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Sedimentary cyclicity and dolomitization of the Raikküla Formation in the Nurme drill core (Silurian, Estonia)

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Abstract. Medium-rank sedimentary cyclicity of the Raikküla Formation (Raikküla Regional Stage, middle Llandovery) is first described in the Nurme drill core (mid-western Estonia). Four shallowing up submesocycles are distinguished, which group in pairs into two mesocycles, treated as the Lower and Upper Raikküla subformations. In the Nurme drill section, totally dolomitized carbonate rocks of the Raikküla Formation are sandwiched between the unaltered limestones of the Juuru and Adavere regional stages. It suggests early dolomitization during the Raikküla Age soon after deposition. X-ray diffraction, X-ray fluorescence, and titration analyses were applied to study the composition of rocks and lattice parameters of dolomite. Geochemistry of rocks is consistent with dolomitization by normal marine water. No evidence of hypersalinity of the sedimentary environment or inflow of external fluids has been observed. The results of d_{104} measurements showed a trend of growing dolomite stoichiometry towards the top of the regressive, shallowing up submesocycles. The most completely ordered dolomite (< 51 mol% $CaCO_3$) occurs in the intervals of primarily bioclastic, winnowed sediments, which indicates the promoting role of the agitated-water environment in the dolomitization process. Extensive pervasive dolomitization associated with regressive phases of the evolution of the Baltic Palaeobasin in the Silurian and was related to a zone of shallow normal-saline inner shelf facies, migrating in space in accordance with sea level changes.

Key words: Silurian, dolomitization, sedimentary cyclicity, dolomite, X-ray diffraction, Estonia.

INTRODUCTION

Complex studies of massive pervasive secondary dolomitization of Silurian rocks (Teedumäe et al. 1999, 2001, 2003) have shown that dolomitization associates with the regressive phases of the evolution of the Baltic Palaeobasin and is related to the restricted area of inner shelf facies. Dolomitization has changed the primary composition of the sediments but relict lithological textures suggest its diagenetic origin. Studies of spatial dolomitization of primarily normal-saline calcareous sediments in the same area (Vishnyakov 1956; Jürgenson 1970; Kiipli 1983; Bityukova et al. 1996, 1998; Shogenova 1999) in general support its diagenetic origin with differences in details. Various sources of magnesium have been supposed for the Silurian secondary dolomites: Devonian sediments (Jürgenson 1970), hypersaline lagoonal waters of the Devonian basin (Vishnyakov 1956), mixed fresh ground water and marine water of the Devonian basin (Kiipli 1983), and contemporaneous Silurian seawater (Teedumäe et al. 1999, 2001, 2003).

An extensive body of dolomites cross-cuts depositional sequences of the Raikküla and Nurmekund formations of the Raikküla Regional Stage (middle Llandovery). The pervasively dolomitized rocks of the Raikküla Stage are commonly underlain by the limestones of the Juuru Regional Stage. To the south the rocks are overlapped by the limestones or secondary dolostones of the Rumba Formation of the Adavere Regional Stage, or by the Devonian clastic sediments (Figs. 1, 2). In several drill sections (e.g. Nurme, Valgu) the totally dolomitized carbonate rocks of the Raikküla Stage are sandwiched between the undolomitized limestones of the Juuru and Adavere stages. This suggests the Raikküla Age for the diagenetic dolomitization, as was also supposed in previous studies on separate outcrop sections (Teedumäe et al. 2001, 2003).

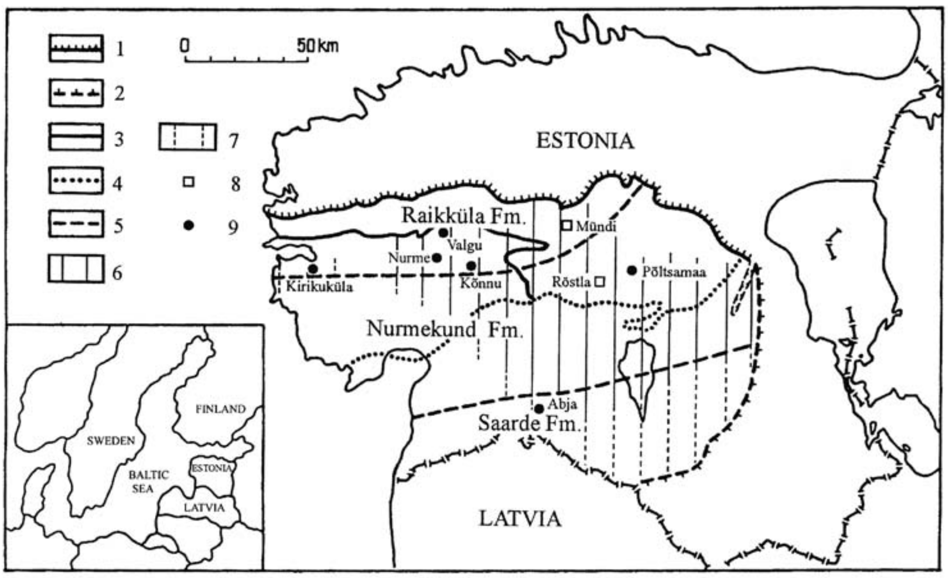


Fig. 1. Location map, showing the distribution of the formations of the Raikküla Stage in mainland Estonia and the area of pervasive dolomitization. 1, outline of the outcrop area; 2, outline of the subsurface distribution of the stage; 3, limit of the overlying Adavere Stage; 4, limit of the Devonian cover; 5, boundary between the distribution areas of formations; 6, area of replacive (pervasive) dolomitization; 7, dolomitization only at the upper contact of the stage; 8, outcrop; 9, borehole.

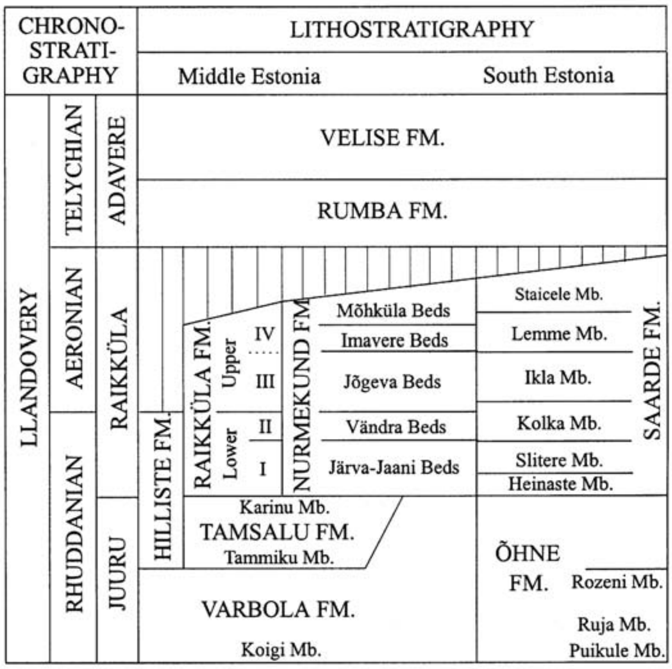


Fig. 2. Stratigraphical chart of the Juuru and Raikküla stages in Estonia (after Nestor et al. 2003).

In the Nurme drill core the whole section of the Raikküla Formation is dolomitized and lies between the unaltered limestones of the Juuru and Adavere stages. The main objective of the research was complex study of the variability of the composition of dolostones, stoichiometry of dolomite and the nexus of these properties with the facies and sedimentary cyclicity in order to understand the evolution and timing of the process of dolomitization.

A. Teedumäe studied the aspects of dolomitization, H. Nestor prepared the description of the studied section, made stratigraphical research and cyclicity analyses, T. Kallaste performed the X-ray diffractometry and X-ray fluorescence measurements. Titration and gravimetric analyses were made at the Central Laboratory of the Geological Survey of Estonia.

MATERIAL AND METHODS

Forty samples (Table 1) were collected from the Nurme drill core for complex study of the chemical composition and X-ray diffractometry. Of these, 33 samples are from the middle Llandovery Raikküla Formation, 3 samples from the underlying Tamsalu Formation, and 4 samples from the overlying Rumba Formation (Fig. 3). Samples were collected by lithological varieties, considering the changes within a variety.

CaO and MgO were analysed by titration, insoluble residue (henceforward i.r.) was determined by gravimetric analyses. Fe_2O_3 (total), Mn, and Sr were analysed by the X-ray fluorescence method with the VRA-30 analyser using an X-ray tube with Mo anode at 50 kV and 20 mA. Calibration of Mn and Fe was based on internationally intercalibrated dolomite reference materials Es-4 and Es-11 without matrix corrections. In calibration of Sr some additional silicate and limestone reference materials were used and, accordingly, matrix corrections (Compton scattering method) were applied. The precision of analyses was determined from 10 replicate measurements: Fe_2O_3 ±0.005%, MnO ±0.005%, Sr ±2 ppm.

XRD measurements were carried out on a diffractometer HZG4, using Fe-filtered Co radiation. The rock powder was mixed in a mortar with Si in the ratio of 8 : 2, some drops of ethanol were added, and the mixture was evenly spread on a glass slide. The measured angular range 32–38 °2θ reveals the 104 reflection of dolomite (d_{104}) and calcite and 111 reflection of Si. The positions of reflections were calculated as weighted average. The instrumental shift was corrected according to the Si reflection (3.1355). The precision of the measurement of d_{104} is ±0.0005 Å.

The molar concentration of $CaCO_3$ (m_{Ca}) in dolomite (Table 1) was calculated by measuring the displacement of the d_{104} peak relative to a standard (e.g. Lippmann 1973). The formula (1)

$$m_{\mathrm{Ca}} = \frac{(d_{104} - 2.8840)}{0.003} + 50 \qquad (1)$$

expresses the linear dependence of d_{104} reflection with respect to the fix-point of ideal stoichiometric dolomite, the value of which (2.8840 Å) was calculated (Teedumäe et al. 1999) on the ground of the composition of two standards, Es-4 (Estonia) and SI-1 (former USSR). No siderite or rhodochrosite was revealed. Some samples showed slight traces of calcite (Table 1).

GEOLOGICAL SETTING

During the Silurian, Estonia was situated at the northern flank of the pericratonic Palaeobaltic basin characterized by the carbonate to fine-clastic type of sedimentation (Nestor & Einasto 1977, 1997). According to the latest stratigraphical charts (Nestor 1995, 1997) the Raikküla Regional Stage (middle Llandovery) is represented in Estonia by the Hilliste (partially), Raikküla, Nurmekund, and Saarde formations, laterally replacing one another in the southern direction (Figs. 1, 2). The most characteristic features of this stratigraphical interval are: (1) preservation of very shallow-water, nearshore deposits at the northern margin of the basin, which suggests a regressive phase of the basin development (Nestor & Einasto 1997); (2) low content of the clay material; (3) cyclic alternation of comparatively pure, almost barren micritic (micro- to cryptocrystalline) limestones

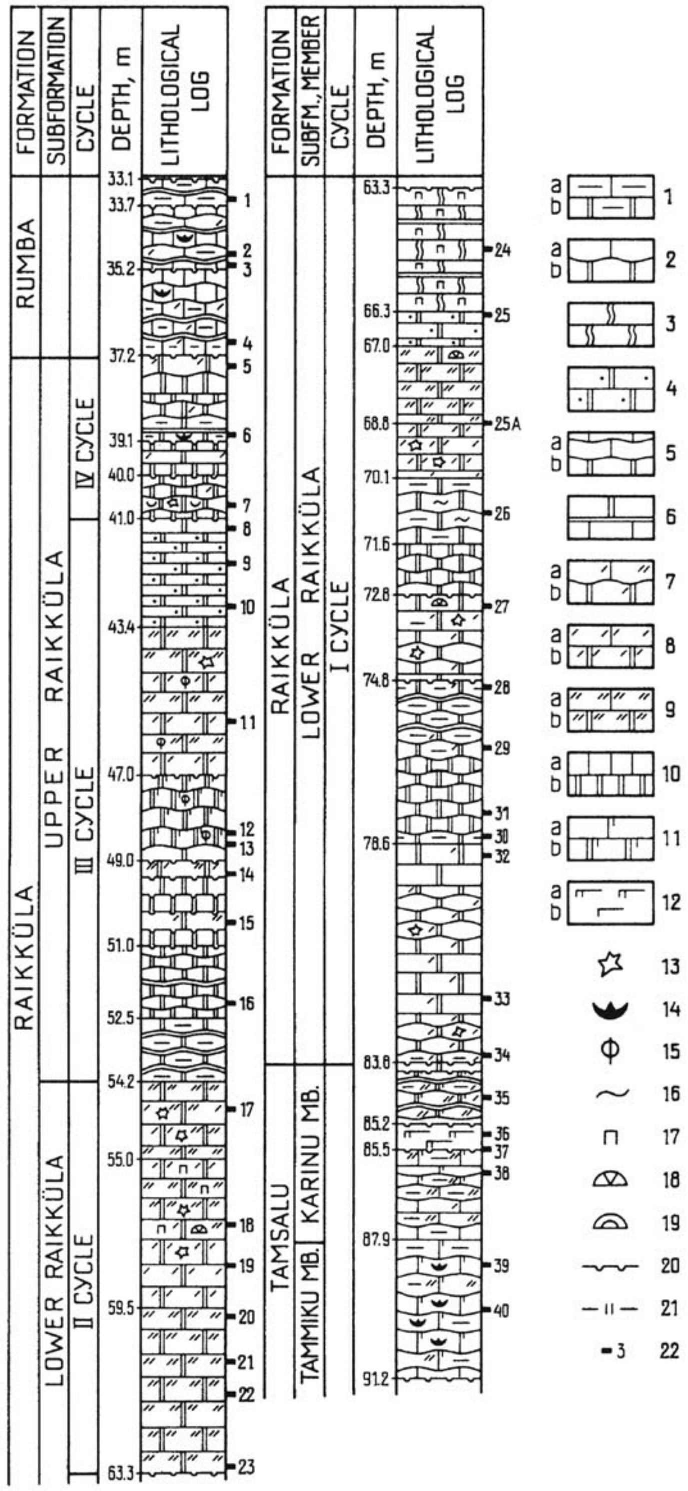
Table 1. Major, minor, and trace element composition of rocks and d_{104} values of dolomite

Sample number	Depth, m	Titration analyses		Gravimetric analyses	X-ray fluorescence analyses				X-ray diffraction analyses			Rock type (Vingisaar et al. 1965)
		CaO, %	MgO, %	Insoluble residue, %	Fe_2O_3, %	Mn, ppm	Br, ppm	Sr, ppm	d_{104}, Å	$CaCO_3$, mol%	Calcite	
1	33.60	39.75	3.01	20.86	1.39	172	4	184			++	Clayey limestone
2	34.80	32.01	3.44	33.10	1.67	150	3	169			++	Marlstone
3	35.10	42.25	3.09	15.60	1.04	142	4	243			++	Clayey limestone
4	36.90	36.49	3.27	25.46	1.45	165	3	273			++	Marlstone
5	37.30	30.37	16.93	8.62	1.09	505	5	67	2.8973	54.4	(+)	Dolostone
6	39.00	27.54	15.99	13.50	5.46	482	3	52	2.8954	53.8	+	Clayey dolostone
7	40.70	29.19	19.60	4.26	0.69	466	15	37	2.8875	51.2	(–)	Dolostone
8	41.10	28.95	18.22	7.98	0.79	445	18	41	2.8888	51.6	(–)	Dolostone
9	42.00	29.31	19.00	5.68	0.66	389	23	37	2.8872	51.1	+	Dolostone
10	43.00	29.54	20.37	4.28	0.56	417	27	38	2.8864	50.8	(–)	Dolostone
11	45.70	28.72	20.28	3.64	0.67	453	15	31	2.8863	50.8	(–)	Dolostone
12	48.28	36.25	14.70	3.84	0.61	433	11	46	2.8866	50.9	++	Calcitic dolostone
13	48.40	29.01	20.34	4.32	0.65	462	16	32	2.8859	50.6	(–)	Dolostone
14	49.40	29.78	19.85	3.56	0.56	508	15	31	2.8860	50.7	(+)	Dolostone
15	50.40	29.66	20.20	3.42	0.65	568	12	36	2.8860	50.7	(–)	Dolostone
16	52.30	25.66	17.02	15.78	1.21	476	20	51	2.8873	51.1	(–)	Clayey dolostone
17	54.80	29.43	20.71	2.42	0.66	601	16	29	2.8861	50.7	(–)	Dolostone
18	57.50	29.54	19.85	4.52	0.60	452	22	35	2.8863	50.8	(–)	Dolostone
19	58.50	29.31	20.97	2.06	0.43	517	34	37	2.8862	50.7	(–)	Dolostone
20	59.70	29.90	20.71	1.84	0.45	522	26	41	2.8871	51.0	(–)	Dolostone
21	60.60	28.37	19.94	6.80	0.61	580	25	31	2.8863	50.8	+	Dolostone
22	61.50	29.66	20.63	2.08	0.46	527	33	44	2.8866	50.9	(–)	Dolostone

Table 1. *Continued*

Sample number	Depth, m	Titration analyses		Gravimetric analyses	X-ray fluorescence analyses				X-ray diffraction analyses			Rock type (Vingisaar et al. 1965)
		CaO, %	MgO, %	Insoluble residue, %	Fe_2O_3, %	Mn, ppm	Br, ppm	Sr, ppm	d_{104}, Å	$CaCO_3$, mol%	Calcite	
23	63.20	29.90	21.14	1.16	0.69	507	19	31	2.8857	50.6	(–)	Dolostone
24	64.80	28.95	20.37	4.58	0.66	445	20	39	2.8863	50.8	+	Dolostone
25	66.40	29.07	19.17	5.76	0.59	460	18	36	2.8866	50.9	+	Dolostone
25A	68.80	30.48	20.63	1.78	0.41	516	23	40	2.8875	51.2	(–)	Dolostone
26	70.90	28.37	19.51	6.20	0.77	445	19	39	2.8870	51.0	(–)	Dolostone
27	73.00	28.95	20.43	4.26	0.49	509	15	32	2.8858	50.6	(+)	Dolostone
28	74.90	27.66	15.99	14.52	1.47	444	20	47	2.8881	51.4	(+)	Clayey dolostone
29	76.30	29.07	17.96	9.50	1.06	420	20	42	2.8874	51.1	+	Dolostone
31	77.90	28.38	20.46	3.58	0.48	387	13	30	2.8866	50.9	(–)	Dolostone
30	78.50	27.50	19.53	7.74	0.89	383	27	39	2.8870	51.0	(–)	Dolostone
32	78.90	27.06	19.61	8.38	0.95	470	28	54	2.8883	51.4	(–)	Dolostone
33	82.20	27.94	19.53	4.92	0.85	366	32	44	2.8882	51.4	(–)	Dolostone
34	83.70	29.15	17.05	8.28	1.05	434	5	57	2.8958	53.9	+	Dolostone
35	84.60	33.77	10.70	14.46	1.08	270	6	77	2.8959	54.0	++	Clayey calcitic dolostone
36	85.40	23.98	8.14	34.82	2.69	248	2	74	2.8962	54.1	++	Dolomitic marlstone
37	85.80	26.62	9.22	28.02	2.26	237	3	73	2.8953	53.8	++	Dolomitic marlstone
38	86.35	36.52	6.05	16.62	1.52	194	5	109	2.8961	54.0	++	Clayey dolomitic limestone
39	88.50	39.16	5.66	14.24	1.30	157	6	158	2.8955	53.8	++	Clayey dolomitic limestone
40	90.50	40.26	5.50	12.32	0.98	153	6	208	2.8939	53.3	++	Clayey dolomitic limestone

(–) below the detection limit; (+) <0.5%; + order of values 1%; ++ high in calcite.



FORMATION
SUBFORMATION
CYCLE
DEPTH, m
LITHOLOGICAL LOG
RUMBA
RAIKKÜLA
UPPER RAIKKÜLA
LOWER RAIKKÜLA
IV CYCLE
III CYCLE
II CYCLE
33.1
33.7
35.2
37.2
39.1
40.0
41.0
43.4
47.0
49.0
51.0
52.5
54.2
55.0
59.5
63.3
FORMATION
SUBFM., MEMBER
CYCLE
DEPTH, m
LITHOLOGICAL LOG
RAIKKÜLA
LOWER RAIKKÜLA
I CYCLE
TAMSALU
KARINU MB.
TAMMIKU MB.
63.3
66.3
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or secondary dolostones with bioclastic limestones or marlstones, or their dolomitized analogues; (4) extensive pervasive dolomitization of rocks in central and eastern Estonia.

The barren micritic limestones or secondary dolostones cyclically alternate with marlstones in the Saarde Formation and with bioclastic limestones (packstones-wackestones) or their dolomitized analogues in the Nurmekund Formation (Nestor et al. 2003). Besides sea-level fluctuation, the sedimentary cyclicity of the meso-cycle level was probably induced also by alternation of arid and humid climate states (Nestor et al. 2003). Cyclicity in the Raikküla Formation is expressed by the alternation of micritic and bioclastic limestones (or their dolomitized analogues), and sedimentary dolostones, more thoroughly characterized below. The Hilliste Formation, which spreads only in northwestern Estonia, is dominated by pelmatozoan grainstones with bioherms. It corresponds to the uppermost Juuru and lower half of the Raikküla stages (Nestor 1995, 1997).

The stratigraphical subdivision of the formations in the Raikküla Stage is based on the cyclic alternation of rock types. The Nurmekund Formation is subdivided into more heterogeneous informal stratigraphical units: the Järva-Jaani, Vändra, Jõgeva, Imavere, and Mõhküla beds (Fig. 2), each corresponding to a submeso-cycle. The Saarde Formation consists of more homogeneous lithostratigraphical units: the Heinaste, Slitere, Kolka, Ikla, Lemme, and Staicele members representing parts of the mesocycles (Nestor et al. 2003). The Raikküla Formation is divided only into the Lower and Upper Raikküla subformations (Nestor 1995, 1997), which correspond to the shallowing upward sedimentary mesocycles. The Lower Raikküla Subformation belongs to the Rhuddanian, the Upper Raikküla Subformation to the Aeronian (Nestor 1995).

The end of the Raikküla Age marked the termination of the differentiation stage in the development of the Baltic Palaeobasin, and also the end of the Lower–Middle Llandovery macrocycle of sedimentation accompanied by extensive regression (Nestor & Einasto 1997). It culminated with an extensive sedimentation break and denudation along the basin margins, in particular in western Estonia, where the gap increases in the northwestern direction so that the entire Upper Raikküla Subformation, corresponding to the Aeronian, is missing in drill cores of northern Saaremaa. The gap embraces also the distribution area of the Hilliste Formation on Hiiumaa Island and adjacent mainland. A pronounced unconformity

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Fig. 3. Geological log of the Raikküla Formation in the Nurme core. Lithological legend: 1, horizontal-bedded clayey (argillaceous) limestone (a) and dolostone (b); 2, nodular limestone (a) and dolostone (b); 3, bioturbated, micritic dolostone; 4, microlaminated dolostone; 5, wavy-bedded limestone (a) and dolostone (b); 6, intercalation of limestone (or dolostone) and marlstone, ca 1 : 1; 7, varigrained skeletal packstone (a) and fine dolowackestone (b); 8, fine grainstone (a) and dolograinstone (b); 9, coarse grainstone (a) and dolograinstone (b); 10, micritic limestone (a) and dolostone (b); 11, dolomitic limestone (a) and calcitic dolostone (b); 12, dolomitic marlstone (a) and marlstone (b); 13, vugs, coarse pores; 14, brachiopod coquina; 15, silica nodules; 16, burrows; 17, pyrite mottles; 18, corals; 19, stromatoporoids; 20, discontinuity surfaces; 21, lower limit of total dolomitization; 22, number of sample.

is developed also on the opposite, southeastern flank of the Palaeobaltic Basin in eastern Lithuania, where the early and middle Llandovery deposits were subjected to denudation all over the carbonate shelf area (Nestor & Einasto 1997). The break had probably a glacio-eustatic origin as it temporally coincided with the Panuara unconformity in New South Wales (Jell & Talent 1989) and elsewhere, as well as with a glaciation in South America (Caputo 1998; Nestor & Nestor 2002a, 2002b). However, the variable span of the deposition break in different areas suggests that partly the break and denudation might have been influenced also by tectonic upheaval, induced by the beginning of the collision of the Laurentia and Baltica continents (Nestor & Einasto 1997).

CYCLICITY OF THE SEQUENCE

Cyclicity in the Nurmekund and Saarde formations of the Raikküla Stage was recently discussed by Nestor et al. (2003). The studied section of the Nurme drill core shows a medium-rank cyclicity, which is rather characteristic of the Raikküla Formation. In this core the rocks of the Raikküla Formation are entirely dolomitized. The formation is under- and overlain by undolomitized limestones of the Tamsalu and Rumba formations, respectively (Fig. 3). Secondary dolomitization has changed the composition of rocks, but the primary lithological characteristics (skeletal remains, etc.) are more or less recognizable and enable identification of the primary origin of the carbonate rocks. This allows application of the textural classification of carbonate rocks (Dunham 1962), by adding the prefix “dolo-”. However, due to dolomitization, the microfossils are very scarce (see V. Nestor 1994, fig. 12/1) and exact determination of the stage boundaries is difficult.

Three main rock types (lithofacies) alternate cyclically in the studied sequence. The lower part of a complete cycle is represented by rough- or wavy-bedded, fine-crystalline, micritic dolostone with a low, vertically variable content of relict skeletal detritus (dolomudstone to dolopackstone), and thin marly intercalations or partings at bedding planes. A more argillaceous layer (up to 1 m thick) of clayish dolostone commonly occurs at the base of such cycles. The type of bedding and content of skeletal and clay material are variable, causing obscure lower-rank cyclicity, but in general the role of the relict skeletal detritus increases upward in the sequence. The rocks of this part of the cycle were formed in the low-energy open shelf environment (facies zone III according to Nestor & Einasto 1977).

The middle part of the cycle is represented by horizontal-bedded, fine- to coarse-crystalline, porous dolograinstone with larger vugs and solution cavities after corals and stromatoporoids in coarse-crystalline intervals and by microlamination in fine-crystalline intervals. The uneven bedding planes are covered with discontinuous, wavy or stylolitic, greenish marly partings. Deposits of the middle part of the cycle were formed in the high-energy shoal environment (facies zone II by Nestor & Einasto 1977) and originally consisted mainly of pelmatozoan skeletal particles.

The upper part of a complete cycle is represented by thin-bedded to laminated, fine-crystalline dolostone with wavy to stylolitic greenish marly partings. The laminated dolostone, in places containing mud cracks, may be replaced by thicker-bedded bioturbated dolomudstone ("pattern dolomite"). This lithofacies was probably formed shorewards of the shoal facies zone, where calcareous silt (or pellets) and mud were alternately deposited in low-energy intertidal conditions (facies zone I by Nestor & Einasto 1977).

In the Nurme section, the Raikküla Formation consists of two complete shallowing upward cycles (I and III in Fig. 3), which contain all three cycle parts described above. They alternate with incomplete cycles (II and IV in Fig. 3) which are lacking some cycle elements.

The first cycle comprises the interval from 63.3 to 83.8 m. Its lower part (70.1–83.8 m) is represented by clayish and micritic dolostones (mainly dolowackestone). The middle part (67.0–70.1 m) consists of pelmatozoan dolograinstone, fine-grained in the lower and coarse-grained in the upper half. The upper part (63.3–67.0 m) of the cycle is formed by laminated and bioturbated dolostones; probable mud cracks occur at a depth of 66.3 m. The first cycle obviously correlates with the Järva-Jaani Beds of the Nurmekund Formation (Fig. 4). The difference is that the cycle includes dolograinstone in the middle part and laminated dolostone in the upper part, but the Järva-Jaani Beds consist of micritic limestone or dolomudstone with only rare interlayers of grainstone tempestites in the uppermost part.

The second cycle, occurring in the interval of 54.2–63.3 m, is incomplete in the studied section. It is represented only by relict pelmatozoan dolograinstone and corresponds to the middle part of a complete cycle. Distinction of the interval as a separate cycle is rather arbitrary and is based on the change of the shallowing upward trend of deposition at both its boundaries. The second cycle is correlatable with the Vändra Beds of the Nurmekund Formation, which consist of nodular wacke- and packstones (or their dolomitized analogues), formed in the low-energy environment of the open shelf facies zone (Nestor & Einasto 1977), whereas the rocks of the present cycle were formed in the high-energy shoal environment. On the other hand, the unit may be treated as a tongue of the Hilliste Formation, consisting of pelmatozoan and reef limestones and spreading in northwestern Estonia (Aaloe & Nestor 1977; Nestor 1997).

The third cycle, in the interval of 41.0–54.2 m, is quite analogous to the first cycle, comprising wavy-bedded micritic dolostones in the lower (47.0–54.2 m), dolograinstones in the middle (43.4–47.0 m), and laminated dolostones with mud cracks in the upper part (41.0–43.4 m). This cycle is correlatable either with the Jõgeva Beds or with the Jõgeva and Imavere beds of the Nurmekund Formation, which both consist of wavy-bedded to nodular micritic dolostones (Nestor et al. 2003). In the latter case, the interval with numerous discontinuity surfaces at 49.0–51.0 m refers to a shallowing event, which may correspond to the regressive, upper part of the Jõgeva Beds.

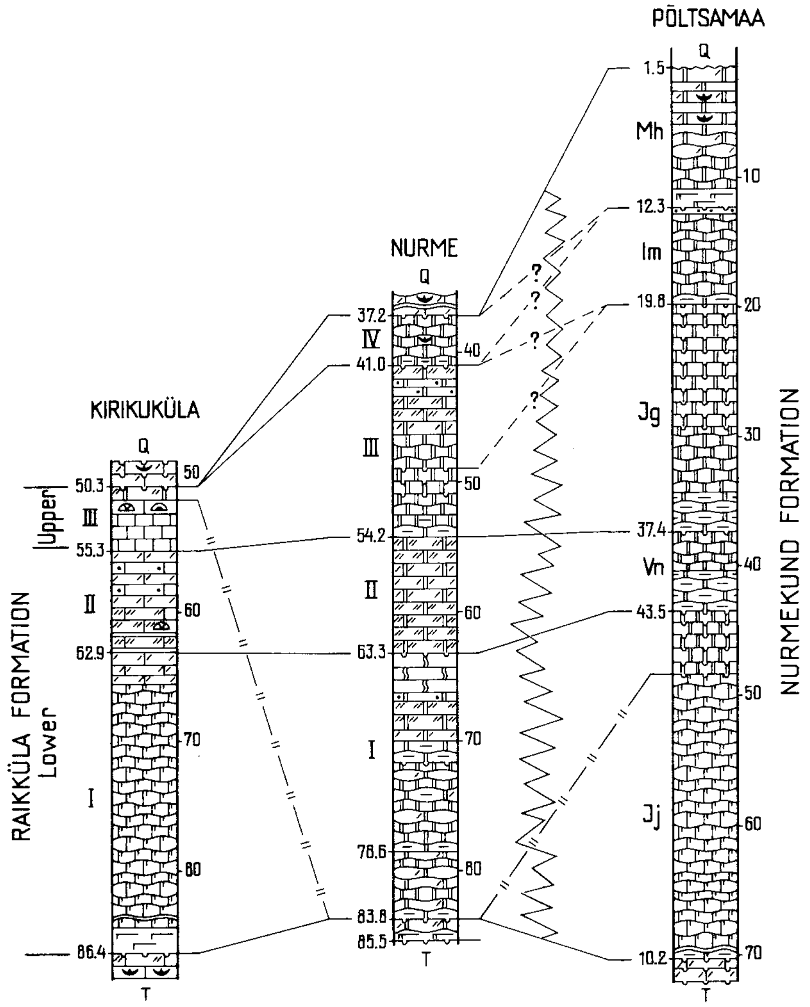

Fig. 4. Correlation of sections of the Raikküla and Nurmekund formations. For lithological legend see Fig. 3. Abbreviations of stratigraphical units: T, Tamsalu Formation; Jj, Järva-Jaani Beds; Vn, Vändra Beds; Jg, Jõgeva Beds; Im, Imavere Beds; Mh, Mõhküla Beds; R, Rumba Formation; Q, Quaternary deposits: Arabic numerals show depth in metres; Roman numerals – submesocycles in the Raikküla Formation.

The fourth cycle, in the interval of 37.2–41.0 m, is represented only by dolomudstones characteristic of the lower part of a cycle. The interval contains pentamerids, which suggest correlation of this cycle with the Mõhküla Beds of the Nurmekund Formation.

The described four medium-scale cycles, based on the changes in the shallowing upward trend of development, are treated as units of the submesocycle rank (Nestor et al. 2001, 2003). They may be grouped into pairs and in such case they form two cycles of the mesocycle rank (Nestor & Einasto 1997), widely used in the East Baltic stratigraphical practice and commonly termed as “Beds” (e.g. Aaloe et al. 1976). In the present case the two mesocycles are treated as subformations of the Raikküla Formation (Nestor 1997): the first and second submesocycles forming the Lower Raikküla Subformation and the third and fourth submesocycles forming the Upper Raikküla Subformation. The mesocycles reveal almost identical recurrence of the main cycle elements. They begin with a more argillaceous basal layer, followed by micritic, pelmatozoan, and laminated dolostones in submesocycles I and III. Only the uppermost parts of both mesocycles, corresponding to submesocycles II and IV, are different, in the first case being represented by pelmatozoan, in the second case – by micritic dolostone.

DISTRIBUTION OF DOLOSTONES

In central and eastern Estonia the rocks of the Nurmekund and Raikküla formations, in particular of their upper parts, are commonly dolomitized (Fig. 1), often also silicified. Dolomitization cross-cuts depositional sequences and is related to the inner shelf facies. The rocks of the Saarde Formation, formed in the deeper-water environment, are mostly undolomitized, except for the topmost metres below the contact with the overlying Devonian sediments. Judging by the spatial distribution and the degree of the alteration of the primary textures and by chemical analyses, the extension and intensity of dolomitization increase upward in the sequence. This coincides with the general regressive trend of the basin development during Raikküla time. The micritic limestones and wackestones in the lower parts of the Nurmekund and Raikküla formations are usually unaltered or only slightly dolomitized, grading upwards into totally dolomitized rocks. The lower limit of the totally dolomitized rocks is rather changeable in space and ranges stratigraphically from the base of the Tammiku Member of the Tamsalu Formation (e.g. Kõnnu core) to the uppermost part of the Ikla Member of the Saarde Formation (e.g. Abja core). Commonly the totally dolomitized sequence begins approximately near the base of the Jõgeva Beds of the Nurmekund Formation, or the Upper Raikküla Subformation, and reaches about 30–50 m in thickness. It means that extensive pervasive dolomitization embraced mainly the upper half of the Raikküla Stage, which is not completely preserved in the denudated sections of western Estonia and at the margins of the present-day distribution area of the stage. Therefore it is difficult to estimate the primary lateral extent of the dolomitized rocks. It is likely that during Raikküla time the zone of dolomitization gradually migrated southwestwards in accordance with progressive shallowing and progradation of the carbonate shelf, especially in the second half of the time. The thickness of the dolomitized rocks may increase in the zones of tectonic disturbances and dolostones may occur there on lower stratigraphical levels. The very complicated picture of the temporal and spatial distribution of dolostones shows that probably there existed different types and sources of dolomitization.

RESULTS OF ANALYSES AND ASPECTS OF DOLOMITIZATION

The $\boldsymbol{d_{104}}$ value of secondary dolomite throughout the section of the Raikküla Formation varies from 2.8857 to 2.8973 Å (Table 1; Fig. 5a). The variability of the d_{104} spacing has distinct regularities. The highest d_{104} values (>2.89 Å) of dolomite occur near (within ca 2 m) the contacts with the over- and underlying marl and limestone. Through the rest part of the studied sequence d_{104} is quite constant, varying between 2.8859 and 2.8888 Å. The increase in the d_{104} spacing of dolomite contacting with limestone is typical of secondary (Vingisaar & Utsal 1978; Kallaste & Kiipli 1995) as well as primary dolomite (Teedumäe et al. 2003). At both boundaries of the Raikküla Formation the content of insoluble residue increases abruptly (Fig. 5b; Table 1). The following transgression of the Adavere Age ended the process of dolomitization and also inhibited the crystallographic ordering of contacting dolomite within about 2 m (Table 1). The contact of the Raikküla dolostone with the underlying limestone of the Tamsalu Formation (Figs. 2, 3) is less distinct and has a transitional character, showing the interbedding of highly clayey calcitic dolostone, dolomitic limestone, and marl. The d_{104} value of dolomite coexisting with calcite in these interlayers is high and equals to that of dolostone contacting with limestone (Table 1; Fig. 5a). This regularity has been widely observed since Lippmann (1973).

The XRD results show the excess of Ca. Provided that additional Ca replaces Mg, the growth of the d_{104} value calculated by the formula (2)

$$\Delta d_{104} = m_{\mathrm{Ca}} \cdot (3.035 - 2.742) = m_{\mathrm{Ca}} \cdot 0.293 \ \text{Å} \tag{2}$$

is 0.0023–0.0047 Å.

As the calculated and measured d_{104} values are, in general, in good accordance, it is most likely that additional Ca is bound in dolomite structure and expands the lattice parameters. The calculated d_{104} spacing for 50 mol% $CaCO_3$ is 2.884 Å, for 52 mol% – 2.890 Å, for 54 mol% – 2.896 Å.

The presence of Fe and Mn in the dolomite lattice may also affect the value of d_{104}. The possible concentration of Fe in the dolomite lattice (Fig. 6, trendline) is 0.42% Fe_2O_3 (0.45 mol% $FeCO_3$), which corresponds to the variation of the d_{104} value of 0.0002 Å, calculated by the formula (3),

$$\Delta d_{104} = m_{\mathrm{Fe}} \cdot (2.79 - 2.742) = m_{\mathrm{Fe}} \cdot 0.048 \ \text{Å} \tag{3}$$

and is below the precision of the X-ray diffractometry.

The impact of Mn on replacement of Mg and Ca is calculated by the formulas (4) and (5), respectively, where m_{Mn} is the molar concentration of $MnCO_3$ in dolomite:

$$\Delta d_{104} = m_{\mathrm{Mn}} \cdot (2.85 - 2.742) = m_{\mathrm{Mn}} \cdot 0.108 \ \text{Å}, \tag{4}$$

$$\Delta d_{104} = m_{\mathrm{Mn}} \cdot (2.85 - 3.035) = -m_{\mathrm{Mn}} \cdot 0.185 \ \text{Å}. \tag{5}$$

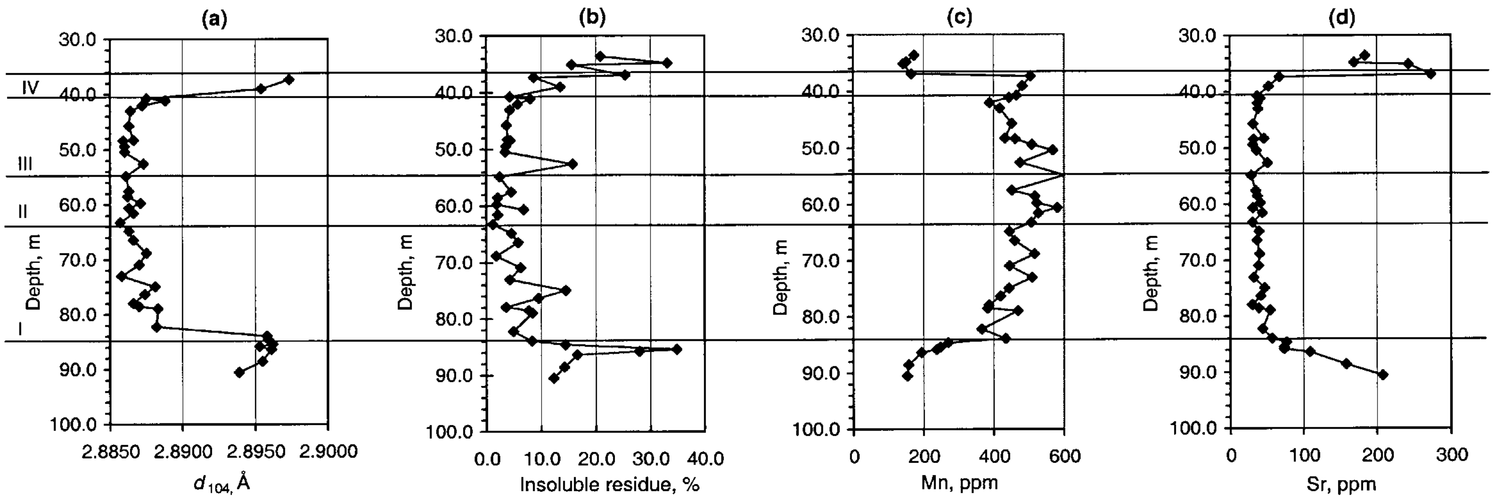

Fig. 5. Variation of d_{104} (a), insoluble residue (b), Mn (c), and Sr (d) *vs.* depth in the Nurme drill core: I, II, III, IV – numbers of submesocycles (see Fig. 3).

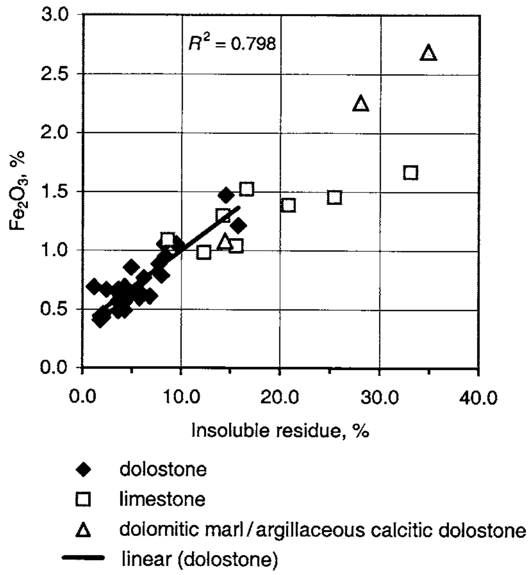

Fig. 6. Correlation between the content of Fe_2O_3 (total) and insoluble residue.

The concentration of Mn in dolostone is low. The maximum content of Mn (600 ppm; Table 1) in dolostone corresponds to 0.10 mol% $MnCO_3$. The calculated variations of d_{104} of mineral dolomite, if Mn replaces Mg as well as Ca, are below the precision of the measurement, being respectively 0.0001 Å (formula (4)) and 0.0002 Å (formula (5)).

As follows from above, the changes in lattice parameters of the studied dolomite are mainly induced by the Ca/Mg ratio in the dolomite lattice. Two general groups of dolomite, based on stoichiometry, can be distinguished (Fig. 7), showing bimodal distribution of Ca^{2+}. One group clusters between 2.885 and 2.888 Å (50–52 mol% $CaCO_3$) and the other between 2.939 and 2.973 Å (53.3–54.4 mol% $CaCO_3$). The first group includes secondary dolomite in dolostone not contacting with limestone. The second group comprises the dolomite in dolostone contacting with limestone and dolomite coexisting with calcite in limestone and marl. The same regularities have been established in a previous study of dolomites of different genesis (Teedumäe et al. 2003). The Ca/Mg ratio of dolomite reflects environmental factors of dolomitization and its bimodal set would generally be interpreted as the reflection of environmental changes during diagenesis.

The stoichiometry can be used for the distinction of different types of dolomite (Goldsmith & Graf 1958; Searl 1994; Kallaste & Kiipli 1995) and it correlates well with the genesis (Teedumäe et al. 2003). The results of previous (Teedumäe at al. 1999, 2001, 2003) and present studies have shown that the secondary dolomite, formed in the course of Silurian pervasive dolomitization, is the most completely ordered dolomite so far known in the Estonian sequence. This may point, besides

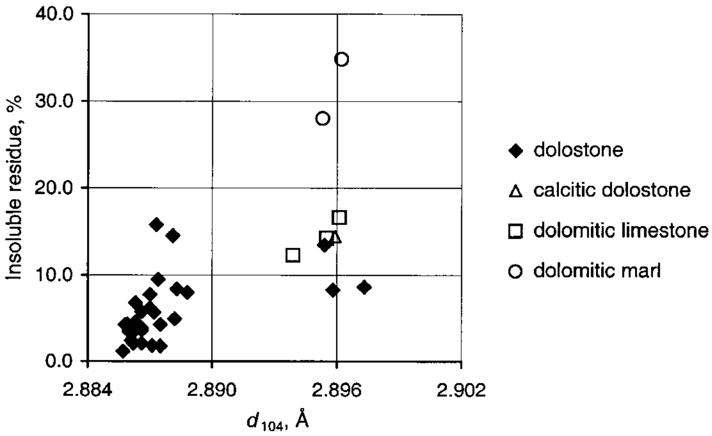

Fig. 7. Correlation between d_{104} of dolomite and insoluble residue of rocks.

the environmental characteristics and total recrystallization, to the role of the crystallization rate, as very slowly growing crystals are closer to the stoichiometric composition (Morrow 1982).

The stability of the lattice of Ca-rich dolomite depends on the character of the ordering of Ca ions within Mg layers. Bimodal ranges of Ca/Mg variation reflect the preferred levels of Ca uptake of the most stable ordering.

The content of **insoluble residue** of dolostone has a positive correlation with the growth of d_{104} spacing (Fig. 5a,b) for the mineral dolomite with d_{104} < 2.890 Å (<52 mol% $CaCO_3$). For dolomite with the expanded lattice (53 mol% $CaCO_3$) no correlation could be revealed (Fig. 7).

Stabilization of calcium-rich dolomite to a more ideal type will mostly take place by dissolution and reprecipitation, since solid-state diffusional processes operate only at the submicron scale and are slow (Tucker & Wright 1994). The lower content of insoluble residue, characteristic of the agitated water sediments, suggests the promoting role of the activity of seawater in the dolomitization process. This supposition is supported by the fact that the most stoichiometric dolomite (<51 mol% $CaCO_3$) in general belongs to the intervals of primarily bioclastic sediments (Table 1; Figs. 3, 5a), which are more permeable to dolomitizing fluids.

The content of **Fe** compounds (Fe_2O_3 total) shows a positive correlation with the content of insoluble residue for all types of the studied rocks (Fig. 6). This regularity, observed also in all previous studies (Teedumäe et al. 1999, 2001, 2003), indicates that iron compounds are mainly of primary, sedimentary origin. Their distribution is controlled by the facies pattern (Jürgenson 1988).

The concentration of **Mn** in dolostone is low (366–601 ppm; Table 1). In general, there is a covariant trend of increasing concentration with increasing stoichiometry (Fig. 5a,c) of mineral dolomite. Mn is highly soluble in an anoxic environment and, if available, should be readily incorporated into the dolomite lattice, but in the present case its impact on the dolomite structure was below the detection limit. There is a negative correlation between the contents of Mn and insoluble residue of dolostone (Figs. 5c, 8). Lower Mn concentrations would suggest a lower primary supply of Mn and fluctuation in Eh. For other types of rocks no correlation is observed.

Low concentrations of Fe and Mn, characteristic of seawater, support the idea of the absence of external dolomitizing fluids and early diagenetic dolomitization. Early, near-surface dolostones, tend to have low Fe and Mn contents, since most near-surface fluids are oxidizing, in contrast with late, deep-burial dolomites, which might have high Fe and Mn concentrations through precipitation from negative Eh porefluids in which Mn and Fe are in solution (Tucker & Wright 1994).

The content of **Sr** in dolostone is low, ranging from 29 to 74 ppm (Table 1). The Sr concentration covaries positively with the d_{104} spacing of mineral dolomite (Fig. 5a,d). The highest concentrations associate with the intervals near the contact of dolostone with limestone (Table 1), where the poorly ordered dolomite occurs. The concentrations (30–40 ppm) are the lowest in the intervals of the coarser-crystal dolostones, which means that Sr is lost in the recrystallization process of dolomite. The mineralogy of precursor carbonate may also have an important role. The Sr contents are high when aragonite is being dolomitized, whereas calcite with its much lower Sr content will be replaced by very Sr-depleted dolomite (Tucker & Wright 1994). It suggests that carbonate sediments might have undergone early diagenetic stabilization to low magnesian calcite before dolomitization.

The components discussed above and their interrelations refer to early diagenetic dolomitization (Einsele 2000) in a normal-saline environment, where the only source for Mg ions was seawater. All components discussed above and their

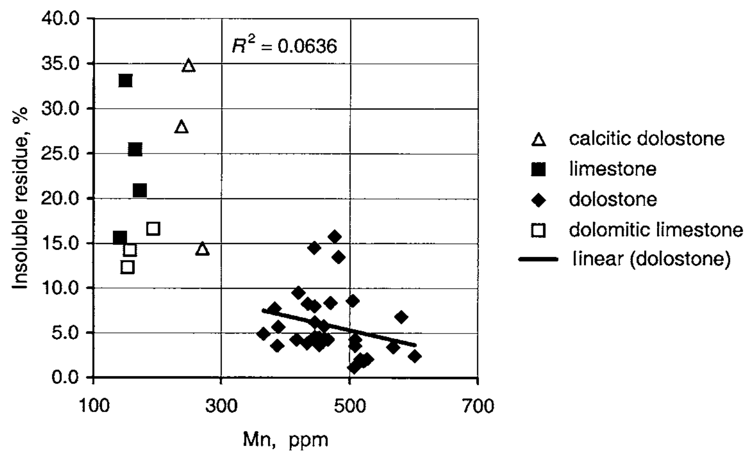

Fig. 8. Correlation between the content of Mn and insoluble residue.

interrelations refer to the early diagenetic near-surface dolomitization of the Raikküla Formation. The only possible source for Mg ions for the dolomitization of these primarily normal-saline sediments could have been seawater. Bacterial sulphate reduction, which has been supposed to have a potential role in the dolomitization process (Garrison et al. 1984; Baker & Burns 1985; Slaughter & Hill 1991; Tucker & Wright 1994; Wright 1999; Einsele 2000; experimentally demonstrated by Warthman et al. 2000), can be treated as the main dolomitizing factor also for the studied dolomites.

Massive pervasive dolomitization, cross-cutting the Silurian depositional sequences, was associated with the regressive phases of the evolution of the Baltic Palaeobasin and related to the restricted migrating zone of the normal-saline shallow-water inner shelf facies (Teedumäe et al. 1999, 2001, 2003). The Raikküla Stage represents such a regressive part of the Early–Middle Llandovery macrocycle (Nestor & Einasto 1997) that ended with extensive local sedimentation brakes and denudation.

The Nurme drill section, represented by the sediments of the shallow, inner shelf facies, was located in the zone of diagenetic (pervasive) dolomitization during the whole Raikküla Age. In the course of general regression and shallowing in Raikküla time, this zone expanded and migrated southwards, where dolomitization commenced somewhat later, approximately in the middle of Raikküla time. The occurrence of totally dolomitized carbonates of the Raikküla Formation between the undolomitized carbonates of the underlying Tamsalu and overlying Rumba formations (Fig. 3) limits the time span of diagenetic dolomitization in the Nurme section within the Raikküla Age.

CONCLUSIONS

Four shallowing up submesocycles were distinguished and first described in the Raikküla Formation of the Nurme drill core. They in turn group into two mesocycles, treated as the Lower and Upper Raikküla subformations. The intensity of dolomitization increases upwards in the cycles in accordance with the shallowing up trend of development. Such a trend is more conspicuous in the lower submesocycles (I and III) of both Raikküla subformations, where stoichiometry of dolomite increases upwards in the sequence. The trend is less distinct in the incomplete submesocycles (II and IV) that were formed in more uniform facies and bathymetrical conditions. In the latter case (submesocycle IV) the dolomitization process at the top of the Raikküla Stage was obviously inhibited by the succeeding, transgressive calcitic sedimentation environment of the Adavere Age.

Intense pervasive dolomitization associated with the regressive phases of the development of the Baltic Palaeobasin, including the middle Llandovery Raikküla Age. Initially dolomitization could have been related to the restricted area of the inner shelf facies, which expanded in accordance with the progressive shallowing of the basin towards the end of the Raikküla Age.

The studied Nurme drill section was located in the zone of pervasive dolomitization during the whole Raikküla Age. The occurrence of totally dolomitized carbonates of the Raikküla Formation between the undolomitized carbonates of the underlying Tamsalu and overlying Rumba formations limits the time span for early diagenetic dolomitization within the Raikküla Age.

The whole set of the studied components is consistent with the normal-marine water environment of that time. There are no signs of possible inflow of external dolomitizing fluids.

Stoichiometry of dolomite correlates with the cyclicity of the sedimentation environment, increasing in accordance with the regressive, shallowing up trend of development. The most completely ordered dolomite is associated with the primarily bioclastic sediments in the upper part of regressive sedimentary cycles, formed in a high-energy environment. It shows the promoting role of the activity of marine water, and primary porosity of sediments in the process of dolomitization.

ACKNOWLEDGEMENTS

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Raikküla kihistu settetsüklid ja dolomiidistumine Nurme puursüdamikus (Silur, Eesti)

Aada Teedumäe, Heldur Nestor ja Toivo Kallaste

Artiklis on esmakordselt kirjeldatud Raikküla kihistu keskmist järku settetsükleid Nurme puursüdamikus. Eristatakse nelja regressiivset submesotsüklit, mis on paarikaupa grupeerunud kaheks mesotsükliks. Viimaseid käsitatakse Alam- ja Ülem-Raikküla alamkihistuna.

Nurme puursüdamikus on Raikküla kihistu kivimid täies ulatuses sekundaarselt dolomiidistunud. Nende lasumi (Adavere lade) ja lamami (Juuru lade) moodustavad dolomiidistumisest puutumata karbonaatkivimid, mistõttu võib arvata, et dolomiidistumine toimus Raikküla ea vältel. Dolokivide koostis ja mikroelementide kooslus sarnaneb normaalmerelistes tingimustes tekkinud karbonaatkivimitega, kusjuures pole täheldatud suurenenud soolsuse või basseiniväliste lahuste juurdevoolu ilminguid. Dolomiidi võre täiustumise aste suureneb regressiivsete settetsüklite (nii meso- kui submesotsükli) ülaosa suunas. Kõige täiuslikuma võrega dolomiit on algselt bioklastilisest lubisettest moodustunud dolokivis, mis näitab aktiivselt liikuva merevee soodustavat mõju dolomiidistumisele. Kõige enam laienenud võrega dolomiit esineb lubjakivis ja sellega kontakteeruvas dolokivis.

Siluri-aegses Balti Paleobasseinis toimunud lausaline dolomiidistumine on seotud basseini arengu regressiivsete faasidega. Dolomiidistumiseks soodne ala paiknes šelfi piires ja selle asukoht muutus vastavalt veetaseme muutustele. Regressiooni arenedes nihkus ala süvamere suunas, mistõttu lõunapoolsetes puuraukudes on dolomiidistunud valdavalt vaid Raikküla lademe ülemine osa. Kogu Raikküla ea vältel on Nurme puuraugu asukoht paiknenud dolomiidistumise vööndis.