

# Hazardous radioactivity levels and heavy mineral concentrations in beach sediments of Lake Peipsi, northeastern Estonia

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## Abstract

The present study discusses results of heavy mineral analyses and radioactivity of beach sediments of Lake Peipsi. Such analyses are commonly done globally, but had not yet been conducted for the fourth largest lake in Europe. The average heavy mineral content in Lake Peipsi beach sediments along the northern and western coast is higher than usual for Estonian coastal and Quaternary sediments. Concomitantly, elevated radioactivity levels have been measured in several places, with the highest concentrations observed at Alajõe (1885.5 Bq/kg), which is over five times more than the recommended limit. The aim of the present study is to find sites with higher radioactivity levels, because the northern coast of Lake Peipsi is a well-known recreational area.

**Key words:** elevated radioactivity, risk management, beach sediments, heavy minerals, Baltic States

## 1. Introduction

High radioactivity levels as a result of heavy mineral content in coastal sediments have been detected in many parts of the world (Mohanty et al., 2004; Vassas et al., 2006; Carvalho et al., 2011). Aero-geophysical measurements have previously suggested that high radioactivity levels are present in different parts of Estonia (Ylätaalo et al., 1996) where major sources of elevated radioactivity are graptolite argillites (marinite-type alum shales, sapropelic in origin) and the *Obolus* phosphorite. Phosphorites are light yellowish or dark-grey, fine- or coarse-grained, slightly cemented sandy deposits that straddle the Upper Cambrian/Lower Ordovician boundary (Raudsep, 1997). In addition, there is granitoid material that was transported to Estonia by glaciers

from basement rocks in Finland. Some varieties of Devonian sand- and siltstones have higher radioactivity as well. Elevated concentrations of heavy minerals have been identified in different parts of the Estonian coast (Järvelill et al., 2015). Due to complicated cross-shore and alongshore migration of sedimentary material in the coastal zone, enrichment of heavy minerals, including zircon, monazite and other radioactive minerals, has occurred, for example in the Lemme area of southwestern Estonia (Raukas et al., 2014).

Systematic mineralogical studies of Estonian bedrock and Quaternary deposits started at the beginning of the 1960s (Raukas, 1961a, b, 1965; Viiding, 1962, 1968) and specifically for the Lake Peipsi area at the start of the 1980s (Raukas, 1981; Raukas & Rähni, 1982; Tavast, 1984), but to date there have

not been any studies of radioactivity of Lake Peipsi beach sediments. The aim of the present study is to document sites with higher radioactivity levels and take these into account when assessing risks to human health, in view of the fact that the northern coast of Lake Peipsi is a well-known recreational area and even rather low levels of radioactivity represent a considerable threat.

## 2. Description of Lake Peipsi

Lake Peipsi, situated to the south of the Finnish Gulf on the border between Estonia and Russia, is the fourth largest lake in Europe, having a surface area of 3,555 km<sup>2</sup>. The lake consists of three parts: the northernmost, largest and deepest part is Lake Peipsi (*sensu stricto*) (Fig. 1), the southernmost is Lake Pihkva (Pskov) and the narrow, strait-like body of water that connects these two, is called Lake Lämmijärv (Warm Lake). The average depth of the lake is 7.1 m, the maximum depth being 15.3 m. It is approximately 47 km wide and 152 km long and is a remnant of proglacial lakes from the deglaciation period. Parameters of the lake can fluctuate widely with a changeable water level. The catchment basin of the lake (including the lake's surface) has an area of 47,800 km<sup>2</sup>, of which 16,323 km<sup>2</sup> is on Estonian, 27,917 km<sup>2</sup> on Russian and 3,650 km<sup>2</sup> on Latvian territory, with a tiny marshy corner extending into the south of Belarus (Haberman et al., 2008).

### 2.1. Geology of the study area

With the retreat of the continental ice sheet into the Finnish Gulf, a glacial lake, Lake Peipsi, came into being. This retreat was most likely drained to

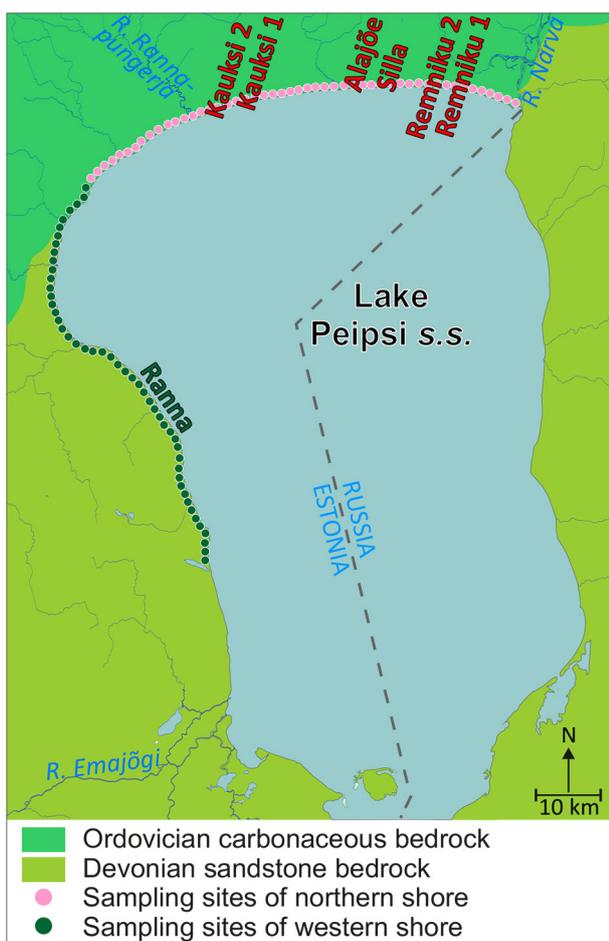
the northeast, but drainage reconstruction down to its minimum level, i.e., c. 20 m a.s.l. (Hang, 2001), was not resolved (Rosentau et al., 2004). Glaciolacustrine sedimentation in Lake Peipsi, which took place between 13,500 and 13,100 varve years BP, correlates with the Pandivere (located to the west of Lake Peipsi) ice marginal zone of late Weichselian glaciation (Hang, 2003; Kalm et al., 2011; Amon et al., 2016).

The crystalline basement of the lake depression is found at a depth of 200–300 m, while the depression itself comprises two sedimentary units: Devonian terrigenous rocks on top of Ordovician carbonates (Fig. 2). Ordovician and Silurian carbonates crop out in the northern part of the depression (Raukas, 2003). The Ordovician–Devonian sequence shows an inclination towards the south to southeast. In the northern and central parts, this amounts to 2–2.2 m per km and in the southern part of the lake 4.5–4.9 m per km; the central part is 50–60 m deeper than the surroundings. The southern part is characterised by a thicker succession of Devonian terrigenous rocks and in the northwestern part of the lake lies a buried valley in Devonian sedimentary bedrock (Miidel et al., 2001).

The thickness of the Quaternary cover is highly variable, ranging from a few metres in the northwest to over 50 m in the central part (Noormets et al., 1998). The Pleistocene sequence is rather simple, the main deposits being tills of the last glaciation and glacial meltwater deposits (Raukas, 2003). The northern part of the lake sediment cover consists of till and glaciolacustrine clay, overlain by Holocene lake deposits (Noormets et al., 1998; Hang, 2001). In the northern part of the lake, Holocene deposits, represented by a lacustrine marl up to 2 m thick and overlain by up to 4 m of gyttja, are missing from many parts of the bottom (Hang, 2001).



Fig. 1. Location of Lake Peipsi (*sensu stricto*).



**Fig. 2.** Sampling locations along the northern (pink dots) and western (green dots) shores of Lake Peipsi (*sensu stricto*), with a simplified geological map of bedrock outcrops (Vaher, 2008; modified by Tiit Vaasma) below Quaternary strata. The seven sites for radioactivity measurements were Remniku 1 and 2, Silla, Alajõe, Kauksi 1 and 2 and Ranna.

Due to land uplift and changing coastal erosion aeolian sands accumulated. Parabolic dunes formed here with blowouts moving downwind and

the low-lying arms fixed with vegetation lag behind. Dune inclination is 10–20° on the windward and 20–30° on the leeward slope. On the Lake Peipsi coast are spread “basket-trap dunes” (according to Orviku, 1933) that formed behind parabolic wind ditches where the vegetation cover was damaged by waves and ice action or by clearing, grazing, burning or trampling (Raukas, 2011).

The lake has an open eastern and a swampy and overgrown western shore. The prevailing south-westerly, southerly and westerly winds caused active erosional shores in the northern and eastern parts of the lake (Raukas & Tavast, 2011). Waves commonly are 60–70 cm high, with the highest up to 240 cm (Jaani & Raukas, 1999). Sedimentation and resedimentation of bottom and coastal deposits is influenced by lake ice, over a period of 114 days on average. Ridges of pressure ice can be up to 10 m in height (Raukas & Tavast, 2011).

On the northern coast of the lake, i.e., the main object of our study, beach sediments are mostly well sorted (1.2–1.3), fine and medium-grained (0.15–0.47) sands. Medium-grained, less well-sorted sands can be found from Kauksi to the River Narva (median 0.38–0.44 mm; sorting 1.29–1.7) (Tavast & Raukas, 1996). The mineral composition of the Lake Peipsi (*sensu stricto*) sediments is impacted by till and local bedrock (Raukas, 1999). The sandy beaches, for example Kauksi, on the northern coast are very popular among tourists and locals (Fig. 3).

### 3. Material and methods

The main study sites of Remniku, Silla, Alajõe, Kauksi and Ranna (Fig. 2) are important recreational areas that belong to the Estonian National Environmental Monitoring Programme. The prime task of this programme is to monitor long-term and large-scale environmental changes and to recognise situations



**Fig. 3.** Sandy beaches on the northern shore of Lake Peipsi. A – Silla; B – Kauksi.

which require counter measures and identify problems and solutions, which call for further complementary studies. During this study of the shores of the large lakes in Estonia (i.e., Peipsi and Võrtsjärv), which have been studied every two years since 1993, changes in coastal morphology, granulometric sediment composition and assessment of impacts of human activities on the coast have been measured (Raukas & Tavast, 2011; Vaasma et al., 2016).

In our study, 99 samples were analysed; 50 from the northern and 49 from the western coast. Samples were collected from beach sediments, backshore and beach face (Fig. 4), with a distance of 1 km between samples (from Kauksi and Remniku were collected two samples after every 200 m). Grain size analyses needed for the analyses of mineral content and radioactivity were conducted with an Analysette 3 Vibratory Sieve Shaker PRO using a sieve set with nine fractions (>2,000, >1,000, >500, >250, >125, >100, >63, >36 and 0–36  $\mu\text{m}$ ) or with a Retsch Vibratory Sieve Shaker type AS200 Basic for radioactivity analysis, using a sieve set with seven fractions (>500, >250, >150, >106, >75, >38 and 0–38  $\mu\text{m}$ ). Grain size is not discussed in the results and discussion chapters to preclude deviation from the topic. The light and heavy (over 2.89  $\text{g}/\text{cm}^3$ ) mineral suites were separated. The immersion method was applied, making use of liquids with refractive indexes of 1.54 and 1.64 (Larsen & Berman, 1934; Stoiber & Morse, 1994). About 300 mineral grains from the light subfraction and 500 grains of the heavy subfraction were counted in each mineral suite on glass slides using a polarising microscope.

The sieved sample fractions from Remniku, Silla, Alajõe, Kauksi and Ranna, as well as integral (bulk) sand samples, were studied using a gamma spectrometer. These samples were first dried at 105 °C



**Fig. 4.** The site of Ranna, with the zone of sample collection marked: backshore and beach face.

and then weighed. From each sample, an aliquot was taken and placed in an airtight 55  $\text{cm}^3$  metal beaker and sealed. When dried, the remaining sample material was sieved into fractions (>2,000, >1,000, >500, >250, >150 or >125, >106 or >100, >75 or >63, >38 and 0–38  $\mu\text{m}$ ). The sealed beakers were stored for at least 28 days to form a secular equilibrium between Rn-222 and its progeny. Two types of metal beakers were used: 55  $\text{cm}^3$  beakers made from steel if there was sufficient material to fill them and 100  $\text{cm}^3$  aluminium beakers with an epoxy resin if there was only a small amount of sample material. For the gamma analyses, this small amount of material was mixed with 5 gr of epoxy resin. Afterwards, this 100  $\text{cm}^3$  aluminium beaker was sealed. After 28 days, these hermetically sealed beakers were analysed by gamma-ray spectroscopy. The measurements were performed by a coaxial, high-purity germanium detector (HPGe) (GEM-35200, EG&G Ortec). The resultant spectra were analysed using the International Atomic Energy Agency (IAEA) analysis programs, Gamma- and Neutron- Activation Analysis Software (GANAA) and Cambio. The spectrometer was calibrated using IAEA standard uranium ore (RGU-1), thorium ore (RGTh-1) and potassium sulphate (RGK-1). The activity concentration of U-238 in the sample was calculated using a gamma line of Th-234 (63.3 keV) and gamma lines of the Ra-226 daughter nuclides Pb-214 and Bi-214, under the assumption that U-238 is in secular equilibrium with the daughter nuclide of Ra-226. This is consistent with how it was considered in previous studies in the Lemme area coastal sands in southwestern Estonia (Koch, 2006; Raukas et al., 2014). The activity concentration of Th-232 in the sample was determined using the gamma lines of Th-232 daughter nuclides as Ac-228, Bi-212, Pb-212 and Tl-208. The activity concentration of K-40 was determined using the 1460.7 keV emission gamma line, while that of U-235 was not calculated separately. The influence of the Ac-228 gamma peak of 1459.19 keV, with a gamma yield of 1.06% to the K-40 gamma peak of 1460.7 keV, with gamma yield of 10.67%, have not been taken into account (De Meijer et al., 2001). It should be noted that 1 Bq of activity concentration for Th-232 would contribute less than 0.1 Bq to the K-40 activity concentration.

The radium equivalent index  $\text{Ra}_{\text{eq}}$  is given by the following formula (Beretka & Mathew, 1985):

$$\text{Ra}_{\text{eq}} = A_{\text{Ra226}} + 1.43 A_{\text{Th232}} + 0.077 A_{\text{K40}}$$

where  $A_{\text{Ra226}}$ ,  $A_{\text{Th232}}$  and  $A_{\text{K40}}$  are the activity concentrations in Bq/kg of the corresponding radionuclides in the sample. The recommended limit for  $\text{Ra}_{\text{eq}}$  values is less than 370 Bq/kg (UNSCEAR, 1982).

## 4. Results

### 4.1. Mineral composition

Mineral composition differentiates samples from the northern and western shores (i.e., areas marked in different colours; Fig. 2) with a greater heavy mineral content in northern shore samples (Table 1). On the northern shore, a lower feldspar content and higher content of heavy minerals, in particular those for ore minerals and zircon, are observed. On the western shore, iron hydroxide content is higher. The zircon content in the north is 4.2% and in the west decreases to the medium level of 2.6%. The garnet content is 27.9% in the north, and nearly identical in the west, 27.2%. The content of ore minerals is higher on the northern shore (28.1%) than on the western shore (22.1%).

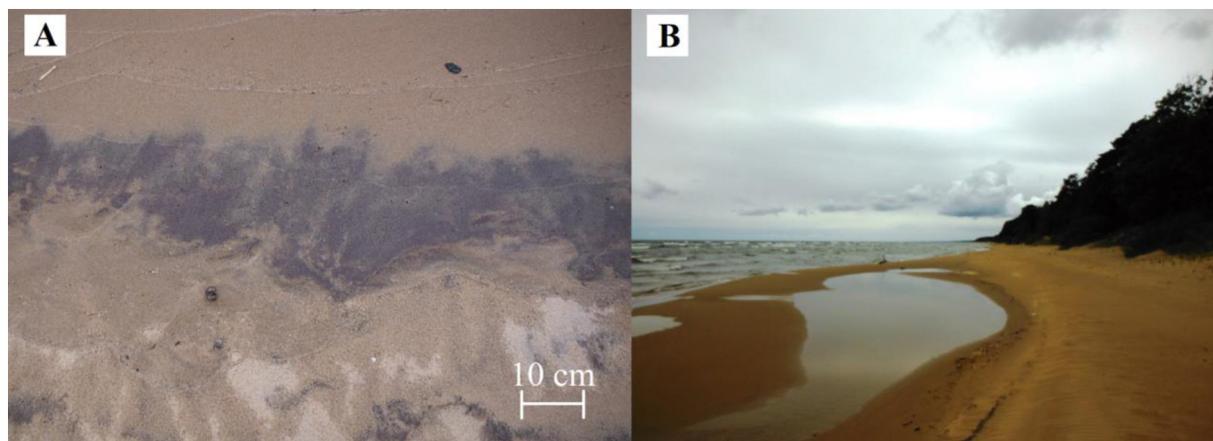
The average heavy mineral content in 92 samples is 1.1%, but when results for seven anomalously enriched samples are included, this value increases to 3.6% (Table 2). The highest heavy mineral content compared to other samples from Peipsi coast was found in samples from Remniku 1, Alajõe and Silla (Table 3).

In all samples, in the fraction 125–250  $\mu\text{m}$ , quartz (83.7%) predominates over feldspars (16%). Among the seven anomalous samples, the K-feldspar content is 15.3%. In the heavy subfraction ore minerals (25.7%) and garnets (27.6%) are the prevailing minerals, while the median zircon content was 3.6% (Table 2).

In the finer fractions (100–125  $\mu\text{m}$ ), the heavy mineral content is higher in comparison to the coarser fraction (125–250  $\mu\text{m}$ ). Samples most suitable to analysis were collected from the sites of Remniku 1 (99.6% of heavy minerals) (Fig. 5) and Ranna (82.2% of heavy minerals).

**Table 1.** Comparison of the mineral content on western and northern shores of Lake Peipsi (*sensu stricto*) (content lower than 0.1% marked with “+”).

	Western beaches (%)	Northern beaches (%)
Light minerals		
Quartz	79.2	86.6
Feldspars	20.2	13.2
Micas	0.2	0
Carbonates	0.4	+
Heavy minerals		
Iron hydroxides	3.5	0.9
Pyrite	+	0.5
Leucoxene	0.9	1
Anatase	+	0
Biotite	3.2	0.2
Chlorides	0	+
Ore minerals	22.1	28.1
Garnets	27.2	27.9
Zircon	2.6	4.2
Tourmaline	1.3	1.3
Apatite	1.9	0.7
Rutile	0.1	+
Titanite	0.1	0.2
Epidote	1.8	2.7
Andalusite	+	0.4
Amphiboles	27.4	26.3
Pyroxenes	2.6	4.1
Dolomite	0.6	0.1
Kyanite	0.1	0.1
Olivine	0.1	0.1
Zoisite	+	+
Staurolite	0.5	0.4
Sillimanite	0.2	0.8
Biogenic apatite	3.8	+
Topaz	0	+
Fluorite	+	0
Sphalerite	+	0
Heavy subfraction	1.7	4.9



**Fig. 5.** Higher heavy mineral concentrations at the Remniku 1 site (A, B). Heavy minerals are visual (dark zone distinguished, A).

**Table 2.** Percentages of minerals in the samples studied; fraction 125–250 µm (content lower than 0.1% marked with “+”).

	Mean (%)	Min (%)	Max (%)	Median	Variance (n-1)	Standard deviation (n-1)
Light minerals						
Quartz	83.7	68.1	96	0.843	0.004	0.065
Feldspars	16	4	31	0.156	0.004	0.063
Micas	0.1	0	2.7	0	0	0.004
Carbonates	0.2	0	2.6	0	0	0.005
Heavy minerals						
Iron hydroxides	2	0	21.2	0.011	0.001	0.03
Pyrite	0.3	0	30.6	0	0.001	0.031
Leucosene	1	0	39.2	0	0.002	0.04
Anatase	0.01	0	0.4	0	0	0
Biotite	1.4	0	56.6	0	0.005	0.068
Chlorides	+	0	0.5	0	0	0
Ore minerals	25.7	0.4	61.2	0.248	0.017	0.132
Garnets	27.6	6.6	47.8	0.294	0.01	0.102
Zircon	3.6	0	13.7	0.032	0.001	0.026
Tourmaline	1.3	0	6.1	0.01	0	0.011
Apatite	1.1	0	4.6	0.01	0	0.011
Rutile	0.1	0	0.7	0	0	0.001
Titanite	0.1	0	1	0	0	0.002
Epidote	2.4	0	11.6	0.02	0	0.021
Andalusite	0.3	0	7.3	0	0	0.009
Amphiboles	26.7	0	75.7	0.238	0.028	0.168
Pyroxenes	3.5	0	35.2	0.028	0.002	0.04
Dolomite	0.3	0	2.4	0	0	0.005
Kyanite	0.06	0	2.9	0	0	0.002
Olivine	0.1	0	3.8	0	0	0.005
Zoisite	0.02	0	0.6	0	0	0.001
Staurolite	0.4	0	3	0.004	0	0.005
Sillimanite	0.5	0	4.4	0.002	0	0.008
Biogenic apatite (broken fossil shells)	1.5	0	20.8	0	0.002	0.039
Topaz	+	0	0.2	0	0	0
Fluorite	0.01	0	0.4	0	0	0.001
Sphalerite	+	0	0.2	0	0	0
Heavy subfraction	3.6	0.04	71.6			
Light subfraction	96.4	28.4	99.96			

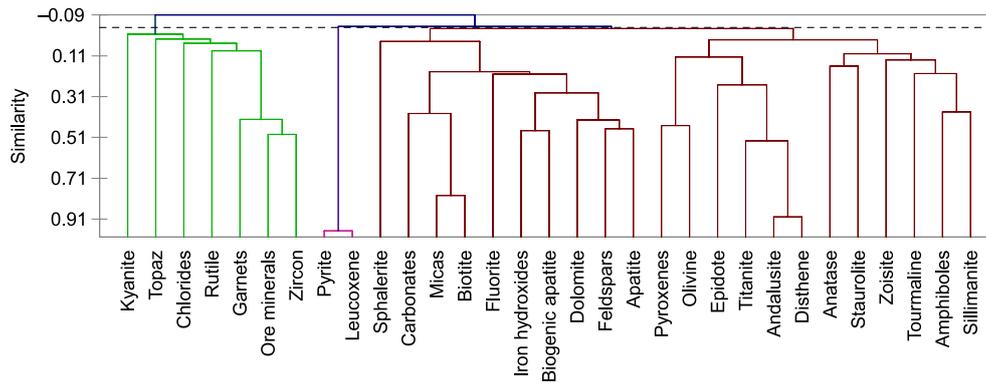
The seven anomalous samples (Table 3) were also analysed for finer fractions, 100–125 µm and 63–100 µm. In the 100–125 µm fraction, the zircon content was found to be present at an average level

**Table 3.** Percentages of heavy and light minerals in anomalously enriched samples (fraction 125–250 µm).

Site	Heavy minerals (%)	Light minerals (%)
Remniku 1	71.6	28.4
Remniku 2	17.5	82.5
Silla	57.4	42.6
Alajõe	61.6	38.4
Kauksi 1	9.4	90.6
Kauksi 2	2.0	98.0
Ranna	23.9	76.1

of 7.6% (six samples), with a maximum found in the Kauksi 1 sample with a zircon content of 15.8%. In the 63–100 µm fraction the zircon content increases to an average of 9.6%, with the highest content found in the Remniku 1 sample, i.e., 21.1%. The content of ore minerals is higher in the fraction 100–125 µm at 50.3% (the highest ore mineral content was found in an Alajõe sample at 62.4%). In the fraction 63–100 µm the ore mineral content is lower at 37.6%, with the highest content found in the Silla sample at 65.5%. It should be noted that these samples are from levels enriched in heavy minerals and they do not characterise the entire sample site.

The assemblage of heavy minerals shows high contents of semi-resistant minerals (27.6% garnet,



**Fig. 6.** Agglomerative hierarchical clustering analysis of minerals in beach deposits of Lake Peipsi (*sensu stricto*). Final clusters emerge above the dotted line.

2.4% epidote), although the non-resistant mineral content is also high (26.7% amphiboles) and the resistant minerals content quite low (3.6% zircon, 0.4% staurolite, 0.06% kyanite, 1.3% tourmaline, 0.1% rutile, 0.3% andalusite, 1.1% apatite, 0.1% titanite).

There are three groups of minerals according to an agglomerative hierarchical clustering (AHC) analysis (Fig. 6) used for assessment of the mineral content of samples. In the first group, the heaviest minerals are observed, such as zircon, garnets, rutile and ore minerals. Pyrite and leucoxene form the second group and the third group, which is the largest, comprises various types of minerals.

## 4.2. Radioactivity analysis

Fractions for which activity concentration of U-238, Th-232 and K-40, as well the radium equivalent ( $Ra_{eq}$ ) and ratio of thorium to uranium from seven sampling sites were calculated, are listed in Table 4.

The activity concentration of Th-232 in bulk samples varies from 6.5 Bq/kg (Kauksi 2) to 1,007.3 Bq/kg (Alajõe), which is a difference of about 155 times. The activity concentration of U-238 in bulk samples varies from 7.96 Bq/kg (Kauksi 2) to 427.55 Bq/kg (Alajõe), which is a difference of more than 53 times. The activity concentration of K-40 in bulk samples

**Table 4.** Grain size distribution of beach sand samples of Lake Peipsi and activity concentrations of radionuclides (Th-232, U-238 and K-40) in different fractions.

Sites, grain size ( $\mu\text{m}$ )	Content in the sample (%)	Th-(Bq/kg)	U-(Bq/kg)	K-(Bq/kg)	Ra-eq.	Th/U
Remniku 1	bulk sample	140.27(4.00)	219.83(2.32)	130.26(0.01)	357.208	0.932
>500	0.707					
>250	54.324	20.41(0.97)	32.44(0.78)	183.26(0.01)	75.748	0.629
>150	36.776	278.48(6.60)	474.00(3.80)	69.64(0.01)	877.591	0.588
>106	1.622	1372(155)	1639(17)	114.66(0.01)	3608.3	0.837
>75	0.023					
>38	0.003					
Remniku 2	bulk sample	37.12(1.13)	28.03(0.97)	259.97(0.01)	92.294	1.288
>500	0.007					
>250	80.462	6.10(0.49)	6.01(0.47)	262.53(0.01)	4.94	1.015
>125	19.354	174.76(3.96)	125.30(2.12)	228.74(0.01)	392.82	1.395
>100	0.128	2337.4(69.9)	848.31(51.31)	373.19(0.01)	4219.51	2.755
>63	0.042	894.77(65.95)	384.80(100)	344.19(0.01)	1690.83	2.325
>36	0.004					
Silla	bulk sample	44.483(1.05)	80.79(0.83)	138.30(0.01)	155.038	0.551
>500	2.066	6.54(4.6)	8.01(1.00)	254.18(0.01)	36.94	0.817
>250	81.127	11.84(0.60)	14.07(0.65)	143.37(0.01)	42.04	0.842
>125	16.78	189.51(5.76)	388.69(5.45)	70.09(0.01)	665.085	0.488
>100	0.018					
>63	0.004					
Alajõe	bulk sample	1007.3(21.5)	427.55(4.18)	227.00(0.01)	1885.51	2.356
>500	14.218	9.90(0.57)	8.26(0.68)	354.88(0.01)	49.746	1.199
>250	27.758	138.90(4.39)	66.90(1.96)	303.02(0.01)	288.862	2.076

Sites, grain size ( $\mu\text{m}$ )	Content in the sample (%)	Th-(Bq/kg)	U-(Bq/kg)	K-(Bq/kg)	Ra-eq.	Th/U
>150	45.404	1427.2(30.8)	630.90(6.16)	158.51(0.01)	2684.053	2.262
>106	5.396	5089.4(107.4)	362.4(14.4)	391.24(0.01)	8670.33	3.736
>75	0.096	10724(272)	004.6(107)	578.03(0.02)	18384.41	3.569
>38	0.006	1185(381)	673.34(193.34)	0.00(0.01)	2367.245	1.759
Kauksi 1	bulk sample	44.73(1.20)	35.55(0.95)	229.54(0.01)	117.196	1.258
>500	0.077	0.000(0.0)	25.10(6.06)	211.78(0.01)	41.4	0
>250	75.837	20.66(0.80)	19.30(0.93)	244.95(0.01)	67.706	1.07
>125	23.985	107.13(2.51)	83.77(1.54)	173.11(0.01)	250.287	1.279
>100	0.094	4985(147)	2329(101)	263.72(0.01)	9478.504	2.141
>63	0.005					
Kauksi 2	bulk sample	6.50(0.55)	7.96(0.66)	411.50(0.01)	48.937	0.816
>500	0.948	11.80(2.83)	11.72(2.23)	394.59(0.01)	58.977	1.006
>250	92.559	6.04(0.50)	6.11(0.51)	385.53(0.01)	44.433	0.989
>125	6.465	12.59(0.61)	15.97(0.80)	279.44(0.01)	55.486	0.789
>100	0.014					
>63	0.006					
Ranna	bulk sample	47.81(1.66)	40.44(1.09)	670.40(0.01)	160.439	1.182
>500	27.689	19.52(1.00)	17.33(0.80)	1170.37(0.01)	135.364	1.126
>250	41.212	22.49(1.18)	16.68(0.84)	614.24(0.01)	96.137	1.348
>150	20.514	337.61(8.10)	237.94(2.73)	122.07(0.01)	730.123	1.419
>106	1.433	519.20(19.90)	409.86(3.97)	168.57(0.01)	1165.291	1.267
>75	0.246	822.22(26.50)	820.7(29.0)	436.97(0.02)	2030.074	1.002
>38	0.063	609.55(29.52)	458.3(38.4)	983.45(0.03)	1405.691	1.33
0–38	0.03	95.61(18.34)	78.41(21.6)	1033.71(0.04)	294.736	1.219

varies from 130.26 Bq/kg (Remniku 1) to 670.40 Bq/kg (Ranna), which is a difference of more than five times. The activity concentration of Th-232 and U-238 is highly dependent on particle size. The highest activity concentration for Th-232 (10,724 Bq/kg) was found in the 75–106  $\mu\text{m}$  fraction from the Alajõe sampling site and the highest activity concentration for U-238 (3,004.6 Bq/kg) was measured in the same sample. The maximum K-40 activity concentration (1170.37 Bq/kg) was found in the >500  $\mu\text{m}$  fraction from the Ranna sampling site. A high concentration of potassium (> 1,000 Bq/kg) was observed for samples from Ranna (1,170.37 Bq/kg) in the >500  $\mu\text{m}$  fraction and (1,033.71 Bq/kg) in the 0–38  $\mu\text{m}$  fraction, whereby activity concentrations for Th and U in these samples were rather low. The thorium/uranium ratio for bulk samples is less than 2.0, except for the sample from Alajõe (2.356), whereby for samples from Remniku 1, Silla and Kauksi 2, the value is below 1.0. The radium equivalent index for bulk samples was calculated from the  $\text{Ra}_{226}$ ,  $\text{Th}_{232}$  and  $\text{K}_{40}$  concentrations, and varied from 48.94 Bq/kg (Kauksi 2) to 1,885.5 Bq/kg (Alajõe).

The mean measured values of thorium, uranium and potassium contents from bulk samples ranges between 6.50 Bq/kg (Kauksi 2) and 1,007.3 Bq/kg (Alajõe) for Th; from 7.96 Bq/kg (Kauksi 2) to 427.55 Bq/kg (Alajõe) for U; and from 130.26 Bq/kg (Remniku 1) to 670.40 Bq/kg (Ranna) for K. The world-

wide average concentrations of these radionuclides are: 35 Bq/kg for U (Ra-226), 30 Bq/kg for Th-232 and 400 Bq/kg for K-40 (UNSCEAR, 2000). The average nuclide concentrations in Estonian soils are as follows: 510 Bq/kg for K-40, 35 Bq/kg for Ra-226 and 27 Bq/kg for Th-232. The maximum concentration of Th-232 in our bulk samples exceeds the Estonian mean value by about 34 times, while the maximum concentration of U-238 exceeds the mean

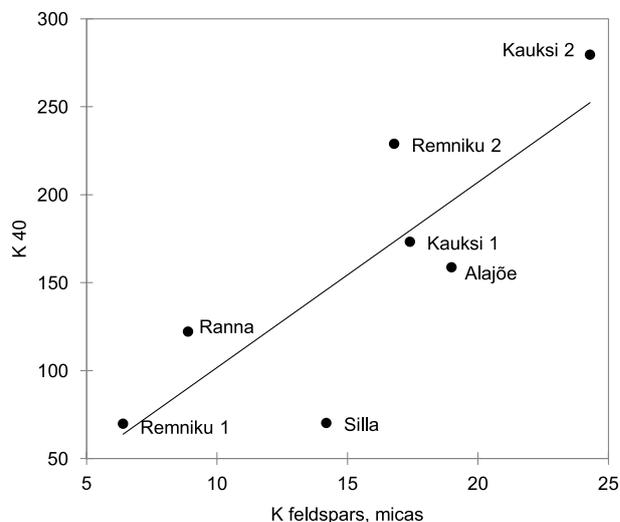


Fig. 7. Scatter plot for K-40 vs K-feldspars and content of micas.

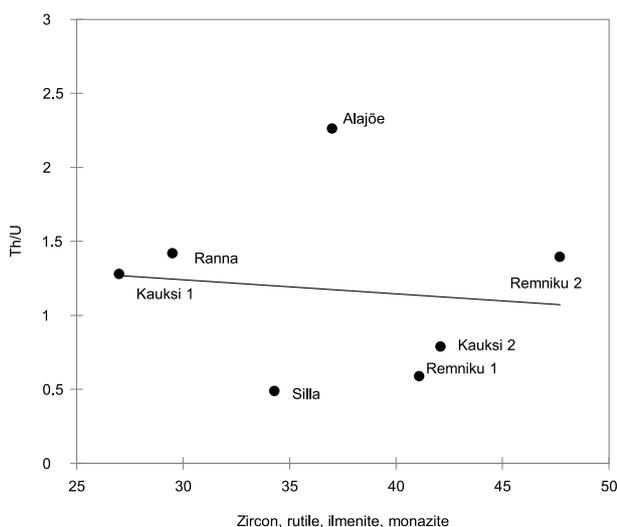


Fig. 8. Scatter plot for Th/U vs content of zircon, rutile, ilmenite and monazite.

value by about 12 times, and about 1.3 times for the concentration of K-40. Considering that 1 ppm of eU and eTh corresponds to 12.3 Bq/kg and 4.0 Bq/kg, the calculated maximum contents of Th-232 and U-238 in samples (from the Alajõe fraction >75 µm) is 2,638 ppm for eTh and 243 ppm for eU.

The thorium/uranium ratio for bulk samples is less than 2.0, except for the sample from the Alajõe sampling point, whereby for samples from Remniku 1, Silla and Kauksi 2, the Th/U ratio remains below 1.0. The radium equivalent index for bulk samples, calculated from the  $Ra_{226}$ ,  $Th_{232}$  and  $K_{40}$  concentrations, varied from 48.94 Bq/kg (Kauksi 2) up to 1,885.5 Bq/kg (Alajõe), which exceeds the recommended limit of 370 Bq/kg by over five times (UNSCEAR, 1982).

## 5. Discussion

### 5.1. Mineral composition

Beach sediments have bedrock and glacial sources and are modified by coastal hydraulic processes connected with wave action, storms, wind and water level changes. Therefore, higher heavy mineral concentrations on the northern shore (4.9%) than the western shore (1.7%) are observed.

In India, Kurian et al. (2001) identified that the cause of heavy mineral enrichment was monsoon activity (swells) and winnowing which had an influence on the swell period. They noted that mineral content of denser heavy minerals such as opaques (ilmenite and magnetite), monazite and zircon con-

tent decreased, while lighter minerals such as sillimanite, hypersthene (pyroxene) and hornblende increased in an offshore direction. Other authors, e.g., Pupienis et al. (2013), documented that the heavy mineral content on the surface of the beaches was influenced by wave and wind action. These tendencies are also observed in our samples. The highest contents of opaques and zircon were found near the shoreline at backshore and beach face. Heavy mineral concentrations are connected to the sites affected by heavy winds and more pronounced water level changes. Lighter minerals are eroded by wind and waves. Heavy mineral accumulation is characterised by an increase of ore minerals, garnets, amphiboles and zircon. Also, in Lake Peipsi (*sensu stricto*) already half a metre of water level rise can lead to serious damage and erosion.

Beach sediment composition is characterised by mineral concentrations with similar densities. Minerals with higher densities mostly appear together, e.g., garnets, zircon and ore minerals (Fig. 6). These denser minerals are more concentrated on the northern shore of Lake Peipsi (*sensu stricto*).

Kalińska-Nartiša et al. (2015) showed in their work near Lake Peipsi that the aeolian sediments contained 13–28% of feldspars in the fraction of 0.5–0.8 mm and 12–34% of feldspars in the fraction of 0.8–1.0 mm. Our results are 4–31% of feldspars in the 0.125–0.25 mm fraction, thus showing more equally distributed results and decreased contents.

Near the Rannapungerja River (Fig. 2), some changes in the mineral content due to the influence of the river can be observed. The heavy mineral content here is only 0.6%. The fluvial processes influence the heavy mineral content by decreasing it. It is assumed that the heavy minerals are flushed away by the river flow.

Garnet, which is highly resistant to mechanical abrasion and medium resistant to chemical weathering (Morawski, 1968), is abundant in the samples. This content is increased in the eroded and redeposited beaches (Racinowski, 1974, 1992; Marcinkowski & Mycielska-Dowgiałło, 2013). The high content of amphiboles indicates that mechanical abrasion has not been high (Goździk, 1980; Manikowska, 1985; Krzyszkowski, 1995; Mycielska-Dowgiałło, 1995; Marcinkowski & Mycielska-Dowgiałło, 2013), but they are slightly more resistant to chemical weathering than are pyroxenes. This means that amphibole content should decrease in deposits affected by long-lived aeolian processes, but in our samples the content is high. Where Racinowski (1974, 1992) and Marcinkowski & Mycielska-Dowgiałło (2013) noted that the beach and nearshore sands had the lowest content of amphiboles and the highest content of

garnets in the 0.12–0.25 mm fraction, this does not apply to our samples. The beach should be enriched by minerals that are resistant to mechanical abrasion, but in our samples from Lake Peipsi amphibole content is high.

## 5.2. Radioactivity analysis

Thorium, uranium and potassium contents are influenced by the mineral compositions of sediments and rocks. There is a strong dependence of radionuclide concentration on grain size in the sand samples, which has been previously observed by other authors (De Meijer, 1998; De Meijer et al., 2001; Seddeek et al., 2005; Koch, 2006; Madruga et al., 2014). Smaller grain size generally is associated with higher activity concentrations. The content of Th and U in samples from Lake Peipsi is closely related to the grain size of the sample, this being significantly higher than in coarser fractions. For all samples with an elevated content of Th and U, this is observed only for fractions with a grain size less than 250  $\mu\text{m}$ . However, this does not apply in the case of K-40 activity concentrations.

The average Th/U ratio for bulk samples is similar to that found in coastal sands of western Estonia (Koch, 2006) and varies from 0.55 to 2.36, which is less than the well-known literature value of 2–2.5 (Mahdavi, 1963; Abdel-Halim & Saleh, 2016).

The potassium content is mostly strongly correlated (Pearson correlation coefficient 0.819) with the presence of K-feldspars and micas (Fig. 7). Potassium still occurs in rock-forming minerals after disintegration of source rocks. The sample from the Ranna sampling point indicates an elevated K-40 content of 1,170.4 Bq/kg in the >500  $\mu\text{m}$  fraction, 983.5 Bq/kg in the >38  $\mu\text{m}$  fraction and 1,033.7 Bq/kg in the 0–38  $\mu\text{m}$  fraction. Uranium and thorium atoms are mostly bound to accessory and dark-coloured minerals (heavy minerals with red, purple or black colour; compare Carvalho et al., 2011). Our samples show a negative correlation between the U/Th concentration and the content of zircon, rutile, ilmenite, monazite (Fig. 8) (Pearson correlation coefficient –0.113).

## 6. Conclusions

At several sites on the beaches of Lake Peipsi (*sensu stricto*), elevated heavy mineral concentrations have been found. Among these heavy mineral accumulations we found also higher concentrations of Th and U, highest in Alajõe and higher than the rec-

ommended (UNSCEAR, 1982) and usual concentrations in Estonia. The radium equivalent index  $Ra_{eq}$  for the sample from Alajõe exceeds the recommended limit more than five times (UNSCEAR, 1982). These concentrations should be taken into account, as they may present a risk to human health. This hazard, however, is decreased by the fact that these concentrations are found only in enriched samples, which are located within narrow-accumulated stripes of sand.

The high content of heavy minerals is due to large fluctuations in the water of the lake and rather heavy winds. In addition, fluvial processes have some influence by inversely decreasing the heavy mineral content. The sites studied that belong to the National Monitoring Programme have rather high concentrations of heavy minerals. At the Ranna site, the content of heavy minerals in the 100–125  $\mu\text{m}$  attains up to 82.2% at some levels.

## Acknowledgements

We are grateful to Kalev Järvelill for assistance during fieldwork and the Tallinn University Centre of Excellence “Natural Sciences and Sustainable Development” for funding the research.

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Manuscript submitted: 9 August 2017

Revision accepted: 17 February 2018