lower Loparian Horizon (sample K294) and the middle Labaz Formation (sample 410), as well as several narrow polarity intervals in the middle Labaz Formation, mostly based on single samples (among samples 531-846) (Fig. 2, new and changed polarity intervals indicated by arrows; see also Table 1 of Supplementary material). Most of the sampling gaps larger

than 10 m are shown in the new version by hiatuses in the magnetostratigraphic column (Fig. 2).

Data from the Orakta Formation were obtained from samples collected in 1998 and yielded a very weak and noisy paleomagnetic signal. Isolation of characteristic paleomagnetic components from most of these samples was either impossible or very difficult. Nevertheless, some samples show a paleomagnetic two-component record (Fig. 6a) suggesting a direction of the characteristic magnetization (Fig. 6c). Some samples show three components of magnetization: less stable, recent (low-temperature) and trap-related (middle-temperature) ones, and a more stable, characteristic (high-temperature) component. The mean paleomagnetic direction of the isolated characteristic component is close to that which is well defined from the Labaz and Kulyumbe formations. Some samples from the Orakta Formation do not allow determination of the direction of the characteristic component (Fig. 6b), but their behavior during analyses indicates the polarity of the ancient magnetization (marked 'great circle' in Table 1 of Supplementary material). Given the overall low quality of the paleomagnetic record in the Orakta Formation, a corresponding polarity sequence needs to be confirmed by data from other sections and should therefore be considered as preliminary (indicated as 'low-quality signal' in Fig. 2).

Thus, a succession of 19–20 magnetic intervals from the Orakta and lower Kulyumbe formations can be added herein to 54–80 intervals previously reported by Pavlov & Gallet (2005) from the Middle Cambrian of Siberia. This number is probably again a minimum estimate, as many magnetic intervals have a small stratigraphic thickness of less than 1 m and are based on single samples. Our combined results confirm observations (Pavlov & Gallet, 2001; Gallet, Pavlov & Courtillot, 2003) that during the

Middle Cambrian the geomagnetic reversal frequency was as high as 5–10 reversals per Ma, assuming a total duration of ~10 Ma and up to 100 magnetic intervals in the Middle Cambrian. By contrast, the sequence attributed herein to the Upper Cambrian on the grounds of chemostratigraphic correlation contains only 10–11 magnetic intervals, whereas there are 2–3 intervals in the lowermost Ordovician (lower Tremadocian) part (Fig. 2). Hence, magnetostratigraphic data from the Upper Cambrian strata of the Kulyumbe section obtained by Pavlov & Gallet (1998) and adjusted herein show a one-order-of-magnitude lower number of magnetic reversals than in the Middle Cambrian.

Upper Cambrian–Lower Ordovician magnetostratigraphic results from several sections, such as the Black Mountain in Australia (Ripperdan *et al.* 1992), Dayangcha (Ripperdan, Magaritz & Kirschvink, 1993) and Tangchan (Yang *et al.* 2002) in China, Green Point in western Newfoundland, Bartyrbay in Kazakhstan, and from Texas (Ripperdan & Kirschvink, 1992) do not, however, provide a standardized magnetostratigraphic scale through the Cambrian–Ordovician interval (Cooper, Nowlan & Williams, 2000). A tentative correlation of magnetostratigraphic zones of the Upper Cambrian–Lower Ordovician of China and Australia with the Kulyumbe section was provided by Pavlov & Gallet (2005). It has been also reported that in general the geomagnetic field had a predominantly reversed polarity through the Cambrian–Ordovician boundary interval, but with episodes of normal polarity in the late *Proconodontus*, late *Eoconodontus* Biozones of the Upper Cambrian, and in the Early Ordovician (Cooper, Nowlan & Williams, 2000). As for the Middle Cambrian and most of the Upper Cambrian, paleomagnetic zonation is known only from the Siberian Platform (Pavlov & Gallet, 1998, 2001, 2005 and herein).

9. Conclusions

The global SPICE excursion with amplitude of ca. +5‰ is found in the Yurakhian Horizon, in the middle part of the Kulumbe Formation (Fig. 2). It terminates an interval with low-amplitude carbon isotopic oscillations in the Middle Cambrian upper Mayan Stage of Siberia. According to the position of the SPICE excursion in the Kulumbe section, the Middle–Upper Cambrian boundary is placed within an interval no lower than the base of the Yurakhian Horizon of the Kulumbe Formation.

A fall of carbon isotope values in the upper Nyajan Horizon agrees with its biostratigraphic correlation with the lowermost Ordovician conodont biozones and consistent with a proposition herein that the most probable position for the Cambrian–Ordovician boundary would be in the middle portion of the Nyajan Horizon.

Our integrated isotopic study further confirms some intriguing aspects of the Late Cambrian ocean, when a major global positive $\delta^{13}\text{C}$ excursion (SPICE) coincided with all-time maximum $^{87}\text{Sr}/^{86}\text{Sr}$ values in marine carbonates. High $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in seawater during this interval relate to uniquely high rates of chemical weathering on the continent.

A continuous reversed polarity interval is observed through the SPICE event, and a zone of normal polarity near the Middle–Upper Cambrian boundary (Fig. 2). The frequency of reversals in the Upper Cambrian was about one order-of-magnitude lower than in the Middle Cambrian. The magnetic polarity sequence presented here further supports the conclusion (Gallet & Pavlov, 2001, Gallet *et al.* 2003), that the geomagnetic reversal frequency was very high during the Middle Cambrian, probably the highest in the Phanerozoic.

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Figure captions

Fig. 1. Location of the Kulyumbe section (Ku) at the northwestern margin of the
Siberian Platform, and other sections discussed. 7320 ... numbering of sampling profiles.

Turukhansk-Irkutsk-Olyokma, Anabar-Sinsk, and Yudoma-Olenyok facies regions in the upper Cambrian correspond respectively to the inner shelf, transitional (with reefs, banks and bars), and slope/basin depositional environments (Sukhov, 1997).

Fig. 2. $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ profiles, $^{87}\text{Sr}/^{86}\text{Sr}$ data, magnetostratigraphy, and changes in associations of trilobites through the upper Middle Cambrian–lowermost Ordovician strata of the Kulyumbe section. 7320 ... as in Figure 1. **Outliers with $\delta^{18}\text{O} < -11\text{‰}$ are considered herein to be probably altered and are shown as void circles ($\delta^{13}\text{C}$ curve) and rectangles ($\delta^{18}\text{O}$ curve). $^{87}\text{Sr}/^{86}\text{Sr}$ values refer to ‘best-preserved’ samples with $\text{Mn}/\text{Sr} < 0.5$ and $\text{Mg}/\text{Ca} < 0.04$ only (see text). * Rozova (1968, 1984) placed the Cambrian-Ordovician boundary at the base of Nyajan Horizon (names of horizons given by Rozova 1968, 1984).**

Fig. 3. Mg/Ca (a) and Mn/Sr (b) against $\delta^{18}\text{O}$ plots showing association of increasing dolomite content with higher $\delta^{18}\text{O}$ and elevated Mn/Sr.

Fig. 4. Global correlation of the Upper Cambrian – lowermost Ordovician part of the Kulyumbe section. **Void circles show samples probably altered by diagenesis as indicated by the authors or/and by $\delta^{18}\text{O} < -11\text{‰}$ data, where available. Occurrence of *Cordylodus proavus* (Cp) and *C. angulatus* (Ca) in the Kulyumbe section according to Tesakov *et al.* (2003).**

Fig. 5. Stratigraphy of the Cambrian part of the Ogon’or Formation at Khos-Nelege (see location KN in Fig. 1), after Krasnov *et al.* (1983) and Rozanov *et al.* (1992).

Fig. 6. a - Zijderveld diagrams, open (solid) circles – projections on vertical (horizontal) plane. b - demagnetization behavior of natural remnant magnetization which indicates the normal polarity of the characteristic component. Solid circles – projections on lower hemisphere. c – distribution of isolated directions of the characteristic component (stratigraphic system of coordinates). Solid circles – projections on lower hemisphere.

Supplementary material (available online at <http://pangaea.de>)

Table 1. $\delta^{13}\text{C}$, $\delta^{18}\text{O}$, elemental, and palaeomagnetic data from the Kulyumbe section.

Table 2. Sr-isotopic data from the Kulyumbe section and associated C- and O-isotopic and trace elemental data.

Table 3. Strontium isotope results of leaching experiments. Samples were leached for analysis using 1) 2.5 N HCl on bulk carbonate powder; 2) 1 M acetic acid on bulk carbonate powder; 3) 0.01 N HCl preleach and 4) 1 M acetic acid on remaining powder after preleach. The leachate from technique 4 was systematically the least radiogenic and presumably most pristine of all the leachates, and so was used for all the reported analyses.

Table 4. List of trilobite species from the Kulyumbe section.