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Radon in the soil air of Estonia

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ABSTRACT

Several investigations in Estonia during 1996¬-1999 have shown that permissible level (200 Bq/m^3) of radon (222Rn) in indoor air is exceeded in 33% of the inspected dwellings. This makes Estonia one of the five countries with highest radon risk in Europe (Fig 1). Due to correlation between the soil radon risk level and radon concentration in houses, small scale radon risk mapping of soil air was carried out (one study point per 70–100 km^2). It turned out that one-third of Estonian mainland has high radon risk potential, where radon concentration in soil air exceeds safe limit of 50 kBq/m^3.

In order to estimate radon content in soil air, two different methods developed in Sweden were used simultaneously. Besides measuring radon content from soil air at the depth of 80 cm with an emanometer (RnM), maximum potential content of radon in soil (RnG) was estimated based on the rate of eU (226Ra) concentration in soil, which was acquired by using gamma-ray spectrometer.

Mapping and following studies revealed that simultaneously measured RnG and RnM in study points may often differ. To inspect the cause, several monitoring points were set up in places with different geological conditions. It appeared that unlike the RnG content, which remains close to average level in repeated measurements, the RnM content may differ more than three times periodically. After continuous observations it turned out that concentration of directly measured radon depended on various factors being mostly controlled by mineral composition of soil, properties of topsoil as well as different factors influencing aeration of soil.

The results of Rn monitoring show that reliable level of radon risk in Estonian soils can only be acquired by using calculated Rn-concentration in soil air based on eU content and directly measured radon content of soil air in combination with interpreting specific geological and geochemical situations in the study points.

1. Introduction

Several studies by the Estonian Radiation Protection Centre and the Swedish Radiation Protection Institute in 1996–1999 showed that average concentration of radon (^{222}Rn) was up to 120 Bq/m^3 (UNSCEAR, 2000) in indoor air of single-story dwellings in Estonia. The permissive limit of radon concentration of indoor air (200 Bq/m^3) was exceeded in 33% of inspected dwellings, reaching up to 10 000 Bq/m^3 in some cases (Pahapill et al., 2003). Accordingly, Estonia is one of the top five countries in Europe with the highest indoor radon risk in EU (UNSCEAR, 2000).

It was presumed that high concentration of radon in indoor air is caused by its excessive amount in soil air. Therefore, in collaboration with Swedish colleagues, the Geological Survey of Estonia started collecting data to solve this issue in 2001. The first map of radon risk in Estonia was compiled in 2001–2004, basing on modification of the method used in Sweden by the Swedish Radiation Protection Centre (Åkerblom, 1994; Clavensjö and Åkerblom, 1994). Mapping revealed that almost one third of the Estonian mainland has high (>50 kBq/m^3) or very high (>250 kBq/m^3) level of radon risk. Same areas have relatively high uranium concentration (>3.5 ppm) either in soil, basement rocks, or both.

1.1. Sources of radon in Estonia

Estonia is situated in the north-western part of the East-

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European Platform overlaying the southern slope of the Fennoscandian Shield. The Estonian basement is formed by Proterozoic crystalline rocks, which are covered by terrigenous Ediacaran and Cambrian sedimentary rocks followed by Ordovician terrigenous and carbonaceous and Silurian carbonaceous sedimentary rocks with Devonian sandstones and siltstones on top. The uppermost Quaternary deposits consist of different types of sediments.

Up to 56 m high and subparallel to Gulf of Finland, The North Estonian Klint is a dominant feature along the north Estonian coast. However, it may also occur as several small escarpments or be entirely buried and is therefore easily overlooked at some locations (Suuroja, 2006).

Overall thickness of the sedimentary rock cover varies from 120 to 160 m in north Estonia to 400–550 m in the south. Loose Quaternary sedimentary layer is mostly between 1 and 20 m in thickness, except for areas of alvars where soil layer is thin or entirely missing, or primeval valleys, uplands and heights with sediment cover of 100–143 m (Raukas and Kajak, 1997).

Most important radon sources are the Lower Ordovician uranium-rich Obolus sandstone (phosphorite), which overlays the Cambrian system and graptolite argillite (also called as Dictyonema shale, an analogy of alum shale in Sweden, Andersson et al., 1985) on top of it. They are both exposed or crop out along the Klint and in the slopes of the valleys cutting it between the city of Narva and Pakri peninsula. Both rock layers occur in north Estonia in extensive areas as sub-parallel beds in the lower part of the Ordovician sequence and dip southwards about 3 m per km. Thickness of graptolite argillite varies between 0.2 and 6 m increasing from east to west. Its uranium concentration ranges mainly from 30 to 210 ppm, exceeding the limit of 300 ppm at some areas (Petersell, 1997). Thickness of the phosphorite bed (Obolus sandstone) underlying the graptolite argillite varies, being at its maximum of 12 m in the Rakvere region. Uranium concentration of the bed depends directly on phosphorus concentration in the rock and varies mainly between 4 and 40 ppm (Loog and Petersell, 1990). In addition, signs of hydrothermal uranium mineralization (>200 ppm, U) is known in sedimentary rocks in north-eastern part of Estonia.

Interlayers or lenses of dark clay and siltstone containing uranium (<10 ppm) with thickness of up to 3 m can be found in the Devonian sandstones, which in general are poor in clay content. Uranium concentration of similar sandstones occurring about 15 km east of Estonia in Russian territory may reach to uranium concentrations of up to 300 ppm (Sammet et al., 1974). In addition, Devonian sedimentary rocks may be locally enriched with uranium-rich zircon and contain orthite, monazite, apatite and xenotime (Viiding, 1961, 1984; Viiding et al., 1983; Raukas et al., 2014).

The crystalline fine material, pebbles and boulders, originated from the basement, are widely present in the Quaternary system. The most common are *rapakivi* granite cropped out from the bottom of the Gulf of Finland, south Finland and Karelia. However, pegmatite granite boulders are also present (Koistinen, 1996). These rocks are widespread and their uranium concentration varies within 3–10 ppm (Koljonen, 1992). Rocks of the same formations exist also in the crystalline basement in north Estonia at depths of 120–300 m (Metsur et al., 2012).

Glacial movements crushed and relocated material from the crystalline basement and sedimentary rocks during recurring ice ages (Kalm, 2012). As a consequence, fragments of those rocks now occur in various concentrations in moraines, but also in other deposits formed during the Quaternary period in Estonia. On the plain between the Klint and the Gulf of Finland, uranium-rich debris can be found which is relocated from the escarpment by the sea.

2. Methods

To estimate radon (^{222}Rn) concentration in soil air, modification of the method developed in Sweden was used (Petersell et al., 2005, 2012). Radon content in study points was described using two different approaches. Besides measuring with emanometer (RnM) from soil air at the depth of 80 cm, maximum potential content of radon in soil (RnG) was estimated based on the concentration of eU (^{226}Ra) in soil, which was acquired using gamma-ray spectrometer.

When choosing the study points, areas with signs of pollution were left out. Measurements of eU concentration were carried out using the gamma-ray spectrometer GR 320 (Detector model GPX-21A; Exploranium) at the bottom of approximately 20 cm wide holes at depths of 80 cm. Simultaneously measured eTh and eK content allows to estimate the amount of generated thoron (^{220}Rn) and, together with eK level, natural radiation.

As gamma spectrometer is adjusted for 2π configuration describing infinite flat surface, acquired results required conversion from 4π to 2π configuration using the factor 0.63. Concentration of radon in soil air (RnG) was calculated using the following formula: (Clavensjö and Åkerblom, 1994)

$$RnG = Ae\sigma\ (1-p)p^{-1}$$

where:

RnG – Maximum radon content developed, Bq/m^3;
A – eU concentration, Bq/kg;
e – Radon emanation factor;
σ – Compact volume weight (specific weight), kg/m^3;
p – Porosity.

Radon emanation factor, specific weight and porosity were calculated for different types of sediment lithologies based on the database of more than 500 measurements excluding any anomalous values.

Simultaneously with gamma-ray measurement, radon in soil air (RnM) was determined using the emanometer Markus-10 (Gammadata, Sweden). Based on the soil type and radon diffusion dependence graph (Clavensjö and Åkerblom, 1994), results of directly measured radon concentrations were calculated to standard depth of 1 m.

3. Results and discussion

3.1. Radon content in soil air

Soil in Estonia is mostly represented by north (mp) and south Estonian moraines (ml) with glaciolacustrine silt (lga) or sand covering them at some places. Sediments lying near the slope of the Klint are roughly of the same material, but may contain uranium-rich phosphorite or graptolite argillite debris and fine grained material so that they can be classified as a separate lithological type (kla).

RnG and RnM content varied significantly in areas with different soil types (Fig. 1). Values of RnG were between 1 and 1802 kBq/m^3 with a geometric mean of 31 kBq/m^3. RnM at the same time reached from 1 to 2112 kBq/m^3 with a geometric mean of 27 kBq/m^3. While the highest ratio between the geometric averages of RnM and RnG was typical to measurements carried out on south Estonian moraines, the lowest ratio appeared in results collected near the Klint slope. Although the RnM and RnG results remained altogether roughly between same boundaries, they often differed up to three or more times in single study points.

In accordance with Estonian standards (EVS 840, 2009), radon

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