Pb-Pb zircon dating of tuff horizons in the Cyrtograptus Shale (Wenlock, Silurian) of Bornholm, Denmark

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Waterlain fallout ashes are interbedded in the upper part of the Cyrtograptus Shale of Bornholm, the youngest preserved member of the Lower Palaeozoic sequence at the southern coast of the island. Graptolite faunas indicate that these tuffaceous sediments belong to the *Cyrtograptus lundgreni* Zone deposited during Late Wenlock. A $^{207}Pb/^{206}Pb$ mean age of 430 ± 1.9 Ma obtained by evaporation of idiomorphic single zircons from the tuff layers supports this observation. Geochemical studies of the pyroclastic rocks point to an explosive, calc-alkaline magmatic arc volcanism which probably occurred along or slightly south of the Tornquist-Teisseyre Lineament, and could have been induced by the collision of Avalonia with the southern margin of Baltica during the Silurian. This assumption is supported by the contemporaneous deposition of bentonites on the Swedish island of Gotland which might represent a distal facies of these fallouts. Further, the subduction-related volcanic activity is interpreted as a fingerprint for closing of the Tornquist Ocean during the Caledonian orogeny.

Key words: Bornholm, Tornquist Ocean, Wenlock, Silurian, tuff, graptolites, Pb-Pb isotopes, single zircon evaporation.

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Several bentonite layers between a few centimetres to 2 m thick of Ordovician and Silurian age occur in many places in Baltoscandia (Batchelor & Jeppsson 1994, 1999; Batchelor et al. 1995, Bergström et al. 1995, 1997a, 1999, Huff et al. 1998), the British Islands (Huff et al. 1991, Batchelor & Clarkson 1993, Batchelor 1995, 1999, Fortey et al. 1996) and the Appalachians of North America (e.g. Bergström et al. 1997b, 1998). They indicate long lasting explosive volcanic activity, and are widely used for stratigraphic correlation. The sources of the Ordovician ash beds were rhyolitic or dacitic melts erupted explosively from a magmatic arc (Bergström et al. 1995). They are mostly interpreted as fingerprints for the closing of the Iapetus Ocean between Laurentia and Baltica to form the Scandinavian Caledonides. However, especially for the pyroclastic horizons in the Lower Silurian beds of the Swedish island Gotland another origin is proposed by Batchelor & Jeppsson (1999). They suggest an area along the Tornquist-Teisseyre Zone as the most likely location for the source volcanoes. Remnants of waterlain ash fall deposits are also known from the southern coast of the Danish island Bornholm. They are inbedded in graptolite-bearing shales of Wenlock age (Bjerreskov & Jørgensen 1983). These tuffs were analysed for major and trace elements using XRF techniques. The geochemical data are compared with data of other Silurian bentonite layers in Scandinavia, and results of radiometric age determinations using evaporation of single zircons are presented and discussed in this paper.

Geological Setting

The Danish island of Bornholm lies in the Baltic Sea at the southwestern margin of the Fennoscandian Shield, which is tectonically marked by several NW–SE trending faults forming the Tornquist-Teisseyre Lineament (TTL). This dominating tectonic structure can be traced from the central North Sea towards SE-Europe. A northwestern branch named the Sorgenfrei-Tornquist Zone

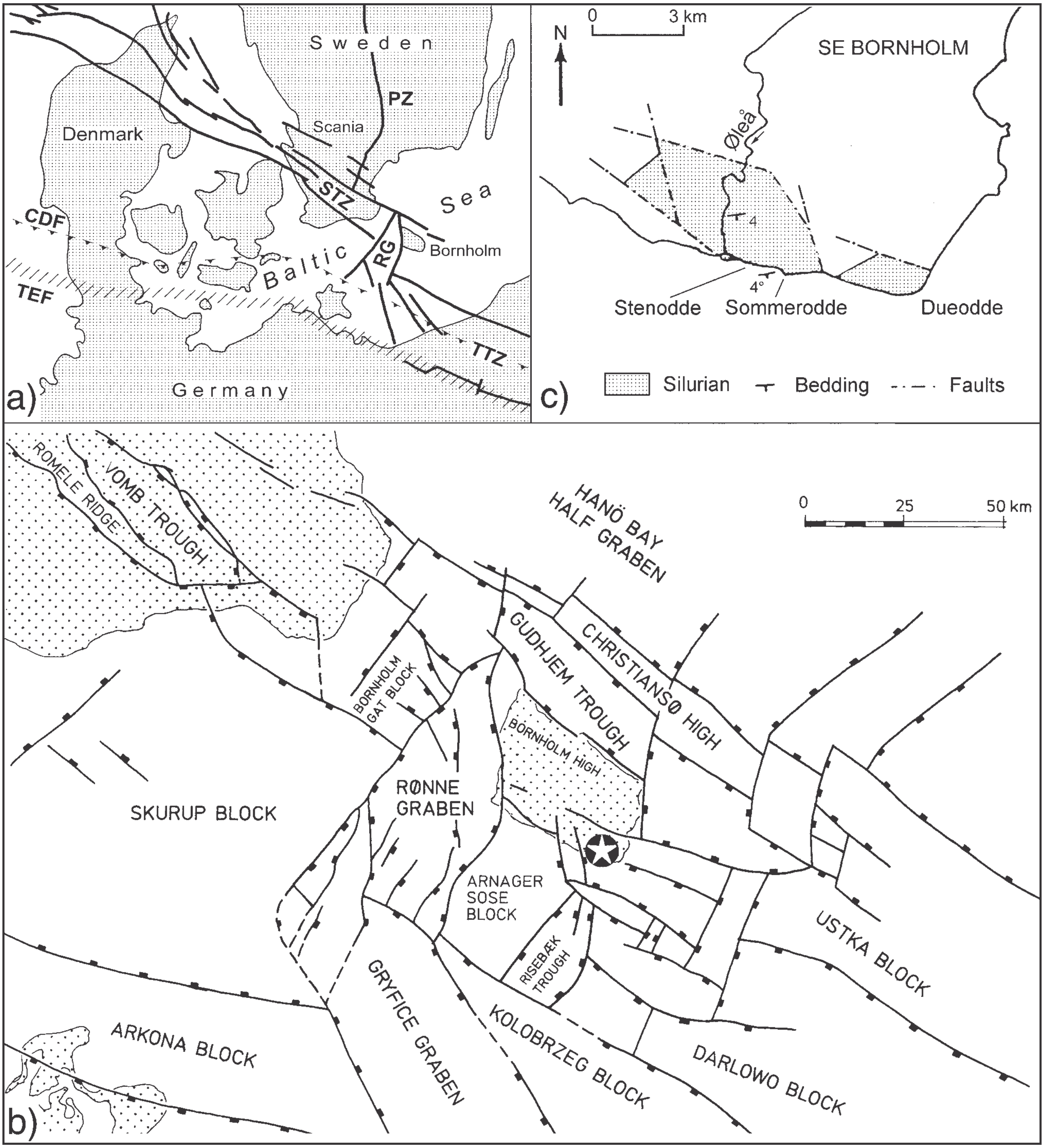

Fig. 1. (a) Simplified sketch showing the major fault pattern at the southwestern margin of the Fennoscandian Shield. CDF = Caledonian Deformation Front, PZ = Protogine Zone, RG = Rønne Graben, STZ = Sorgenfrei-Tornquist Zone, TEF = Trans-European Fault, TTZ = Tornquist-Teisseyre Zone. (b) Structural framework of Bornholm and surroundings (based on Vejbæk 1985, Norling & Bergström 1987). The asterisk marks the occurrence of Silurian rocks on the island. (c) The detail sketch shows strike and dip of the Silurian deposits of southeastern Bornholm (Bjerreskov & Jørgensen 1983, modified). The Wenlock tuff beds probably occur offshore east of Cape Stenodde.

(STZ) is separated from the southeastern Tornquist-Teisseyre Zone (TTZ) by the Rønne graben (RG) west of Bornholm (Fig. 1a). While the Tornquist-Teisseyre Zone marks the border between the Precambrian crust of the East European Craton and younger crustal parts in the SW, the Sorgenfrei-Tornquist Zone only separates the Fennoscandian Shield from the Danish Basin under which the crystalline basement of the East European Craton can be traced as far as the Trans-European Fault (TEF) or Trans-European Suture Zone (TESZ). This southwest-dipping suture was formed by the Early Palaeozoic collision of the palaeocontinent Baltica with the terrane Avalonia. During the Caledonian orogeny crust mainly of Avalonian affinity was deformed and thrusted onto the southwestern margin of Baltica. The Caledonian deformation front (CDF) outlines the recent northern rim of this thrust and fold belt (cf. Berthelsen 1998, Pharaoh 1999, Beier & Katzung 2001).

Bornholm itself is a slightly southwards-inclined horst structure bordered by NW and NNE trending faults (Fig. 1b). The crystalline basement is exposed in the northern and central parts of the island. It consists of Mesoproterozoic gneisses and granites, which are about 1.5 to 1.4 Ga old (Larsen 1980, Tschernoster et al. 1997), cut by younger dolerite dykes mostly of Precambrian age (cf. Obst 2000). Remains of the Lower Palaeozoic and Mesozoic cover sediments are only preserved in faulted blocks of the southern and southwestern parts of the island, respectively.

The Early Palaeozoic sequence on Bornholm includes more than 500 m of Cambrian to Silurian mostly marine sediments, deposited on the relatively stable Baltoscandian shelf (Poulsen 1966, Vejbæk et al. 1994). Sedimentation processes were mainly controlled by opening and closing of the Tornquist Ocean and subsequent collision of the Gondwana-derived terrane Avalonia with Baltica. Evolution of an accretionary wedge and its thrusting onto the southwestern margin of Baltica induced the formation of a foreland basin in the Late Ordovician indicated by strong subsidence (Poprawa et al. 1999). Deep-water sediments were accumulated until sedimentation exceeded subsidence rate, which led to a gradual shallowing of the basin (Beier et al. 2000).

The Silurian of Bornholm starts with a 135 m thick transgressive dark grey mudstone of Llandovery age. Sedimentation of this Rastrites Shale is continuously followed by deposition of a 25 m thick dark grey mudstone sequence of uppermost Llandovery and lowermost Wenlock age (basal Cyrtograptus Shale). While deposits of Middle Wenlock are missing on the island, a ~25 m thick grey silty mudstone (upper part of the Cyrtograptus Shale) occurs on the seafloor at the southern coast (Gravesen & Bjerreskov 1984).

Several medium-grained ashes are interbedded with these youngest preserved Silurian shales, and are documented in numerous loose, angular boulders at the southern beach near the mouth of the Øleå stream, probably eroded from off-shore rocks east of cape Stenodde (Fig. 1c). They are assigned to the *Cyrtograptus lundgreni* Zone by the conspicuous graptolite fauna in the shales, which sometimes border the pyroclastic rocks (Bjerreskov & Jørgensen 1983, Böhnke & Katzung 2001).

Wenlock ashes in the Cyrtograptus Shale

The youngest preserved member of the Palaeozoic succession is the Silurian Cyrtograptus Shale. It can be dated by graptolite faunas of Late Llandovery to Wenlock age (Bjerreskov 1982). The Cyrtograptus Shale generally consists of dark grey shales with silt-sandy and calcareous intercalations. Several tuff horizons seem to be embedded in the uppermost part of this sequence. Although the two graptolite species *Pristiograptus dubius pseudodubius* Boucek and *Monograptus flemingi* Salter occur in great numbers in the tuff horizons, they are not sufficient to determine any precise stratigraphic level. However, the shales which border the tuffs on both sides of loose boulders contain graptolites indicating that the sediments belong to the *Cyrtograptus lundgreni* Zone (Late Wenlock). The complete graptolite fauna list is given by Bjerreskov & Jørgensen (1983). Slightly older faunas are reported from the same area by Bjerreskov (1992) and from the Pernille-1 well (Bjerreskov 1993). It is not possible from the Bornholm material to define a more precise level within the *Cyrtograptus lundgreni* Zone, especially as the fauna is not from *in situ* strata and no biostratigraphic ranges can be given.

On fresh surfaces the tuffs are of light grey colour, but they often show a change to yellow due to intense weathering. The thickness of the individual pyroclastic layers varies and ranges from several mm to about half a metre. The volcanic rocks are described as fine- to medium-grained primary crystal-rich vitric tuffs, which have suffered from sea-floor weathering and subsequent diagenesis as indicated by extensive replacement and authigenic mineral formation (Bjerreskov & Jørgensen 1983). Phenocrysts are feldspar (sanidine and plagioclase), quartz, biotite, apatite, zircon, Ti-Fe oxides. Secondary alteration of feldspar into kaolinite and calcite is common. The biotite flakes are also often completely chloritized. Bjerreskov & Jørgensen (1983) distinguish between clast-bearing type and

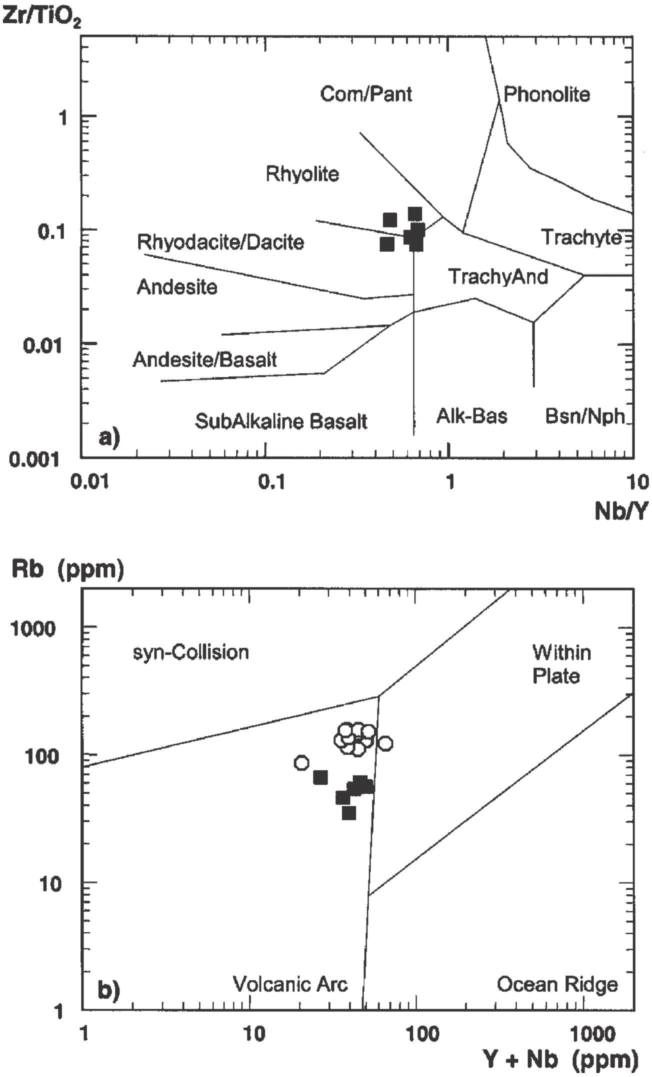

Fig. 2. (a) Nb/Y-Zr/TiO_2 diagram (Winchester & Floyd 1977) for the Silurian tuff samples of Bornholm which can mainly be classified as rhyodacitic and rhyolitic rocks. (b) (Y+Nb)–Rb diagram (Pearce et al. 1984) for discrimination of the tectonic setting of intermediate to acid rocks. Nearly all tephra samples from Bornholm (squares) and Gotland (circles; data from Batchelor & Jeppsson 1999) plot in the field for volcanic arc rocks.

laminated type tephras. The laminated texture of the most tuff samples and the occurrence of vesicular glass shards suggest that they were erupted under subaerial conditions (phreatomagmatic eruptions). Generally, the pyroclastic horizons are interpreted as waterlain airfall ashes, partly redeposited by debris flows (Bjerreskov & Jørgensen 1983; Böhnke & Katzung 2001). Most of the tephra layers are silica cemented and contain framboidal pyrite, often concentrated in discrete bands, and formed during early weathering and cementation processes.

Geochemistry of the pyroclastic rocks

Major and trace element compositions of 6 tuff samples collected on the beach between Cape Stenodde and Cape Sommerodde were determined by using XRF spectrometry on glass beads (Table 1). Sample preparation was done using a WC jaw-crusher and an agate mill in Greifswald. The analyses were performed at the Geochemical Laboratory of the University of Göttingen. Using the Nb/Y-Zr/TiO_2 diagram of Winchester & Floyd (1977) for classification of altered volcanic rocks, the tuff samples plot mainly in the fields for rhyodacitic and rhyolitic rocks (Fig. 2a). Volcanic rocks of similar composition (acid andesites) often occur in areas of volcanic arc magmatism at converging plates (cf. Gill 1981). This scenario is supported by use of the (Y+Nb)–Rb diagram (Pearce et al. 1984) established for determination of the tectonic setting of intermediate to acid rocks (Fig. 2b). All samples plot in the field for volcanic arc rocks. Low Nb/Ce ratios (≤0.20) indicate a negative Nb anomaly, also suggestive of arc magmatism. The contemporaneous, but distal Wenlock bentonites from Gotland are also characterized as calc-alkaline dacites and rhyolites (Batchelor & Jeppsson 1999). These very fine-grained ashes might be co-genetic to the coarser-grained tephras of Bornholm but they have much higher K_2O contents (about 4%). They show very similar Y+Nb values, but

Table 1. Major and trace element geochemistry of Silurian tuff samples from SE Bornholm. *Fe is analysed as total Fe_2O_3. #Sample BH 185b is enriched on pyrite framboides.

	BH 184	BH 185a	#BH 185b	BH 189	BH 206	BH 221
SiO_2 (wt%)	66.4	55.7	47.7	48.5	57.4	62.7
TiO_2	0.42	0.60	0.59	0.38	0.39	0.50
Al_2O_3	12.76	16.82	14.55	13.23	13.10	15.78
Fe_2O_3*	2.72	2.72	8.08	2.35	2.86	3.75
MnO	0.12	0.17	0.22	0.48	0.27	0.08
MgO	2.14	2.59	2.21	2.09	2.16	2.83
CaO	4.52	6.73	7.72	14.10	8.79	3.24
Na_2O	2.56	3.23	2.93	3.24	2.36	2.45
K_2O	1.20	1.65	1.33	0.88	1.35	1.75
P_2O_5	0.12	0.17	0.14	0.14	0.13	0.14
LOI	7.36	9.83	13.02	14.49	10.91	7.13
Total	100.33	100.21	98.50	99.87	99.72	100.36
Ba (ppm)	184	210	212	125	208	188
Ce	87	90	87	100	105	75
Co	5	8	14	2	7	6
Ga	11	12	9	9	10	14
Nb	13	17	17	11	12	10
Ni	10	11	17	7	9	14
Rb	43	55	48	30	48	62
Sc	7	10	13	9	7	7
Sr	84	110	106	140	115	71
V	41	57	57	36	36	51
Y	21	25	26	23	26	15
Zn	20	21	26	20	22	27
Zr	366	607	816	458	296	372

their Nb/Ce ratios are slightly higher. Further determination of REE, and isotopic studies are necessary to testify a common source of both occurrences.

Results of radiometric age determinations

Pb-Pb isotope ratios of the Wenlock ashes from southern Bornholm were determined by evaporation of single zircons to compare absolute ages to those obtained by biostratigraphically well constrained graptolite faunas. The major population are euhedral, long prismatic zircons in the tuff horizons (Fig. 3). Three optically clear crystals without inclusions were separated to determine the $^{207}Pb/^{206}Pb$ isotope ratios on a MAT 262 mass-spectrometer equipped with an ion counting system (Philips 6665) using evaporation techniques (Kober 1987). These ratios measured at the Geochemical laboratory at the University of Freiberg were ^{204}Pb-corrected by means of a program based on the two-stage model from Stacey & Kramers (1975). The obtained isotope ages of the zircons (quoted with 2σ-mean errors) are very similar: 430.7 ± 1.9 Ma, 430.2 ± 5.1 Ma and 428.9 ± 3.1 Ma (Fig. 4). Thus, the mean age of the tuff eruption is 430 ± 1.9 Ma.

Discussion of Silurian time scales

Despite some precise isotope datings improving the Pre-Carboniferous chronology (Roberts et al. 1995, Tucker & McKerrow 1995), the Early Palaeozoic time scale is still uncertain and vague if compared with the more precise data available from the Mesozoic and Cenozoic. Latest summaries are given by Odin (1994) and Gradstein & Ogg (1996) in general and by Young & Laurie (1996) for Australia.

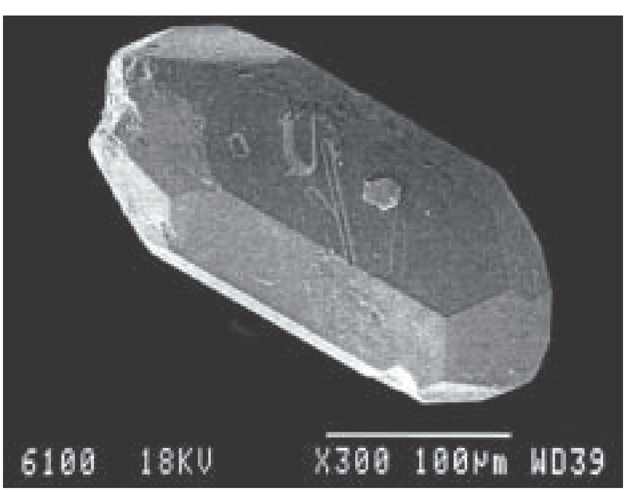

Fig. 3. SEM-photograph of a typical euhedral zircon of the Silurian tuffs with apatite crystals on the surface.

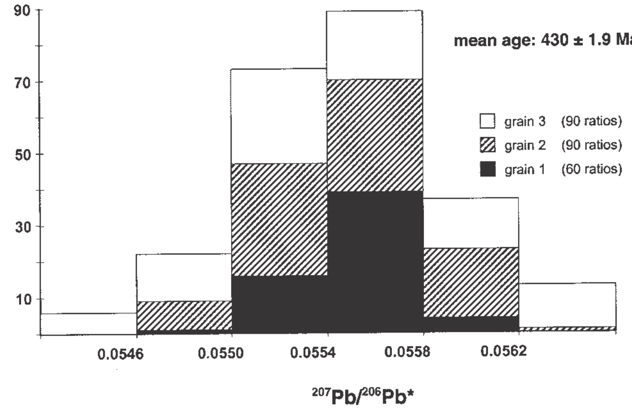

Fig. 4. Histogram of the corrected $^{207}Pb/^{206}Pb^*$ isotope ratios of three euhedral zircon crystals from tuff horizons interbedded within the upper part of the Cyrtograptus Shale (Wenlock, Silurian) of Bornholm.

The time scale for the Silurian presented by Gradstein & Ogg (1996) is updated with the help of precise U-Pb zircon ages for stage boundaries by Tucker & McKerrow (1995). According to the latter authors, the base of the Llandovery can be identified at about 443 Ma, the base of the Wenlock at 428 Ma and the base of the Ludlow at about 423 Ma. The data leave a time interval of about 5 Ma for the whole Wenlock Series.

Incorporating data from new zircon dating techniques (SHRIMP), but following in general the compilation for the Silurian given by McKerrow et al. (1985), Young & Laurie (1996) give lower ages for most series boundaries in the Silurian, if compared with the data discussed by Odin (1994) and Gradstein & Ogg (1996). The base of Llandovery is identified at 434 Ma, and the base of the Ludlow at 420 Ma. According to Young & Laurie (1996) the Wenlock lacks convincing age constraints. By extrapolation, the base of the Wenlock is, therefore, estimated to be close to 425 Ma.

The slight disagreement between the time scales in the Silurian presented in Fig. 5 is rather due to differing age values gained by different analysing techniques instead of a misinterpretation of the geological field relationships. The herein presented Pb-Pb isotope age of 430 Ma for the tuff horizons embedded in the uppermost part of the Cyrtograptus Shale of Bornholm fits best within 2σ-errors the time scale proposed by Odin (1994), but differs slightly from those suggested by Gradstein & Ogg (1996) and Young & Laurie (1996). The value could be used to extrapolate the ages of the base of the Wenlock and Ludlow Series to be 433 Ma

Sub System	Series	Stages	Standard Graptolite Biozones Koren et al. (1996)	Bornholm		Radiometric ages			
				Lithological division	Graptolite zones	Odin (1994)	Gradstein & Ogg (1996)	Young & Laurie (1996)	this paper
UPPER SILURIAN	LUDLOW	GORSTIAN	scanicus						
			nilssoni						
						425±5	423	420±2	428
LOWER SILURIAN	WENLOCK	HOMERIAN	ludensis						
			praedeubeli-deubeli						
			parvus-nassa						
			lundgreni	Cyrtograptus Shale	* C. lundgreni				430±1.9
		SHEINWOODIAN	rigidus-perneri		?				
			riccartonensis-belophorus						
			centrifugus - murchisoni		C. centrifugus				
						430±5	428	425	433
	LLANDOVERY	TELYCHIAN	lapworthi-insectus		C. lapworthi				
			spiralis interval		M. spiralis				
			griestoniensis - crenulata	Rastrites Shale	M. griestoniensis				
			turriculatus - crispus		M. crispus				
			guerichi		M. turriculatus				

* zone with tuff horizons

Fig. 5. Graptolite biostratigraphy of the mid-Silurian, including graptolite biozones, lithological division on Bornholm, and various geochronological time scales. * = level of intercalated tuff horizons; their radiometric age characterizes the time of their deposition.

and 428 Ma, respectively, taking into consideration the estimated duration of a standard graptolite biozone of slightly less than one million years. But such biozones are not equal in duration, and they often differ considerably, based on the ranges of their index species and local interpretations. Carter et al. (1980) interpreted the time intervals of Ordovician and Lower Silurian graptolite zones by sedimentation rates and showed the varying length of the individual biozones. Zalasiewicz (1990) discussed some of the British biozonations yielding an average length of a graptolite biozone in the Llandovery to Ludlow interval of about 0.45 Ma. Similar data for the duration of graptolite zones were provided by Hughes (1995), showing ranges between 0.44 Ma for the Wenlock to 1.43 Ma for the Pridoli. The most detailed biozonation is probably known from the Telychian of Wales (Loydell 1992), reaching a faunal differentiation into intervals of ca. 100 000 years in the former *Spirograptus turriculatus* Zone.

Origin of the tuff horizons

Previously, it was generally thought that the source of Ordovician and Silurian bentonites in Baltoscandia is magmatic arc volcanoes in Norway or further westwards, which formed during subduction and closing of the Iapetus Ocean between Laurentia and Baltica. Although Bergström et al. (1992) recognized the varying trace element compositions of different Silurian bentonites, another source area than those along the destructive plate margin between both palaeocontinents was not discussed. Batchelor & Jeppson (1999), however, studied Wenlock metabentonites from the Swedish island of Gotland, and pointed out that these tuff beds were generated by distal ash fallouts from evolved silicic and alkaline explosive volcanism located along the Tornquist-Teisseyre Zone. This assumption is further supported by the fact that most Caledonian volcanic activity in northwestern Europe appears to have ended towards the top of the Llandovery (Pedersen et al. 1991).

The pyroclastic rocks from southern Bornholm, now dated to be 430 ± 1.9 Ma old, can also be interpreted as a fingerprint for the closing of the Tornquist Ocean during the Caledonian orogeny. The turbiditic nature of the Silurian sediments on Bornholm and the intercalated tuff horizons indicating explosive arc volcanism suggest the existence of a subduction-controlled tectonic regime at the southern margin of Baltica in the later Wenlock. During the collision of Baltica and Avalonia, which led to the subsequent filling of a Silurian foreland basin, subaerial ashes produced by magmatic arc volcanoes south of Baltica could have been easily transported and deposited onto the shelf of this continent. Rather coarse-grained tephra layers representing the proximal facies of such fallouts were deposited on Bornholm, less than 300 km away from a possible eruption centre (Bjerreskov & Jørgensen 1983), while fine-grained ash horizons were formed further north on Gotland. Batchelor & Jeppson (1999) refer to a possible source location near the Caledonian suture zone at Klodsko in Poland (Oliver et al. 1993), but exclude extrusive silicic volcanism of the same age in the Neuvillesous-Huy area of Belgium (Verniers & van Grootel 1991) because of the distance.

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