Correlation of Telychian (Silurian) altered volcanic ash beds in Estonia, Sweden and Norway

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Concentrations of eight trace elements were determined in 80 samples representing 70 thin, altered volcanic ash beds collected from seven drill cores in Estonia. All of the samples studied come from the Adavere Stage (Telychian), except for one which belongs to the lower Jaani Stage (lowermost Sheinwoodian). The source magma composition of these volcanic ash beds varies from moderately to highly evolved. By combining the results of geochemical correlations with biostratigraphical data, based on the distribution of conodonts, 31 individual volcanic events were identified, which in turn could be grouped into four distinct volcanic episodes.

A bimodal distribution of trace elements generated during the third episode of volcanic activity corresponds to a similar bimodality in the composition of a metabentonite suite from Garntangen in Norway. Comparison of these data with those previously published from Norway and Sweden indicates that the majority of volcanic beds from Garntangen can be ascribed to the *Pterospathodus amorphognathoides lithuanicus* Zone in Estonia. The volcanic beds from the Ireviken section on Gotland can be correlated with the upper part of the studied interval in Estonia and occur in strata corresponding to the uppermost *P. amorphognathoides* Zonal Group.

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Introduction

The Silurian was characterised by severe environmental perturbations recorded in lithological sequences as rapid facies changes, extensive gaps (particularly in shallow water regions) (Jeppsson *et al.* 1994), extinctions of biota (Kaljo *et al.* 1995) and isotopic variations (Kaljo et *al.* 1997, 1998). Climatic changes and oceanic circulation overturns have been considered to be the causes of these variations (Jeppsson 1990, 1993; Jeppsson *et al.* 1995; Samtleben *et al.* 1996; Bickert *et al.* 1997). Tillites from multiple glaciations (the youngest of them dated as latest Telychian to earliest Sheinwoodian) are recorded in the Llandovery in Brazil (Grahn & Caputo 1992; Caputo 1998).

In order to study and understand the nature of ancient environmental changes, detailed correlation of sections is essential. Without this, the true succession of events (e.g. extinctions of various faunal groups, isotopic anomalies, and sea-level fluctuations) is not possible to ascertain. In the present paper, the correlations based on the trace element characteristics of thin, altered volcanic ash beds are compared with those based on the distribution of conodonts.

The lithology and mineralogy of Silurian volcanic beds in Estonia were first studied by Jürgenson (1964), the trace elements characteristics by Kiipli & Kallaste (1996), and facies-related compositional variations by Kiipli *et al.* (1997a, b). Correlation of Silurian volcanic beds based on their geochemical composition has not previously been attempted in Estonia.

Silurian volcanic beds in Scandinavia have been investigated, among others, by Spjeldnæs (1959), Snäll (1978), Bergström *et al.* (1992), Batchelor & Jeppsson (1994, 1999) and Batchelor *et al.* (1995), in Scotland by Batchelor (1995), Batchelor & Weir (1988), Batchelor & Clarkson (1993), and in the south-western part of the East European Platform by Huff et al. (2000) and Kiipli et al. (2000). At the time of writing, only one Silurian volcanic bed, the Osmundsberg K-bentonite ("O"), has been correlated over large areas (Bergström *et al.* 1998; Huff *et al.* 1998).

Clay-rich interbeds of probable volcanic origin in Palaeozoic sedimentary sections are commonly called K-bentonites or metabentonites. Since many samples studied by us consist of predominantly potassium feldspar, they cannot be strictly classified as bentonites (neither K- nor meta-), but are better described as feldspathites. As the volcanic origin of these beds is well proven in many of the above-listed publications, and confirmed by the present study, we prefer the term volcanic bed (indicating its structural form) and altered volcanic ash (indicating its material).

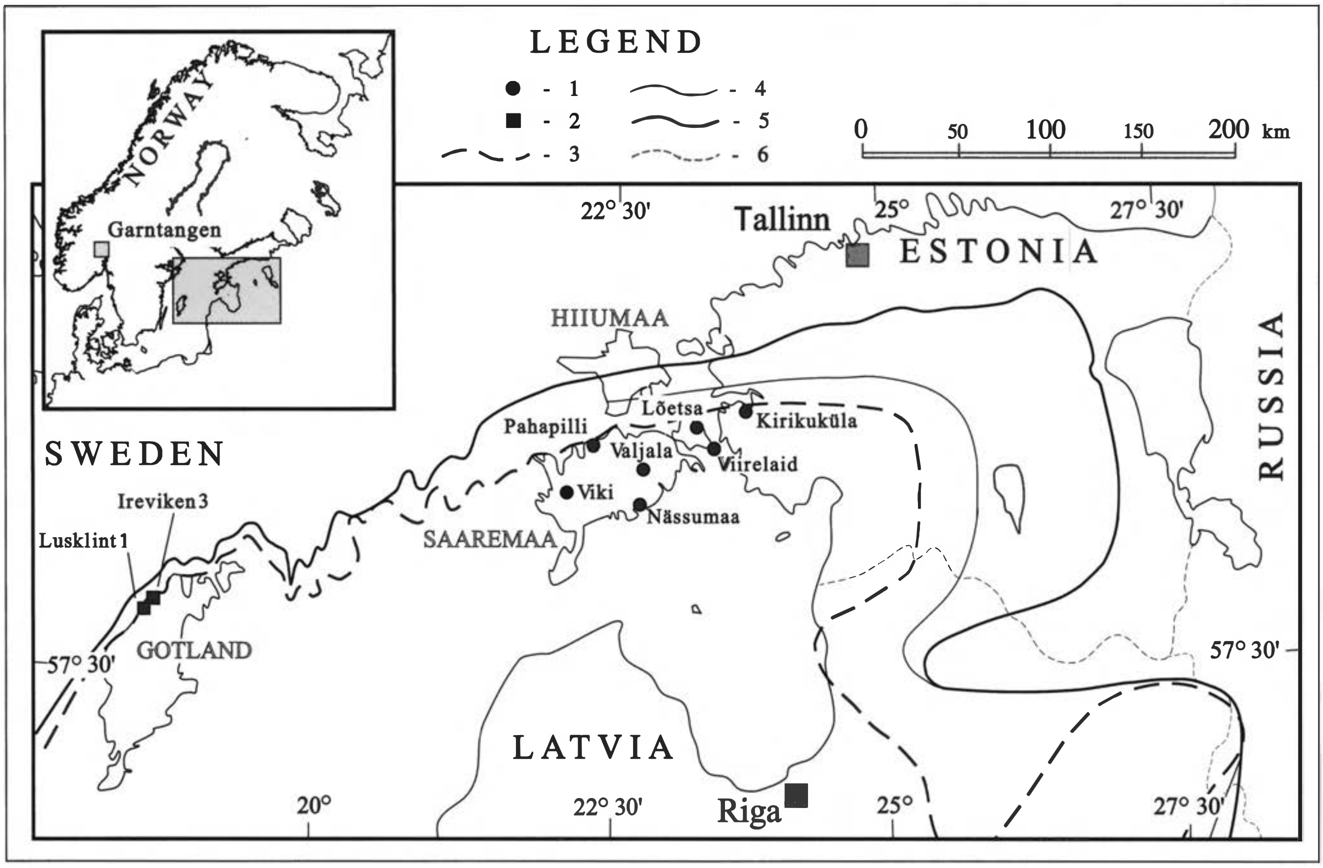


Figure 1. Location of studied core sections from Estonia, and outcrops from Sweden (Gotland) and Norway. 1 – core; 2 – outcrop section; Lines 3–5 indicate the contour of the distribution area of the Sheinwoodian (3), Telychian (4) and Rhuddanian–Aeronian (5); 6 – border line of countries.

Material and methods

Sixty-two samples representing 54 volcanic beds were collected from the Valjala, Viki, Pahapilli, Nässumaa, Lõetsa, Viirelaid and Kirikuküla cores (Figure. 1). Additionally, 18 samples from 17 volcanic beds from the Viki and Kirikuküla cores, formerly studied by Kiipli & Kallaste (1996), were re-studied. In total, 80 samples from 70 individual beds were collected. The depth levels given in the tables and figures indicate the positions of the lower boundaries of the volcanic beds.

The content of Nb, Zr, Y, Ga, Rb, Sr and Ni in these samples, prepared as pressed powder pellets, was analysed with a wavelength dispersive X-ray fluorescence (XRF) spectrometer VRA-30, using an X-ray tube with Ag anode at 25 mA and 45 kV. Background radiation was measured on both sides of the characteristic peaks. The counting time was 30–40 seconds per point. The sample matrix influence was reduced by using peak/background intensity ratios in calibration (variant of the Compton scatter method for matrix corrections; Lachance & Claisse 1994). Major elements were analysed from fused discs using Li-tetraborate as the flux (sample/flux ratio was 1:5), applying an X-ray tube with Cr anode at 20 mA and 30 kV. Counting times varied from 20 s for Ti to 100 s for Na. The precision of analyses of particular elements was: Nb – 2, Rb – 2, Y – 4, Ga – 4, Sr – 4, Zr – 10 and Ni – 10 ppm (determined from 6 replicate measurements, 2 standard deviations). The precision of major component analyses was MnO – 0.02, TiO_2 – 0.006, Fe_2O_3 – 0.08, K_2O – 0.15, CaO – 0.015, Na_2O – 0.8, MgO – 0.3, Al_2O_3 – 0.4, SiO_2 – 1.2 and P_2O_5 – 0.15 % (10 replicate measurements, 2 standard deviations). In order to compare data from two different datasets (Batchelor et al. 1995 and the present study), it was necessary to check the analytical method. The majority of the analyses were carried out in the Institute of Geology (Tallinn Technical University),

Table 1: Results of comparative analyses from the Viki core (ppm)

Element	Viki 185.1		Viki 147.5	
	Tallinn	St Andrews	Tallinn	St Andrews
Nb	9	11	40	46
Zr	194	181	692	703
Y	7	4	35	37
Ga	18	16	17	17
Rb	95	92	88	82
Sr	106	111	131	124
Ni	11	12	86	95
Ti	2400	2338	5460	5276

but selected samples from the Viki core were also analysed in St. Andrews, Fife (Table 1).

From bulk-rock samples, prepared as slurry mounts, X-ray diffractograms were obtained on an HZG-4 diffractometer using Co radiation from 5 to 45 degrees. Coarse fractions were separated from ultrasonically suspended samples and studied under a microscope to establish euhedral magmatic minerals.

Table 2: Field and **analytical** data of altered volcanic ash

Bed No.	CORE	DEPTH m	Thickness cm	DESCRIPTION AND MAIN MINERALS	Nb ppm	Zr ppm	Y ppm	Ga ppm	Rb ppm	Sr ppm	Ni ppm	Ti ppm
29	VALJALA	113,80	1,5	I-s,B,F	14	263	19	14	99	103	37	2803
28	VIKI	115,00	5,0	Grey fat clay. I-s,	34	596	24	20	65	175	22	4391
27	VIKI	121,05	5,0	Grey clay, hard in u.p., I-s,F	21	388	18	22	77	188	74	4313
27	VALJALA	119,80	3,0	I-s, traces of F and B.	22	466	27	22	83	150	90	3894
27	PAHAPILLI	20,85	10,0	Grey clay, I-s	21	412	25	19	82	115	21	3200
26	KIRIKUKÜLA	12,60	?	F, traces of D, I-s, Q	8	217	25	12	75	80	24	4109
26	VIKI	145,75	5,0	Yellow hard feldspathite, F.	13	305	25	10	70	169	67	4469
26	NÄSSUMAA	199,70	2,0	Hard feldspathite, traces of I-s.	11	217	28	9	64	178	30	4337
25	KIRIKUKÜLA	15,50	?	F, I-s.	35	393	36	14	76	48	16	3300
24	KIRIKUKÜLA	15,80	?	F, I-s	34	556	40	17	85	71	11	3940
24	VIKI	147,50	5,0	Grey fat clay, I-s, F.	40	692	35	17	88	131	86	5460
23	VIIRELAID	65,90	4,5	grey clay	42	756	39	18	81	108	17	5410
23	LÕETSA	44,40	4,0	Green clay, I-s, traces of F, B.	51	800	44	21	90	93	18	5942
23	NÄSSUMAA	200,70	5,0	White, I-s.	44	760	42	19	88	108	16	5283
23	PAHAPILLI	40,30	2,0	White, I-s, F, traces of B.	37	624	37	23	99	98	23	4780
22	KIRIKUKÜLA	17,20	?	F.	11	215	21	11	68	281	41	5139
21	VIKI	148,00	1,5	Grey fat clay, I-s, F.	41	627	34	17	98	116	29	5936
20	VIKI	148,80	0,5	Yellow hard, F, traces of A.	14	243	39	12	95	366	133	7290
19	VIKI	149,40	?	F, I-s.	37	297	9	19	101	75	6	1990
19	NÄSSUMAA	201,80	5,0	White, I-s, F.	42	320	11	21	99	65	10	1983
19	LÕETSA	45,10	4,0	White, I-s.	44	354	14	17	88	70	24	2288
19	KIRIKUKÜLA	17,85		Hard white, F, I-s.	37	283	22	12	85	42	10	1851
19	VIIRELAID	66,60	3	white clay	37	382	15	14	80	81	8	2110
18	VIKI	151,80	?	F traces of A.	12	246	23	11	82	309	47	4710
18	VIIRELAID	67,75	1	feldspathite	15	226	33	13	70	306	76	5548
17	LÕETSA	47,20	3,0	Yellow, F, traces of I-s, A, Q.	15	303	59	8	65	235	33	5924
17	PAHAPILLI	45,20	1,5	Yellow hard, F, traces of I-s, A.	14	272	64	10	70	320	55	5175
17	VIIRELAID	68,30	0,7	feldspathite	11	229	83	10	52	245	11	3950
16	VIKI	152,10	0,5	Yellow hard feldspathite, F.	21	306	24	11	79	390	57	7949
16	PAHAPILLI	46,20	1,0	Grey hard, F, traces of I-s, A.	16	289	25	12	72	272	27	7050
15	KIRIKUKÜLA	28,25	?	Grey clay, I-s, F, traces of A.	12	234	35	17	84	212	49	4876
15	PAHAPILLI	51,50	0,5	White greenish, I-s,F,traces A.	13	259	27	18	76	249	92	5325
15	VIKI	156,80	2,5	Yellow hard, F, I-s, traces of A.	21	308	47	18	70	182	38	4642
14	VIIRELAID	76,45	2	grey clay	14	137	43	16	106	121	11	1720
13	VIIRELAID	76,70	2,5	grey clay	14	259	38	22	123	73	13	2150
13	VIKI	169,60	10,0	Grey clay, B, I-s.	18	253	37	33	140	92	49	2696
13	LÕETSA	56,80	4,0	Purple clay, B, I-s.	11	235	40	24	125	58	15	2276
13	NÄSSUMAA	219,40	?	Purple clay, B, I-s.	14	183	30	32	134	86	29	2306
13	PAHAPILLI	61,70	5,0	Grey purplish clay, B, I-s.	13	210	36	30	135	91	36	2438

I-s - illite-smectite, B - biotite, Q - quartz, M - magnetite, F - feldspar, A - apatite, H - hematite

Table 2: Field and analytical data of altered volcanic ash (continued)

Bed No.	CORE	DEPTH m	Thickness cm	DESCRIPTION AND MAIN MINERALS	Nb ppm	Zr ppm	Y ppm	Ga ppm	Rb ppm	Sr ppm	Ni ppm	Ti ppm
12	LÕETSA	57,70	6,0	Green clay, B, I-s	23	402	13	24	107	77	10	3480
12	PAHAPILLI	64,60	5,0	Yellow to red, B, I-s.	21	413	15	23	108	121	25	3534
12	VIKI	171,95	3,0	Red clay, I-s, H, traces of F.	21	254	18	22	87	114	20	2743
12	VIIRELAID	77,60	4	green clay	17	343	11	18	89	93	23	2364
11	VIKI	173,10	10,0	Purple hard, I-s,F	37	351	25	22	97	110	18	2850
11	PAHAPILLI	65,65	8,0	Red, I-s,F	39	385	33	27	102	113	10	2648
11	PAHAPILLI	65,70	8,0	Yellowish white, I-s,F	42	410	25	25	99	105	22	3091
11	VALJALA	153,40	2,5	Red, I-s,F	37	358	26	26	105	89	29	2654
11	VALJALA	153,40	2,5	Yellowish white, I-s,F	41	443	20	25	107	83	37	3768
11	VIIRELAID	78,11	12	feldspathite	33	392	29	16	85	82	9	2300
10	VIKI	174,40	3,0	Violet, I-s,H traces of F,Q	34	208	12	22	99	109	37	2378
10	VIIRELAID	78,20	1,5	green clay	33	298	24	21	111	97	36	2300
9	PAHAPILLI	67,00	5,0	Red clay, I-s	26	574	18	24	96	110	34	7300
9	VIKI	175,55	2,0	Red clay, I-s,H	20	451	21	18	86	106	44	6152
9	VIIRELAID	78,48	4	feldspathite	24	485	16	24	91	101	15	4920
8	PAHAPILLI	68,50	5,0	Red clay, I-s traces of F,Q,B,M	18	288	22	25	99	113	34	3348
7	VIIRELAID	79,13	1	green clay	24	357	31	21	93	99	12	3080
6	VIIRELAID	79,38	6	green clay	23	222	7	26	93	76	14	1030
5	VIKI	178,80	1,5	Violet clay, I-s,F, traces of Q	41	657	45	20	96	113	31	6733
4	VIKI	181,80	0,5	Bluish grey hard, I-s,B,Q,F,D	15	253	21	20	104	86	34	4373
4	VIIRELAID	79,44	1	grey clay	17	271	27	21	99	135	16	3110
3	PAHAPILLI	72,20	5,0	Green and purple clay, I-s	57	932	58	21	90	137	33	5600
3	VIKI	182,30	1,5	Violet clay, I-s, traces of F,Q	44	769	40	21	83	115	16	4756
2	VIKI	184,35	<1	Grey clay, I-s, traces of Q,F,D	22	192	17	16	121	81	55	3756
1	VIIRELAID	80,90	0,7	Yellow hard feldspathite, F	14	354	19	20	90	75	6	1890
0	KIRIKUKÜLA	38,15	16,0	Yellow hard feldspathite, F	3	174	6	17	69	80	1	1500
0	KIRIKUKÜLA	38,15	16,0	Yellow hard feldspathite, F	5	161	6	15	63	69	7	1500
0	VIKI	185,10	10,0	Yellow hard, F, I-s	9	194	7	18	95	106	11	2400
0	LÕETSA	65,90	25,0	Yellow hard feldspathite, F	7	103	12	20	69	55	18	2400
0	LÕETSA	65,95	25,0	Yellow hard feldspathite, F	4	167	6	21	71	79	0	2700
0	LÕETSA	65,95	25,0	Yellow hard feldspathite, F	6	115	10	21	59	55	8	1800
0	LÕETSA	66,05	25,0	Yellow hard feldspathite, F	4	160	11	22	61	58	4	1971
0	LÕETSA	66,05	25,0	Yellow hard feldspathite, F	5	156	13	21	67	76	8	2400
0	VALJALA	165,40	17,0	Yellow hard feldspathite, F	6	113	9	20	67	50	4	1539
0	VALJALA	165,40	17,0	Yellow hard feldspathite, F	5	152	9	18	72	74	1	2000
0	NÄSSUMAA	235,05	20,0	Yellow hard feldspathite, F	7	140	9	26	80	74	7	2400
0	NÄSSUMAA	235,10	20,0	Yellow hard feldspathite, F	7	184	7	20	83	80	9	1700
0	NÄSSUMAA	235,15	20,0	Yellow hard feldspathite, F	7	186	9	17	79	83	0	2400
0	PAHAPILLI	78,35	15,0	Yellow hard feldspathite, F	7	144	9	17	72	60	18	1725
0	VIIRELAID	85,00	20	Yellow hard feldspathite, F	6	207	7	15	60	74	0	1900
-1	PAHAPILLI	83,85	2,0	Yellow hard feldspathite, F	14	249	28	12	80	58	11	4612

I-s - illite-smectite, B - biotite, Q - quartz, M - magnetite, F - feldspar, A - apatite, H - hematite

Most of the studied volcanic beds are only a few centimeters in thickness (Table 2). As the amount of material in every studied sample was limited (all samples come from core sections) in most cases we could only analyse a single sample per bed. Only 15 analyses from the lithologically well correlated Osmundsberg bed firmly characterize within-bed variations in trace element contents. We realize that this method reduces the reliability of geochemical correlations proposed below, but still, we believe that most of them are real as our samples mainly represent the whole thickness of thin ash beds, averaging within-bed variability. Biostratigraphy and the vertical succession of beds in a single core section offered further helpful constraints eliminating all cross-correlations that could be indicated by the geochemical compositions. Further studies are planned to expand the geochemical database and check the correlations.

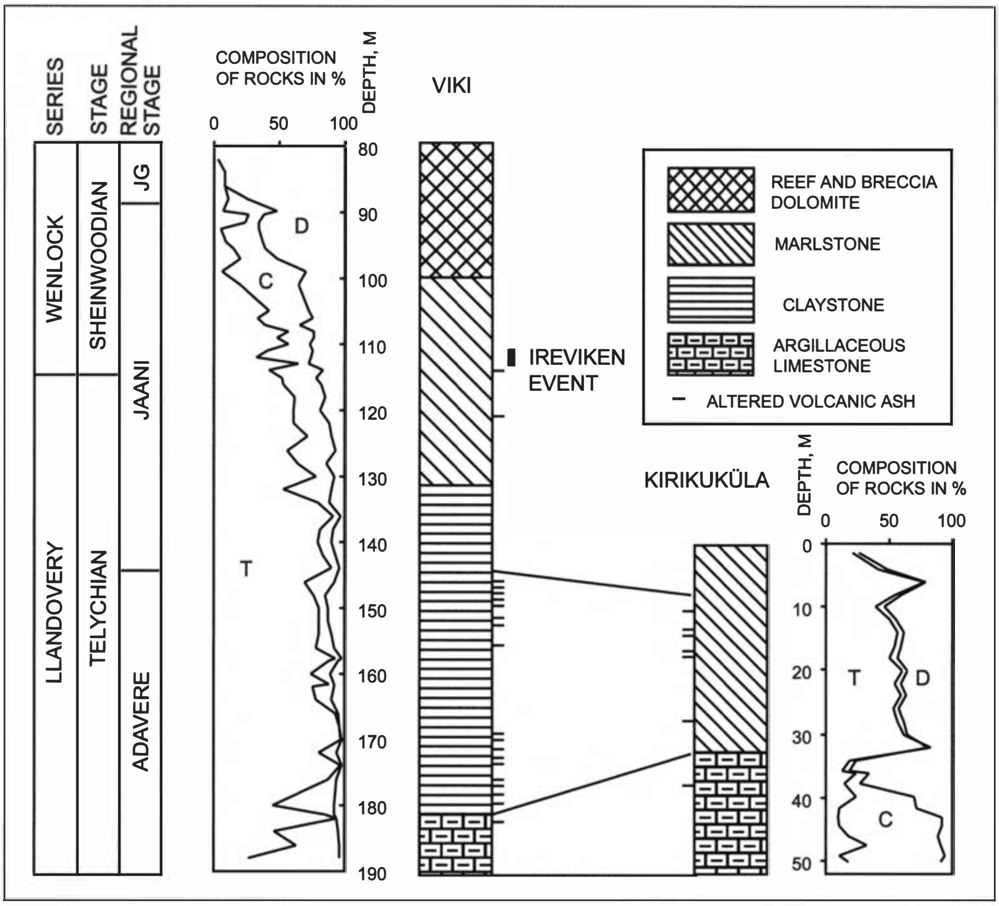


Figure 2. Stratigraphy and lithology of the Viki and Kirikuküla sections. T – terrigenous material, C – calcite, D – dolomite (calculated from main components of X-ray fluorescence analyses as in Kaljo et al. 1997). Note the rocks of the Kirikuküla section contain more carbonate than the Viki rocks.

Stratigraphy

The studied interval corresponds to the Rumba and Velise formations of the Adavere Stage (Telychian), and to the lowermost part of the Jaani Formation of the Jaani Stage (lowermost Sheinwoodian). The boundary between the Adavere and Jaani stages has been considered to correspond to the Llandovery–Wenlock boundary (Nestor 1987).

In general, the Rumba Formation is represented by horizontally-bedded to nodular biomicritic limestones (wackstones, packstones) with clayey partings (Nestor 1997). Scattered shells or tempestite accumulations of *Pentamerus oblongus* are characteristic of the formation in its outcrop area in central Estonia. The formation consists of up to 12 low-grade sedimentary cycles beginning with argillaceous rocks (marlstones, argillaceous limestones), and ending with a layer of pure, hard limestone (Einasto et al. 1972). Westwards the clay content of the rock increases, and on Saaremaa marlstones are prevailing in the sequence of the Rumba Formation. The Velise Formation (which corresponds to the main part of the Adavere Stage) and the lowermost Jaani Formation are dominated by various greenish- to bluish-grey marlstones, mudstones and plastic clays. In central and western Saaremaa (Viki, Valjala and Nässumaa cores), the lower part of the Velise Formation is partly reddish. The stratigraphy and lithology of the two marginal sections at Viki and Kirikuküla are represented in Figure. 2. The lithostratigraphy and biostratigraphy of some of the investigated cores have been published in Jürgenson (1966), Jeppsson & Männik (1993) and Nestor (1994).

The biostratigraphical framework of this paper is based on the conodont sequence. The studied interval is mainly represented by the rich, highly variable *Pterospathodus* fauna. Only the Rumba Formation contains rare conodonts and probably represents the *Distomodus staurognathoides* Zone.

A high-resolution conodont zonation is available for the *Pterospathodus* interval (Männik 1998a, b). Starting from the uppermost Rumba Formation (from the 12th cycle according to Einasto *et al.* 1972), the *P. eopennatus*

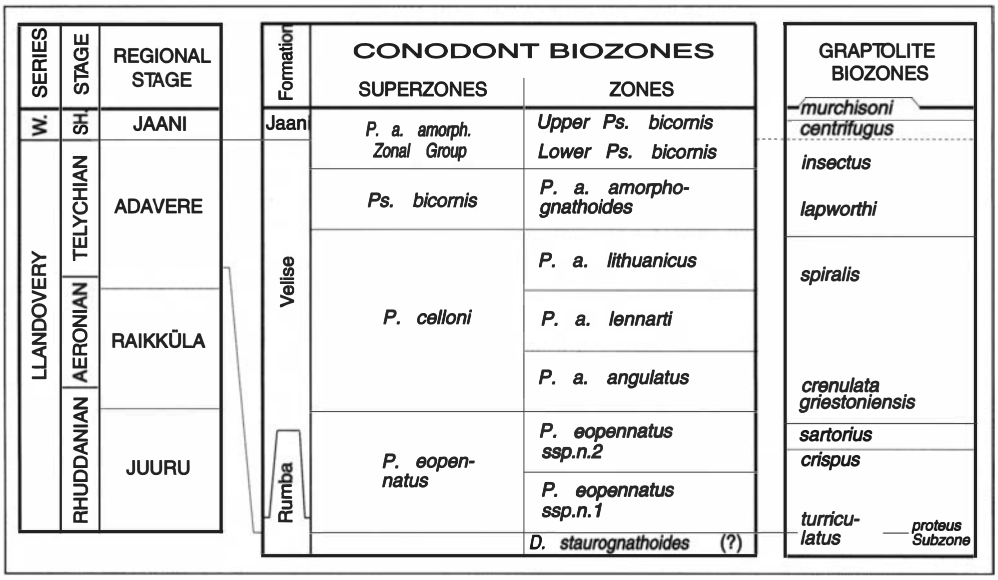


Figure 3. Conodont zonation (from Männik 1998b). Correlation with the graptolite zonation based on Loydell et al. (1998). W. = Wenlock, SH. = Sheinwoodian.

and *P. celloni* superzones, and the *P. amorphognathoides amorphognathoides* Zone, the last one corresponding to the main (lower) part of the *P. amorphognathoides* Zonal Group of Jeppsson (1997), are recognized. *P. eopennatus* ssp. n. 1 and *P. eopennatus* ssp. n. 2 zones were described in the *P. eopennatus* Superzone, and *P. a. angulatus*, *P. a. lennarti* and *P. a. lithuanicus* zones in the *P. celloni* Superzone (Figure. 3; Männik 1998a, b). However, due to inadequate sampling density of sections, the precise positions of several zonal boundaries are not known. In Figure. 4, the zonal boundaries are not marked as lines but, instead, the boundary intervals are indicated. The duration of the Adavere Stage (Telychian) was about 4 million years (Kaljo *et al.* 1998).

Mineralogy of altered volcanic ash in Estonia

The mineralogy of clay-rich interbeds must be considered in order to discriminate between terrigenous and volcanogenic origin. Euhedral biotite, which is detectable visually, is very common in volcanic beds. Variable amounts of euhedral quartz (bipyramid forms or broken crystals) and apatite, detected microscopically in many samples, are good indicators of primary volcanic origin. The most characteristic feature of volcanic beds in Estonia is the absence of terrigenous quartz as a main component and the presence of mixed-layer illite-smectite and K-sanidine in variable proportions. In contrast to volcanogenic clays, terrigenous claystones consist of illite, chlorite, abundant quartz and less K-feldspar. Instead of a broad reflection of illite-smectite at 11–12 Å, which is characteristic of most volcanic beds, a sharp reflection occurs at 10 Å in terrigenous claystones, which represents illite. Pyrite, a random accessory mineral, is of authigenic origin. The rare occurrence of weak reflections of chlorite, calcite and dolomite in X-ray diffractograms most probably represents contamination (resulting from natural bioturbation or impure sampling) by fragments from adjacent sediment, although an authigenic origin of these minerals in volcanic beds is known in other regions of world.

An interesting phenomenon is noticed in several volcanic beds from Saaremaa; the lower part of the bed is yellowish-white and the upper part is red, caused by authigenic hematite (Kiipli et al. 2000). Usually, the trace element contents are more variable in volcanic beds than in terrigenous claystones. In some cases, however, the content of trace elements in a volcanic bed is quite similar to that in the terrigenous material, and mineralogical analysis is needed to positively identify if there is a volcanic origin of the bed.

Vitric ash is thermodynamically unstable and recrystallizes in exogenic environments. During deposition, and on the seafloor, the composition of volcanic ash undergoes substantial changes depending upon facies and burial conditions. Although the effects of late diagenesis on sedimentary rocks in Estonia are minimal (as indicated by the low colour alteration index of conodonts – CAI=1, which shows that the sediments were never affected by temperatures higher than 50 °C), halmirolysis and early diagenesis caused great variation in the final composition of volcanic beds (Kiipli *et al.* 1997a, b). Shallow-water limestones (facies model of Nestor & Einasto 1977) and calcareous marlstones host volcanic beds dominated by K-feldspar, whereas at intermediate marine palaeodepths volcanic beds consist mainly of illite-smectite. Deep-shelf claystones contain volcanic beds dominated by kaolinite. In samples studied in this work only K-feldspar and illite-smectite dominate the mineralogy. In all studied cores the main part of the Adavere Stage is represented by carbonate terrigenous sediments of open shelf origin (Velise Formation, see above). However, the changes in the general composition of sediments and faunas indicate a deeper, more distal environment of sedimentation in the western part of the studied area than in the east (Kaljo & Jürgenson 1977). Accordingly, in most volcanic beds from the Kirikuküla and Lõetsa cores, K-feldspar dominates over illite-smectite (K_2O more than 12 %). In contrast, the content of K-feldspar in most beds from the Viki core is considerably lower (K_2O < 12 %) than that of illite-smectite (Table 3).

Table 3: Major components (in %) in altered volcanic ash from the Kirikuküla, Lõetsa and Viki cores

Depth m	Bed No.	LOI	P_2O_5	SiO_2	Al_2O_3	MgO	Na_2O	CaO	K_2O	Fe_2O_3	TiO_2	MnO
KIRIKUKÜLA												
12,59	26	2,06	0,01	62,4	18,4	1,18	0,7	0,92	13,6	1,14	0,69	0,009
15,50	25	4,58	0,00	58,9	17,6	2,40	0,5	1,71	12,7	1,30	0,55	0,014
17,20	22	1,95	0,04	62,9	17,8	0,64	0,6	1,12	14,3	1,32	0,86	0,003
17,85	19	2,66	0,00	62,1	18,9	1,64	0,3	0,80	13,3	0,99	0,31	0,005
28,25	15	3,51	0,07	58,2	19,7	2,10	1,2	0,63	11,0	3,50	0,81	0,014
LÕETSA												
44,40	23	5,95	0,00	55,0	22,2	3,70	0,4	1,16	7,8	2,52	0,99	0,008
45,10	19	3,97	0,05	60,0	22,3	2,74	0,5	0,58	10,1	1,41	0,38	0,003
47,20	17	2,74	0,56	60,9	18,9	1,19	0,3	1,60	12,9	1,48	0,99	0,019
56,80	13	3,32	0,03	59,2	20,8	2,66	0,3	0,54	10,1	2,78	0,38	0,013
57,70	12	4,76	0,10	57,5	21,5	4,16	0,2	0,78	8,3	3,02	0,58	0,012
66,05	0	0,57	0,03	65,4	17,6	0,52	0,5	0,27	15,3	1,25	0,33	0,016
VIKI												
115,00	28	5,53	0,06	55,3	24,3	2,42	0,3	0,48	7,9	1,33	0,73	0,001
121,05	27	4,97	0,00	56,8	24,0	2,35	0,0	0,55	8,2	1,37	0,72	0,007
145,75	26	2,95	0,03	59,7	18,2	0,77	0,6	1,87	13,0	1,37	0,75	0,013
147,50	24	5,80	0,08	54,8	22,3	3,18	0,5	1,05	7,2	2,77	1,12	0,013
148,00	21	6,25	0,02	54,6	22,1	3,48	1,0	1,63	7,0	3,18	0,99	0,010
148,80	20	2,63	0,36	58,9	18,7	1,35	0,9	1,19	12,3	2,26	1,22	0,014
152,10	16	2,52	0,19	60,2	19,3	1,12	0,3	0,68	12,7	1,85	1,33	0,001
156,80	15	4,10	0,05	56,8	21,3	2,18	0,8	0,64	10,3	2,51	0,77	0,004
169,60	13	4,36	0,05	56,3	22,0	2,83	1,0	0,65	7,8	4,62	0,45	0,020
171,95	12	6,18	0,07	51,4	20,4	3,20	0,4	2,22	6,5	9,38	0,46	0,023
174,40	10	4,99	0,11	54,4	20,9	3,12	0,6	0,59	7,5	6,07	0,40	0,009
175,55	9	4,98	0,09	52,5	21,2	2,99	0,9	0,72	7,1	8,84	1,03	0,019
178,80	5	4,50	0,06	54,6	21,3	2,89	0,9	0,70	7,7	7,28	1,12	0,027
181,80	4	4,68	0,24	55,8	19,8	3,44	0,2	1,29	8,2	5,51	0,73	0,056
182,30	3	5,61	0,00	55,2	21,0	3,21	0,6	0,52	7,2	5,95	0,79	0,023
184,35	2	5,35	0,02	56,8	17,3	2,81	0,3	2,04	9,6	3,15	0,63	0,033

Correlation of volcanic beds in Estonia

The concept of chemical correlation is based on the well-known variation of trace element concentrations in volcanic rocks (Winchester & Floyd 1977). When a sufficient number of samples are available from each volcanic bed, then multivariate statistical analyses can be applied for proving correlations (Huff & Kolata 1989).

The variation of trace elements is considerably higher in volcanic beds than in terrigenous claystones (Fig. 6). Commonly volcanic beds contain more Nb, Zr, Y and less Ni than terrigenous claystones, often also more Ga and Sr, and in some cases more Rb. As well as the major elements and some minerals, some trace elements underwent changes during diagenesis. For that reason, immobile elements should preferably be used in correlations. According to Winchester & Floyd (1977), Nb, Zr, Y, Ga and Ti could be considered as immobile under weathering processes. Based on the positive correlations of Sr and Ni with Ti (correlation coefficients respectively 0.57 and 0.44 – Kiipli & Kallaste 1996), we assume a relatively low mobility of Sr and Ni in these particular environmental conditions. The contents of Sr and Ni may reflect the composition of the source magma to some extent. Rb correlates with Mg and Fe, suggesting that biotite is the main mineral control of these elements (Kiipli & Kallaste 1996). Biotite, represented by euhedral crystals, is considered to be a primary magmatic mineral, and therefore Rb could also be used for correlations within these restricted facies conditions. The ranges of trace element concentrations representing about 30–40 % of the samples varies as follows: Nb 15–30, Zr 270–450, Y 20–34, Ga 16–22, Rb 80–100, Sr 100–180, Ni 20–45 and Ti 3200–4800 ppm. Lower and higher contents were considered as potential discriminating parameters for individual eruptions.

Cluster analysis was used for grouping previously uncorrelated samples. Discriminant analyses cannot be applied due to the small number of samples from most beds. Only the lithologically well-correlated Osmundsberg bed is represented by 15 samples (up to 5 from one core). Comparison of trace element content revealed that this bed can be confidently distinguished from all others on the basis of its low Nb content, and from many others by its low Zr, Y and Ti content (Table 2, Fig. 6). Chemical correlations between other beds are less certain and additional geological information must be considered.

Cluster analysis revealed groups of samples characterized by similar geochemical composition, suggesting possible correlation between them. However, it also appeared that several clusters included beds from the

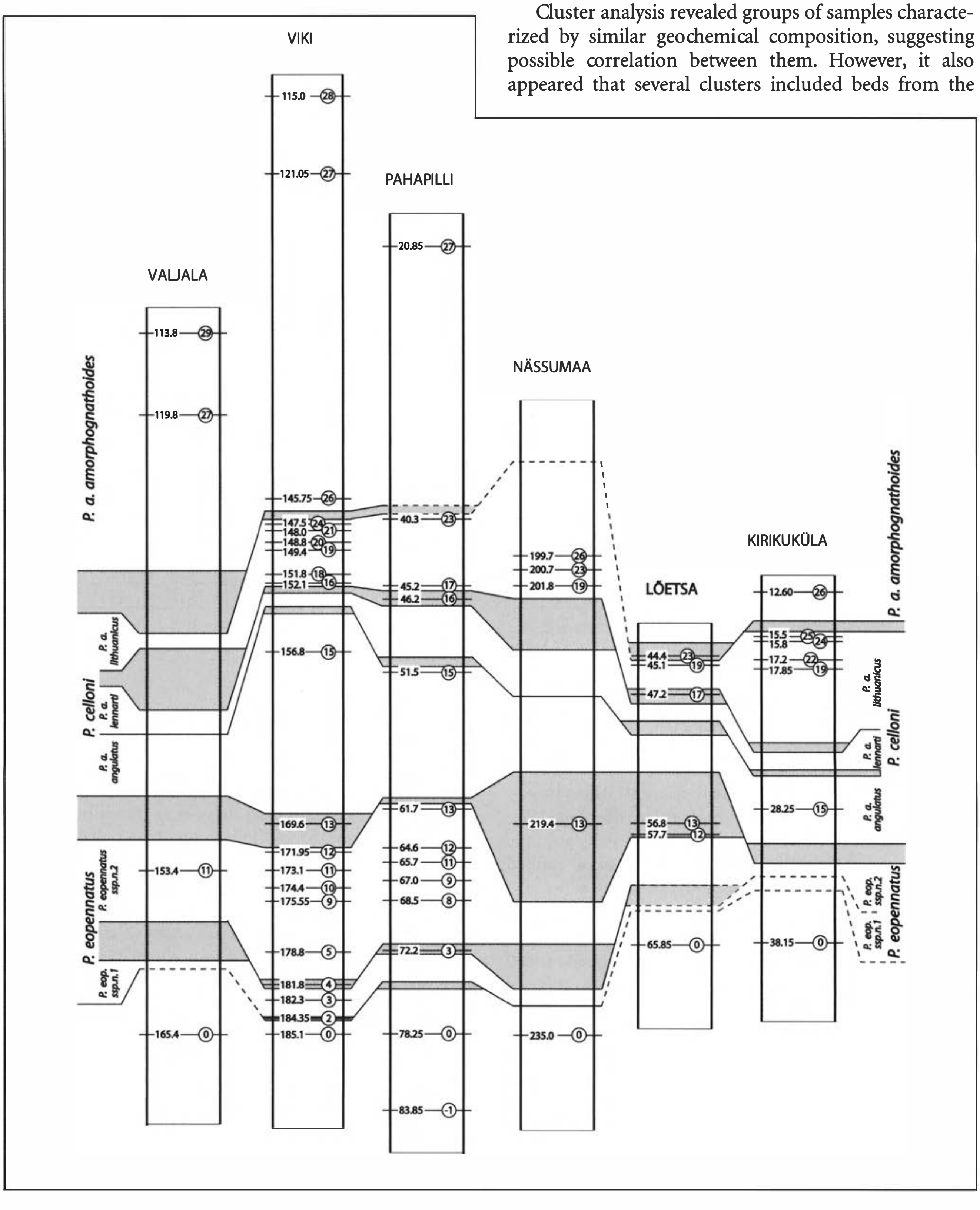


Figure 4. Correlation of volcanic beds in the Valjala, Viki, Pahapilli, Nässumaa, Lõetsa and Kirikuküla cores. Depths in metres and numbers of volcanic beds (in circles) attributed to distinct eruptions are marked on the sections. Shaded depth intervals represent unsampled boundary intervals of the biozones.

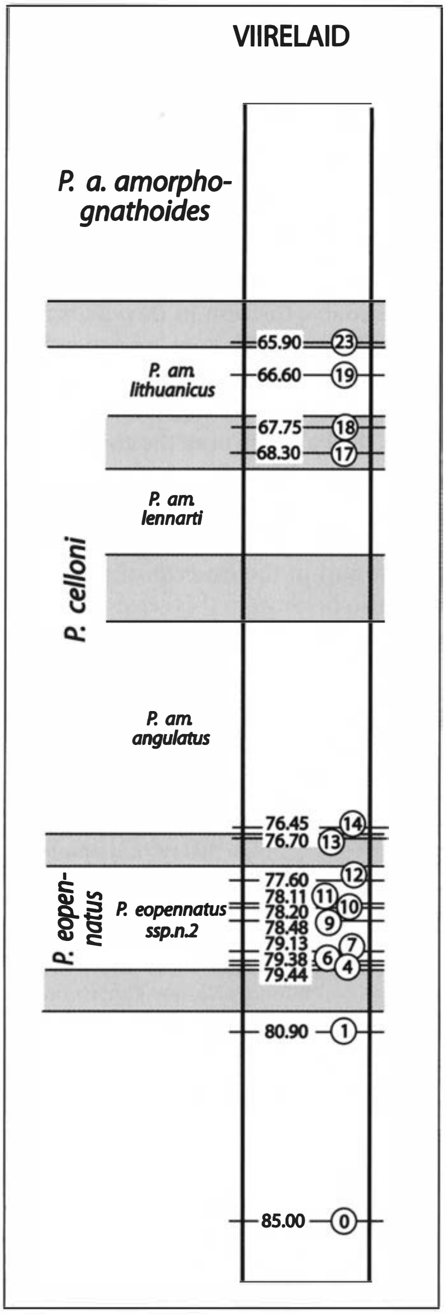
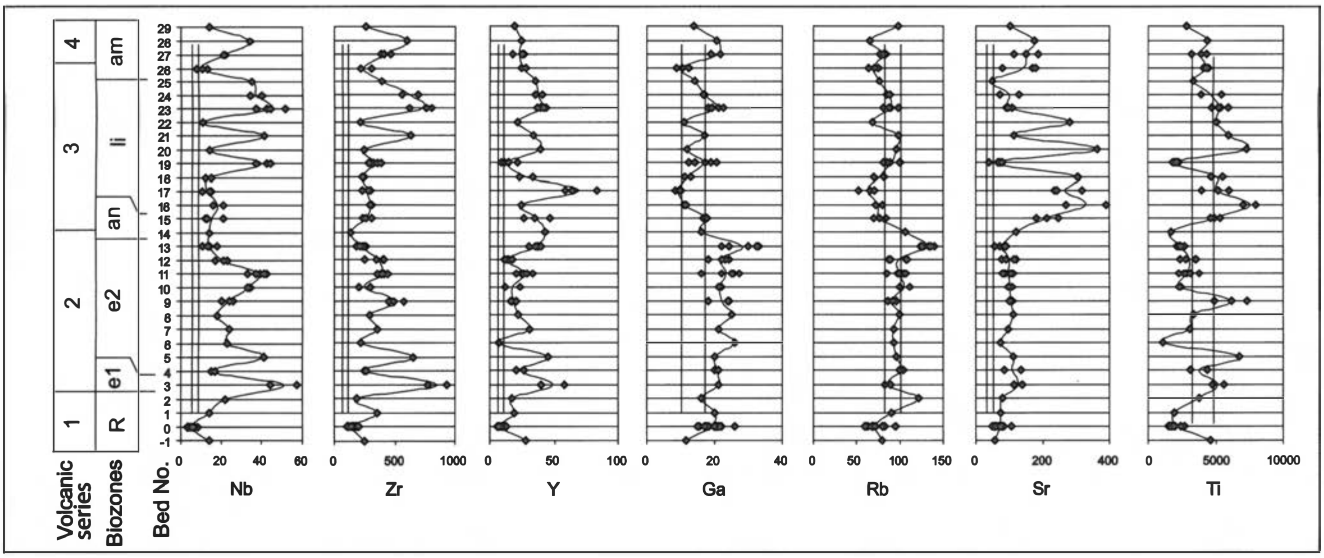


Figure 5. Volcanic beds and conodont biozones in the Viirelaid section (see Figure 4 for explanations).

same core and clearly from different stratigraphical levels. Accordingly, ash from different eruptions may be characterised by similar chemical features and cannot be distinguished on the basis of studied trace elements only. The results were plotted against the conodont zonation established in the studied cores to test the correlations suggested by geochemical data (Figs 4 and 5). Additional restrictions arise in the process of correlation: each pair of correlated samples between two cores eliminates many geochemically potentially acceptable correlations due to the illegality of cross-correlations.

As a result of careful chemical and stratigraphical correlation, we distinguished 31 separate volcanic eruptions, although the maximum number of individual volcanic beds recognized in one section is 20 (Viki core – Fig. 4). Several volcanic beds with distinct chemical composition were identified in one section only, and it is not yet possible to establish their exact stratigraphical position in the sequence relative to other cores. The volcanic beds can be absent in sections due to sedimentary hiatuses, or these beds can be lost in the drilling process or missed during the description and sampling of cores. Beds are numbered from -1 to 29. Zero (0) is assigned to the 10–25 cm thick bed in the upper part of the Rumba Formation. This bed can be easily traced in all investigated cores and is well known among Estonian geologists as the "O" bed. Bergström *et al.* (1998) called this bed the Osmundsberg K-bentonite. All volcanic beds can be grouped according to their stratigraphic positions into four series occurring roughly in the Rumba Formation (1), in the *P. eopennatus* Superzone (2), in the *P. a. lithuanicus* Zone (3) and in the upper part of the *P. a. amorphognathoides* Zonal Group (4) (Figure 6).

Figure 6. Trace element abundance (in ppm) in volcanic beds ordered according to the succession of eruptions. The vertical axis indicates the numbers attributed to the volcanic beds. Two vertical lines mark the interval of range of trace elements in 8 samples of terrigenous claystones from the Viki core. R-Rumba formation, el-eopennatus 1, e2-eopennatus 2, an-angulatus, li-lithuanicus, am-amorphognathoides.



1. In the Rumba Formation four separate volcanic beds were found, although no more than two were present in one core section. In all studied sections the thick Osmundsberg bed (up to 25 cm) can be traced readily. Others were considerably thinner (0.5–2 cm) and were discovered only in a single core. The stratigraphic position of these beds is clear relative to the Osmundsberg bed, which occurs in all cores. The relative positions of beds 1 and 2 could not be proven as the beds occur in different cores. All beds, especially the Osmundsberg one, are characterized by low contents of trace elements. With the exception of a sample from the Viki core 184.35 m, all beds were represented by hard, light-coloured feldspathites.
2. The second volcanic bed series starts with bed 3 in the *P. eopennatus* ssp. n.1 Zone. This bed, characterized by maximum contents of Nb and Zr, was discovered only in the Viki and Pahapilli sections located in the northwestern part of the study area. This corresponds well to biostratigraphical data, indicating that the *P. eopennatus* ssp. n.1 Zone thins out eastward. Higher up these sections, beds with relatively high Nb and Zr occur repeatedly (beds 5, 9, 10, 11). Other beds are characterized by low content of all trace elements, similar to the beds from the Rumba Formation. Bed 13, found in five cores close to the upper boundary of the *P. eopennatus* Superzone, can be easily distinguished from others by very high Rb values, exceptionally strong biotite reflections on bulk sample diffractograms and a high content (about 20 %) of the coarse fraction in clay. Beds 5, 6, 7 and 8 are found only in a single core, and therefore their real succession between beds 4 and 9 is not proven. Beds 7 and 8 have very similar trace element composition, but in the Pahapilli core the coarse fraction of bed 8 (68.5 m) contains abundant magnetite, which is not discovered in other beds. We therefore consider that these beds originated from different eruptions. Bed 14, found in the lowermost part of *P. a. angulatus* Zone, also belongs to this series. All beds in this series consist of soft clay, except two from Viirelaid core.

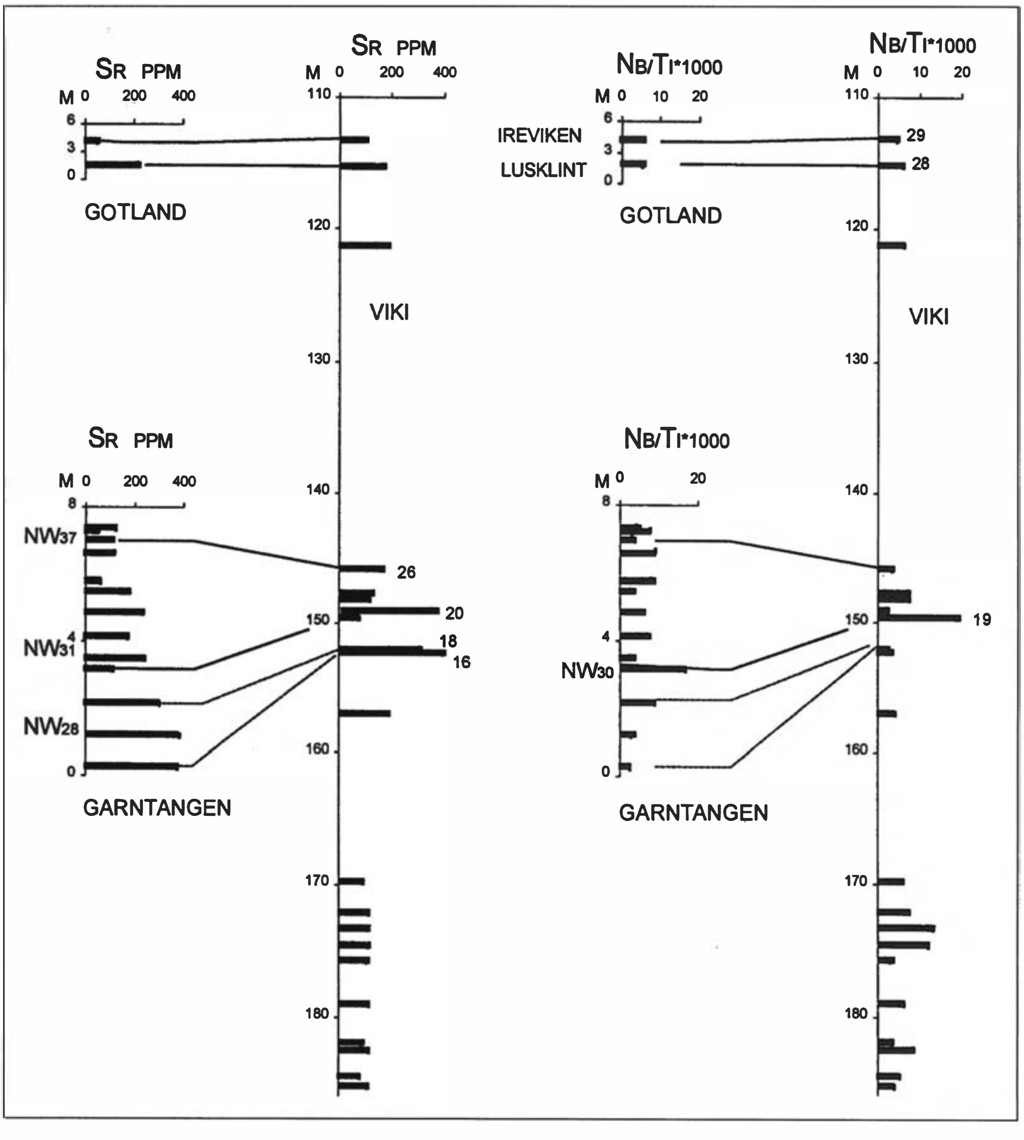


Figure 7. Geochemical correlation between Viki, Garntangen and Gotland. Note that vertical scale in Garntangen is double of that in Viki. High Sr content in the lower part of the Garntangen section and uniqely high Nb, low Ti, Y, Zr bed (NW30 – bed 19) indicate correlation to the middle part of Viki section. Bed 29, not found in Viki, is taken from the Valjala core (113.8m) and placed to the depth 113m in Viki, where it must be according to biostratigraphy and comparison with Gotland. Stippled lines correlate groups of beds, solid lines single beds. Bed numbers for Garntangen according to Batchelor et al. (1995).

3. The third volcanic bed series begins with bed 15 in the upper part of *P. a. angulatus* Zone and ends with bed 26 in the lower part of *P. a. amorphognathoides* Zone. All other beds occur within the *P. a. lithuanicus* zone. In this group of beds two very distinctive chemical types can be distinguished. The lower part of the series is dominated by beds with high Ti and Sr, and low Nb and Zr content (beds 15, 16, 17, 18, 20, 22 and 26). On the bulk sample diffractograms clear apatite reflections commonly occur, and this fact is supported by high phosphorus content in the bulk analyses. In the discrimination diagram of Winchester & Floyd (1977) these beds cluster near the andesite/dacite boundary (Fig. 8). Bed 17 exhibits distinctively high Y content and bed 26 has the lowest Sr content of this type. With the exception of bed 15, these beds occur in cores as hard light-coloured feldspathites. The other bed type is characterized by high Nb and Zr content and occurs as soft clays in the middle and upper part of this series. We distinguished four beds of this type in the upper part (21, 23, 24 and 25), although in a single section only two beds were established. It must be noted that unquestionable correlation of these beds with others of the same chemical type is not possible due to similar composition, although they are clearly distinctive from other types within this series. Bed 19 is chemically unique, being characterized by high Nb, but low Zr, Y and Ti content. This bed was recognized in five sections.
4. The fourth volcanic bed series includes three beds (27, 28 and 29) occurring in the upper part of *P. a. amorphognathoides* Zonal Group. Beds 27 and 28 are chemically similar exhibiting moderate concentrations of trace elements, but bed 29 differs considerably having low contents of trace elements. Bed 29 comes from the Ireviken Event interval.

Correlation with Gotland and Norway

The chemistry of volcanic beds in the Llandovery–Wenlock boundary interval from outcrops in Gotland was described by Batchelor & Jeppsson (1994). Two volcanic beds, named the Lusklint Bentonite and the Ireviken Bentonite, separated by a vertical distance of about 2.5–3 m in northern Gotland, were studied. On the basis of trace element composition, they can be easily recognized in several outcrops. The Ireviken Bentonite is characterized by low content of Nb, Zr, Y, Ti and Ga, and is geochemically closest to our bed 29 (Valjala, 113.8 m). The Lusklint Bentonite has high Ti, Zr and Sr, indicating similarities with bed 28 (Viki, 115.0 m). The comparison, based on the results of St. Andrews' analyses of Zr/Nb, Ti/Nb, Zr/Ti, Nb/Y, Ga*10/Y, Rb*100/(Rb+Sr) ratios, confirms the correlation between the Lusklint Bentonite and bed 28 in Estonia. However, bed 27 in the Viki core (121.05 m) is also similar to the Lusklint Bentonite. Biostratigraphically, both beds (Viki, 115.0 m and Viki, 121.05 m), as well as the Lusklint bed in Gotland come from the upper part of the *P. a. amorphognathoides* Zone and, therefore, both correlations are possible.

The composition of volcanic beds in the Garntangen section (Oslo region, Norway) was investigated by Batchelor *et al.* (1995). Thirteen metabentonites were described from a 7 m section. The contents of trace elements in bulk sample and in apatite phenocrysts allowed two source magmas to be distinguished: an evolved magma with high content of Nb, Zr and Th, and a less evolved ('primitive') magma with low Nb, Zr and high Ni content. The Garntangen Member brachiopods *Pentameroides* and *Costistricklandia* suggest correlation with the low C5 of the British sequence (Worsley et al. 1983). Data from the other regions in Norway indicate that the upper part of the Garntangen Member, which hosts the metabentonites, correlates with the lower part of the *P. a. amorphognathoides* Zone (Batchelor *et al.* 1995). However, according to the revised taxonomy of *Pterospathodus* (Männik 1998a), the taxa identified in Norway as the oldest representatives of *P. a. amorphognathoides* (Nakrem 1986) in reality belong to the *P. a. lithuanicus.* Accordingly, it is most probable that the metabentonites in Garntangen come from the interval corresponding to the *P. a. lithuanicus* Zone.

A recent study of Sr isotopes in apatite from Garntangen metabentonites suggests a correlation of this section with the *M. spiralis* graptolite Zone (Batchelor & Evans 2000). It is a further indication that the strata in the Garntangen section are no younger than the *P. a. lithuanicus* Zone as the *M. spiralis* Zone corresponds to the upper part of the *P. celloni* Superzone (Loydell et al. 1998). In Estonia this stratigraphical interval contains 10 volcanic beds (Fig. 4).

The possibility that the Garntangen metabentonites belong to the *P. a. lithuanicus* Zone is also indicated by the geochemical data. Two different source magmas have been recognized in this interval in Estonia. The lower parts of both sections (from the base up to 3.6 m in Garntangen and from 151.8 to 148.8 m in the Viki core) are characterized mainly by the source magma of the 'less evolved' type, whereas the upper part comprises volcanic beds belonging to the 'evolved' set. A comparision of Sr concentrations in Garntangen and Viki is shown in Fig. 7. High Sr content uniquely characterizes volcanic beds from the third volcanic series of the Viki section and compare favourably with the lower part of the Garntangen section. A unique sample with high Nb, low Zr, Y, Ti (bed 19 in Estonia) can be related to sample NW30 in the Garntangen section (Fig. 7). The geochemical similarity (less evolved type with low Sr) of bed NW37 from Garntangen with bed 26 in Estonia indicates that the upper part of the Garntangen section most probably correlates with the lowermost *P. a. amorphognathoides* Zone.

Although two eruption types can be recognized in both sections, more detailed correlation of the beds under discussion is complicated due to poorly studied within-bed variations and some general differences in trace element contents between Estonia and Norway. The main difference is the content of Y and Rb are twice to three times higher in Norway. These differences may be caused by: 1) different diagenesis, 2) different distances from the source volcanoes causing differentiation of minerals in the dust cloud or 3) different volcanic sources for some beds. It is not yet possible to tell which of the above reasons is responsible for the differences noted.

Petrogenesis of magmas

A broad classification of the Estonian altered volcanic ashes can be achieved by plotting the data on the Winchester & Floyd (1977) volcanic rock discrimination diagram (Fig. 8). The source magma compositions suggested from the diagram range from rhyodacite to trachyandesite and alkaline trachyte. Regular variations through the Telychian occur in the altered volcanic ash composition (Figs. 6 and 8).

Llandovery volcanism took place at a time when Eastern Avalonia was colliding with Baltica and the Tornquist Sea was closing, and the Wenlock is regarded as the time when the Iapetus and Tornquist Oceans finally closed (Williams *et al.* 1992; Trench & Torsvik 1992; Soper *et al.* 1992).

Alkaline magmas are commonly generated in an extensional environment associated with localized strike-slip tectonics at the continental margins. Rhyolite, comendite and trachyte clasts have been described from a basal Wenlock conglomerate from the Hagshaw Hills Silurian inlier in the Midland Valley of Scotland (Heinz & Loeschke 1988). An alkaline quartz keratophyre is known from the Llandovery–Wenlock succession at Lough Mask, Co. Mayo, Ireland, and could be related to the sinistral strike-slip tectonics described by Williams & Harper (1988).

The chemical composition of apatite phenocrysts in the Telychian Lusklint Bentonite from Gotland suggested that potassic magma may have been responsible for its formation (Batchelor & Jeppsson 1994). A waning subduction zone, with thickening crust, can generate melts which interact extensively with thick crust during their ascent, generating magmas enriched in K and lithophile elements, such as the HFSE (Zr, Nb, Th). This scenario could also have given rise to the alkaline source magma suite. In contrast, the rhyodacite magmatism represents a collisional tectonomagmatism generating calc-alkaline magmas.

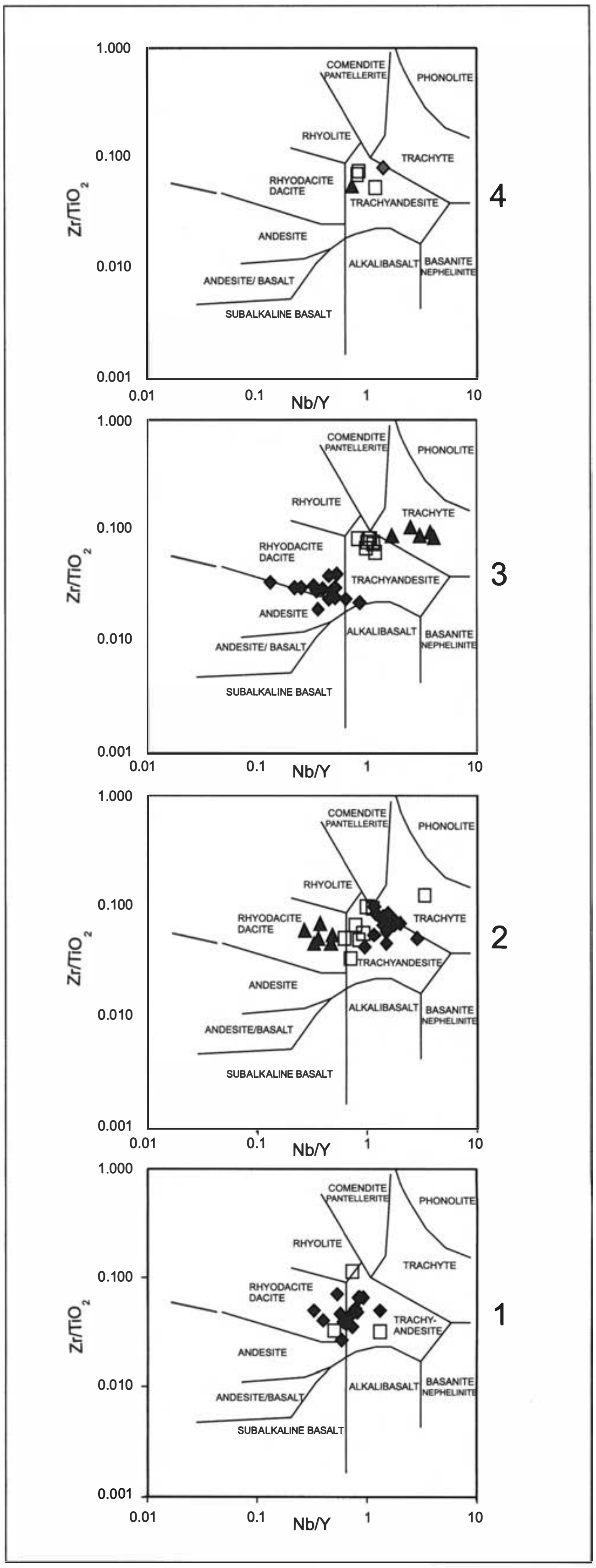


Figure 8. Classification diagrams for volcanic rocks according to Winchester & Floyd (1977). Numbers to the right represent volcanic bed series. 1. Rumba series, filled rhombs mark the Osmundsberg bed, open quadrangles other beds. 2. P. eopennatus series, open quadrangles represent beds 3–7, filled rhombs beds 8–12, filled triangles beds 13 and 14. 3. P. a. lithuanicus series, open quadrangles represent evolved magma type, filled triangles bed 19, and filled rhombs represent beds derived from a less evolved magma source. 4. Upper P. a. amorphognathoides Zonal Group series, open quadrangles bed 27, filled triangle bed 29 and filled rhomb bed 28.

Volcanic sources

Silurian volcanism is known in the Brabant massif of Belgium where calc-alkaline volcanism is believed to have ended in the Wenlock with an acidic ash fall (André *et al.* 1986). Extrusive silicic volcanism is also recorded in late Llandovery – early Wenlock times in the Neuville-sous-Huy area of Belgium (Verniers & Van Grootel 1991). However, being at a distance of approx. 1400 km to the SW, these Belgian sources may be unlikely candidates for providing the ash falls on Saaremaa. Ludlow volcanism has been described along the closing Tornquist Suture Zone at Klodzko in Poland (Oliver *et al.* 1993), about 1050 km SSW of Saaremaa. The Tornquist–Teysseyre Zone, the northernmost expression of the Trans-European Suture Zone, lies 650 km SW of Saaremaa, while the Elbe Line, which is now believed to mark the SW margin of Baltica, is 650 km further away to the SW (Cocks *et al.* 1997). Llandovery volcanic activity in Norway is a feasible source for some volcanic beds (Pedersen *et al.* 1991), and Wenlock volcanic beds are known from the Steinsfjord Formation, near Oslo (Jørgensen 1964).

The most likely source area for the Baltic volcanic beds is located in the Tornquist–Teysseyre zone, which lies to the south and south-west of Saaremaa. An unknown factor in determining the source of ash production is the influence of wind direction on volcanic ash distribution. Generating isopach maps of volcanic ash bed distribution is fraught with problems; thickness can be affected by post-depositional reworking or tectonic attenuation, so it can be difficult to trace a single clay band for any distance. A mean thickness for many beds from the same volcano can provide a clue to a relative distance from the ash source. It has been established that there were two sources for the Estonian and Garntangen (Norway) volcanic beds: one designated as 'less evolved' and the other as 'evolved' magma. Considering only the beds assigned to the *P. a. lithuanicus* and to the lowermost *P. a. amorphognathoides* zones, a mean thickness is calculated for both localities (Table 4). Assuming a common volcanic source, this suggests that the source of "less evolved" magma was closer to Garntangen than to Saaremaa whereas the source of "evolved" magma appears to have been equidistant from both localities. Bergström *et al.* (1998) correlated the Osmundsberg K-bentonite over North Europe. Considering that this bed reaches up to 1 m in thickness in central Sweden, 10–25 cm in Estonia, and has not been identified in Latvia (supposedly thin), one might assume a source located to the north-west.

Conclusions

1. In total, 31 different volcanic eruptions were recognised. Fourteen of them are represented by volcanic beds, each of which was recognized in only one of the sections. Fifty-seven volcanic beds, representing the remaining fifteen eruptions, can be identified in two or more sections.
2. Although volcanic beds occur throughout the studied interval, it is evident that during *P. eopennatus* and *P. a. lithuanicus* times, the volcanic activity in the Northern Iapetus region was considerably higher than in other times during the Telychian. Twenty-two volcanic beds out of a total of 31 come from these intervals.
3. Concentrations of Nb, Zr, Y, Ga, Rb, Sr, Ti and element ratios appear to be useful correlation tools in the Estonian sections. Comparison with Norway revealed 2–3 times higher Rb and Y values there. Possibly some fractionation of material in air transport or different diagenetic pathways occurred. Cluster analysis is useful for revealing correlations. However, ash from different eruptions could be characterized by similar chemical features and, therefore, cannot be distinguished on the basis of the studied trace elements only. Biostratigraphical control is necessary to constrain the volcanic bed correlations.
4. Two types of eruptions were recognized in both Garntangen, Norway (Batchelor *et al.* 1995) and in the *P. a. lithuanicus* Zone in Estonia, allowing limited correlation. With caution, two beds near the Llandovery–Wenlock boundary can be correlated between Estonia and Gotland.
5. Comparison of the mean thicknesses of volcanic beds reveals that the source of the "evolved" magma was probably equidistant from Garntangen and Estonia. The source of the "less evolved" magma was closer to Garntangen than to Estonia.

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Table 4: Comparision of average thickness of volcanic beds from Norway and Estonia

	Garntangen, Norway		Saaremaa, Estonia	
	Less evolved	Evolved	Less evolved	Evolved
Number of beds	5	6	9	9
Mean thickness (mm)	39	39	18	38

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