Development of a methodology at European level for dose-assessment to the members of public following use of NORM sludge in agriculture.

C. Nuccetelli (Italian National Institute of Health - ISS)

Co-Authors: F. Trotti, R. Ugolini (ARPAV), R. Trevisi, F. Leonardi (INAIL), G. Venoso, C. di Carlo (ISS), L. Urso (BfS)

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Introduction

At European level the general obligation to recycle pushes for increased reuse of NORM residues. This includes the reuse in agriculture of sludge produced by groundwater filtration facilities and by sewage treatment plants receiving backwash waters from the filtration facilities.

Exposure scenarios relevant to agricultural workers have been identified to be modelled by RESRAD v. 7.2, a software which implements all the exposure pathways relevant to farmland and is widely accepted as a standard for conservative exposure assessments.

The goal is to develop a generic model and obtain tabulated dose coefficients per 1 Bq g\(^{-1}\) of natural radionuclides in sludges and determine Clearance Levels with dose criterion of 1 mSv per year, together with analogous parameters relevant to discharge of backwash water in sewers.
In present study RESRAD ONSITE is used and some approaches are different compared to RP122/2 in order to describe exposure scenarios:

- subchains are considered instead of secular equilibrium for U and Th-chains
  - Th-232 and Th-228 go together since chemically separated
- Cumulative doses over 35 years for workers or 70 for residents

In RP122/2 (and RP 135) these scenarios are not considered
Pathways considered in RESRAD

- In RP a dose-assessment is carried out whenever compliance with dose limits is to be verified.
- Radioecological models are used to carry out dose-assessments.
- Conservative approach: calculated dose overestimates real dose.
- Updated DCFs for external exposure from ICRP 144.
- Specific segments of decay chains in secular equilibrium (different from RP122/2).
Scenarios considered

1 Farmer working for 35 years
   a) starting to spread out sludges at time 0 - sludge application for 35 years.
   b) after annually application of sludge for 35 years - sludge application for 35 years.

Pathways:
External irradiation, inhalation of suspended soil, accidental ingestion of soil

2 Farmer working and living for 70 years
   a) starting to spread out sludges at time 0 - sludge application for 70 years.
   b) after annually application of sludge for 35 years - sludge application for 70 years.

Pathways:
Same of scenario 1, Ingestion of contaminated food not water dependent
In scenario 2, Rn exposure is not accounted for since the bulk amount of soil is not contaminated.
Case 1

Use of sludge directly from waterworks treatment plant
Radionuclide activity concentration in soil-sludge mixture

Specific parameters to be considered:

- Application rate of dry sludge matter per hectar per year ($S$)
- Tillage depth ($d$)
- Soil density ($r$)

$$A_{\text{soil}} = A_{\text{sludge}} \times S / (d \times r)$$

All the analyses were performed @ $A_{\text{sludge}} = 1 \text{ Bq/g}$

Values used for deterministic analysis:

- $S = 5 \text{ tons/hectare} (= 0.5 \text{ kg/m}^2) \text{ per year}$
- $d = 0.3 \text{ m}$
- $r = 1300 \text{ kg/m}^3$

$A_{\text{soil}} = 0.00128 \text{ Bq/g}$
Considered radionuclides for dose calculations by RESRAD-ONSITE

Taking into account that sludges directly from waterworks (or from sewages) are contaminated from NORMs not at secular equilibrium, only subchains chosen on basis of chemical behaviour and decay time were considered in calculations using RESRAD ONSITE:

- $^{238}\text{U}$, $^{234}\text{U}$, $^{235}\text{U}$ (4.6%) and their short decay products

- $^{230}\text{Th}$

- $^{226}\text{Ra}$: Decay chain to $^{214}\text{Bi}$ at equilibrium

- $^{232}\text{Th}$ & $^{228}\text{Th}$: Chemically separated thorium from $^{232}\text{Th}$ series

- $^{228}\text{Ra}$: Decay chain to $^{208}\text{TI}$ at equilibrium

- $^{210}\text{Pb}$: $^{210}\text{Pb}$, $^{210}\text{Bi}$

- $^{210}\text{Po}$
This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 900009.

Expected dose trends

Single spreading of sludges for 1 year

Pb-210+

y = 4.2E-05e^{-4.6E-02x}

R² = 1.0E+00

Annual dose (mSv/y)

All the analyses were performed @ $A_{\text{soil}} = 0.00128$ Bq/g

Spreading of sludge repeated every year for a number of years

Pb-210+

Year
### Results of RESRAD Modelling

#### $^{238}$U sub chains

<table>
<thead>
<tr>
<th>Segment of chains</th>
<th>Farmer works for 35 years</th>
<th>Farmer works for 35 years starting from contaminated soil</th>
<th>Farmer works and lives for 70 years</th>
<th>Farmer works and lives for 70 years starting from contaminated soil</th>
<th>Max dose per Bq g$^{-1}$ of dry sludge (mSv y$^{-1}$ per Bq g$^{-1}$)</th>
<th>CL (Bq g$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>years of spreading</td>
<td>mSv y$^{-1}$</td>
<td>years of spreading</td>
<td>mSv y$^{-1}$</td>
<td>years of spreading</td>
<td>mSv y$^{-1}$</td>
</tr>
<tr>
<td>U$_{nat}$</td>
<td>35</td>
<td>1.39E-03</td>
<td>70</td>
<td>1.95E-03</td>
<td>70</td>
<td>2.01E-03</td>
</tr>
<tr>
<td>Th-230</td>
<td>35</td>
<td>5.52E-04</td>
<td>70</td>
<td>1.38E-03</td>
<td>70</td>
<td>3.91E-03</td>
</tr>
<tr>
<td>Ra-226+</td>
<td>35</td>
<td>4.82E-02</td>
<td>70</td>
<td>7.59E-02</td>
<td>70</td>
<td>1.23E-01</td>
</tr>
<tr>
<td>Pb-210+</td>
<td>35</td>
<td>7.44E-04</td>
<td>70</td>
<td>8.93E-04</td>
<td>70</td>
<td>1.47E-02</td>
</tr>
<tr>
<td>Po-210</td>
<td>35</td>
<td>9.60E-06</td>
<td>70</td>
<td>9.60E-06</td>
<td>70</td>
<td>1.54E-04</td>
</tr>
</tbody>
</table>

Dose criterion to determine Clearance levels (CL) is **1 mSv per year**
## Results (cont.)

### $^{232}$Th sub chains

<table>
<thead>
<tr>
<th>Segment of chains</th>
<th>Farmer works for 35 years</th>
<th>Farmer works for 35 years starting from contaminated soil</th>
<th>Farmer works and lives for 70 years</th>
<th>Farmer works and lives for 70 years starting from contaminated soil</th>
<th>Max dose per Bq g$^{-1}$ of dry sludge (mSv y$^{-1}$ per Bq g$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>years of spreading</td>
<td>mSv y$^{-1}$</td>
<td>years of spreading</td>
<td>mSv y$^{-1}$</td>
<td>years of spreading</td>
</tr>
<tr>
<td>$^{232}$Th&amp;$^{228}$Th</td>
<td>35</td>
<td>4.18E-02</td>
<td>70</td>
<td>9.29E-02</td>
<td>70</td>
</tr>
<tr>
<td>$^{228}$Ra+</td>
<td>35</td>
<td>1.10E-02</td>
<td>70</td>
<td>1.17E-02</td>
<td>70</td>
</tr>
<tr>
<td>$^{228}$Th+</td>
<td>35</td>
<td>2.70E-03</td>
<td>70</td>
<td>2.70E-03</td>
<td>70</td>
</tr>
</tbody>
</table>

### $^{40}$K

| K-40               | 35                  | 5.43E-04     | 70                  | 5.43E-04     | 70                  | 4.07E-03     | 100                  | 6.24E-03     | 160               |
Discussion

- From the scenario of farmer who starts to work and live on soil contaminated CL for $^{226}$Ra+ and $^{232}$Th&$^{228}$Th are of the order of Bq g$^{-1}$

- The most important pathways is the external irradiation by $\gamma$ rays, followed by plant ingestion (water indipendent)
Case 2

Use of sludge from WWTP
Activity concentration in sludge from WWTP

• Population equivalent (PE): it is the most used term to describe the size of Waste Water Treatment Plant (WWTP)
• Annual sludge production: **20 kg** dry matter per person (IAEA, Safety Reports Series n. 19)
• Activity in sludge may reside on treatment plant site for weeks or months before being applied to land.

• Activity concentration in sludge (dry matter) produced in a WWTP for radionuclide \(i = \text{Unat}, \text{Ra-226}, \text{Ra-228}, \text{Th-228}, \text{Th-230}, \text{Th-232}, \text{Pb-210}, \text{Po-210}, \text{K-40}\)

\[
A_{i, \text{WWTP-sludge}} = \frac{Q_i}{SR_i (20 \times PE)}
\]

<table>
<thead>
<tr>
<th>(A_{i, \text{WWTP-sludge}})</th>
<th>activity concentration of radionuclide (i) in the sludge (dry matter) produced in WWTP (Bq/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SR(_i)</td>
<td>sludge retention factor for radionuclide (i)</td>
</tr>
<tr>
<td>(Q_i)</td>
<td>annual activity of radionuclide (i) released from waterworks into sewage system (Bq/y)</td>
</tr>
<tr>
<td>(20xPE)</td>
<td>annual sludge (dry matter) produced per year (kg/y) in a WWTP with capacity of served inhabitants expressed in PE</td>
</tr>
</tbody>
</table>

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SRF: Sludge retention factors

- Solid-solution partitioning of radionuclides occurs during sewage treatments.
- SRF: sludge retention factor is the proportion of total activity entering the WWTP that becomes associated with sludge and solids, the remaining being discharged with treated effluent.
- The red values were used in calculations

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>SRF * (from exp.)</th>
<th>SRF § (from Kd)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-234, U-235, U-238</td>
<td>0.01</td>
<td>0.1</td>
</tr>
<tr>
<td>Th-228, Th-230, Th-232</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>Ra-226, Ra-228</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td>Pb-210</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>Po-210</td>
<td>-</td>
<td>0.9</td>
</tr>
</tbody>
</table>

**Dose calculation**

- **Aim of this study**: dose calculation where applied sludge comes from a WWTP.
- **Results from Resrad modelling**: can be applied to evaluate dose to farmers.
- **Dose can be expressed** as a function of annual activity discharged from the waterwarks and capacity of the WWTP.

\[
Dose_{WWTP-sludge}^{i} = (Max\ dose)_{i} \times A_{WWTP-sludge}^{i}
\]

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Dose_{WWTP-sludge}^{i})</td>
<td>Effective dose (mSv/y) for radionuclide (i)</td>
</tr>
<tr>
<td>((Max\ Dose)_{i})</td>
<td>Max dose per Bq/g of dry sludge for radionuclide (i) from Resrad modelling (mSv/y per Bq/g)</td>
</tr>
<tr>
<td>(A_{WWTP-sludge}^{i})</td>
<td>Activity concentration of radionuclide (i) in the dry sludge from WWTP</td>
</tr>
</tbody>
</table>
A case – study: Rakvere Water Works (Estonia)

Rakvere waterwork
15,000 people
5 background water wells
water treatments*

Activity concentration in back wash water (Bq/l)

<table>
<thead>
<tr>
<th>Activity concentration in back wash water (Bq/l)</th>
<th>RESRAD modelling</th>
<th>From RESRAD to Rakvere waterwork</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Max Dose (mSv/y)/(Bq/g)</td>
<td>CL (Bq/g)</td>
</tr>
<tr>
<td>Ra-226</td>
<td>1.77</td>
<td>1.40E-01</td>
</tr>
<tr>
<td>Ra-228</td>
<td>1.80</td>
<td>3.06E-02</td>
</tr>
</tbody>
</table>

*water treatment to remove Fe and Mn

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Doses from the sludge use in agriculture are negligible in this case study.

In the past, studies on this use of sludge were already carried out based on NRPB 13/2 document.

Results were very different since the “dose factor” (dose per unit annual activity release) for this scenario were obtained with very conservative assumptions

- very small waterworks for 500 PE
- retention factor equal to 1, i.e. all radioactivity in the sludge

From the NRPB 13/2 document no information about Ra-228 because it was not considered.
Conclusions

The use of RESRAD allowed to model all the pathways relevant to the use of waterworks sludges in farmland

The results of modelling indicate a possible critical situation for use of sludges directly taken from waterwork treatment plants

When sludges are sent to waste waster treatment plants they don’t seem to determine situation of radiological concern

Perspectives:
– dose for infants (mother milk and cow milk),
– different diets
– age dependent doses
– up date of discharge levels of the RP 135 report
– impact of water treatment technology

In RadoNorm NORM e-survey there is a special section for sludge use in farmland. We invite you to participate to the survey → https://www.radonorm.eu/norm-e-survey/
Thank you for your attention!

cristina.nuccetelli@iss.it