Research paper

NORM-related industrial activities in Estonia – Establishing national NORM inventory

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ABSTRACT

There is incomplete information available concerning NORM-related (naturally occurring radioactive material) industries in Estonia. In order to fill the gap in this knowledge, a nationwide study was carried out between 2015 and 2017 to determine and radiologically characterize potential NORM-related industries. The study included compiling available literature and studies as well as on-site measurements (external dose rate and radon) and samplings in multiple industries, which had been determined to be potential NORM creators. The results from this study concluded that there are 3 industries in Estonia where the activity concentrations of naturally occurring radionuclides can reach an increased level which may require further regulatory control. Radionuclide exemption values were clearly exceeded in the filter materials of drinking water treatment plants that use water from Cambrian-Vendian aquifer. $^{226}$Ra and $^{228}$Ra values in filter materials reached over 40 kBq kg$^{-1}$. Additionally, the gross radionuclide activity concentrations of $^{238}$U and $^{232}$Th in waste from a rare metal production industry reached up to 191 kBq kg$^{-1}$. Clinker dust from the cement industry showed elevated concentrations of $^{210}$Pb, rising to over 2 kBq kg$^{-1}$ in fine clinker dust.

The creation of a national NORM inventory can be the basis for establishing a national NORM strategy for the management of NORM residues. This work provides a thorough overview of the radiological parameters of the potential NORM-related industries, describes the radionuclides that have elevated concentrations, provides estimations on their yearly creation amounts and produces input for determining possible NORM management options.

1. Introduction

Radionuclides of natural origin can be found in all minerals and raw materials. For the purposes of radiation protection, the most important are radionuclides of the $^{238}$U and $^{232}$Th day series. Numerous industrial activities that involve naturally occurring radioactive material (NORM) can increase workers’ or the public’s exposure to radiation, which is why there is a need for regulation and control from a radiation safety perspective.

The European Union (EU) Member States (MS) were obliged to transpose the requirements from 2013/59/EURATOM (Directive) (European Parliament, 2014) to national legislation in order to ensure protection against ionizing radiation at the beginning of 2018 (6th of February). The Directive foresees the dangers arising from natural radiation sources to be fully integrated within the overall requirements. Thus activities that involve, for example, processing materials containing naturally occurring radionuclides are to be managed within the same regulatory framework as other practices.

Although the concerns and questions related to NORM have been present in the majority of the EU MS for an extended period of time, there has not been detailed legislation at EU level, prior to the 2013 EU BSS that deals with these issues. The topicality of the creation and management of NORM among scientists and regulators has clearly been on the rise over recent years, which can be seen through an increased number of initiatives, such as networks and programmes (EU ALARA networks, among others, is the European Alara network for NORM; COST Action NORM4Building, EUNORM network; CONCERT European joint programme for the integration of radiation protection research), organized conferences and workshops (NORM, ENA conference series; ENVIRA; ICRER; INCO-PoPb, TREICEP) and EU-wide projects (REDMUD; TERRITORIES; ENGAGE) and the IAEA initiative (ENVIRONET NORM project), which all keep the problem in focus.

In order to determine existing NORM-related industrial activities, a nationwide study was carried out in Estonia between 2015 and 2017. The selection of industrial activities was carried out according to the list of industries potentially generating NORM as given in ANNEX VI of the
for NORM management (IAEA, 2003, 2013). The results obtained can also be supported by the IAEA Safety Standards and Requirements for NORM management. It is essential to establish suitable and economic data to be compiled in order to radiologically characterize the technical data of the industrial process as well as the amounts of the available information from previous research and publications (Jantsikene, Kiisk, Suursoo, Koch, & Lumiste, 2014; Kiisk, Suursoo, Isakar, & Koch, 2011; E. Realo, Realo, & Jõgi, 1996; Vaasma, Bityukova, Kiisk, Özden, & Tkaczyk, 2016; Vaasma, Kiisk, Meriste, & Tkaczyk, 2014a, 2014b). This led to the investigation of the following industries:

- Oil shale industry – combustion of oil shale for electricity production; maintenance of large combustion boilers and production of shale oil.
- Cement industry and maintenance of clinker ovens.
- Rare metal processing (production of niobium and tantalum).
- Groundwater treatment plants.
- Central heating stations using solid and gaseous fuels.
- Underground oil shale mines.

The nationwide study enabled the necessary radiological and technical data to be compiled in order to radiologically characterize the potential NORM-related industrial activities within the country. This included estimating the radionuclide activity concentrations in raw materials, both by-products and end-products, evaluating NORM amounts and carrying out dose measurements. A national NORM inventory can also be the basis for establishing a national NORM strategy for NORM management. It is essential to establish suitable and economically sound approaches to managing NORM with long-term goals. This is also supported by the IAEA Safety Standards and Requirements as well as by IAEA technical documents that provide recommendations for NORM management (IAEA, 2003, 2013). The results obtained can also be added to the large scale NORM4Building database (Schroeyers et al., 2018), which provides supporting information to legislators and industry.

2. Methods and materials

2.1. Approach to create a national NORM inventory

A multistep approach was taken in order to establish a comprehensive national NORM inventory. The steps are listed below.

2.1.1. Determining the industries where NORM can be processed or produced

The selection of industrial activities that can be connected to NORM was initially based on Annex VI of the 2013/59/EURATOM Directive. NORM-containing raw materials are used in the industrial activities listed within this directive and thus can give rise to enhanced doses due to NORM discharge, NORM accumulation in certain processes/products and the management/disposal of NORM. It was also foreseen that the MS should determine additional NORM related activities within their countries, which can then be added to this list. The available data (publications, projects, national monitoring data, expert assessments and relevant reports) was compiled and analyzed to estimate the NORM creation potential within different industries.

2.1.2. Gathering input on the technological set-up of the industries and compiling a sampling plan

Additional samplings and measurements were planned in cases where the available literature data did not provide a sufficient overview to draw qualitative or quantitative conclusions on the creation of NORM within an industrial activity. Firstly, contact was made with selected companies to introduce the study and obtain input on the technical data of the industrial process as well as on the amounts of the raw material used and the by-products and end-products created. Additional information was obtained from companies’ environmental licenses. In close coordination with the industry’s contact points, a sampling and measurement plan was compiled according to which visits to companies’ premises were conducted.

2.1.3. On-site visits, measurements and sample collection

On-site visits (organized in all companies included, except drinking water treatment plants and Eesti Energia (EE) power plants, within this activity) included a tour of the company’s premises to get a better overview of the industrial process. On-site dosimetric measurements (ALNOR and DGM dosimeters) and radon measurements (Radon Monitor RM3-B (Alnor) and AlphaGUARD PQ 2000PRO (Saphymo)) were carried out in locations occupied longest by the workers and in some sample collection points. Long-term radon measurements (3 months) were carried out in an underground oil shale mine with plastic detectors CR-39. These detectors were then analyzed in the Radiation Department of the Environmental Board using accredited methodology.

The solid samples collected were mostly in the range of a few kilograms. Liquid samples had a volume between 0.5 and 1 L. The sample collection points were chosen with the aim of describing the whole production unit – input fuel, various by-products and end-products, and waste. This provides an understanding of radionuclide concentration processes within the system.

2.1.4. Sample preparation and measurement

Solid samples did not require additional grinding or sieving prior to measurement. These samples were dried at 105 °C for 24 h. The dried samples were compacted by a hydraulic press and placed into containers with a volume of 55 cm³. Containers were hermetically sealed to avoid radon (222Rn) exhalation. The samples were measured approximately 1 month after sealing in order to achieve secular equilibrium between 226Ra and 222Rn within the sample. Samples were measured with a Canberra BeGe BE 3830-P high purity germanium detector. The activity concentrations of the following radionuclides from the 238U and 232Th series were determined: 238U (234Th 63.3 keV), 226Ra (214Bi 1764.9 keV and 214Pb 242.0 keV, 295.2 keV and 351.9 keV), 226Ra (46.5 keV), 232Th (via 228Ra; 212Pb; 238.6 keV), 228Ra (via 228Ac; 911.2 keV, 964.8 keV and 969.0 keV) and 40K (1460.8 keV). RGU-1 source (IAEA, 1987) where secular equilibrium is between the 238U series radionuclides, was used to calibrate the measurement system. Due to the partial absorption of gamma photons (especially under < 100 keV) within the sample matrix, a self-absorption correction was applied according to Cutshall, Larsen, & Olsen, 1983. A more precise description of the gamma spectrometric measurement approach can be found in Vaasma, Loosaar, Gyakwaa et al., 2017.

Liquid samples were placed into the same dimension containers, sealed and measured after approximately one-month waiting period. The measurement values provided in the tables represent the following: i) U-238 decay chain (without Pb-210) – values for 226Ra and 234Th; ii) Th-232 decay chain – values for 228Ra and 228Th. If not provided separately, then the values for the given radionuclides coincided within the limits of measurement uncertainty.

3. Results and discussion

Pre-existing information and measurement results were compiled to provide an exhaustive overview concerning the radiological characteristics of the selected industrial activities. The radionuclide activity concentration values obtained were compared to the exemption and clearance levels in the Directive and in existing national regulations (both 1 kBq kg⁻¹ for the 238U and 232Th series in equilibrium and 10 kBq kg⁻¹ for 40K). This was used as the basis to decide if the industry is producing NORM material.

3.1. Oil shale industry

3.1.1. Oil shale combustion for electricity production

Two of the world’s largest oil shale-fired power plants (PPs) are operated in the North East of Estonia by a company called Eesti Energia (EE). These PPs produce over 70% of the electricity used in Estonia. The amount of oil shale used by these PPs is currently between 12 and 15
million tonnes per year. Due to the high ash content of this fuel (∼50%), approximately half of it is eventually left behind as residual material. The majority of this oil can be taken to nearby landfills via hydro transport. Only a small amount (1–2%) finds secondary use, e.g. in building materials, as mass stabilizer in road construction and in agriculture.

The activity concentrations and behaviour mechanisms of radionuclides within the combustion boilers of these PP's have been studied extensively during the past 6 years and information is available in numerous publications. This information also includes radionuclide emissions, atmospheric dispersion as well as deposition fluxes (Vaasma, Karu, Kiisk, Pens, et al., 2017; Vaasma, Loosaar, Gyukwa, et al., 2017; Vaasma et al., 2014a, 2014b, 2016; Vaasma, Kaasik, Loosaar, Kiisk, & Tkacyz, 2017; Vaasma, Loosaar, Kiisk, et al., 2017). Oil shale can be compared to coal, where numerous studies have indicated potentially high concentration values of naturally occurring radionuclides in coal combustion residues (Charro & Peña, 2013; Flues, Camargo, Figueiredo Filho, Silva, & Mazzilli, 2007; Karangelos, Petropoulos, Anagnostakis, Hinis, & Simopoulos, 2004; Ozden et al., 2017; Suhana & Rashid, 2016; Vreček & Benedik, 2003; Zeevaert, Sweeck, & Vanmarcke, 2006).

The average radionuclide concentration in raw oil shale has mostly remained between 20 and 30 Bq kg⁻¹ for the ²³⁸U and ²³²Th series radionuclides. Radionuclide activity concentrations are elevated in various fly ash fractions. The magnitude of enrichment strongly depends on the type of combustion system (pulverized fuel or circulating fluidized bed boiler) and the purification system installed (mostly either electrostatic filters or bag filters with a novel integrated desulfurization system). The highest radionuclide activity concentrations in the combustion system have been found in filter ashes, where ²¹⁰Pb (volatile radionuclide) has reached up to 140 Bq kg⁻¹. The values in ash for uranium, radium, and thorium are below 100 Bq kg⁻¹. The highest values for ⁴⁰K were below 2000 Bq kg⁻¹ (Table 1). These values are below the clearance and exemption levels currently stated in national legislation and the values given in the Directive. Questions remain concerning the radiological burden that the PP's have placed on the surrounding environment due to their long operational period of approximately 50 years. The fly ash emission rates have fluctuated significantly throughout these years, with the most impacted areas being within 2 km of the PP's (Vaasma, Kaasik, Loosaar, Kiisk, et al., 2017). The same study also concluded that considering that the most enriched fraction of fly ash is escaping the purification system, calculation of the cumulative doses which the most exposed members of the public are exposed to may still be useful in order to obtain a more quantitative idea of public exposure, even if it is not of regulatory concern as clearance and exemption values are not exceeded.

Additional on-site sampling and measurements were done in a smaller scale oil shale-fired plant (belonging to a company Kiviõli Keemiatööstus – KKT) that produces electricity and heat. This PP uses below 25 000 tonnes of oil shale (as its main fuel) per year and has a power output of 48.6 MW. Samples of oil shale, ash from multicyclone and ash from the boiler furnace were collected. The combustion boiler was temporarily shut down for maintenance, which allowed sampling and radon measurements to be conducted.

²¹⁰Pb, measured in ash samples obtained from the furnace's interior walls, had the highest concentration of ²³⁸U and ²³²Th series radionuclides. These values reached up to 500 Bq kg⁻¹ (Table 1). Due to its high combustion temperatures, over 1000 °C, ²¹⁰Pb is often volatilized and can be partially deposited on the walls of a combustion system. The activity concentrations of other elements did not demonstrate such an enrichment trend. The values for ⁴⁰K are also below 2000 Bq kg⁻¹, which is similar to the large oil shale-fired PPs.

On-site radon measurements in the operator's room and in the combustion furnace during its maintenance at KKT demonstrated average radon concentrations of 72 ± 12 and 59 ± 10 Bq m⁻³, respectively. This was well below 300 Bq m⁻³, which is considered the reference level for indoor radon in workplaces. Dosimetric measurements in the company's premises near the working units and in the furnace were between 0.07 and 0.15 μSv h⁻¹. The average natural background levels remain around 0.1 μSv h⁻¹, thus no considerably elevated doses were recorded.

The results demonstrate that oil shale combustion for electricity and heat production does not create residual material that could be considered as NORM. However, the technological set-up of combustion systems may create a situation (as described later with cement production) where elevated enrichment of especially volatile radionuclides can occur, potentially exceeding the general exemption levels for NORM material. Therefore, it is important to consider such effects when modifications or renovations are performed on combustion units.

### 3.1.2. Shale oil production

On-site sampling was conducted from two large oil processing industries in the North-East of Estonia, KKT and Viru Keemia Grupp – VKG. KKT employs different technologies to produce shale oil – gas generator stations and solid heat carriers produce crude shale oil that will later be processed into commercial oil. According to the environmental licence issued to the company, the annual oil production capability of crude oil can be up to 157 000 tonnes. The largest solid residual materials created through these processes are semi coke and ash from the solid heat carrier. These are mostly deposited in nearby hazardous waste landfills, with limitations of approx. 470 000 tonnes of

<table>
<thead>
<tr>
<th>Company</th>
<th>Radionuclide</th>
<th>Total no. of samples</th>
<th>Raw material</th>
<th>Products</th>
<th>Semi coke and ash from shale oil production</th>
<th>Ashes from the PP</th>
</tr>
</thead>
<tbody>
<tr>
<td>KKT</td>
<td>U-238 decay chain (without Pb-210)</td>
<td>20-37</td>
<td>&lt; 1-15</td>
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<td>Pb-210</td>
<td>16</td>
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<td>7-21</td>
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<tr>
<td>VKG</td>
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* Values obtained from previous studies and in the aforementioned publications.

* Various filter ashes.

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semi coke and 174 000 tonnes of ash per year.

During the visit, oil shale, ash, semi coke and various crude oil samples (16 in total) were collected and measured (Table 1).

The other company (VKG), where shale oil and other fine chemistry is produced, also uses gas generator stations and solid heat carrier stations. In 2014, the company produced around 420 000 tonnes of different oil products and generated approx. 500 000 tonnes of semi coke and 600 000 tonnes of bottom/furnace ash. Similarly to KKT, various solid and liquid (oil) samples (15 in total) were collected and measured (Table 1).

Similarly to KKT, the on-site radon measurements taken in the VKG operator’s room were modest – 57 ± 6 Bq m⁻³. Dosimetric measurements at various points of the company’s premises and near the operating units were below 0.12 μSv h⁻¹.

In general, the results from the KKT and VKG sample measurements (Table 1) indicate that the shale oil production processes do not generate NORM. Most of the measured radionuclides are found in various by-products and the radionuclide activity concentrations in the end-products are mostly very low (indicated as products in Table 1), often remaining below the detection limit. This is an important factor as some of the crude oil can be used as combustion fuel (in power stations, marine machinery, etc.). Due to the low ash content of such combustion fuels, significant radionuclide enrichment can occur in the residues during combustion processes.

At this time, mass balance calculations of radionuclide concentrations were not conducted, as it was not possible to obtain information about the ratios of ash created at various steps or the proportions of oil products produced.

3.1.3. Radon measurements in an oil shale mine

Long-term (3-month) radon measurements were conducted in an oil shale mine which belongs to VKG. The mine is located 30 m below ground surface and the annual amount of oil shale that is permitted to be extracted is 3.5 million tonnes. 7 sets of paired CR-39 plastic detectors (Fig. 1) were placed at various locations of the mine – in repair shops, workers’ break rooms and active mining areas. The detectors were set up in the mine in two ways, one with a cover and one without a cover. The detector without a cover was placed upside down to avoid the accumulation of dust near the air inlet of the detector. The covered detector (with gauze fabric) was positioned right side up. The aim of the fabric was to determine any variations in radon concentrations caused by the loose dust in the mine.

The results (Fig. 2) demonstrate that the ²²²Rn values were very low, fluctuating between 15 and 35 Bq m⁻³. These low concentrations can be explained by the effective ventilation system employed in the mine. The average air exchange time in the mine is between 30 and 60 min, depending on the location.

In most cases, the radon measurement values of the covered and uncovered detectors stayed within the limits of uncertainty. There was no constant trend of higher or lower values that could be connected to the covering or uncovering of the detectors. Also, the radon concentrations did not have any location based dependences. The ventilation seems to be effectively keeping radon concentration low, regardless of the location.

3.2. Cement production

The company AS Kunda Nordic Cement has the capacity to produce up to 1.4 million tonnes of cement per year. The company makes use of three large rotating ovens, so called rotary kilns, to create cement clinker. A mixture of limestone, clay and water is heated in the rotary kilns by using oil shale as the main fuel. Temperatures can reach up to 1450 °C during clinker formation. A large portion of the oil shale ash mixes with the material to form cement clinker, only around 6% of the fine ash (clinker dust) is removed from the system via electrostatic filters and transported to a silo. Approximately 20 000 tonnes of fine clinker dust is removed from the system yearly and the majority of it is taken to a nearby industrial landfill. This material has also been partially used in road construction as a stabilizing material.

The samples collected included raw material – limestone and clay; fuel – oil shale, coal and refuse-derived fuel (the latter two are used in small quantities); clinker dust and samples from various cement products. It should be noted that a specific type of oil shale ash from EE’s
power plants is used in the production of some cement products. The mass ratio of oil shale ash added to these cement products can vary between 16% and 23%.

The measurement results (Table 2) indicate the radionuclide activity concentrations in oil shale are similar to the ones in EE, VKG and KKT, although they are mined from a different location in Estonia.

Limestone and clay demonstrate up to 2-fold higher radionuclide concentration values compared to oil shale. In the cement products, the values are relatively modest, below 80 Bq kg$^{-1}$ for $^{238}$U decay chain and below 40 Bq kg$^{-1}$ for the $^{232}$Th decay chain. The activity concentration for $^{210}$Pb in the sample obtained from the furnace (rotary kiln) was below the detection limit. There is significant depletion of $^{210}$Pb in the rotary kiln compared to the values in various ashes. This could be due to high combustion temperatures during which Pb can be partially vaporized and transported towards the back end of the combustion system via flue gases. This is also supported by the relatively high $^{210}$Pb values in the clinker dust, over 2300 Bq kg$^{-1}$. However, the concentrations of other radionuclides in the $^{238}$U and $^{232}$Th series are in a similar range as in the raw material. High enrichment is also observable for $^{40}$K – activity concentrations in the clinker dust are around 10 times higher than those in the raw material.

These elevated concentrations can raise radiological concerns regarding the workers who are in contact with this material (collection, transportation and landfill) as well as questions regarding the safety of the material in other areas, e.g. in construction. The value for $^{210}$Pb in clinker dust exceeds the clearance and exemption levels brought out in the Directive. The Directive, however, states that these clearance and exemption values are directly applicable in the case of a preserved secular equilibrium in the decay series. In order to determine if the same values (1 kBq kg$^{-1}$) are appropriate in cases where only some radionuclides have elevated concentrations, dose risk assessments should be carried out. Current national legislation does not provide exemption values for separate radionuclides that would be properly applicable to NORM. The European Commission however has issued practical guidance in its publication (European Commission, 2001) for establishing higher exemption levels for different radionuclides, including $^{210}$Pb and $^{210}$Po (for which the exemption level is 5000 Bq kg$^{-1}$). This study demonstrates that the radionuclide enhancement processes, even when using the same type of fuel, can vary significantly, depending on the technological set-up of the industrial process.

On-site $^{222}$Rn measurements in the operator’s room and inside the rotary kiln, which was temporarily shut down for maintenance, were 24 ± 13 Bq m$^{-3}$ and 73 ± 15 Bq m$^{-3}$, respectively. Dose rate measurements in the premises were between 0.07 and 0.12 μSv h$^{-1}$. Significantly higher doses were measured within the rotary kiln (furnace end), where values close to the interior wall (approx. 5–10 cm) reached 0.8 μSv h$^{-1}$. This exceeds the natural background value by up to 10 times. This may be explained by the deposition onto the inner walls or due to refractories used in the kiln. Conservative estimations on the potential doses received by a worker who would be involved during the whole maintenance work were calculated to be up to 0.14 mSv. This only includes doses from external exposure to gamma radiation.

Another study (Leier & Kiisk, 2015) was initiated in 2014 in order to understand how common the generation of NORM in drinking water treatment plants (WTP) is. 18 WTPs agreed to participate, some of which were designed for the removal of Fe, Mn, Ra, and dissolved gases. Most of the WTPs were designed only for Fe, Mn and dissolved gases removal. These 18 WTPs covered 47.7% of the national Cm-V groundwater productivity and 48.8% of consumers who use a Cm-V aquifer, whether entirely or partially.

Exemption and clearance levels, as given in the Directive, for $^{226}$Ra were exceeded in 15 WTPs out of 18 and $^{228}$Ra levels were exceeded in 16 WTPs (Fig. 3). Filter material that exceeds the exemption and clearance levels has a total volume of 300 tonnes. It is noteworthy that two of the big WTPs contribute more than 50% of the total identified NORM volume.

These results clearly show that even when a WTP is not designed for radium removal, NORM generation may still take place due to co-precipitation and the adsorption of radium with Fe and Mn flocks. This was previously described in a published research (Kivimäe, & Eensalu, 2012) on radium removal technology for Cm-V groundwater. It should be noted that the co-precipitation of radium with Mn(OH)$_4$ is more effective than with Fe(OH)$_3$. New technology to

### Table 2

Radionuclide activity concentrations (Bq kg$^{-1}$) in samples from the cement industry. The total number of samples was 15. Measurement uncertainty ($k = 2$) remains between 5% and 20%, if not stated differently.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Limestone/clay</th>
<th>Fuel</th>
<th>Residues from furnace</th>
<th>Products</th>
<th>Ash for cement production</th>
<th>Clinker dust</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}$U decay chain</td>
<td>55 ± 61</td>
<td>25–27</td>
<td>52–64</td>
<td>60–76</td>
<td>58–64</td>
<td>50–58</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>56 ± 10</td>
<td>21 ± 7</td>
<td>&lt; 4</td>
<td>9–27</td>
<td>82–103</td>
<td>1830–2320</td>
</tr>
<tr>
<td>K-40</td>
<td>331 ± 14</td>
<td>310 ± 17</td>
<td>113 ± 10</td>
<td>350–600</td>
<td>1350–1525</td>
<td>2200–3200</td>
</tr>
</tbody>
</table>
remove radium from drinking water, but at the same time avoid radium accumulation in the filter material, is being experimented in an ongoing EU project under the LIFE programme (LIFE Alchemia – https://www.lifealchemia.eu/en/).

The radium accumulation rate depends on the technology used in the WTP, the filter material employed and the quality of the raw water. At present, there is no full overview of the activity concentrations of filter materials in all WTPs using a Cm-V aquifer. Both the WTPs’ operators and the regulatory body have the responsibility of gathering such information.

3.4. Rare metal production

NPM Silmet AS has the capacity to produce 360 tonnes of niobium and 84 tonnes of tantalum per year. The company has been issued a radiation practice licence to process raw material – up to 550 tonnes of columbite and 175 tonnes of tantalite per year. These raw materials contain elevated concentrations of $^{238}$U and $^{232}$Th series radionuclides and the processing of this material creates approx. 73 tonnes of radioactive waste annually. Both the WTPs’ operators and the regulatory body have the responsibility of gathering such information.

3.5. Central heating stations

In 2017, renewable energy sources provided close to 17% of the total electric energy demand in Estonia. 54% of renewable-based electric energy was produced from biomass and waste. Woodchips and peat are often the main form of solid biomass used to produce heat and electricity. Some available studies (Erlandsson, Hedvall, & Mattsson, 1995; Rantavaara & Moring, 2001; Vaičiukyniene et al., 2018) have indicated that the ash from solid biomass combustion can contain elevated concentrations of naturally occurring and anthropogenic radionuclides. This provided the basis to include local biomass based stations in order to have a more precise investigation. A local power plant (Tartu PP) that operates on peat, woodchips and low value wood and a central heating station (CHS) (Anne CHS) that is also using the same solid biomass as well as biogas during higher demand periods were included in the study. In 2015, the total amounts of woodchip, peat and biogas burned within these plants were approx. 190 000 tonnes, 60 000 tonnes and 400 000 m$^3$, respectively.

The higher concentration rate of radionuclides in peat and wood ash is due to the low ash rate of these fuels, which are around 6% for peat and 3% for wood. To determine the radionuclide activity concentration in various points of the boiler systems, the following samples were obtained: peat, bottom ash, fly ash and ash samples from the walls of the peat combustion furnace (Table 3).

A previous study (Realo & Realo, 1999) has indicated that the radionuclide activity concentrations in woodchips is low, often remaining below detection limits. The current study shows that the radionuclide values in peat are below average concentration expected in soils world-wide (UNSCEAR, 2010) with the exception of $^{210}$Pb. The $^{210}$Pb values depend directly on the depth which the peat was collected. Considerably higher values can be expected in the top layers of peat (Vaasma, Karu, Kiisk, Pensa, et al., 2017). Due to its low ash content, the $^{210}$Pb enrichment in fly ash can be up to 6 times higher than in the original fuel. Significantly higher enrichment values can be seen for $^{40}$K, which stays below the detection limit in peat, but reaches over 300 Bq kg$^{-1}$ in the ashes. The $^{137}$Cs values, however, were low in the raw material as well as in the ash products, between 2 and 36 Bq kg$^{-1}$. On site radon measurement in the operators’ room was 47 ± 10 Bq m$^{-3}$ and dose rate measurements were from 0.05 μSv h$^{-1}$ to 0.14 μSv h$^{-1}$ (in the stopped combustion unit). The values are in the same range as the background dose rates measured near the company.

The values indicated in Table 3 are below the clearance and exemption levels indicated in the Directive. However, if the source of the fuel should change (currently mostly locally derived fuel is used), additional measurements should be conducted of the source material as the radionuclide activity concentrations will vary in peat depending on its origin and location.
3.6. NORM management in Estonia

Currently there is no existing practice for NORM treatment in Estonia, although awareness on the NORM-related issues (e.g. the creation of NORM and its amounts) has significantly increased. To date NORM has been handled as hazardous or non-hazardous waste, which has in some instances led to uncontrolled management of this material. Further options to manage this material are connected to determining reuse possibilities or finding optimal and safe disposal options. The reuse options for such materials are often limited due to their physical and mechanical properties or due to chemically and radiologically hazardous components. To consider reuse as an option for NORM material, radiation safety assessments to determine exposure to radionuclides through various exposure pathways would be required. Additionally, economic and environmental assessments are needed to determine if there is a demand and market for such reusable materials and what the environmental constraints that must be taken into consideration prior to its use are.

Landfills (industrial and municipal) as NORM treatment options have been considered the most in Estonia. This is due to the relatively small amounts of NORM created each year as well as the radionuclide activity concentrations in them (mostly below 100 kBq kg\(^{-1}\)). Clinker dust from the cement industry is currently directed to a near-by industrial landfill, however radiation safety assessments have not been conducted to determine potential doses to workers and the general public. Direction towards municipal landfills for filter material from WTP has been taken by the authorities as well. A similar approach has also been adopted in other countries (e.g. Belgium) with extensive experience in NORM treatment practices (Pepin, Biermans, Dehandschutter, & Sonck, 2016; Pepin, Dehandschutter, Poffijn, & Sonck, 2013). On the other hand, no uniform national strategy on NORM waste management exists in Estonia that would provide NORM producers and treatment facilities with the necessary regulation and guidelines to operate with. Therefore, the next relevant steps would include actions to address such concerns, starting with the development of a national NORM strategy, which would provide clear understanding and guidelines to address NORM creation, treatment and management related issues.

### 4. Conclusions

The nationwide study provided the necessary information to determine and radiologically characterize NORM-related industries in Estonia. The sampling and on-site measurement results combined with previously available data indicated 3 industries where by-products and waste with elevated radionuclide concentrations are created. The management of this material is complicated, partly, due to its different physical, chemical and radiological properties as well as the amounts of NORM materials created yearly. The material from WTP contains only small amounts of NORM created each year as well as the radionuclide have been considered the most in Estonia. This is due to the relatively standards.

### Acknowledgements

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### References


European Commission LIFE programme project LIFE16 ENV/ES/000437 Alchemia.

### Table 3

Radionuclide activity concentrations (Bq kg\(^{-1}\)) in the collected samples. The total number of samples was 7. Measurement uncertainty (k = 2) remains between 5 and 20%, if not stated differently.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Raw material</th>
<th>Bottom ash</th>
<th>Fly ash</th>
<th>Ash from furnace walls</th>
<th>Ash mix from silo (87% woodchips, 13% peat)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb-210</td>
<td>31 ± 13</td>
<td>35 ± 7</td>
<td>171 ± 9</td>
<td>116 ± 17</td>
<td>558 ± 20</td>
</tr>
<tr>
<td>Th-232 decay chain</td>
<td>&lt; 1–1</td>
<td>9 ± 1</td>
<td>12–14</td>
<td>16 ± 2</td>
<td>34–37</td>
</tr>
<tr>
<td>K-40</td>
<td>&lt; 8</td>
<td>314 ± 14</td>
<td>252 ± 10</td>
<td>327 ± 13</td>
<td>1461 ± 40</td>
</tr>
<tr>
<td>Cs-137</td>
<td>2 ± 0.7</td>
<td>3 ± 0.4</td>
<td>17 ± 1</td>
<td>36 ± 1</td>
<td>36 ± 1</td>
</tr>
</tbody>
</table>

Woodchips U-238 decay chain (without Pb-210) – 18–19 64–96 – –

Pb-210 – 72 ± 17 630 ± 50 – –

Th-232 decay chain – 14–15 32–34 – –

K-40 – 953 ± 30 1291 ± 60 – –


