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Implications of a new, biostratigraphically well-controlled, radio-isotopic age for the lower Telychian Stage of the Llandovery Series (Lower Silurian, Sweden)

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*Radio-isotopic analysis of single zircons from two early Telychian K-bentonites, one of which is among the most widespread Lower Paleozoic volcanic ash falls in northern Europe, yields overlapping weighted mean $^{206}\text{Pb}/^{238}\text{U}$ ages of 438.7 ± 1.0 Ma and 437.8 ± 0.5 Ma, respectively. The former age is from zircons of the Osmundsberg K-bentonite from the type locality at Osmundsberget in the Siljan area of central Sweden where it occurs in the lower part of the *Spirograptus turriculatus* Graptolite Zone and in the lower part of the *Angochitina longicollis* Chitinozoan Zone. Zircons giving the latter age are from a bed previously identified as the Osmundsberg K-bentonite at the Kallholn Quarry in the same area. Based on new biostratigraphic data, the latter bed is now considered to be slightly younger than the Osmundsberg K-bentonite. The dated stratigraphic level of the ash layers is slightly younger than the base of the Telychian Stage and thus represents a minimum age for the Aeronian/Telychian Stage boundary. A U-Pb age of > 438 Ma for the base of this stage, however, is older and in conflict with estimates in the most recent compilation of the Silurian time scale. In view of the fact that only three radio-isotopic dates from the entire Llandovery have been previously accepted, this new and biostratigraphically exceptionally well-controlled radio-isotopic date fills an important gap in the Lower Silurian geochronology.*

Introduction

The geologic time scale is calibrated against radio-isotopic ages from different isotopic systems that are not necessarily comparable due to systematic biases arising from miscalibrated decay constants and standards. In addition, a large number of these ages are either hampered by analytical and/or statistical shortcomings or are biostratigraphically poorly constrained and hence of little use for the establishment of a robust geological time scale. As discussed below,

arguably most of the published radio-isotopic ages for late Ordovician to early Silurian times are associated with such complications that are either not or insufficiently discussed in time scale compilations, highlighting the urgent need for new high-resolution radio-isotopic ages and more transparent evaluation in compilations. In the present study we present U-Pb zircon ages for biostratigraphically calibrated (graptolites and chitinozoans) K-bentonites, one of which can be assigned to the lower Telychian *Spirograptus turriculatus* Zone (Upper Llandovery). We also critically discuss some currently published Silurian ages that were included in a widely used compilation (Gradstein et al., 2004).

The Osmundsberget locality

In a regional study, Bergström et al. (1998a) and Huff et al. (1998) described the stratigraphic position, distribution, mineralogy, geochemistry, and various other aspects of the Osmundsberg K-bentonite. It was named for its type locality (Osmundsberget North) along the northern entrance road to the now disused Osmundsberget Quarry ($61^{\circ}1'3''\text{N}$, $15^{\circ}12'4''\text{E}$) in the Siljan region, Province of Dalarna, south-central Sweden (Figure 1). The Osmundsberg bed is 1.15 m thick at this locality and is accompanied in approximately 10 m of section by 8 additional K-bentonite beds that range between 1 and 32 cm in thickness (Huff et al., 1998). It is a bluish-gray clay-rich unit that stands in contrast to the adjacent limestones and graptolite shales (Figure 1). Regrettably, recent infilling of this road has almost completely destroyed this formerly splendid section, and further study of this prominent ash bed now requires considerable excavation. Fortunately, when this bed was fully exposed in the early 1990s, we carried out extensive sampling and the present study is based on these collections.

In the initial paper on the Osmundsberg K-bentonite (Bergström et al., 1998a), the biostratigraphy had to be based on the incomplete information that was available for this locality in the 1990s. Graptolites of the *Stimulograptus sedgwickii* Zone had long been recorded (Jaanusson, 1982; Thorslund and Jaanusson, 1960) from the basalmost part of the Kallholn Shale (Figure 2), which is a few meters below the interval of the Osmundsberg K-bentonite. From higher portions of the Kallholn Shale, including the interval of the Osmundsberg K-bentonite, there were records of *Spirograptus turriculatus* and other graptolites of the *Spirograptus turriculatus* Zone (Hutt et al., 1970). However, the level of the base of the latter graptolite zone remained undetermined until recently when Loydell and Maletz (2002) clarified the graptolite biostratigraphy in the section. Their data show that the interval from about 5 m to 9.5 m in the

Figure 1 The Osmundsberg K-bentonite at its type locality along the northern entrance road to the Osmundsberget Quarry (61°3'3"N, 15°12'4"E) in the Siljan region, Province of Dalarna, south-central Sweden (1992 photograph).

section published by Bergström et al. (1998a, Figure 4) belongs in the *Spirograptus guerichi* Zone, and that *S. turriculatus* and other graptolites of that zone appear about 0.5 m below the base of the Osmundsberg K-bentonite (Figure 2). Loydell and Maletz (2002, Figure 2) also recorded the zonal index *S. turriculatus* about 0.5 m above the Osmundsberg K-bentonite, hence confirming the correctness of the original assignment of this prominent ash bed at its type locality to the *S. turriculatus* Zone. It should be stressed that the recent reference, without explanation, of this ash bed to the Aeronian *S. sedgwickii* Zone (Heatherington et al., 2004) is clearly in error.

Chitinozoans provide additional, although somewhat less precise, biostratigraphic data bearing on the age of the Osmundsberg K-

bentonite at Osmundsberget. In a regional study of Llandovery and Wenlock chitinozoan biostratigraphy of the Swedish mainland, Grahn (1998) investigated 13 samples from the Osmundsberget North (his Osmundsberget 1) section. His data show that the lower 2.5 m of the Kallholn Shale, which corresponds broadly to the 4–8 m interval in Bergström et al. (1998a, Figure 4), is referable to the *Conochitina emmastensis* Zone. The Osmundsberg K-bentonite occupies a position in the lower part of the overlying *Angochitina longicollis* Zone (Figure 2), the lower part of which has been shown to be of early Telychian age elsewhere (Grahn, 1998, Figure 15). Hence, the graptolite and chitinozoan evidence from this section is in excellent agreement and provides a firm biostratigraphic age dating of this volcanic ash bed. The known stratigraphic ranges of selected graptolites and chitinozoans and the chronostratigraphic classification of the Osmundsberg K-bentonite at its type locality are summarized in Figure 2.

The Kallholn Quarry locality

Bergström et al. (1998a) identified a potential Osmundsberg K-bentonite equivalent at another locality in the Siljan region, namely in the outcrop of the Kallholn Shale along the south side of the western entrance road to the inactive Kallholn Quarry (61°10'0"N, 14°41' 60"E). This section, which is about 30 km north–west of Osmundsberget, is clearly the exposure from which Loydell (1991) described taxonomically diverse and beautifully preserved graptolites of the *Monograptus argenteus* Zone. These graptolites, which were collected from limestone concretions in the lower two meters of the Kallholn Shale, are from the middle part of the Aeronian Stage and show that the base of this shale is slightly older biostratigraphically at this locality than at the Osmundsberget North section. The graptolite zone classification of the overlying major part of the Kallholn Shale, which has long been assumed to represent the *S. sedgwickii* and *S. turriculatus* zones, has yet to be firmly established. However, the chitinozoan biostratigraphic data from the Kallholn Quarry published by Grahn (1998) appear to suggest that the ash bed in this section identified as the Osmundsberg K-bentonite by Bergström et al. (1998a) may not be located within the *S. turriculatus* Zone, and hence, it would represent a different volcanic ash bed than the Osmundsberg K-bentonite. According to Grahn (1998), chitinozoans of the *Ramochitina nestorae* Subzone of the *Angochitina longicollis* Zone appear just a few m above the base of the Kallholn Shale in his Kallholn section. Because this subzone is of late Telychian age and interpreted to be no older than the *Octavites spiralis* Graptolite Zone (Grahn, 1998, Figure 15), this interval is significantly younger than that of the Osmundsberg K-bentonite.

However, there were some problems in using Grahn's data for our section. First, his Kallholn section is described as being located “in the south wall of the quarry”, not along the entrance road, and there are at least two Kallholn Shale sections in the quarry. Second, Grahn's (1998, Figure 2) generalized lithologic log shows no similarity to that of our section (Bergström et al., 1998a, Figure 5), including the position of several K-bentonite beds. In view of these conflicting data, we were uncer-

Figure 2 Vertical ranges of selected graptolites (after Loydell and Maletz, 2002) and chitinozoans (after Grahn, 1998) and stratigraphic classification of the Osmundsberget North section. Note the biostratigraphic position of the Osmundsberg K-bentonite in the lower part of the *S. turriculatus* Zone.

tain whether or not Grahn's section is the same as ours, and hence, if his chitinozoan data could be used for interpretation of the biostratigraphy of our section. However, observations made during a visit to the quarry in 2007 by two of us (WDH and SMB) left no doubt that Grahn's (1998) section is the same as that studied by us. Hence, there is biostratigraphic evidence that the studied K-bentonite at Kallholn is younger than the Osmundsberg K-bentonite. This conclusion is consistent with the fact that recent chemostratigraphic fingerprinting studies of the Osmundsberg K-bentonite (Toprak et al., in preparation) show differences to the Kallholn bed.

New U-Pb zircon ages and analytical methods

Samples SWE129 and SWE132 from the Osmundsberg and Kallholn localities, respectively, were subjected to standard mineral separation techniques for zircon extraction at University of Cincinnati. The sample yielded colorless, clear zircons of elongated prismatic or needle-like morphology with few inclusions. The crystals were carefully examined in transmitted light using a petrographic microscope. In order to avoid averaging effects caused by older inheritance, xenocrystic contamination and Pb loss, or a combination of all (Mundil et al., 2001), zircons were analyzed individually by low blank, isotope dilution thermal ionization mass spectrometry (IDTIMS) analytical techniques at the Berkeley Geochronology Center. Zircon crystals were annealed at 850°C and chemically abraded following the techniques described in Mundil et al. (2004) and Mattinson (2005) in order to minimize the effects of Pb loss. Subsequent to chemical abrasion crystal fragments were very small, weighing 0.3 to 0.8 µg, but contained sufficient $^{206}\text{Pb}_{\text{rad}}$ for precise analysis. Analytical protocols follow those described in Mundil et al. (2004). Individual isotopic ratios containing ^{206}Pb are

Figure 3 Concordia diagram and individual $^{206}\text{Pb}/^{238}\text{U}$ ages (sorted by uncertainty) of SWE129 (white) and SWE132 (red)

corrected for excess ^{230}Th during the crystallization of the zircons (with an assumed Th/U of 4 in the host rock) that results in an age bias of +84 Ka in average (Schärer, 1984).

As shown in Figure 3 and Table 1, four out of five analyzed zircons from the Osmundsberg locality (SWE129) yield a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ age of 438.7 ± 1.0 Ma (MSWD 0.2). One analysis is affected by old inheritance at ca 1500 Ma. SWE129 zircons have an average U concentration of 270 ppm, and Th/U ranges from 0.58 to 0.82. Six out of seven zircon analyses from SWE 132 yield a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ age of 437.8 ± 0.5 Ma (MSWD 0.3). The age of crystal Z06 is slightly older (445 Ma) but resolved from the remaining cluster, indicating the presence of xenocrysts. As for SWE129 the average U concentration is 270 ppm and Th/U varies similarly between 0.55 and 0.74.

Table 1 Isotopic ratios and analytical data for Osmundsberg zircons.

Sample	a)	µg ^(b) zirc.	ppm U	Pb _{c.} ^(c) (pg)	Th ^(d) U	isotopic ratios						isotopic ages			
						$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{235}\text{U}$	$\pm$	$^{206}\text{Pb}/^{238}\text{U}$	$\pm$	ρ^g	$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{235}\text{U}$	$^{207}\text{Pb}/^{206}\text{Pb}$	
						^{204}Pb	^{206}Pb	%	^{235}U	%					
SWE129.Z01	CA	0.3	157	2.2	0.66	108	0.05598	10.3	0.5441	11.0	0.070498	0.76	439.2 ± 3.3	441.2 ± 48.4	451 ± 230
SWE129.Z02	CA	0.3	248	1.8	0.76	211	0.05636	4.8	0.5479	5.1	0.070494	0.38	439.1 ± 1.7	443.6 ± 22.8	467 ± 107
SWE129.Z03	CA	0.3	155	2.4	0.66	305	0.09421	1.9	3.3761	2.1	0.259918	0.29	1489.4 ± 4.3	1498.9 ± 31.2	1512 ± 35
SWE129.Z04	CA	0.2	366	2.2	0.58	156	0.05612	6.8	0.5443	7.2	0.070344	0.52	438.2 ± 2.3	441.3 ± 31.7	457 ± 150
SWE129.Z05	CA	0.3	303	2.2	0.82	181	0.05583	5.8	0.5417	6.2	0.070374	0.46	438.4 ± 2.0	439.6 ± 27.3	445 ± 130
weighted mean age												438.7 ± 1.0			
SWE132.Z01	CA	0.3	292	2.3	0.55	196	0.05637	5.3	0.5468	5.6	0.070358	0.43	438.3 ± 1.9	442.9 ± 24.8	467 ± 117
SWE132.Z02	CA	0.8	304	7.7	0.74	162	0.05621	6.5	0.5453	6.8	0.070346	0.50	438.2 ± 2.2	441.9 ± 30.3	461 ± 143
SWE132.Z03	CA	0.7	415	2.4	0.66	544	0.05569	1.8	0.5398	1.9	0.070300	0.23	438.0 ± 1.0	438.3 ± 8.5	440 ± 40
SWE132.Z04	CA	0.4	225	1.8	0.69	224	0.05654	4.5	0.5480	4.8	0.070286	0.37	437.9 ± 1.6	443.7 ± 21.2	474 ± 100
SWE132.Z05	CA	0.5	316	1.9	0.61	403	0.05611	2.5	0.5434	2.6	0.070227	0.21	437.5 ± 0.9	440.6 ± 11.6	457 ± 55
SWE132.Z06	CA	0.3	194	2.3	0.71	137	0.05627	7.8	0.5543	8.3	0.071439	0.56	444.8 ± 2.5	447.8 ± 37.1	463 ± 173
SWE132.Z07	CA	0.7	135	1.6	0.65	269	0.05566	3.7	0.5385	4.0	0.070177	0.34	437.2 ± 1.5	437.5 ± 17.4	439 ± 83
weighted mean age												437.8 ± 0.5			

a) CA = annealed/chemically abraded

b) sample weight is calculated from crystal dimensions and is associated with as much as 50% uncertainty (estimated)

c) total common Pb including analytical blank (analytical Pb blank is 0.8 ± 0.3 pg per analysis). Blank composition is $^{206}\text{Pb}/^{204}\text{Pb} = 18.55 \pm 0.63$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.50 \pm 0.55$, $^{208}\text{Pb}/^{204}\text{Pb} = 38.07 \pm 1.56$ (all 2? of population), and a $^{206}\text{Pb}/^{204}\text{Pb}$ - $^{207}\text{Pb}/^{204}\text{Pb}$ correlation of +0.9

d) present day Th/U ratio calculated from radiogenic $^{206}\text{Pb}/^{206}\text{Pb}$ and age

e) measured value corrected for tracer contribution and mass fractionation (0.15 ± 0.09 ‰/amu)

f) ratios of radiogenic Pb versus U; data corrected for mass fractionation, tracer contribution and common Pb contribution

g) correlation coefficient of radiogenic $^{207}\text{Pb}/^{235}\text{U}$ versus $^{206}\text{Pb}/^{238}\text{U}$

Uncertainties of individual ratios and ages are given at the 2σ level and do not include decay constant errors.

Ratios involving ^{206}Pb are corrected for initial disequilibrium in $^{230}\text{Th}/^{238}\text{U}$ adopting Th/U=4 for the crystallization environment.

Weighted mean $^{206}\text{Pb}/^{238}\text{U}$ ages (in *italics*) are calculated from individual ages in bold.

Silurian, including the *C. cyphus* Zone, is only about 5 m on Esquibel Island. If the same exceptionally slow rate of net deposition continued to prevail up to the base of the sedimentary breccia, the 4 m thick, apparently non-graptolite bearing, succession between the topmost graptolite collection and the base of the breccia could conceivably correspond to one, or even several, graptolite zones younger than the *C. cyphus* Zone. Accordingly, and even ignoring the analytical shortcomings, until better biostratigraphic control has become available, this radio-isotopic date should not be used unconditionally as representative of the age of the *C. cyphus* Zone. It is appropriate to note that this age appears to differ by as much as 4 Ma from that of the Birkhill Shale K-bentonite despite the fact that they have been assumed to represent the same graptolite zone.

Direct radio-isotopic age control is even less robust for the Llandovery/Wenlock boundary, the youngest Llandovery date (430.1 ± 2.4 Ma (± 4.8 Ma including decay constant uncertainties)) being from the *Monoclimacis crenulata* Zone (Tucker, 1991; Tucker and McKerrow, 1995), which is the fourth graptolite zone below the top of this series. As mentioned above, the scatter of the data indicates the presence of Pb loss suggesting that the oldest $^{206}\text{Pb}/^{238}\text{U}$ age of 433 Ma may be a minimum age.

The oldest post-Llandovery radio-isotopic date (426.8 ± 1.7 Ma) (recalculated from 423.7 ± 1.7 Ma in Kunk et al. 1985, see above and Renne et al. (1998)) included in the most recent compilation by Melchin et al. (2004) is from the interval of the early Ludlow *Neodiversograptus nilsoni-Lobograptus scanicus* zones (Kunk et al., 1985), which is 9–10 graptolite zones above the top of the Llandovery. Recalculating this age from the data table given in Kunk et al. (1985) yields 425.0 ± 2.2 Ma (with an assumed uncertainty of 0.2% in J), which must be adjusted to 427.9 Ma using the accepted age for MMhb-1 and ca 432.2 Ma accounting for the additional 1% bias in order to compare it to U-Pb ages. Even in the absence of the discussed complications affecting the accuracy of these ages, the extrapolation to the top of the Llandovery from such stratigraphically widely separated horizons, which in this case amounts to an interval of at least 11 graptolite zones, results in very significant errors. Recently, a multigrain U-Pb zircon age of 433.2 ± 1.6 Ma was briefly reported from the Ireviken K-bentonite (Jeppsson et al., 2005), which is located just above the base of the Wenlock on the Island of Gotland, Sweden. However, the reliability of this age is difficult to assess until full documentation has been published.

Our new radio-isotopic ages, with excellent biostratigraphic control, provide important age control in a previously poorly calibrated stratigraphic interval of significant length (five graptolite zones) in the Llandovery (Figure 4). Furthermore, because the dated Osmundsberg horizon is only one graptolite zone above the Aeronian/Telychian Stage boundary, the new radio-isotopic date suggests that the radio-isotopic age of this stage boundary is only slightly older than 438 Ma. This age is older than hitherto postulated in all compilations, which is due to many published U-Pb ages being affected by Pb loss resulting in inaccurate and slightly younger ages as well as biased $^{40}\text{Ar}/^{39}\text{Ar}$ ages due to a miscalibrated K decay constant and incorrect ages of flux monitors, also resulting in ages which are too young. The quality of the Silurian time scale is still in a highly provisional state and will only be improved when more precise and accurate radio-isotopic ages are available. In view of the many K-bentonites present in this series, particularly in northern Europe and eastern North America (Bergström et al., 1997; Bergström et al., 1998a, b), the potential for obtaining the required radio-isotopic ages is very promising. Until then, considerable caution is warranted in using the currently available radio-isotopic ages, in particular if they are extrapolated to represent true ages of zones and stages.

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