Radioactivity in residues and effluents from Estonian waterworks treatment plants

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THE PROBLEM:

- Some groundwaters in Northern Estonia exhibit rather high natural radioactivity, mainly due to $^{226}$Ra and $^{228}$Ra.
- EC Directive 98/83/EC: Total Indicative Dose 0.1 mSv/year (parametric value)
- Enforced in Estonia national regulation in 2001 (dose limit)
Estimation of concentrations of radionuclides in Estonian groundwaters and related health risks – Final Report

THE TASK:

REVIEWING THE EXISTING KNOWLEDGE
Radiological (and chemical) database
Analytical methods
Waterworks and water distribution structure
Water treatment plants

SUGGESTION ON
Future monitoring campaigns
Dosimetric evaluations
Fit for purpose analytical methods/strategy
Countermeasures
Radioactivity removal and sludge managing
High radium concentrations are found mostly in the deepest aquifer (Cambrian-Vendian): it lays on a crystalline rock basement. It is hardly recharged and the water is very old.

The Cambrian-Vendian aquifer is shallower (about 100 m) near the Baltic sea coastline. In this area it is used to feed waterworks.

The coastal area is the most densely populated.
Total Indicative Dose (TID)

**TID adults**

- Cm-V: 0.1 mSv/year

**TID infants**

- Cm-V: 0.1 mSv/year

**Others**
Average relative contributions of the two Ra isotopes to dose.

M Forte et al. *Radium isotopes in Estonian groundwater: measurements, analytical correlations, population dose and a proposal for a monitoring strategy*  
Journal of Radiological Protection, 30(4), 761-780
Dimensions of Water Supply Zones (Cm-V)

Total: 140 WSZ; 250,000 people

Served population:
- 12 WZS
  - 152,000 people

Wastes and sludges:
- Dosimetric evaluation
- Environmental impact

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**Graph:**
- Frequency of served population
- Categories: <50, 50-100, 100-250, 250-500, 500-1000, 1000-2500, 2500-5000, 5000-10,000, >10,000

- Frequency distribution:
  - <50: 9%
  - 50-100: 23%
  - 100-250: 44%
  - 250-500: 59%
  - 500-1000: 71%
  - 1000-2500: 84%
  - 2500-5000: 91%
  - 5000-10,000: 96%
  - >10,000: 100%

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**Legend:**
- 12 WZS
  - 152,000 people

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**Title:**
- Wastes and sludges:
  - Dosimetric evaluation
  - Environmental impact
Effluents and residues from existing treatment plants

- 4 Estonian aqueducts with waters rich in $^{226}\text{Ra}$ and $^{228}\text{Ra}$ and treatment systems surveyed in the year 2009.

- Data collected for a preliminary evaluation of the potential impact on the environment of the effluents and residues formed during the treatment processes.

- Radium activity concentrations in water and residues were assessed by several laboratories, among them the Estonian Radiation Protection Centre, the Tartu University and the STUK (Finland).
Tallinn waterworks

- **85 wells** afferent to 56 pumping stations, 19 supplied with treatment systems.

- **water** filtered through sand and gravel to remove Fe, Mn and \( \text{NH}_4 \); an aeration stage precedes the filtering.

- **filters** periodically cleaned by backwash water then channelled to sewage.

- **all backwash** conveyed to a single sewer (that of Tallinn city) waters (about 400,000 inhabitants).

- **sludge** used as filling material in landscape construction projects

- **purified water** released into the sea.
## Information on effluents from waterworks treatment plants of Tallinn

<table>
<thead>
<tr>
<th>Treatment Station</th>
<th>Treatment type</th>
<th>Backwash water</th>
<th>( ^{226}\text{Ra} ) Fate</th>
<th>( ^{228}\text{Ra} ) Fate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>(m(^3)/y)</td>
<td>(Bq/m(^3))</td>
<td>(Bq/y)</td>
</tr>
<tr>
<td>Jugapuu</td>
<td></td>
<td>1708</td>
<td>3100</td>
<td>8700</td>
</tr>
<tr>
<td>Raba</td>
<td>Stations with sand/gravel filters for Fe, Mn, NH(_4) removal (pre-aerated)</td>
<td>507</td>
<td>8730</td>
<td>14710</td>
</tr>
<tr>
<td>Toome - Õitse</td>
<td></td>
<td>1018</td>
<td>9350</td>
<td>13650</td>
</tr>
<tr>
<td>Tiskre</td>
<td></td>
<td>581</td>
<td>5320</td>
<td>6040</td>
</tr>
<tr>
<td>All other Stations</td>
<td></td>
<td>17428</td>
<td>n.a.°</td>
<td>n.a.°</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td></td>
<td>21242</td>
<td>6625*</td>
<td>10775*</td>
</tr>
</tbody>
</table>

* Average concentration value; ° not available
Effluents and residues from existing treatment plants (cont. II)

- **Keila waterworks**
  - groundwater from 4 wells mixed before being collected in treatment tanks
  - Fe and Mn removal techniques similar to those of Tallinn
  - waters to sewage - resulting sludge not used in agriculture
  - 2 samples of filter material (sand) analysed to assess $^{226}$Ra, $^{228}$Ra and $^{228}$Th

- **Rakvere waterworks**
  - 5 groundwater wells deliver water to a treatment plant of the same kind as the previous ones
  - backwash water cleared to sewer
  - sludge used as farmland fertilizer
  - purified waters released into a small river
• Viimsi waterworks

- water drawn from 35 independent wells

- pilot treatment device for radium purification operating at well # 412
  ✓ two parallel filtration columns + additional common cleaning stage.

- backwash water conveyed to the same sewer used by the Tallinn waterworks.
### Information on effluents from treatment plants of Keila, Rakvere and Viimsi

<table>
<thead>
<tr>
<th>Water works</th>
<th>Treatment Station</th>
<th>Treatment type</th>
<th>Backwash water (m³/y)</th>
<th>Fate</th>
<th>²²⁶Ra (Bq/m³)</th>
<th>²²⁸Ra (Bq/m³)</th>
<th>²²⁶Ra (Bq/y)</th>
<th>²²⁸Ra (Bq/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Keila</td>
<td>All 4 wells</td>
<td>Sand filters for Fe and Mn removal (preaerated)</td>
<td>1400</td>
<td>1380</td>
<td>1580</td>
<td></td>
<td>1.93E+06</td>
<td>2.21E+06</td>
</tr>
<tr>
<td>Rakvere</td>
<td>All 5 wells</td>
<td>Sand filters for Fe removal (aerated)</td>
<td>36500</td>
<td>1774</td>
<td>1796</td>
<td>Sewer</td>
<td>6.48E+07</td>
<td>6.56E+07</td>
</tr>
<tr>
<td>Viimsi</td>
<td>Well # 412</td>
<td>column I + column II (in parallel) for Ra purification</td>
<td>9.0</td>
<td>1805</td>
<td>2050</td>
<td></td>
<td>1.62E+04</td>
<td>1.85E+04</td>
</tr>
</tbody>
</table>

**In conclusion** $^{228}\text{Ra} \geq \text{ } ^{226}\text{Ra}$
REFERENCE DOCUMENTS FOR CLEARANCE LEVELS AND EFFLUENT DISCHARGE SCREENING LEVELS

- Waterworks treatment processes may be regarded as work activities involving NORM
- the EC BSS draft includes them in the positive list of NORM activities
- Estonian national legislation does not define reference values for NORM wastes or discharges; evaluate the adequacy of processes, reference should be made to available international or national technical guides

Solid residues
- In the analysed waterworks solid residues are mainly sand filters.
- The document Radiation Protection 122 (RP 122) - part II derives General Clearance Levels (GCLs) for natural radionuclides in residues and waste from work activities involving NORM
- it is useful to classify waterworks solid residues
# General Clearance Levels (GCLs) from RP 122 - part II

<table>
<thead>
<tr>
<th></th>
<th>$^{226}$Ra* (Bq/kg)</th>
<th>$^{228}$Ra* (Bq/kg)</th>
<th>$^{228}$Th* (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>All materials</td>
<td>500</td>
<td>1000</td>
<td>500</td>
</tr>
</tbody>
</table>

* In secular equilibrium with short half-life decay products

Residues with activity concentrations < GCLs can be reused, recycled, delivered for disposal with no constraint as for their radiological aspects.

GCLs determined to comply with the exemption-clearance dose criterion of 0.3 mSv/y for the individual effective dose.
### Information on residues from waterworks treatment plants

<table>
<thead>
<tr>
<th>Waterworks</th>
<th>Treatment station</th>
<th>Material</th>
<th>$^{226}$Ra (Bq/kg)</th>
<th>$^{228}$Ra (Bq/kg)</th>
<th>$^{228}$Th (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Tallinn</strong></td>
<td>Merivälja</td>
<td>Sand</td>
<td>8603</td>
<td>8681</td>
<td>5798</td>
</tr>
<tr>
<td><strong>Keila</strong></td>
<td>All 4 wells</td>
<td>Sand filter 1</td>
<td>5524</td>
<td>5754</td>
<td>3817</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sand filter 2</td>
<td>5202</td>
<td>5618</td>
<td>3139</td>
</tr>
<tr>
<td><strong>Rakvere</strong></td>
<td>All 5 wells</td>
<td>Sand filter</td>
<td>3788</td>
<td>3047</td>
<td>1768</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Backwash water sediment</td>
<td>20103</td>
<td>15034</td>
<td>7176</td>
</tr>
</tbody>
</table>

All values > or >> GCL
Comparison between solid residues from existing treatment plants and GCLs of RP 122

<table>
<thead>
<tr>
<th>Waterworks</th>
<th>Treatment station</th>
<th>Material type</th>
<th>Sum index*</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Tallinn</strong></td>
<td>Merivalja</td>
<td>Sand</td>
<td>37</td>
</tr>
<tr>
<td><strong>Keila</strong></td>
<td>All 4 wells</td>
<td>Sand filter (sample 1)</td>
<td>24</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sand filter (sample 2)</td>
<td>22</td>
</tr>
<tr>
<td><strong>Rakvere</strong></td>
<td>All 5 wells</td>
<td>Sand filter</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Backwash water sediment</td>
<td>70</td>
</tr>
</tbody>
</table>

* The sum index is the sum of ratios of single nuclide activity to the respective GCLs; for compliance it should be less than 1.

No material complies with RP 122 levels
Comparison between solid residues from existing treatment plants and GCLs of RP 122 (cont.)

• GCLs selected from the most conservative conditions (material type, reference scenario and population group), i.e. people living in a house whose building materials contain the radioactive residues

• unrealistic scenario for solid residues from drinking water treatment plants

BUT

• also in a different, more realistic scenario (e.g. exposure of workers that use contaminated material for road construction or people living in houses close to a disposal site of contaminated residues), compliance not achieved

• no solid material complies with the reference levels, not even when compared with the clearance levels of the EC BSS draft (1000 Bq/kg individually applied to $^{226}\text{Ra}$, $^{228}\text{Ra}$ and $^{228}\text{Th}$, with or without short half-life progeny in secular equilibrium).
The assessment of the radiological impact of effluent discharges based on three documents giving discharge screening levels from very conservative scenarios.

1) the IAEA Safety Rep. “Generic models for use in assessing the impact of discharges …”
   • reference levels for liquid discharge into small rivers and sewers
   • critical scenario: workers exposure in the sewing plant for discharge into sewer

2) the EC RP 135 “Effluent and dose control from European Union NORM industries:…”
   • screening levels concerning release into rivers of various sizes and into coastal sea.

3) the document NRPB 13 n.2 “Generalised Derived Constraints for….Po, Pb, Ra and U”
   • reference levels for liquid discharge into small rivers and sewers
   • critical scenario: use of sludge on farmland for discharge into sewer
RP 135 and NRPB 13: screening levels calculated with a dose criterion of 0.3 mSv/y (individual effective dose)

EC BSS draft also considers 0.3 mSv/y the dose for public exposure to NORM work activities as a general clearance criterion.

IAEA: individual annual dose for unit discharge of main radionuclides; screening levels calculated depending on dose criterion

IAEA and EC documents suggest reference values at the international level

NRPB document only sets national standards.
CHOICE OF SCREENING LEVELS FOR EFFLUENTS

Tallinn (+ Viimsi) and Keila plants:
• waters from filter cleaning to the sewer
• sludge formed in the process not used in agriculture.
  IAEA values for sewer and screening levels on the basis of the 0.3 mSv/y (levels scaled to account for the 400 000 inhabitants of Tallinn vs. 20 000 modelled; proportional dilution of the radioactivity)
• purified waters from the sewer released into the sea
  an alternative conservative assumption: sewer sludge contains no radium from backwash water, all radium is discharged into the sea - comparison with RP 135 screening levels for coastal sea

Rakvere:
• backwash waters flow to the sewage system
• resulting sludge used in farmland treatment:
  NRPB General Derived Constraint in sewer; agricultural scenario the most critical one in case of discharge into the sewer. Test only carried out for $^{226}$Ra, the $^{228}$Ra GDC not available
• purified waters from the sewer released into a small river:
  an alternative conservative assumption: all radium content of backwash water discharged into the river; comparison with RP 135 screening levels for discharge into a river
### Discharge screening levels for backwash water

<table>
<thead>
<tr>
<th>Compartment</th>
<th>Critical pathway</th>
<th>$^{226}\text{Ra}$ (Bq/y)</th>
<th>$^{228}\text{Ra}$ (Bq/y)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sewer</td>
<td>Sewer workers</td>
<td>$3.8 \times 10^9$</td>
<td>$2.7 \times 10^9$</td>
<td>IAEA 19 (Tallinn)*</td>
</tr>
<tr>
<td>Sewer</td>
<td>Sludge for agriculture</td>
<td>$1 \times 10^7$</td>
<td>n.a.</td>
<td>NRPB 13 n.2</td>
</tr>
<tr>
<td>Coastal sea</td>
<td>Ingestion (fish)</td>
<td>$2.2 \times 10^{13}$</td>
<td>$1.2 \times 10^{13}$</td>
<td>RP 135</td>
</tr>
<tr>
<td>Small river</td>
<td>Ingestion (fish)</td>
<td>$7.5 \times 10^{10}$</td>
<td>$4.2 \times 10^{10}$</td>
<td>RP 135</td>
</tr>
</tbody>
</table>

* Screening levels scaled to account for the number of inhabitants served by the Tallinn sewer
Discharge screening levels applied to backwash waters of existing treatment plants

<table>
<thead>
<tr>
<th>Waterworks</th>
<th>Compartment</th>
<th>Sum index*</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tallinn and Viimsi°</td>
<td>Sewer</td>
<td>1.22E-01</td>
<td>IAEA 19</td>
</tr>
<tr>
<td>Tallinn and Viimsi°</td>
<td>Coastal sea</td>
<td>2.55E-05</td>
<td>RP 135</td>
</tr>
<tr>
<td>Keila</td>
<td>Sewer</td>
<td>2.60E-02</td>
<td>IAEA 19</td>
</tr>
<tr>
<td>Rakvere</td>
<td>Sewer</td>
<td>6.48</td>
<td>NRPB 13 n. 2</td>
</tr>
<tr>
<td>Rakvere</td>
<td>River (small)</td>
<td>2.42E-03</td>
<td>RP 135</td>
</tr>
</tbody>
</table>

* The sum index is the sum of ratios of single nuclide activity to the respective GCLs; for compliance it should be less than 1.

°As backwash waters from Tallinn and Viimsi treatment plants flow into the same sewer (Tallinn city), their contributions were added for the comparison.
Conclusions 1

**Solid residues** (sand filters and backwash water sediments):

- $^{226}\text{Ra}$, $^{228}\text{Ra}$ and $^{228}\text{Th}$ activity concentrations higher than both the general clearance levels suggested by RP122 part II and the exemption and clearance levels set by the EU BSS draft

- Compliance could be demonstrated case by case with dose calculations in specific scenarios, taking into account the actual use and radiological impact of the residues.

**Liquid effluents**:

- All but those from the Rakvere treatment plant (sewer compartment) comply with IAEA and EC discharge screening levels

- Rakvere sewer compartment produces sewer sludge used on farmland and the relevant NRPB assumptions for calculating screening levels are highly conservative
Suggestions:

• need for a more robust assessment:
• more realistic scenarios
• all radionuclides in backwash water accounted for

• special care must be taken when managing solid residues
• more detailed surveys and systematic analyses of radiation protection aspects should be made as soon as the new EC BSS will be transposed in the Estonian legislation.
Thank you for your attention