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Groundwater treatment as a source of indoor radon



Alar Jantsikene ^{a,*}, Madis Kiisk ^a, Siiri Suursoo ^a, Rein Koch ^a, Liie Lumiste ^b

- ^a University of Tartu, Institute of Physics, Riia 142, 51014 Tartu, Estonia
- ^b Tallinn University of Technology, Department of Chemical Engineering, Ehitajate tee 5, 19086 Tallinn, Estonia

HIGHLIGHTS

- This work presents the preliminary results of a ²²²Rn study conducted during a period of one and a half years in a new Viimsi Parish water treatment plant.
- It was quantitatively established that both open filter columns of water treatment plant act as a source of indoor air ²²²Rn.
- ²²²Rn indoor measurements under the normal operation conditions revealed that activity concentrations in the filter hall of water treatment plant fall between 600 and 700 Bq/m³.

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ABSTRACT

New Viimsi Parish water treatment plant (Northern Estonia) was investigated in order to determine whether the open filter columns serve as a source of ²²²Rn generation in the treatment process and whether they influence indoor air ²²²Rn activity concentrations. ²²²Rn measurements of indoor ²²²Rn were performed at different locations of the treatment plant; water samples from incoming raw water, from all the purification stages, consumers water and solid filter material from two filtration stages were analyzed.

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1. Introduction

Increased activity concentrations of inhaled radon (henceforth ²²²Rn) and its daughter nuclei are posing increased lung cancer risk. Collaborative analysis of individual data from 13 European Case-control Studies has shown, that the risk of lung cancer increases by 16% per 100 Bq/m³ increase in ²²²Rn activity concentration (Darby et al., 2005). Indoor ²²²Rn is a dominant contributor to the ionizing radiation dose received by the general population in Estonia. According to the Estonian Standard EVS 840:2009 "Design of radon-safe buildings", the annual ²²²Rn concentration in living-, rest- and workrooms should remain below 200 Bq/m³ (Estonian standard, 2009). As estimated by the United States Protection Agency (EPA), additional cancer risk comes from drinking water containing dissolved ²²²Rn. The guidance level for ²²²Rn in drinking water is 100 Bq/L (European Commission, 2001). In general, indoor air ²²²Rn originates from the ground: either by direct diffusion from soil through the foundation or by being dissolved into groundwater used in households. In some cases, the latter might become a dominant source of indoor air ²²²Rn.

Its inert gas characteristics allow it to enter buildings and move from one room to another through the smallest pores, cracks and poorly isolated wall joints, provided there is any air flow.

Viimsi parish is known for its indoor ²²²Rn problems resulting from the high amounts of Alum Shale that causes high ²²²Rn activity concentrations in soil. Cambrian-Vendian (henceforth Cm-V) water used for drinking exceeds total indicative doses of radium isotopes (226Ra and 228Ra) by manifold (Forte et al., 2010) set in the drinking water EU directive (2001/928/Euratom) (European Commission, 2001). In order to diminish radium content in groundwater, Viimsi parish launched a new water treatment plant in winter 2012. It is situated in Viimsi parish, Northern Estonia, 15 km east from capital of Estonia, Tallinn. Four out of five parallel treatment lines are currently in operation, serving about 15,000 consumers. It is designed to remove iron, manganese, radium and dissolved gases (including ²²²Rn) from groundwater. Equipped with five parallel water treatment lines, it pumps water from nine Cm-V wells. Based on the measured ²²⁶Ra and ²²⁸Ra activity concentrations of all the wells, the estimated annual input of Ra-isotopes into the plant given the average capacity of $4000\,\mathrm{m}^3/\mathrm{d}$ is $440\,\mathrm{MBq}$ for $^{226}\mathrm{Ra}$ and $580\,\mathrm{MBq}$ for ²²⁸Ra. As indicated by the measurements of the output water samples, majority of the input Ra isotopes (below the limit of detection, $\sim 10-15 \text{ mBg/L}$) remain in the treatment plant after treatment (Suursoo et al., forthcoming). Thus, one can expect strong

^{*} Corresponding author. Tel.: +3725226349.

E-mail address: alar.jantsikene@ut.ee (A. Jantsikene).

accumulation of Ra isotopes in the filter materials of the plant and ²²²Rn emanation in large quantities.

Based on the previous studies that have pointed out that zeolite sand and FMH FILTERSORB® are effective adsorbers of radium (Lumiste et al., 2012), Viimsi water treatment Ltd. plant opted technology which is based on these adosrbers. In order to estimate if both open filtration columns of new Viimsi water treatment plant serve as a source of ²²²Rn generation in the treatment process, a series of measurements were conducted. This work presents the preliminary results of a ²²²Rn study conducted during a period of one and a half years in Viimsi water treatment plant with the main focus on the ²²²Rn generated inside the plant.

2. Materials and methods

2.1. Description of the water treatment plant

Equipped with five parallel water treatment lines, Viimsi water treatment plant pumps water from nine Cm-V wells. Average designed capacity is 4000 m³/d (max. 6000 m³/d). The treatment scheme involves several purification steps (see Fig. 1). First, there is an air injector (1) and an oxidation tank (2). The venture-type air injector is used for aeration, which allows later oxidation of Fe^{2+} and Mn^{2+} . The oxidation tank (h=4350 mm, dn=1500 mm) provides required residence time (about 5-7 min) for Fe²⁺ and Mn²⁺ oxidation. Second, there is a special type of patented venturi-type centrifugal degassing separation unit GDT (Gas-Degas Technology, Maazei Corp., USA) (3), which separates gasses in the water (e.g. ²²²Rn, CO₂, H₂S). A ventilation connected with the oxidation tank and GDT-separator draws entrained gasses from the incoming groundwater outside the building and releases it into atmosphere. Finally, after the gas removal process, water undergoes a two-stage purification from Fe, Mn and heavy elements (e.g., Ra-isotopes) in open filter columns. First filtration stage (4) consists of gravel (h=150 mm, dn=3000 mm) non-catalytic quartz sand (h=600 mm, dn=3000 mm) and catalytic filter material (FILTERSORB® FMH) (h=1200 mm, dn=3000 mm), which is coated with MnO₂. Its purpose is to remove Mn, Fe and Ra isotopes. Second filtration stage (5) consists, in addition to the gravel (dn=3000 mm, h=150 mm) and quartz sand (h=400 mm, dn=3000 mm), of non-catalytic zeolite sand (type Zeolith N) (h=1500 mm, dn=3000). Zeolite sand is a known adsorbent that has a negative surface charge, so it is used primarly for the adsorption of excessive cations (e.g. Ra isotopes, Mn, NH₄). It has also a large specific surface area (Mar Camacho et al., 2010). FMH is not porous and has smaller specific surface area compared to zeolite sand. For the cleansing of filters air-water backwash system is devised. For the first year backwash interval was set to 120 h for the first stage filtration and 168 for the second one, with a volume of $35 \, \text{m}^3/\text{cycle}$. After 1 year of monitoring the accumulation of investigated elements the backwash interval for zeolite sand was extended, being now after each 336 h. During the period of study the filter materials were not replaced and their lifespan has not been estimated yet. After passing through all these purification steps (see Fig. 1), water is fed to the consumers through three water reservoirs $(3 \times 2000 \, \text{m}^3)$ (Lumiste et al., 2012).

Along the flow sheet five faucets are present after each step of purification (see Fig. 1), which simplifies sampling. Additional sampling point is the water tap situated in the plant, which represents the water fed to the consumers. The plant building itself is divided into two main sections. One side is fully devoted to the filter system hall, where all the previously mentioned purification processes take place. The other side of the building takes up as much height as the filter system hall, but is divided into two floors. The first floor hosts laboratory and dressing rooms; the second floor is devoted to the offices. Unlike other offices, the conference room and plant operator's room on the second floor is separated from the filter system hall with glass wall instead of fibo blocks.

2.2. Analysis procedure for quantification of ²²²Rn in water

2.2.1. Sample preparation

In this method, an organic exctracting 2-phase scintillation cocktail (Betaplate ScintTM, PerkinElmer) was used. In order to remove dissolved ²²²Rn during the storage, the Betaplate Scint cocktail underwent aeration with argon gas, as commonly mentioned in pertinent literature (Salonen, 2008). Next, 20 ml glass vial was filled with 10 ml of cocktail, sealed tightly with aluminum foil coated caps and transported to the sampling site. In order to quantify the exact amount of each sample, the vial and inserted cocktail were previously weighted.

2.2.2. Sampling

The sampling method is important to avoid ²²²Rn losses during water sample collection (Passo and Floeckher, 1991). Water samples were taken from the following sampling points (see Table 1): incoming raw water, after oxidation, after aeration, after first stage filtration, after second stage filtration and from tap water (consumer's water). It is assumed that the samples are representative right after opening the faucet, since the samples are collected from the system with continuous water flow. To minimize the loss of ²²²Rn during sampling, 10 ml of water was taken with a syringe and infused directly under 10 ml of Betaplate Scint cocktail contained

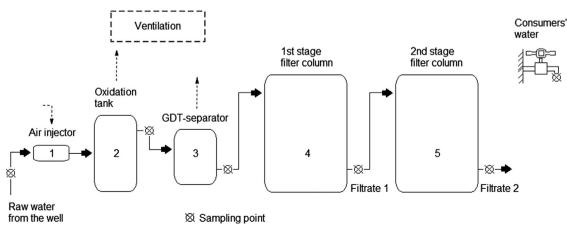


Fig. 1. Viimsi water treatment plant flow scheme and the water sampling points.

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