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Radon emissions in Harju County, North Estonia

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Abstract. The risk of radon emissions is high in Estonia, especially in the northern part of the country where graptolite argillite (Dictyonema shale) crops out close to the ground surface. The behaviour and concentration of Rn in soil air vary considerably due to several reasons. To elucidate these, investigations were carried out in densely populated Harju County where the concentration of Rn is generally high and the risk to human health is the greatest. The investigations of soil air and in soil samples from the bottom of the excavations were carried out in 31 points. The assessments were made on the results of two simultaneously applied methods: calculated from eU concentration in soil measured with a gamma ray spectrometer, and by direct measurements in soil air with a Markus-10 emanometer. The results obtained by the two methods can notably differ.

In the high and very high Rn risk areas the concentration of Rn in the indoor air of dwellings was high as well. The main source of Rn there is the soil underneath the dwellings and/or bedrock, primarily radioactive graptolite argillite lying below the Quaternary deposits. Areas of such high Rn concentrations in the ground did not follow the genetic–lithologic types of Quaternary deposits. In the high Rn-risk areas within the klint zone the content of U, F, Mo, and some other elements exceeded the recommended and even permissible level for residential areas in several investigation points.

Key words: radon, uranium, klint area, soil air, graptolite argillite.

INTRODUCTION

Radon (Rn) is a source of natural ionizing radiation, it is a toxic element causing mutations, especially lung cancer (Naturally …, 2000; Mjönes & Falk, 2005). Radon occurs in air and water as a colourless and odourless noble gas. It solidifies at –71 °C and decays into seven highly radioactive metals.

In the Baltic States the first investigations of Rn were carried out in 1997–1998 by Valter Petersell at the Geological Survey of Estonia. More detailed investigations of Rn in soil were launched in 2000; in 2002–2008 they were continued in cooperation with Swedish geologists in 566 observation points (Petersell et al., 2005).

In Estonia Rn risk is among the highest in Europe. In the process of compiling the small-scale (1 : 500 000) Rn-risk map of Estonia (Petersell et al., 2005) it appeared that grounds with high and very high Rn content (50–250 kBq/m^3 and

>250 kBq/m^3, respectively) are frequent in northern Estonia. Such areas cover several square kilometres, mostly in the klint zone. According to the Estonian Radiation Protection Centre (unpublished data), in areas of high and very high Rn risk the concentration of Rn in indoor air often exceeds the permissible level (200 Bq/m^3), reaching 3000 Bq/m^3, sometimes even 10 000 Bq/m^3.

In Estonia the Rn level in soil air and its behaviour are highly variable, but at the same time the sources of Rn, its formation, reasons of Rn concentrations and dispersion, and regularities are still poorly investigated. The main source of Rn in indoor air is the soil beneath the buildings and the underlying loose sediments and bedrock of variable composition. Therefore, inappropriate assessment of Rn in soil air and its migration regularities may cause health disorders. On the other hand, uncertainty about Rn risk may generate emotional stress and cause unfounded material expenses. To at least partially find solutions to these problems, the authors chose for the study densely populated Harju County in northern Estonia, where the Rn risk level is high and all potential sources of Rn are present in the geological succession. In the klint escarpment radioactive graptolite argillite (Dictyonema shale) and obolus phosphorite crop out. Their clasts and smalls are scattered everywhere. Within the study area with high Rn risk several towns (Tallinn, Paldiski, Keila, Saue, Maardu, Kehra, Loksa), other settlements, and farmsteads are situated.

The factual material of the current paper originates from the databases of Rn risk-maps (Petersell et al., 2005, 2008), but includes also the new data collected by the authors in 2009 and 2010. The content of K, U, P, and Mo in the samples was determined in AcmeLabs, Canada, and that of eU, eTh, and K (^{40}K) in the laboratories of the Estonian Radiation Protection Centre and Geological Survey of Estonia.

MATERIAL AND METHOD

Investigation area

Harju County borders on the Gulf of Finland in the north (Fig. 1). With its area of 4333 km^2 and 522 252 residents (in 2003) it is the largest and most densely (120.8 inhabitants per km^2) populated county in Estonia.

The topography is mostly flat. The North-Estonian Klint, a 10–30 m high west–east oriented escarpment, divides the county into two regions with different geological setting: the Fore-klint Lowland in the north and the Harju Limestone Plateau in the south. The geology is diversified by deep ancient valleys cutting into the sedimentary bedrock and filled with Quaternary deposits (Miidel & Tavast, 1978; Raukas & Tavast, 1987; Vaher et al., 2010).

In the succession three rock complexes overlying one another can be distinguished: Proterozoic crystalline basement is overlain by Palaeozoic sedimentary rocks and these in turn by loose Quaternary deposits (Raukas & Teedumäe, 1997). Quaternary deposits contain to a greater or lesser extent crushed particles of the first two rock complexes. The radioactivity of all these rock types of different age and composition is variable.

Among the crystalline basement rocks, the granites of the rapakivi formation cropping out on the bottom of the Gulf of Finland and in southern Finland are noteworthy. These rocks contain 3–10 g/t of U, 10–50 g/t of Th, and 2.2–3.5% of K (Geological Survey of Finland, 1992). Their crushed varieties are found in till and other Quaternary deposits.

The surface of the crystalline basement and the Palaeozoic sedimentary rocks have a southward inclination of ca 3 m/km. In the Fore-klint Lowland the Quaternary cover is underlain by Cambrian sandstones, siltstones, and clay, and on the limestone plateau, by Ordovician limestones, less frequently by marls and dolostones. Limestones are often karstified.

Between the Cambrian sand- and siltstones and Ordovician limestones occur the major sources of Rn: Lower-Ordovician obolus sandstone with P-rich layers of phosphorite that are overlain by graptolite argillite. Both these rocks are rich in U and have a background Th content. Alongside U, the two rock types contain also other environmentally hazardous elements (Table 1).

Between Narva and the Pakri Islands, in the klint and in the valleys cutting into the bedrock, graptolite argillite and phosphorite rich in U are exposed or spread beneath the Quaternary cover. Further to the west, these rocks crop out on the bottom of the Gulf of Finland. Their clasts and smalls are found in various glacial and marine deposits and serve as the main sources of Rn hazard and elevated natural radiation.

Table 1. Average concentration of radioactive and some other elements in graptolite argillite and phosphorite (Petersell et al., 2008)

Element	Regions				Earth crust average[b]
	Western Harju County	Maardu	Kuusalu	Parent rock of Estonian soil[a]	
In graptolite argillite					
Uranium, g/t	86	36	84	2.1	2.5
Uranium (prevailing content), g/t	30–170	20–90	30–160		
Thorium, g/t	10	8	12	5.8	10.3
Molybdenum, g/t	162	53	210	0.94	1.4
Potassium, %	5.9	6.2	5.1	1.86	2.86
In phosphorite					
Uranium[c], g/t		22	16	2.1	
Thorium, g/t		7.4	12	5.8	
Fluorine, g/t		12 600	8 400	284	611
Potassium, %		<0.2	<0.2	1.86	
P_2O_5, %		13	8.5	0.096	0.150

[a] In this paper, the generally non-weathered Quaternary deposit (horizon C) is considered as the parent rock of Estonian soil.

[b] After Wedephol, 1995.

[c] In deposits, the U content of phosphorites directly correlates with P_2O_5.

The position of the northern boundary of the distribution area of graptolite argillite and obolus sandstone (with phosphorite) prior to the continental glaciation is not known, but based on the palaeogeographic conclusions (Männil, 1966), it was several kilometres or even tens of kilometres northward of their present-day outcrop line in the klint escarpment. Thus, several billions of tonnes of U-rich rock material crushed by glaciers have been carried southwards. As a result of the activity of the continental glacier, the U-rich material was mixed with the material with background U concentration, and when the continental glacier retreated, the mixed material mostly deposited on the bedrock surface as till of variable thickness (Petersell et al., 2005).

Methods

The investigation points were selected so that they would be characteristic of the major potential Rn risk areas and most important lithotypes of the Quaternary cover. Their coordinates were determined by Garmin GPS 76.

Field investigations were carried out in 31 points (Fig. 1). Their position was determined in detail within a circle ca 200 mm in diameter, considering that the ground surface should be flat, without any visible evidence of technogeneous contamination, and the gamma radiation level (determined with СРП-88Н) should be typical for the area. The concentration on Rn in the investigation points was determined simultaneously by two methods: (1) calculated on the basis of the content of eU (ppm) or ^{226}Ra (Bq/kg) measured with a portable gamma ray spectrometer (RnG) and (2) by direct measurements of Rn (kBq/m^3) in soil air with a Markus-10 emanometer (RnM) (Petersell et al., 2005).

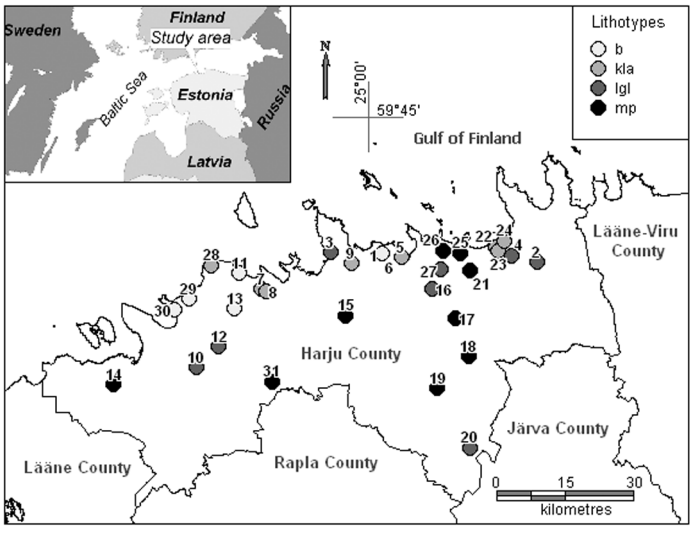

Fig. 1. Study area with investigation points and lithotypes: Holocene marine deposits (b), Fore-klint Lowland and talus deposits (kla), glaciolacustrine deposits (lgl), and till (mp).

The eU concentration in soil was measured with a portable gamma ray spectrometer (Detector model GPX-21A) at the bottom of the excavations (depth 80 cm, cross-section area ca 20 cm × 20 cm). The concentration of Rn in soil air was determined directly with a Markus-10 emanometer at the same depth. Simultaneously, the genetic type of the Quaternary deposit was identified and soil samples were taken from the excavation bottom to determine in the laboratory the concentrations of elements accompanying eU.

Based on the eU concentration measured with the gamma ray spectrometer, the concentration of Rn in soil air balanced with Ra was calculated according to the following formula from Clavensjö & Åkerblom (1994):

$$\mathrm{RnG} = Ae\delta(1-p)p^{-1},$$

where RnG is Rn maximum content developed, kBq/m^3; A is eU concentration, Bq/kg; e is Rn emanation factor; δ is compact volume weight (specific weight), kg/m^3; and p is porosity (1 g/t eU = 12.35 Bq/kg ; Petersell et al., 2005).

Based on the depth, soil type, and Rn diffusion dependence graph (Clavensjö & Åkerblom, 1994), the results of directly measured Rn concentrations were re-calculated to the standard depth of 1 m.

The soil samples collected at the bottom of excavations were dried, the <2 mm fraction was separated and ground into powder by quartering. The powder was then sent to AcmeLabs in Canada where the concentrations of approximately 50 elements were determined, including U, Th, K, P, and Mo, which were of special interest for our investigation team. The concentration of F was determined at the laboratory of the Geological Survey of Estonia. In the investigation area the dispersion of the measured elements was very high and changeable (Table 2), therefore further the geometric mean concentrations and standard coefficients were used to assess the concentrations of elements.

RESULTS AND DISCUSSION

Concentration of eU

Different authors have assessed the average concentration of eU in the Earth's crust to be 2.5–3 g/t (Wedephol, 1995). The average concentration of eU in Estonian Quaternary cover is 2.14 g/t, mostly (68%) 0.9–4.9 g/t (Petersell et al., 2005). Thus in the area under discussion the average eU concentration in soil – 6.11 g/t – more than twice exceeded the average for the whole Estonian territory, the variation in concentrations was very high, and the maximum concentration reached 19 g/t (Fig. 2, Table 2).

The lateral distribution of eU concentrations was very changeable. Unambiguously, the highest eU concentrations occurred in the klint belt, both on its slope and in the Quaternary deposits in front of the escarpment, irrespective of the

Table 2. Results of field measurements and laboratory analyses, arithmetical mean concentrations ($Mean_A$), arithmetical standard deviations ($Std\ dev_A$), geometrical mean concentrations ($Mean_G$), and geometrical standard deviations ($Std\ dev_G$) of the calculated activity and contents of elements in different lithotypes: Fore-klint Lowland and talus deposits (kla), glaciolacustrine deposits (lgl), Holocene marine deposits (b), and till (mp)

Point number	Lithotype	eU content		Rn, kBq/m^3		RnG/RnM	Content of elements				
		g/t	Bq/kg	RnG	RnM		U, g/t	P, %	F, g/t	Mo, g/t	K, %
1	kla	10.10	124.74	149.00	15.00	9.93	8.30	2.00	3710.00	0.70	0.35
2	lgl	3.70	45.70	56.00	11.00	5.09	2.90	0.13	250.00	0.10	1.60
3	lgl	4.20	51.87	64.00	5.00	12.80	3.10	0.46	880.00	0.30	0.86
4	lgl	2.90	35.82	44.00	108.00	0.41	2.50	0.58	310.00	1.90	1.47
5	kla	18.10	223.54	262.00	55.00	4.76	20.40	4.25	6780.00	1.90	0.16
6	b	9.30	114.86	138.00	17.00	8.12	7.80	2.20	4030.00	0.70	0.27
7	kla	8.10	100.04	119.00	98.00	1.21	7.60	0.57	104.00	1.80	1.91
8	kla	5.50	67.93	81.00	31.00	2.61	6.70	0.28	660.00	0.40	2.16
9	kla	12.70	156.85	187.00	86.00	2.17	11.10	1.54	3150.00	2.00	0.23
10	lgl	2.10	25.94	32.00	24.00	1.33	2.60	0.05	470.00	0.20	2.49
11	b	12.40	153.14	184.00	195.00	0.94	12.10	2.17	286.00	5.30	0.30
12	lgl	2.00	24.70	30.00	66.00	0.45	2.00	0.03	260.00	0.50	2.27
13	b	5.10	62.99	76.00	53.00	1.43	5.20	0.49	1030.00	0.20	0.85
14	mp	2.00	24.70	28.00	94.00	0.30	1.30	0.04	200.00	0.10	1.07
15	mp	4.90	60.52	69.00	38.00	1.82	3.00	0.11	270.00	1.00	1.67
16	lgl	3.00	37.05	46.00	35.00	1.31	2.50	0.14	260.00	0.60	2.08
17	mp	1.90	23.47	27.00	47.00	0.57	1.50	0.06	230.00	0.20	1.69
18	mp	0.90	11.12	13.00	12.00	1.08	1.50	0.06	240.00	0.40	1.69
19	mp	1.40	17.29	20.00	11.00	1.82	1.40	0.05	240.00	0.30	1.64
20	lgl	2.50	30.88	38.00	73.00	0.52	2.70	0.06	250.00	0.30	2.49
21	mp	4.10	50.64	58.00	90.00	0.64	3.20	0.21	510.00	0.50	2.08
22	kla	8.70	107.45	128.00	263.00	0.49	6.70	1.03	1730.00	3.20	1.13
23	kla	6.90	85.22	102.00	93.00	1.10	5.80	1.11	2430.00	0.80	0.59
24	kla	12.20	150.67	180.00	184.00	0.98	10.80	1.18	1120.00	7.70	0.96
25	mp	8.20	101.27	116.00	40.00	2.90	6.40	0.76	1490.00	1.90	0.94
26	mp	9.60	118.56	136.00	111.00	1.23	9.70	0.42	910.00	12.50	2.03
27	lgl	2.90	35.82	44.00	82.00	0.54	2.50	0.15	350.00	0.20	1.74
28	kla	19.00	234.65	280.00	198.00	1.41	16.80	1.01	2090.00	14.30	2.84
29	b	2.00	24.70	30.00	54.00	0.56	1.40	0.13	190.00	0.10	0.30
30	b	2.10	25.94	31.00	35.00	0.89	1.70	0.10	250.00	0.40	0.55
31	mp	1.00	12.35	14.00	38.00	0.37	1.20	0.03	300.00	1.10	1.98
	$Mean_A$	6.11	75.49	89.74	72.97	2.25	5.56	0.69	1128.39	1.99	1.37
	$Std\ dev_A$	4.90	60.49	71.63	62.86	2.97	4.79	0.93	1503.18	3.46	0.78
	Min	0.90	11.12	13.00	5.00	0.30	1.20	0.03	104.00	0.10	0.16
	Max	19.00	234.65	280.00	263.00	12.80	20.40	4.25	6780.00	14.30	2.84
	$Mean_G$	4.43	54.74	65.12	49.85	1.31	4.00	0.28	591.43	0.74	1.07
	$Std\ dev_G$	2.32	2.32	2.33	2.61	2.67	2.28	4.29	3.01	3.91	2.26
	Limit of safe concentration	~3.6	~45	50	50		20		450	10	

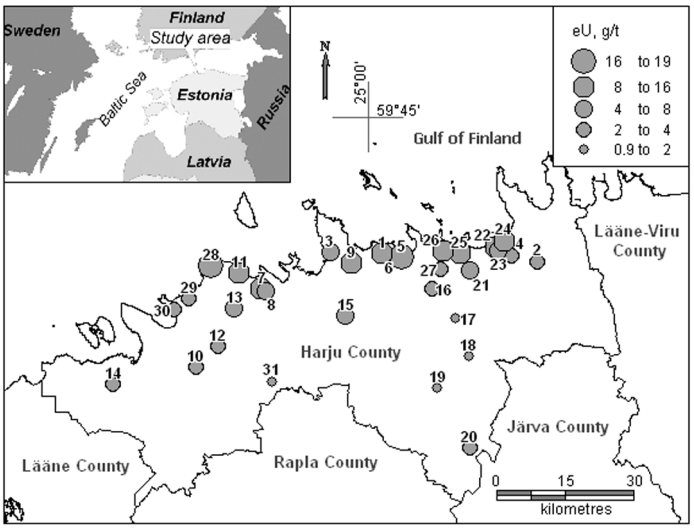

Fig. 2. Concentration of eU in soil air.

lithotype. Southwards, the eU concentration in Quaternary deposits gradually decreased. Near the klint escarpment in the till (investigation point No. 26) the eU concentration was 9.6 g/t, but in the carbonate-rich till in the southern part of the study area (investigation point No. 18) it was less than 1 g/t.

The highest eU concentrations, ranging from 5.5–19.0 g/t, were observed in the talus slope of the klint escarpment and in the deposits of its intermediate escarpments. As for lithotypes, the highest eU concentrations were related to fore-klint and klint slope deposits (Table 3). The concentration of eU in the talus deposit more than twice exceeded the average concentration of the area, characterizing the generally high but extremely variable share of the material originating from graptolite argillite and obolus phosphorite. Besides, the eU concentration was changeable and frequently high in the sand and silt of different Baltic Sea development stages (Table 3), reaching even 12.4 g/t in the sand in front of the klint escarpment (investigation point No. 11). It reflects wave erosion of the klint escarpment, but also an increased content of granitoid material of the rapakivi formation. Generally, the concentration of eU was lower in glaciolacustrine deposits and carbonate-rich till; however, patches with higher eU concentration occurred within these deposits as well.

Radon in soil air

The concentration of Rn in soil air was determined by two methods: calculated on the basis of the eU concentration in soil (RnG) and the Rn preserved in soil air was directly measured (RnM). As Rn is a direct decay product of eU, there is

Table 3. Arithmetical mean concentrations ($Mean_A$), arithmetical standard deviations (Std dev_A), geometrical mean concentrations ($Mean_G$), and geometrical standard deviations (Std dev_G) of the calculated activity and contents of elements in different lithotypes: Holocene marine deposits (b), glaciolacustrine deposits (lgl), till (mp), and Fore-klint Lowland and talus deposits (kla)

Litho-type	Num-ber of points	Geochemical parameter	eU content		Rn, kBq/m^3		RnG/RnM	Content of elements				
			ppm	Bq/kg	RnG	RnM		U, g/t	P, %	F, g/t	Mo, g/t	K, %
b	5	$Mean_A$	6.2	76.3	92	71	2.39	5.6	1.02	1157	1.34	0.45
		Std dev_A	4.6	56.5	68	71	3.22	4.5	1.08	1642	2.23	0.25
		$Mean_G$	4.8	58.9	71	51	1.40	4.1	0.50	563	0.49	0.41
		Std dev_G	2.3	2.3	2.3	2.4	2.82	2.6	4.4	3.6	4.5	1.6
lgl	8	$Mean_A$	2.9	36.0	44	51	2.81	2.6	0.20	379	0.51	1.88
		Std dev_A	0.8	9.3	11	37	4.33	0.3	0.20	216	0.58	0.57
		$Mean_G$	2.8	35.0	43	34	1.25	2.6	0.13	343	0.35	1.78
		Std dev_G	1.3	1.3	1.3	3.0	3.53	1.1	2.8	1.6	2.4	1.4
mp	9	$Mean_A$	3.8	46.7	53	53	1.19	3.2	0.19	488	2.00	1.64
		Std dev_A	3.2	39.8	46	36	0.86	2.9	0.25	438	3.98	0.40
		$Mean_G$	2.7	33.3	38	41	0.93	2.4	0.11	376	0.68	1.59
		Std dev_G	2.4	2.4	2.4	2.3	2.17	2.1	3.1	2.0	4.1	1.3
kla	9	$Mean_A$	11.3	139.0	165	114	2.74	10.5	1.44	2419	3.64	1.15
		Std dev_A	4.7	58.5	69	84	2.98	5.0	1.16	1998	4.56	0.95
		$Mean_G$	10.4	128.6	153	83	1.84	9.6	1.12	1556	2.01	0.77
		Std dev_G	1.5	1.5	1.5	2.5	2.48	1.5	2.1	3.4	3.1	2.8
All	31	$Mean_A$	6.1	75.5	106	90	2.25	5.6	0.69	1128	1.99	1.37
		Std dev_A	4.9	60.5	72	72	2.97	4.8	0.93	1503	3.46	0.78
		$Mean_G$	4.4	54.7	83	65	1.31	4.0	0.28	591	0.74	1.07
		Std dev_G	2.32	2.32	2.33	2.61	2.67	2.28	4.29	3.01	3.91	2.26

an unambiguous correlation between the soil's eU concentration and the generated Rn concentration (RnG), which is influenced only by the variable emanation capacity of lithotypes. Studies have shown that only 20–35% of the Rn formed as a result of Ra decay in grains of soils emanates into soil air (Petersell et al., 2005). The emanation coefficient of finer-grained lithotypes was higher, but in general the concentration of Rn formed in soil air followed the distribution and regularities of eU in soil.

Concentration of Rn formed in soil air by Ra decay

The concentration of Rn in soil air calculated from the eU concentration (RnG) characterizes the Rn concentration level that has developed in a closed system with no soil air aeration or additional Rn inflow from deeper soil or bedrock layers.

In Harju County the RnG concentration in soil air well correlated with the eU concentration in soil (Table 4). The highest concentrations (up to 280 kBq/m^3) occurred on the Fore-klint Lowland and in the distribution area of talus deposits, and the lowest (RnG < 13 kBq/m^3) in carbonate-rich till. Tables 2 and 3 present the concentrations of RnG in different lithotypes of the study area.

Figure 3 presents the areal distribution of RnG concentrations in the study area, which is close to the distribution of eU concentrations (Fig. 2).

The concentration of RnG varied considerably, from 13 to 280 kBq/m^3 with the average being 90 kBq/m^3, which exceeds threefold the average value for Estonian soils (27 kBq/m^3; Petersell et al., 2005) and is 1.8 times higher than the maximum permissible concentration for construction activity without applying Rn protection measures (50 kBq/m^3; Eesti Standardikeskus, 2009).

Residual concentration of Rn in soil air

Although the concentration of Rn formed from eU was stable in soil air, the natural soils are not a closed system and some Rn formed as a Ra decay product

Table 4. Correlation of U with main accompanying contaminants

All	eU	RnG	RM	U	P	F	Mo	K
eU	1							
RnG	1	1						
RnM	0.510	0.514	1					
U	0.979	0.978	0.454	1				
P	0.803	0.803	0.221	0.834	1			
F	0.719	0.716	0.023	0.738	0.895	1		
Mo	0.663	0.660	0.634	0.629	0.212	0.129	1	
K	−0.299	−0.301	0.016	−0.273	−0.609	−0.526	0.199	1

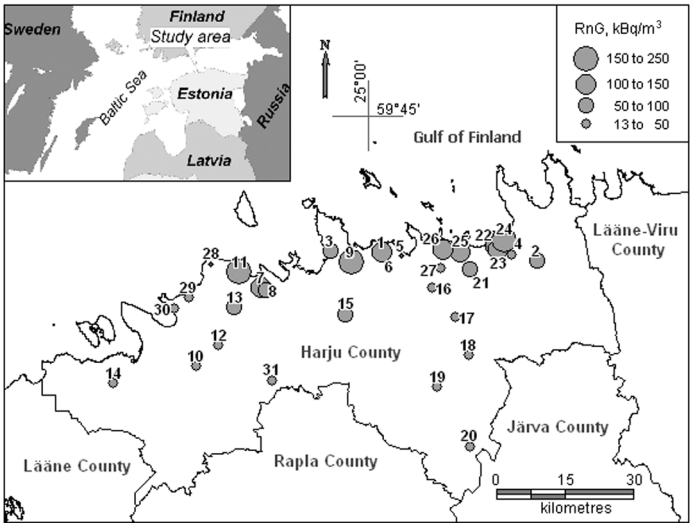

Fig. 3. Concentration of RnG in soil air.

emanates into air. At he same time, Rn may accrue to soil air also from deeper soil/bedrock layers. Such so-called residual concentration of Rn (RnM) generally follows the same regularities as the concentrations formed from eU in soil. Although the average concentrations of RnG and RnM in soil air of the region were generally similar (Fig. 3), differences in their areal distribution are notable (Figs 3–6; Tables 2, 3).

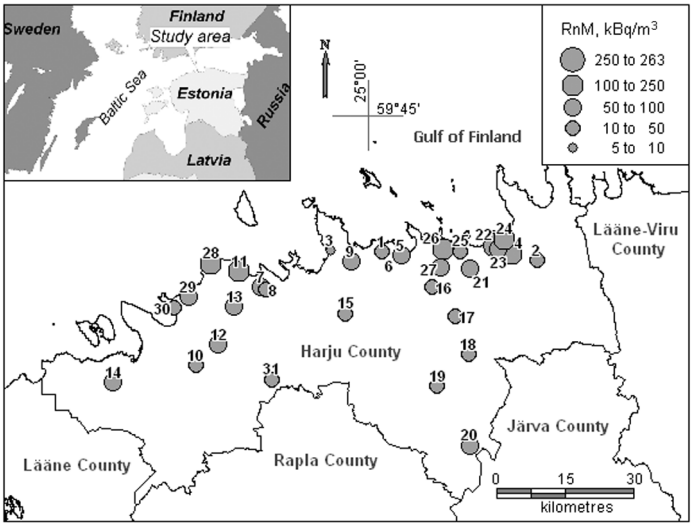

Fig. 4. Concentration of RnM in soil air.

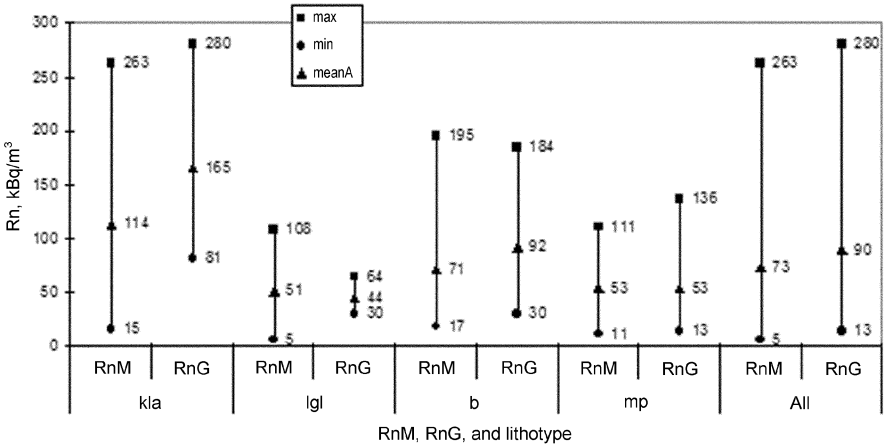

Fig. 5. Maximum, minimum, and mean values of RnG and RnM in different lithotypes: Fore-klint Lowland and talus deposits (kla), glaciolacustrine deposits (lgl), Holocene marine deposits (b), and till (mp).

Investigations carried out in Sweden (Clavensjö & Åkerblom, 1994) showed that when a humus horizon has developed on the ground surface, at a depth of 1 m the concentration of RnM forms 50–90% of the Rn formed from eU. Figure 7 presents the concentrations of Rn in soil air calculated on the basis of eU (RnG) as well as those measured with a Markus-10 (RnM) in various lithotypes. In general, the observation points with high RnM values were located on the flat fore-klint area in front of the klint escarpment, as well as in talus deposits and on

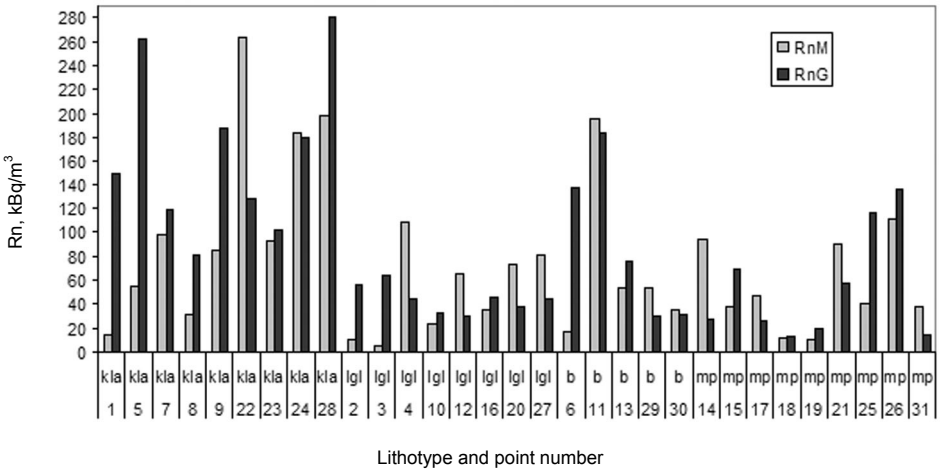

Fig. 6. Concentrations of RnG and RnM in the soil air of different lithotypes: Fore-klint Lowland and talus deposits (kla), glaciolacustrine deposits (lgl), Holocene marine deposits (b), and till (mp).

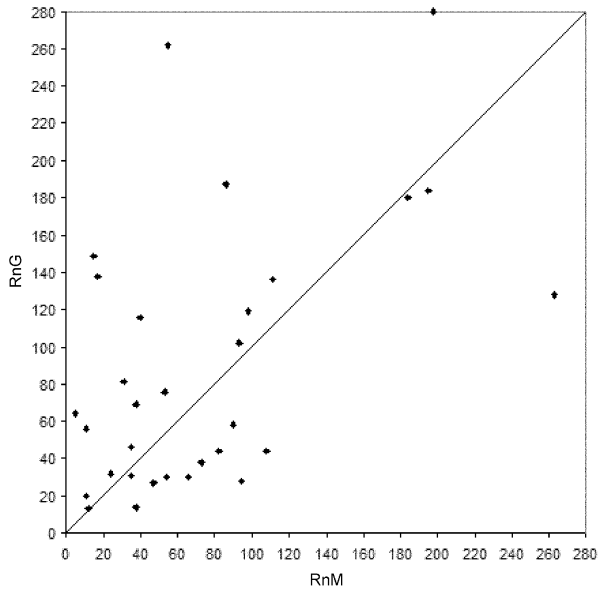

Fig. 7. Concentrations of Rn in soil air calculated after eU (RnG) vs concentrations measured by Markus-10 (RnM).

the slopes of ancient valleys. Some high RnM concentrations in soil air were found also outside these areas.

A characteristic feature was that in some investigation points the RnM concentration in the soil air was several times lower than could be assumed from the concentration of eU, while in others RnM concentrations many times exceeded those suggested by the concentrations of eU (Fig. 6). In the former case, the sandy soil with a high eU concentration was covered by a very poorly developed humus horizon (e.g. investigation point No. 6, RnG = 138 kBq/m^3; RnM = 17 kBq/m^3). The latter case (e.g. investigation point No. 14, RnG = 28 kBq/m^3; RnM = 94 kBq/m^3) was observed when the soil with a background eU concentration was overlain by a dense and thick (>20 cm) humus horizon and there existed an additional inflow of Rn from graptolite argillite and obolus phosphorite lying at a depth of 65–75 m. Our investigation showed that in the soil air of the limestone fissures of the alvar areas on the limestone plateaus in Harju County the concentration of Rn reached up to 96 kBq/m^3.

As mentioned above, the Palaeozoic sedimentary rocks have a southward dipping of about 3 m/km. Consequently, in the distribution area of soils with a background eU concentration high concentrations of Rn of deep origin (investigation point No. 20, RnM = 73 kBq/m^3) were observed even in areas where rocks with a high content of U lie at a depth of more than 100 m below ground surface. Such distribution of eU content by areas and lithotypes indicates its

direct connection with the U-rich rocks exposed in the klint escarpment. Although it is obvious that continental glaciers carried their clasts and smalls to the south and sea abrasion and accumulation to the north, precise elucidation of their distribution in the Quaternary deposits can be done only by detailed geochemical mapping.

The reasons of the above-described phenomena are variable. Often graptolite argillite or phosphorite layers crop out beneath talus deposits in front of the klint as well as in the slopes of valleys and in fore-klint flat areas, or clasts or fines of graptolite argillite or phosphorite occur in deeper layers of deposits. Such areas present health hazards to residents. These are common in the intermediate klint plateaus (e.g. at Suurupi, Rannamõisa, Tiskre, Jõelähtme, etc.). The Rn potential of graptolite argillite and obolus phosphorite can be high, depending on the concentration of eU in rocks, reaching up to 1400 kBq/m^3 in graptolite argillite and up to 400 kBq/m^3 in obolus phosphorite. In massive rock bodies as a maximum 10% of the Rn formed in them is mobile, but in fissured rocks or when crushed in the process of construction activity or when the varieties rich in clasts or fines of graptolite argillite and phosphorite are present, they may become hazardous sources of migrating Rn (e.g. observation point No. 22).

Origin

As was mentioned above, in the study area the main sources of high Rn concentrations in soil air are the Lower-Ordovician U-rich rocks, graptolite argillite, and obolus sandstone (phosphorite), and among the Quaternary deposits, clasts and fines of the above-mentioned rocks and to a lesser extent the Quaternary deposits enriched with erratic granitoid material of the rapakivi formation.

In addition to U, graptolite argillite is also rich in K and environmentally harmful elements Mo, V, As, etc. In phosphorite the content of these elements is small, but the content of F is high.

Consequently, in high Rn-risk areas the concentrations of Mo, F, and other elements listed above exceed the safe limit for residence and can cause health problems. The connections between U and accompanying hazardous elements are reflected in their correlations (Table 4).

There is a distinct positive correlation between RnG, eU, U, P, F, and Mo. The correlation between RnM and RnG and other listed elements is mostly positive, but frequently also neutral, reflecting the discrepancies between RnM and RnG in observation points.

The content of the elements in graptolite argillite and phosphorite associations is closely entwined. Such entwinement, but a rather regular relationship with U, can be seen on the ABC diagrams of elements (Fig. 8).

In soil the characteristic high K content in graptolite argillite is supplemented with K from granite and other rocks. As a result, K is neutral towards the observed environmentally hazardous elements or is negatively correlated.

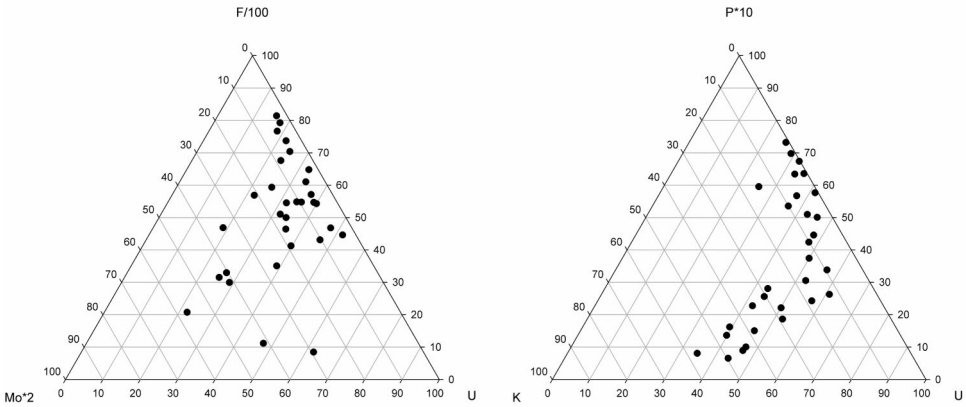

Fig. 8. ABC diagrams of some elements.

CONCLUSIONS

In Harju County the concentration of Rn in soil air was high (>50 kBq/m^3) or very high (>250 kBq/m^3), and its distribution was heterogeneous. The concentration of Rn several times exceeded the average values of Estonian soil and the Earth's crust. The reason is presence of U-rich graptolite argillite and obolus phosphorite in the geological sequence and distribution of their clasts and fines in Quaternary deposits. The concentration of Rn was especially high in the klint zone where graptolite argillite and phosphorite are exposed or directly underlie the Quaternary cover, or the Quaternary deposits rich in clasts and fines of these rocks are present.

The concentration of Rn calculated from eU in soil air was temporally stable and well correlated with the concentration of eU as well as U, and was somewhat variable by lithotypes.

The natural soil in the study area is not a closed system and therefore the Rn formed by Ra decay partly migrates to air. At the same time, an additional inflow of Rn into soil air occurs from lower graptolite argillite and phosphorite beds. Thus, the concentration of Rn measured with a Markus-10 is actually residual concentration. Although it generally followed the same regularities as the concentration of Rn formed from eU in soil, there were considerable discrepancies in the lateral distribution. Depending on the location of the investigation point, type of Rn source, aeration properties of soils, time of measuring, moisture content, and some other factors, the results obtained by the two methods used considerably differed. In case Rn sources are present, additional Rn may migrate into soil air from a depth of 100 m and even from greater depths.

The soils beneath dwellings and their nearest surroundings with a high concentration of Rn or Rn formed in the bedrock under the Quaternary deposits serve as the main sources of Rn in indoor air of dwellings.

The high and very high Rn-risk areas did not follow the distribution of genetic–lithologic types of Quaternary deposits. Outlining these and elucidating their Rn-risk level can be performed only by detailed measurements with applying two parallel methods: calculated from the eU concentration and directly measured with a Markus-10 in soil air.

In the high Rn-risk areas within the klint zone there were some investigation points where the content of U, F, Mo, and other elements exceeded the recommended and even permissible level for residential areas.

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Radooniemissioonid Harjumaal

Krista Jüriado, Valter Petersell ja Anto Raukas

Harju maakonna pinnaseõhu radoonisisaldus on sageli kõrge (>50 kBq/m^3) või väga kõrge (>250 kBq/m^3), ületades mitu korda maailma ja ka Eesti pinnase keskmise. Põhjuseks on kõrge uraanisisaldusega graptoliitargilliidi ja fosforiidi esinemine geoloogilises läbilõikes ning nende purru ja peenese esinemine pinnakattes. Eriti kõrge on radoonisisaldus klindivööndis, kus esineb graptoliitargilliidi ja oobolusfosforiidi paljandeid, nende purru- ning peeneserikkaid kvaternaarisetete erimeid või nende avamus on vahetult pinnakatte all.

eU-sisalduse järgi pinnaseõhus kujunev arvutuslik radoonisisaldus on ajas stabiilne ja heas korrelatsioonis pinnases eU- ning samuti uraanisisaldusega ja litotüüpide lõikes mõnevõrra erinev.

Raadiumi lagunemisel tekkivast radoonist haihtub osa õhku. Samas lisandub radooni pinnaseõhku sügavamal lasuvatest kivimitest. Seega on Markus 10-ga mõõdetud radoonisisaldus tegelikult jääksisaldus. Kuigi see jälgib pinnaseõhus üldjoontes samu seaduspärasusi nagu pinnases eU-st kujunev radoonisisaldus, esineb pindalalises levikus olulisi erinevusi. Sõltuvalt uuringupunkti asukohast, radooniallika tüübist, pinnase aeratsiooniomadustest, mõõtmise ajast, niiskusest ja teistest tingimustest võivad kahe meetodiga saadud mõõtmistulemused oluliselt erineda. Kõrge või väga kõrge radoonisisaldusega majadealune ja selle vahetu ümbruse pinnas ning (või) viimase all aluspõhjakivimites tekkiv radoon on majade siseõhu peamine radooniallikas.

Need kõrge ja eriti kõrge radoonisisaldusega alad ei järgi kvaternaarisetete geneetilis-litoloogiliste tüüpide levilaid. Viimaste kontuurimine ja nende piires radooniriski taseme selgitamine on reaalne ainult detailsete otsemõõtmiste tulemusel, paralleelselt kahe meetodiga, pinnases eU-sisalduse järgi arvutatult ja pinnaseõhus Markus 10-ga otsemõõdetult.

Klindivööndis kõrge radooniriski aladel esineb uuringupunkte, mille pinnases ületab uraani-, fluori-, molübdeeni- jt sisaldus alalistes elurajoonides soovitusliku ja isegi lubatu taseme.