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**ESTIMATION OF CONCENTRATIONS OF RADIONUCLIDES IN
ESTONIAN GROUND WATERS AND RELATED HEALTH
RISKS**

Component 2 – Technical Report

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1. FOREWORD

In the first report a preliminary evaluation of existing data was attempted. Geographic and demographic criteria were applied. The outstanding conclusion were:

- 1) The radiologically monitored water serves 62% of Estonian population.
- 2) The coverage of population is higher in the Northern Estonia: it ranges from 47% to 100% (in Harjumaa district)
- 3) In Southern Estonia the population coverage is rather low (4 % or less), probably due to the expectation of lower radioactivity content.
- 4) Based on the information provided by Estonian authorities, the radioactivity is mainly due to the presence of Ra-226 and Ra-228
- 5) Approximately one half of monitored population in Estonia uses water exceeding the reference level of 0.1 mSv/y
- 6) The average total indicative dose exceeds 0.1 mSv/y in the northern districts (Laanemaa, Harjumaa, Lääne-Virumaa, Ida-Virumaa)
- 7) In the Central and Southern districts the total indicative dose has been mostly calculated from gross alpha and gross beta activities. Resulting values are rather high but hardly reliable.

In the second phase of the project (component 2) more data concerning water supply zone, waterworks, local geology and chemical-physical properties of waters were available. This allowed a more detailed examination of the Estonian database.

In this Technical Report N.2 new topics are considered and discussed:

- Water supply zone sizes and their distribution in connection to radiological risk
- Relationship between water radioactivity and related doses with geological layer of watertables
- Radiological doses in connection with age class of population
- Relationship between radiometric parameters and chemical-physical characteristics of waters

- Preliminary data on effectiveness of operating Estonian treatment plants and perspectives of radium removal techniques.
- Radioactivity content of effluents and wastes from Estonian existing treatment plants and comparison with International Standards.
- Finally a thorough review of international regulation and recommendation is provided.

2. ANALYSIS OF ESTONIAN DATABASE: WATER SUPPLY ZONES, AQUIFERS AND DOSE

by Rosella Rusconi and Elena Caldognetto

2.1. Water Supply Zones size

The analysis on water supply zones (WSZs) size and distribution is useful both to evaluate the feasibility of remedial action and to plan a representative monitoring of water resources (in the following we assume that in the WSZ there are one or more waterworks with the same chemical and physical characteristics).

In Tab. 2.1 and in Fig. 2.1 the water supply zone distribution by size, calculated on the basis of available data, is reported. It accounts for 912 WSZs serving a population of 1,110,215 people. WSZs employing fresh water are included, while private water resources (wells) are not.

Table 2.1 *Distribution of WSZs in Estonia by size*

Class (people served)	Frequency	% Cumulative Frequency
50	146	16,0%
100	162	33,8%
250	269	63,3%
500	177	82,7%
1000	75	90,9%
2500	40	95,3%
5000	20	97,5%
10000	9	98,5%
More than 10000	14	100,0%
N. WSZs	912	
N. people	1110215	

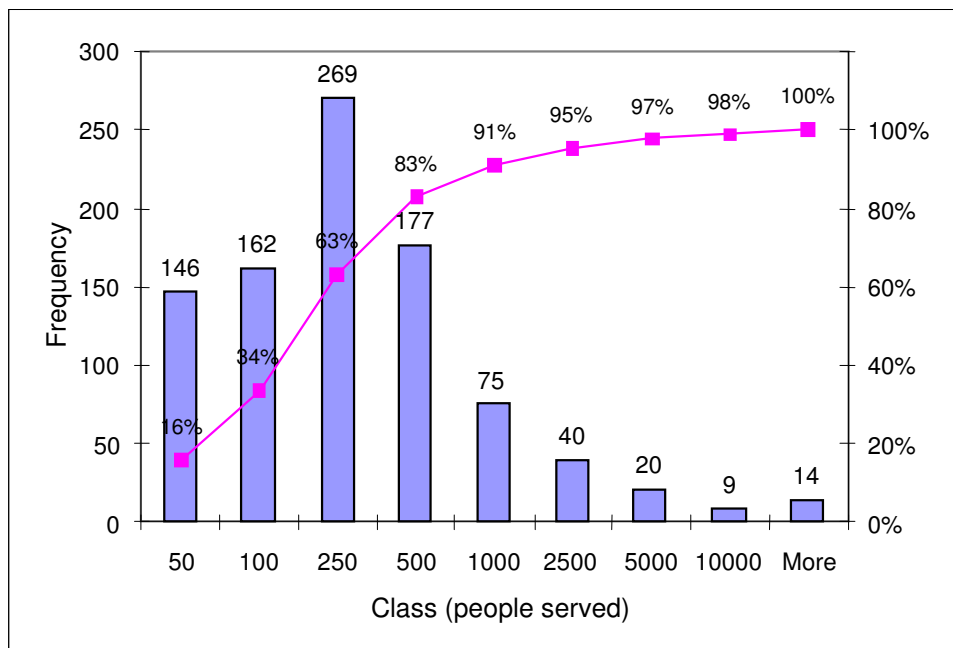


Figure 2.1 *Distribution of WSZs in Estonia by size*

The distribution graph shows that 83% of total WSZs (754) serves 500 people or less.

Most Estonian WSZs are small size ones. As can be seen from the data reported in Tab. 2.2, in every county a similar percentage of small WSZs is found.

Table 2.2 *Distribution of WSZs in Estonian counties by size*

County	People	WSZs (No.)	WSZs ≤ 500 people (No.)	WSZs ≤ 500 people (%)
Hiiumaa	6588	14	11	79
Laanemaa	21870	35	31	89
Harjumaa	510201	172	121	70
Laane-Virumaa	49044	89	78	88
Ida-Virumaa	167833	85	62	73
Raplamaa	22437	51	42	82
Jarvamaa	22527	59	54	92
Jogevamaa	20403	48	39	81
Saaremaa	23260	27	21	78
Parnumaa	55042	59	50	85
Viljandimaa	35782	62	54	87
Tartumaa	120002	74	64	86

County	People	WSZs (No.)	WSZs ≤ 500 people (No.)	WSZs ≤ 500 people (%)
Valgamaa	20314	48	44	92
Polvamaa	15186	42	39	93
Vorumaa	19726	47	44	94
Estonia	1100215	912	754	83

2.2. Available data and aquifers

The overall number of available radiometric data for each Estonian county is summarized in the following table (taken from the first Technical Report):

Table 2.3 *Number of available radiometric data*

County	No. of available data
Hiiumaa	1
Läänemaa	15
Harjumaa	190
Lääne-Virumaa	22
Ida-Virumaa	62
Raplamaa	7
Järvamaa	3
Jõgevamaa	5
Saaremaa	4
Pärnumaa	8
Viljandimaa	10
Tartumaa	10
Valgamaa	6
Põlvamaa	7
Võrumaa	3

In most counties (excluding the Northern counties of Harjumaa and Ida-Virumaa) few analytical data are available.

In this Technical Report a further data analysis was carried out:

1. to test the hypothesis that high radioactivity values can be found only in waters from Cambrian-Vendian aquifer;
2. to assess the actual relevance of the radioactivity problem in the different Estonian counties.

2.2.1 Distribution of high radioactivity values in different aquifers

The following analysis is based on the radiometric database provided by Estonian experts. Gross alpha and beta activity values cannot be used as screening parameters because of the 'interference' of potassium 40 (see par. 3.1); the following analysis is therefore limited to radium isotopes data.

A preliminary analysis of the full data set showed that the number of available data for aquifers different from the Cambrian-Vendian is limited, thus not enough for a reliable statistical analysis based on single aquifers. We therefore decided to cluster the available data in two main groups:

1. "C-V group", including data for Cambrian-Vendian aquifers (C-V, V2gd and V2vr);
2. "non C-V group", including data for the remaining aquifers (O-C, O, D, Q, S, S-O).

The following graphs (Fig. 2.2, 2.3, 2.4, 2.5) show, for each radium isotope and for each group:

- the number of available data,
- the frequency histogram of concentration values,
- the cumulative percentage curve, showing for each size class the percentage frequency of the class added of the percentages in preceding size classes.

The Derived Water Concentration (DWC), that is the activity concentration of each isotope corresponding to an adult ingestion dose of 0.1 mSv/y, is also shown (for more details on DWC see par. 5.3).

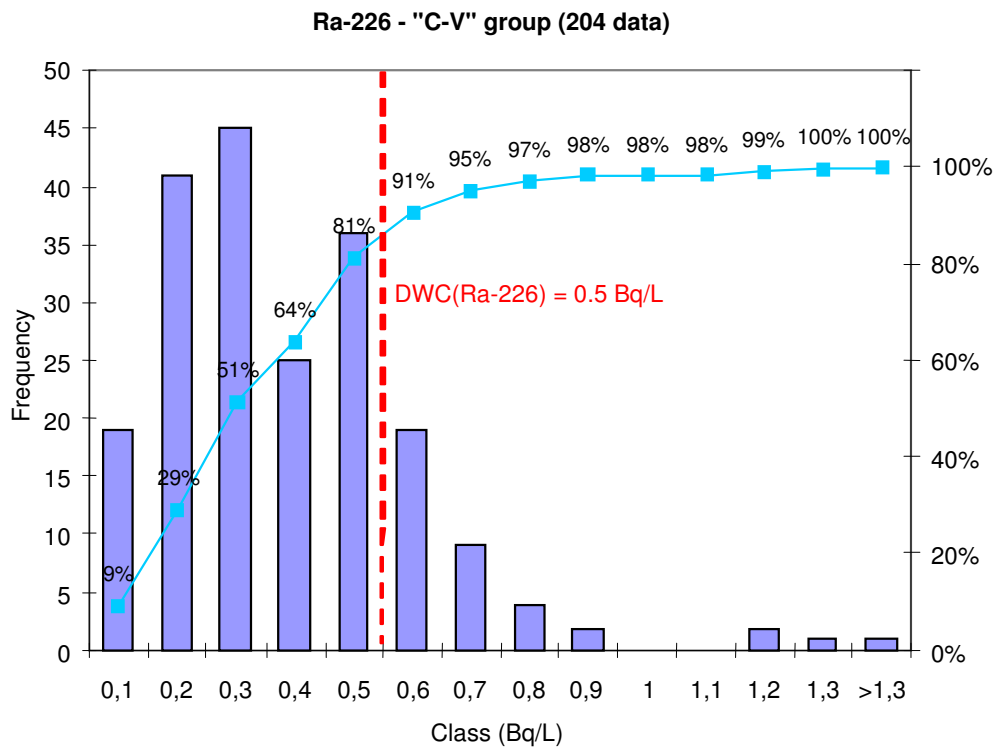


Figure 2.2 Frequency histogram of Ra-226 activity values in "C-V" WSZs

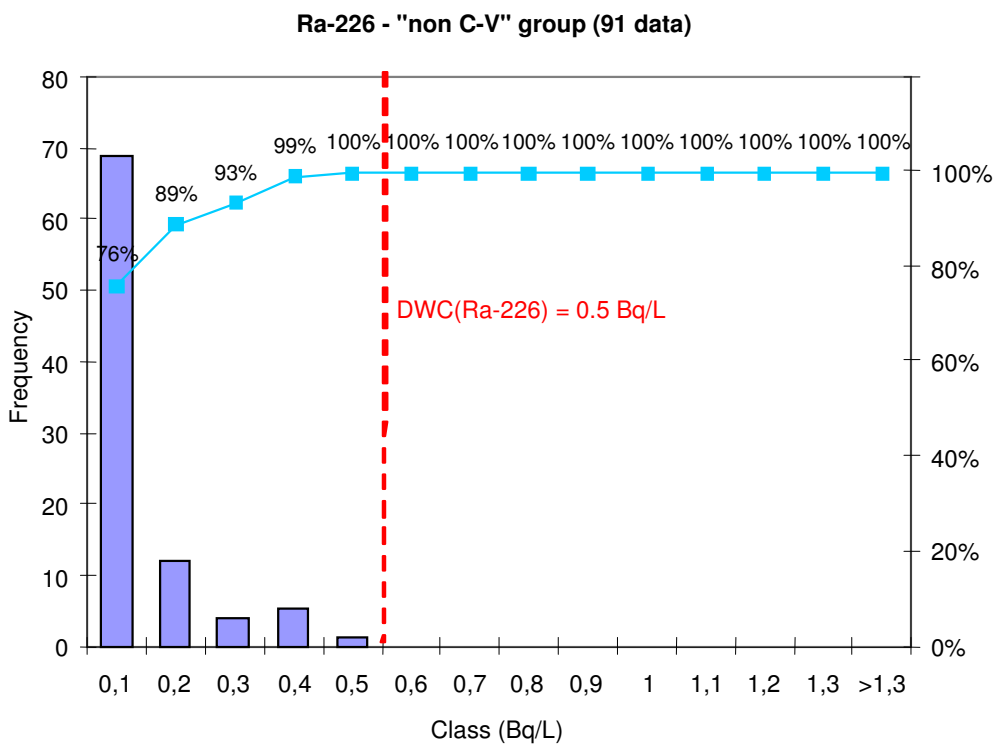


Figure 2.3 Frequency histogram of Ra-226 activity values in "non C-V" WSZs

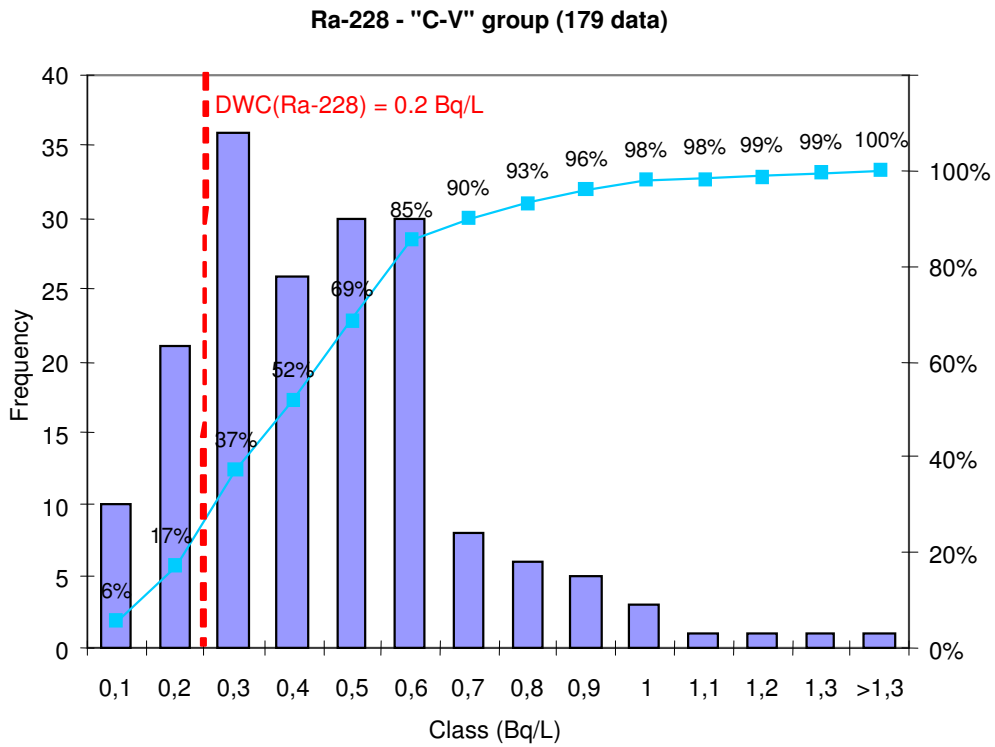


Figure 2.4 Frequency histogram of Ra-228 activity values in "C-V" WSZs

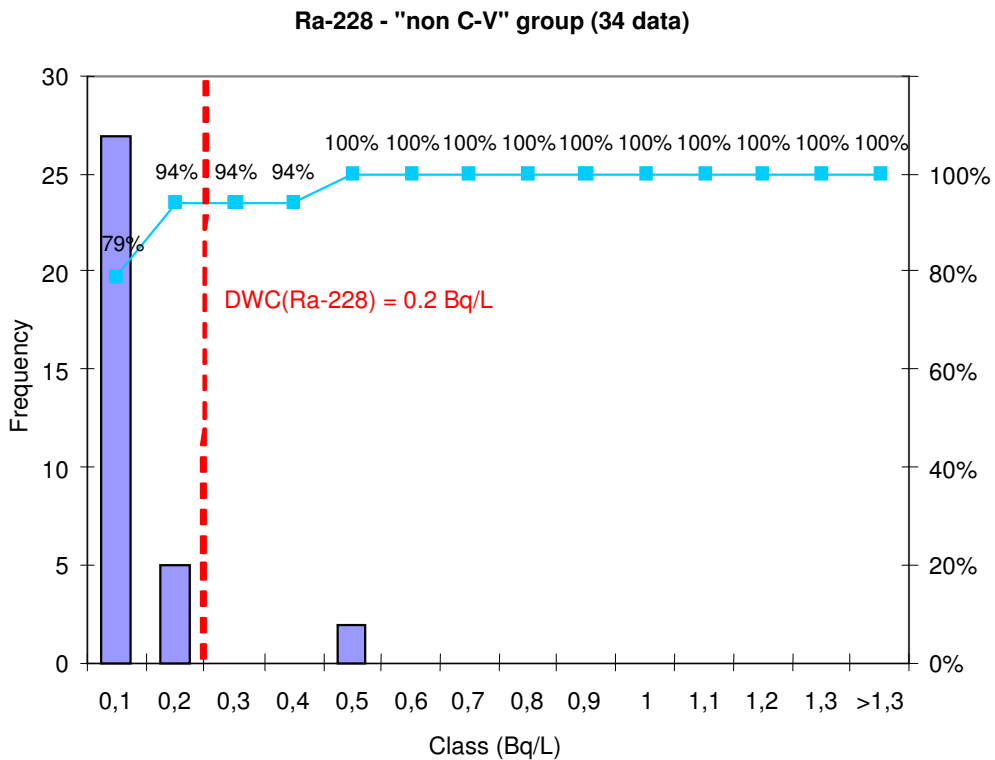


Figure 2.5 Frequency histogram of Ra-228 activity values in "non C-V" WSZs

The most relevant data are summarised in the following Tab. 2.4:

Table 2.4 *Resume of available Radium data*

Group	Ra-226		Ra-228	
	No. of data	% of data > DWC (DWC = 0.5 Bq/L)	No. of data	% of data > DWC (DWC = 0.2 Bq/L)
C-V	204	19%	179	93%
not C-V	91	0%	34	6%

Almost all waters from Cambrian-Vendian WSZs exceed at least one of the DWC values, the main problem is due to high Ra-228 concentrations.

Waters from “non C-V” WSZs look ‘safer’ but the radiation protection problem can’t be completely excluded, as the number of available data is low and not all the values are lower than the DWCs.

2.2.2 Actual relevance of the radioactivity problem in different Estonian counties

The following analysis is based on the “Groundwater complexes database” provided by Estonian experts.

In the whole of Estonia, 912 different water supply zones can be identified. We used the same grouping criteria used for radiometric data (“C-V group” and “non C-V group”) to count the number of supply zones per aquifer group in the different counties (zones taking water from different aquifers are classified using the criteria of the prevalent water source), the results are shown in Tab. 2.5 and in Fig. 2.6:

Table 2.5 *Distribution of “C-V” and “non C-V” WSZs in Estonian counties*

County	No. of “non C-V” WSZs	No. of “C-V” WSZs	% of “C-V” WSZs with respect to the total	Population served by “C-V” WSZs
Harjumaa	88	84	48.8%	134771
Ida-Virumaa	41	44	51.8%	76013

County	No. of "non C-V" WSZs	No. of "C-V" WSZs	% of "C-V" WSZs with respect to the total	Population served by "C-V" WSZs
Laane-Virumaa	80	9	10.1%	23451
Laanemaa	32	3	8.6%	13365
Tartumaa	74	0	0%	0
Viljandimaa	62	0	0%	0
Parnumaa	59	0	0%	0
Jarvamaa	59	0	0%	0
Raplamaa	51	0	0%	0
Valgamaa	48	0	0%	0
Jogevamaa	48	0	0%	0
Vorumaa	47	0	0%	0
Polvamaa	42	0	0%	0
Saaremaa	27	0	0%	0
Hiiumaa	14	0	0%	0

Distribution of "C-V" water supply zones in Estonia (tot. 140)

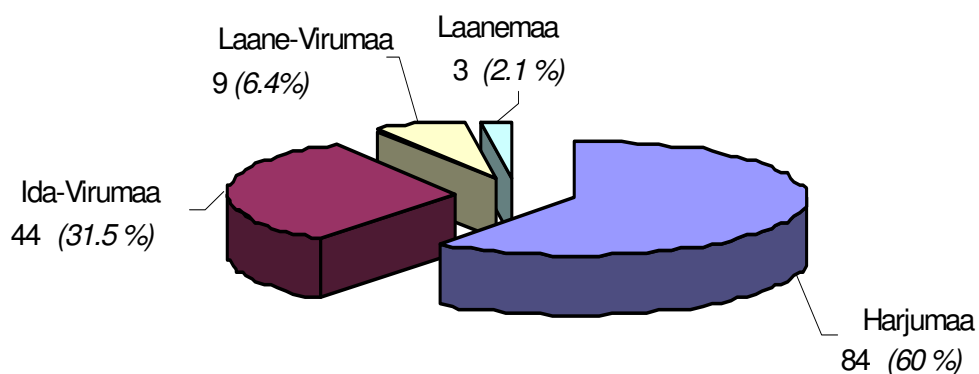


Figure 2.6 Distribution of "C-V" water supply zones in Estonia

In conclusion, C-V aquifers used for drinking purposes are 140 out of 912, all of them in the four Northern counties; they account for about 50% of the whole water supply zones in Harjumaa and Ida-Virumaa, and for about 10% in Laane-Virumaa and Laanemaa.

If we assume that high levels of radioactivity can be found mainly in the “C-V group” water supply zones, we can conclude that the problem is mostly limited to 140 water supply zones to about 250000 people (22% of Estonian population). Actually, the 12 biggest water supply zones (population served > 5000) feed water to about 152000 people (see Tab. 2.6).

Table 2.6 *Twelve biggest “C-V” water supply zones*

Db_no	County	Municipality	Name of water supply zone	Water producer	Production m ³ / day	Population served
6	Harjumaa	Tallinn	Nõmme wells WSZ no 7	AS Tallinna Vesi	4800	33300
835	Ida-Virumaa	Kohtla-Järve town	Kohtla-Järve town Järve district, Tākumetsa and Peeri villages WSZ	Järve Biopuhastus OÜ	2364	19689
742	Lääne-Virumaa	Rakvere town	Rakvere town WSZ	Rakvere Vesi AS	1500	16000
851	Ida-Virumaa	Sillamäe town	Sillamäe town WSZ	Sillamäe Veevärk AS	2053	15680
194	Läänemaa	Haapsalu town	Haapsalu	Haapsalu Veevärk AS	1600	13000
846	Ida-Virumaa	Jõhvi town	Jõhvi town WSZ	Jõhvi Veemajandus OÜ	2590	12400
9	Harjumaa	Maardu town	Kallavere south district (old)	AS Maardu Vesi	1500	9700
16	Harjumaa	Keila town	Keila town WSZ	AS Keila Vesi	1100	9258
849	Ida-Virumaa	Kiviõli town	Kiviõli town WSZ	Kiviõli Vesi OÜ	653	6734
10	Harjumaa	Maardu town	Kallavere north district	AS Maardu Vesi	650	5700
2	Harjumaa	Saue town	Saue wells WSZ no 8	AS Tallinna Vesi	1800	5200
98	Harjumaa	Viimsi municipality	Lääneranniku WSZ	AS Viimsi Vesi	1400	5012

The following graphs (see Fig. 2.7 and 2.8) provide more information about the “C-V group” supply zones: 83 supply zones serve less than 500 people; the 12 biggest zones altogether serve about 152000 people, 10 of them produce more that 1000

m³/day of water. The 71 supply zones producing less than 50 m³/day of water altogether serve about 12400 people.

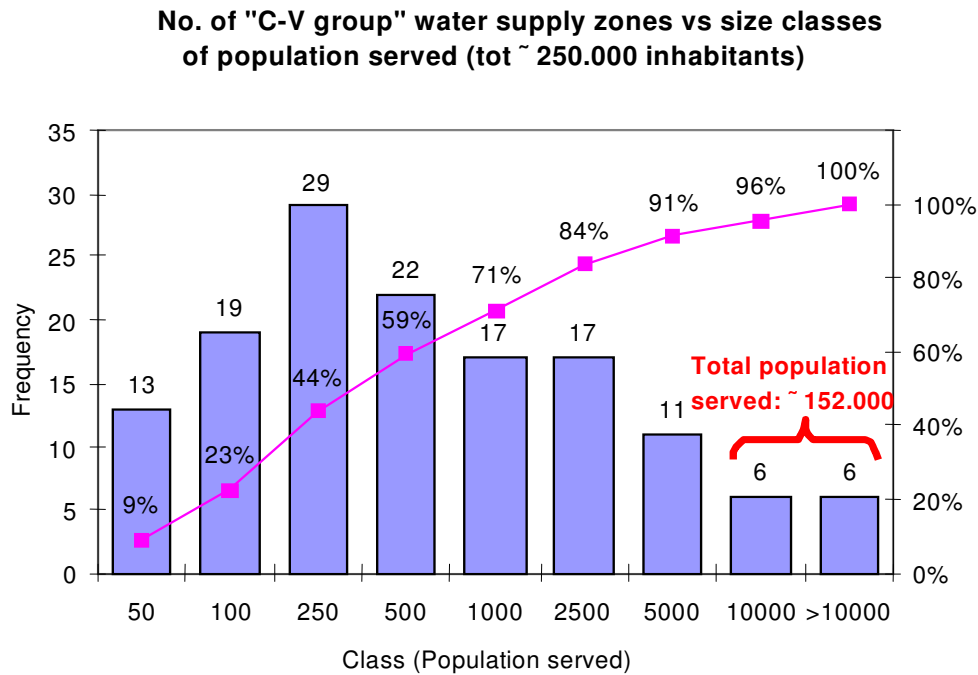


Figure 2.7 Distribution of "C-V" water supply zones by population served

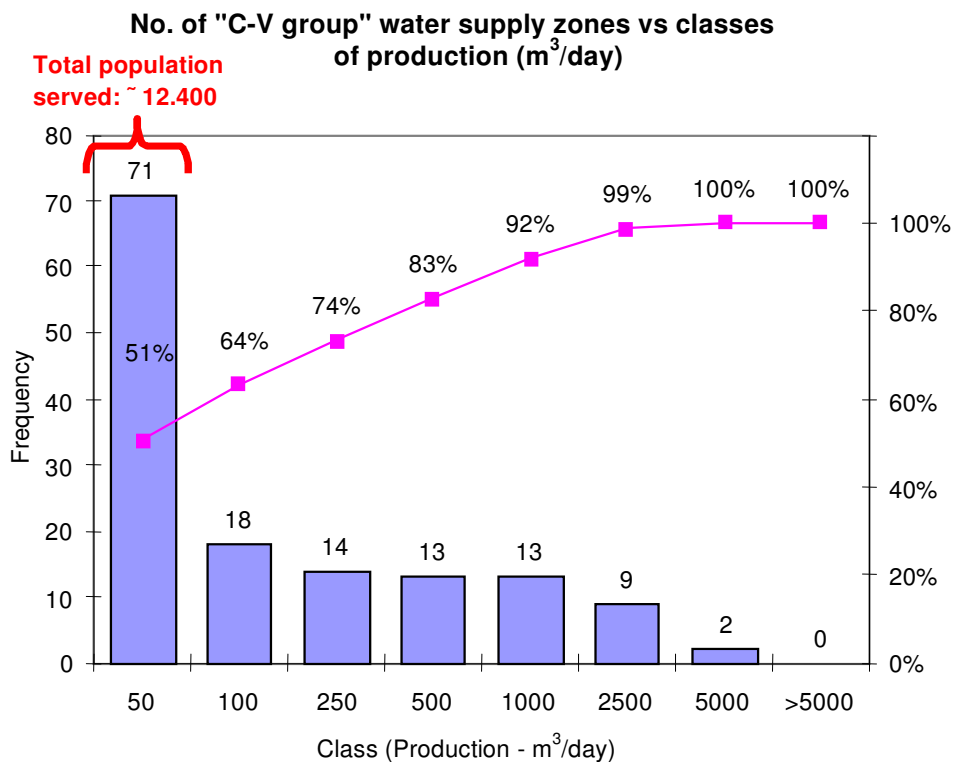


Figure 2.8 Distribution of "C-V" water supply zones by daily water production

Furthermore, in 16 zones out of 140, water from “C-V group” aquifers is mixed with water from “non C-V” aquifers or surface water.

2.3. Total indicative doses

Radium values reported in Estonian radiometric database have been used to calculate the Total Indicative Dose (TID) using the following formula:

$$\text{TID(Sv / y)} = \text{Conc. (Bq/L)} \times \text{Committed eff. dose per unit intake (Sv/Bq)} \times \text{Ann. intake value (L/y)}$$

The TID was evaluated using committed effective doses per unit intake via ingestion (Sv/Bq) for adults and infants (≤ 1 y), reported in the EURATOM Basic Safety Standards, and on the basis of the intake values chosen by the Article 31 working party.

Activity concentration values lower than the minimum detectable activity (MDA) were previously removed in order to avoid undue dose overestimates (actually, the dose estimate analysis gives similar results even considering values $<$ MDA). Only records for which both radium isotope values were available have been used.

The results for “adult” age class are summarised in the following graphs (Fig. 2.9 and 2.10):

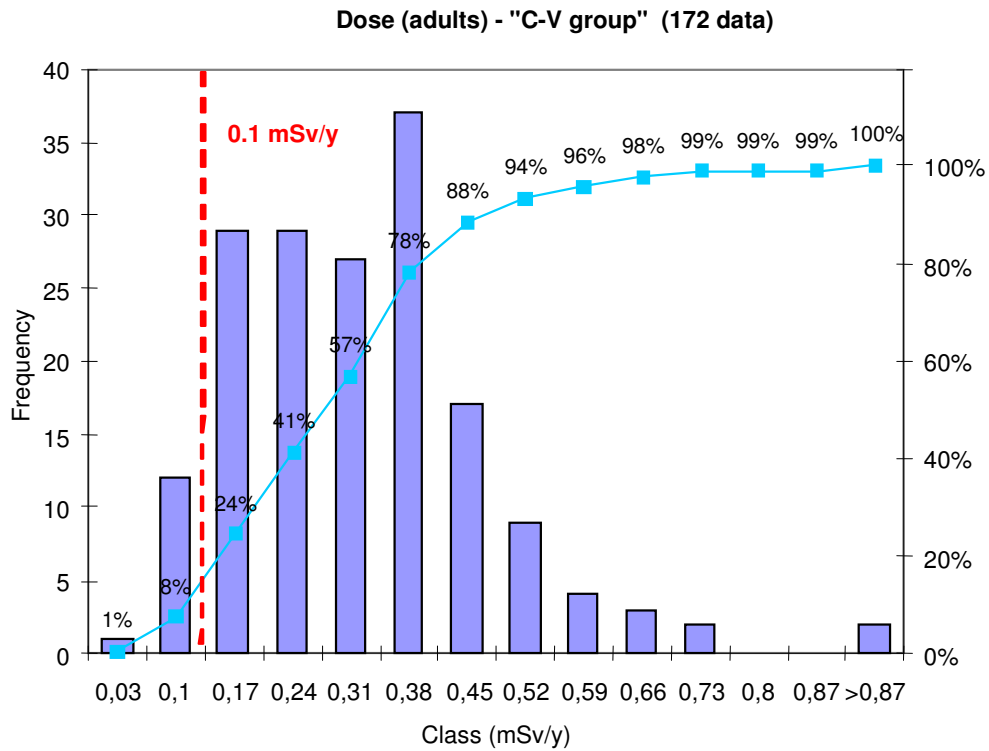


Figure 2.9 Distribution of adult dose - "C-V" group WSZs

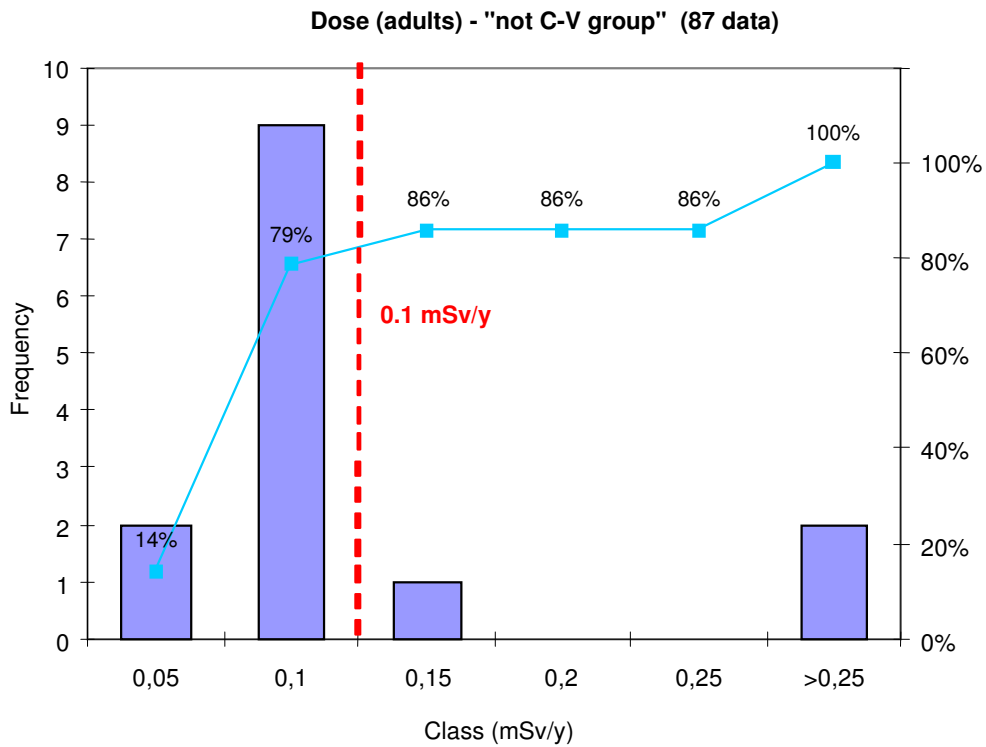


Figure 2.10 Distribution of adult dose - "non C-V" group WSZs

TIDs due to waters from C-V aquifers exceed the dose parametric value of 0.1 mSv/y in 91% of cases, as expected on the basis of radium data analysis.

For waters from “non C-V” aquifers the number of available data is low, nonetheless it is worth to point out that also in this case not all TID values are lower than 0.1 mSv/y. This is even more dramatic if the dose is calculated from the same measured data for the “infant” (≤ 1 y) class age, as shown in the Fig. 2.11 and 2.12.

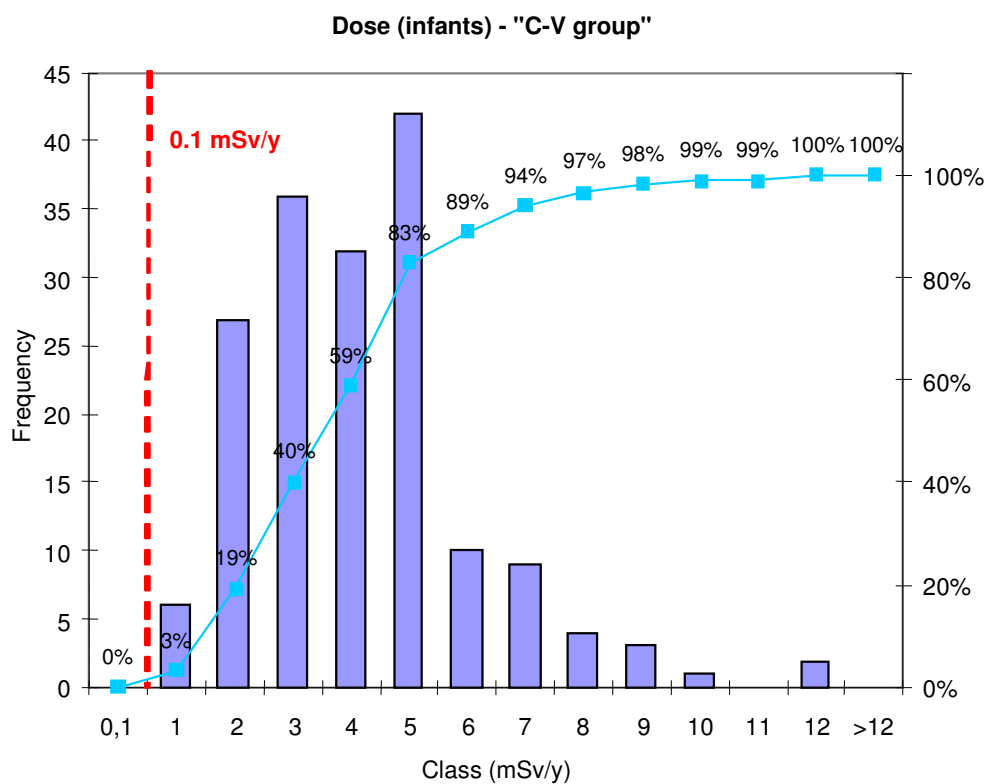


Figure 2.11 Distribution of infants (≤ 1 y) dose - “C-V” group WSZs

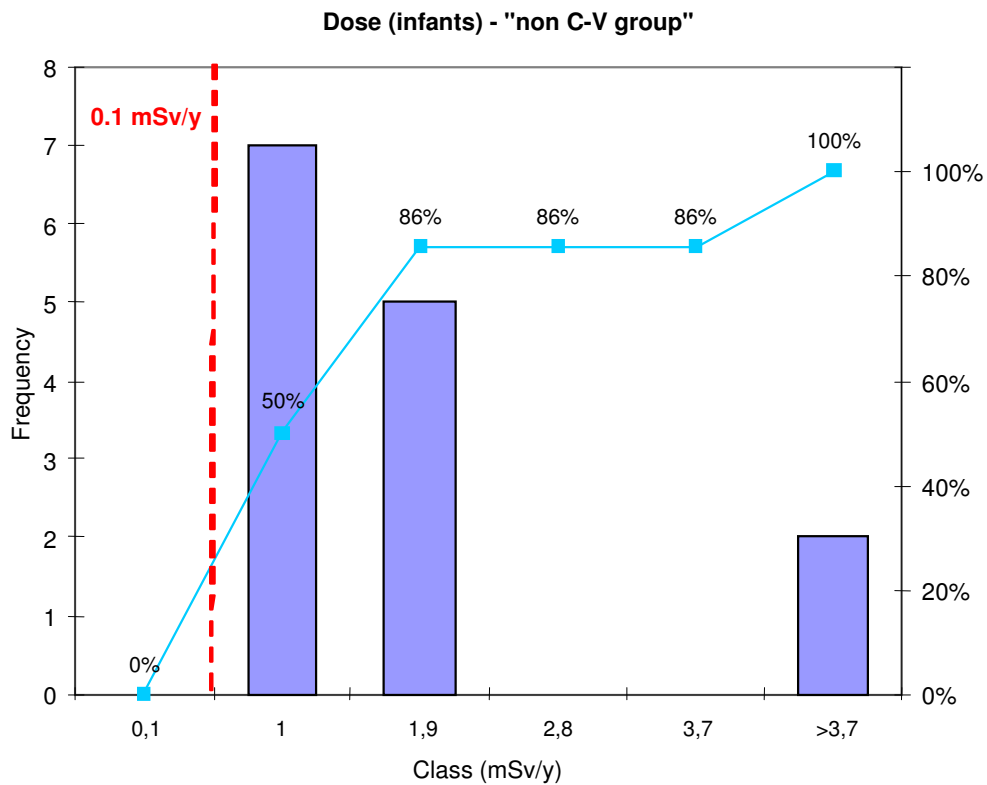


Figure 2.12 *Distribution of infants (≤ 1 y) dose - "non C-V" group WSZs*

In the previous graphs we only considered adults and infants. However, TID values are always lower for adult class age than for any other class; this is especially true for teenagers, due to high radium metabolic absorption in bones (radium has the same chemical behaviour as calcium) during the growing age.

3. ANALYSIS OF ESTONIAN DATABASE: RADIOMETRIC DATA

by Maurizio Forte and Luca Bagnato

3.1. Gross alpha and beta activities

WHO recommends the measurement of gross alpha and beta activities as first screening of water radioactivity. Attention values of 0.5 Bq/L for alpha activity and 1 Bq/L for beta activity are given. Unfortunately, when radium isotopes are the prevailing radioactive components this method may not be precautionary: 0.5 Bq/L of the alpha emitter Ra-226 will lead to a 0.1 mSv/y total indicative dose (TID) by itself and 1 Bq/L of the beta emitter Ra-228 will lead to much higher doses.

On the other hand, if the whole gross beta activity would be used to estimate Ra-228 concentration, the resulting TID would be largely overestimated since an important contribution from K-40 is expected.

K-40 activity concentration can easily be calculated from potassium concentration given by chemical analyses (1 g/L of potassium corresponds to 30.7 Bq/L of K-40, which produces a beta activity of 27.6 β /L). The concentration of potassium in Estonian ground water is relatively high.

By examination of database results, the average contribution of K-40 to the beta activity is 0.24 Bq/L (Fig. 3.1). K-40 is the prevailing contributor to gross beta activity in more than 50% of the samples (Fig. 3.2)

In 76% of examined cases the K-40 activity concentration is higher than 200 mBq/L. If 200 mBq/L were attributed to Ra-228, it would lead to total indicative doses of 0.1 mSv/y.

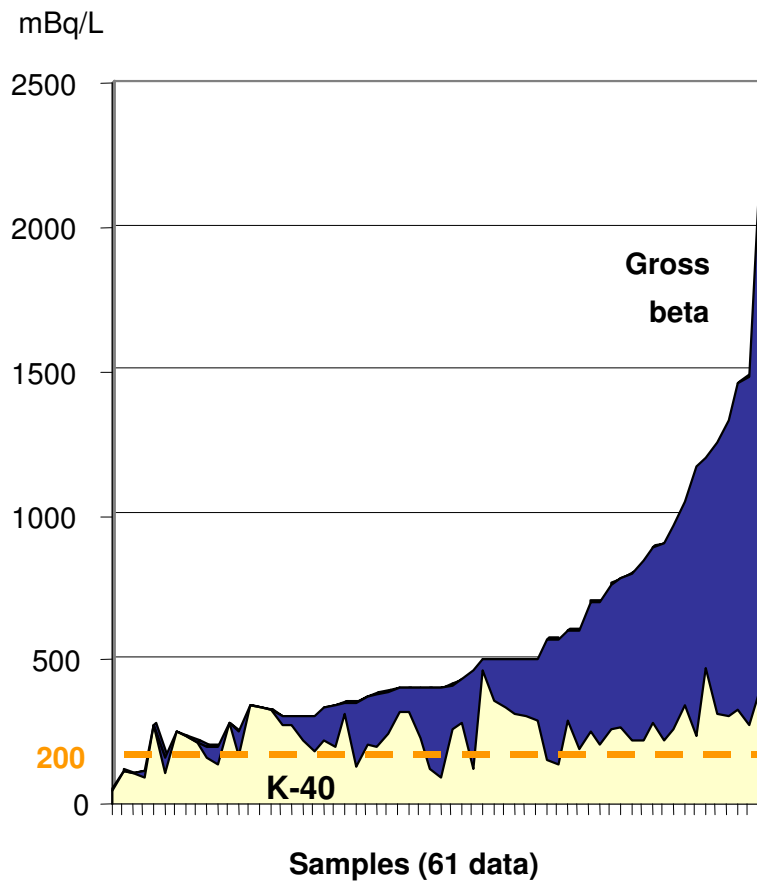


Figure 3.1 Potassium-40 contribution to gross beta activity concentration (mBq/L)

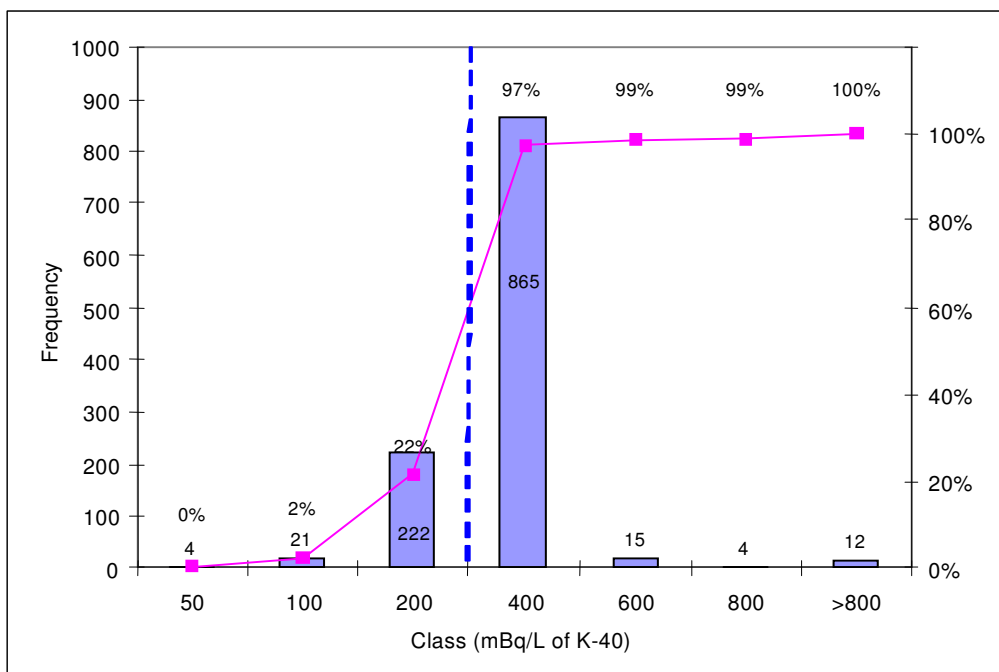


Figure 3.2 Frequency histogram of K-40 activity based on chemical measurement of potassium (1143 input data)

In conclusion, gross alpha and beta activities are not a good radiological descriptor for Estonian water and their use should not be encouraged.

3.2. Statistical analysis

3.2.1 Radium vs chemical parameters

A first attempt was made to look for correlations between chemical and radiometric parameters. Since chemical analyses are currently and easily done on a large number of samples, such a relation would have an obvious advantage of pointing out high radioactivity values without further analytical efforts.

In literature straightforward connections between chemical and radiochemical parameters can hardly be found, nevertheless the peculiarity of Estonian water stimulated such an effort.

Statistical analysis was affected by the limited amount of samples with both radium isotopes data and complete chemical analyses (56 data only). Most of the examined samples belongs to the Cambrian-Vendian aquifers.

The correlation matrix (Fig. 3.3) shows that the strongest correlation ($R=0.82$) is between Ra-228 concentrations and water mineralization (the sum of chemical macro-components, approximately the total salt content). A weaker relation ($R=0.67$) is between mineralization and Ra-226. Other chemical parameters, like total residue and calcium concentration, show a similar behaviour: this is reasonable since both mineralization and total residue are a measure of dissolved salts and calcium is a major component of them.

The amount of dissolved solids is not specific to Cambrian-Vendian aquifers. In Fig. 3.4 - 3.6 it is shown that a similar distribution of mineralization is found in other aquifers. This means that a high degree of mineralization (e.g. > 300 mg/L) is not a specific feature of Cambrian-Vendian waters.

	Ra226	Ra228	Residue	Mineral.	Na	K	NH4	Ca	Mg	Fe2	Fe3	Cl	SO4	HCO3	pH	SiO2	CO2v	Oxidab.	Al	Mn	PO4
Ra226	1,00	0,77	0,67	0,73	0,58	0,53	-0,04	0,73	0,63	0,58	0,21	0,64	-0,41	0,44	-0,57	-0,10	0,34	0,28	0,11	0,11	0,37
Ra228		1,00	0,80	0,82	0,79	0,61	0,07	0,79	0,61	0,57	0,00	0,77	-0,22	0,23	-0,49	0,13	0,16	0,38	-0,10	-0,06	0,20
Residue			1,00	0,99	0,95	0,70	0,28	0,96	0,77	0,56	0,00	0,98	-0,35	0,20	-0,59	-0,01	0,07	0,56	-0,16	-0,09	0,26
Mineral.				1,00	0,92	0,75	0,30	0,99	0,80	0,59	0,01	0,95	-0,42	0,35	-0,63	0,01	0,14	0,58	-0,07	-0,04	0,26
Na					1,00	0,68	0,18	0,87	0,59	0,50	-0,05	0,96	-0,22	0,02	-0,42	-0,02	-0,10	0,49	-0,21	-0,13	0,29
K						1,00	0,19	0,74	0,61	0,41	0,11	0,67	-0,44	0,37	-0,44	-0,02	0,10	0,35	0,20	-0,08	0,25
NH4							1,00	0,34	0,39	0,29	-0,18	0,24	-0,21	0,25	-0,32	0,17	-0,02	0,39	-0,07	0,05	-0,08
Ca								1,00	0,80	0,58	0,00	0,91	-0,45	0,41	-0,62	0,06	0,21	0,62	-0,06	0,00	0,22
Mg									1,00	0,64	0,18	0,73	-0,58	0,47	-0,80	-0,14	0,24	0,44	0,02	0,01	0,27
Fe2										1,00	0,12	0,58	-0,35	0,21	-0,55	0,03	0,13	0,15	0,05	0,13	0,32
Fe3											1,00	0,05	-0,16	0,00	-0,18	-0,45	-0,03	-0,11	0,34	-0,12	0,10
Cl												1,00	-0,32	0,05	-0,54	-0,06	-0,02	0,53	-0,21	-0,13	0,29
SO4													1,00	-0,64	0,57	0,21	-0,21	-0,37	-0,15	-0,36	-0,54
HCO3														1,00	-0,53	0,05	0,50	0,28	0,40	0,33	0,15
pH															1,00	0,27	-0,34	-0,18	-0,11	-0,12	-0,26
SiO2																1,00	0,17	0,11	-0,19	-0,05	-0,50
CO2v																	1,00	0,00	0,05	-0,05	-0,14
Oxidab.																		1,00	-0,10	-0,05	-0,06
Al																			1,00	0,03	0,07
Mn																				1,00	0,26
PO4																					1,00

Figure 3.3 : Correlation matrix among radiometric and chemical parameters

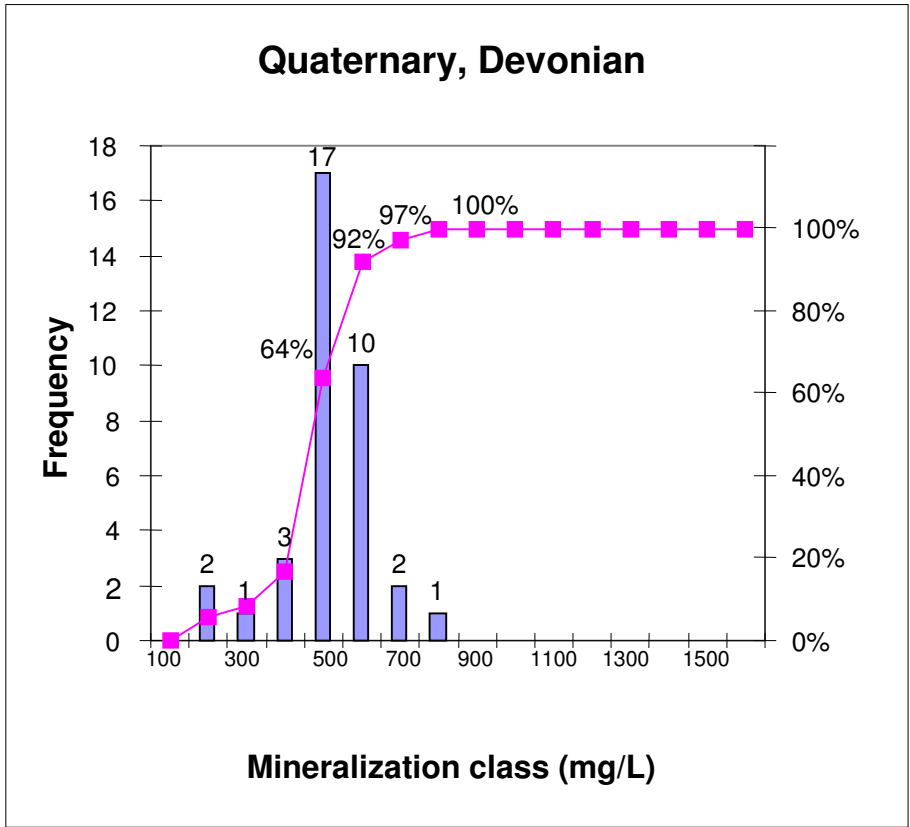


Figure 3.4 Mineralization of Quaternary and Devonian waters

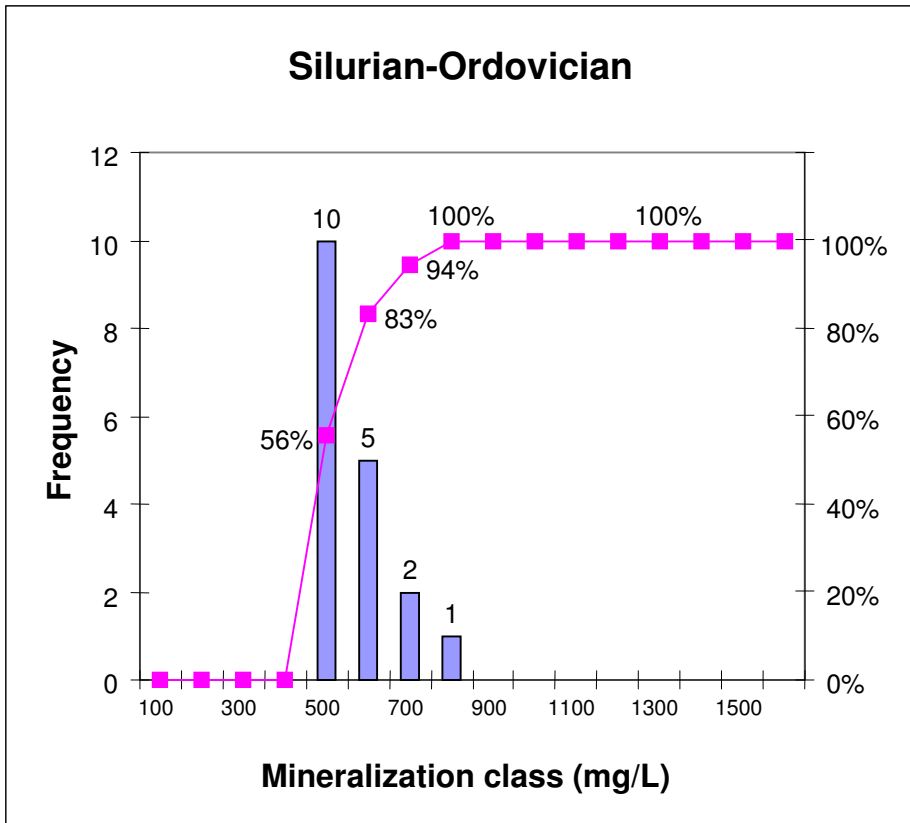


Figure 3.5 Mineralization of Silurian-Ordovician waters

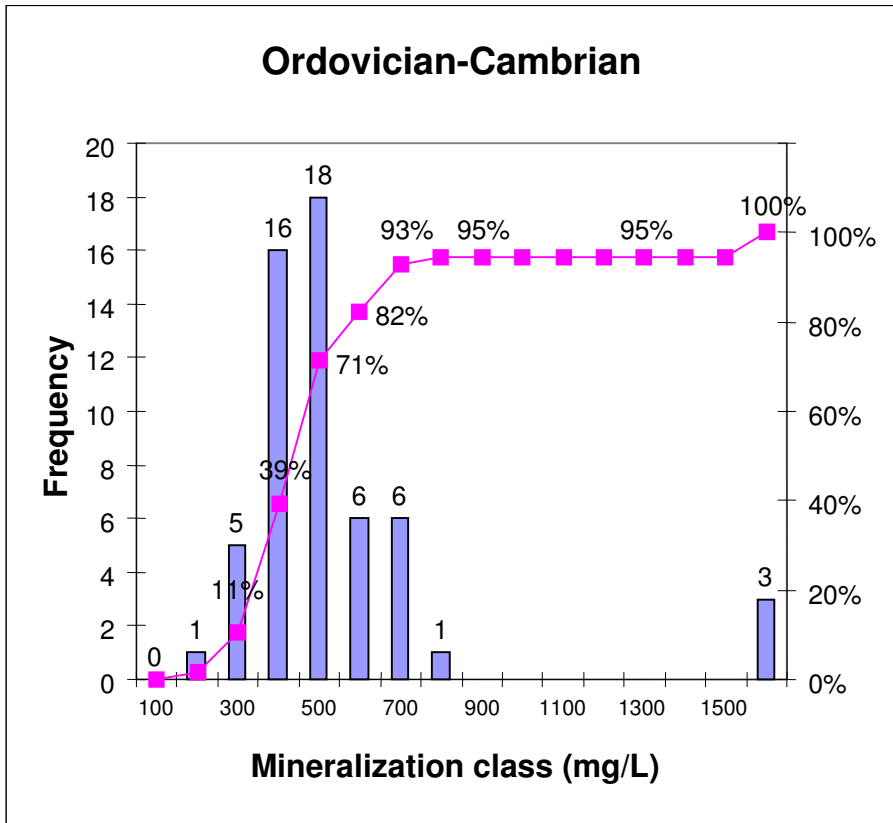


Figure 3.6 Mineralization of Ordovician-Cambrian waters

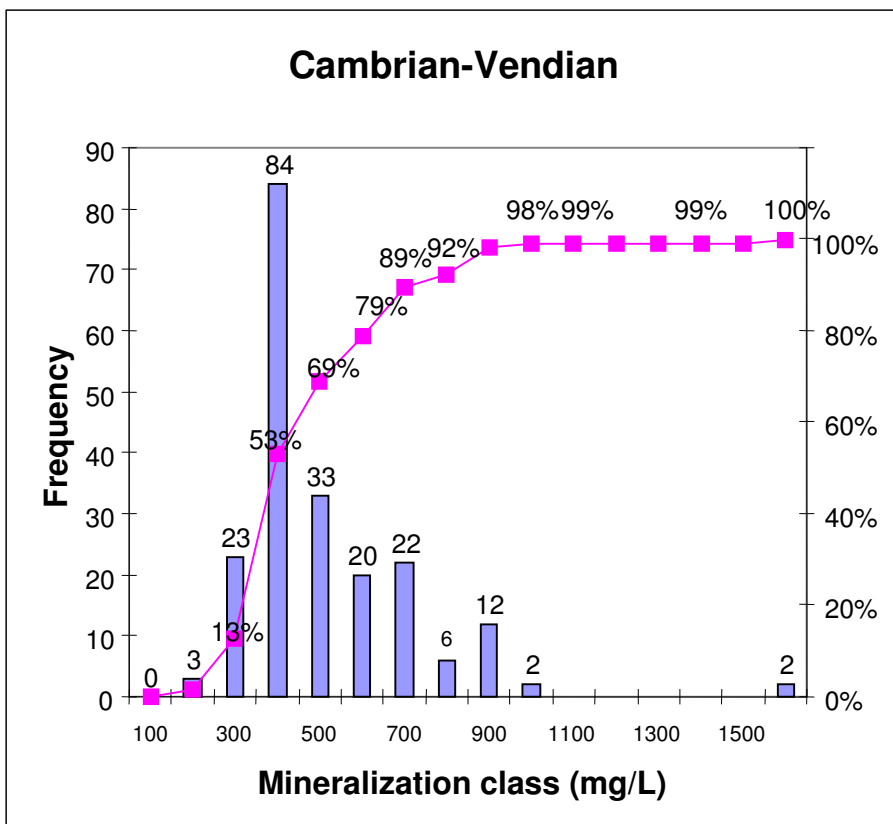


Figure 3.7 Mineralization of Cambrian-Vendian waters

The relationship between radium activity concentration and degree of mineralization (which can be widely variable) within the Cambrian-Vendian could thus be a useful predictive tool if further statistical analysis on a higher amount of data will confirm it.

The correlation graph between Ra-228 activity concentration and mineralization is shown in Fig 3.7.

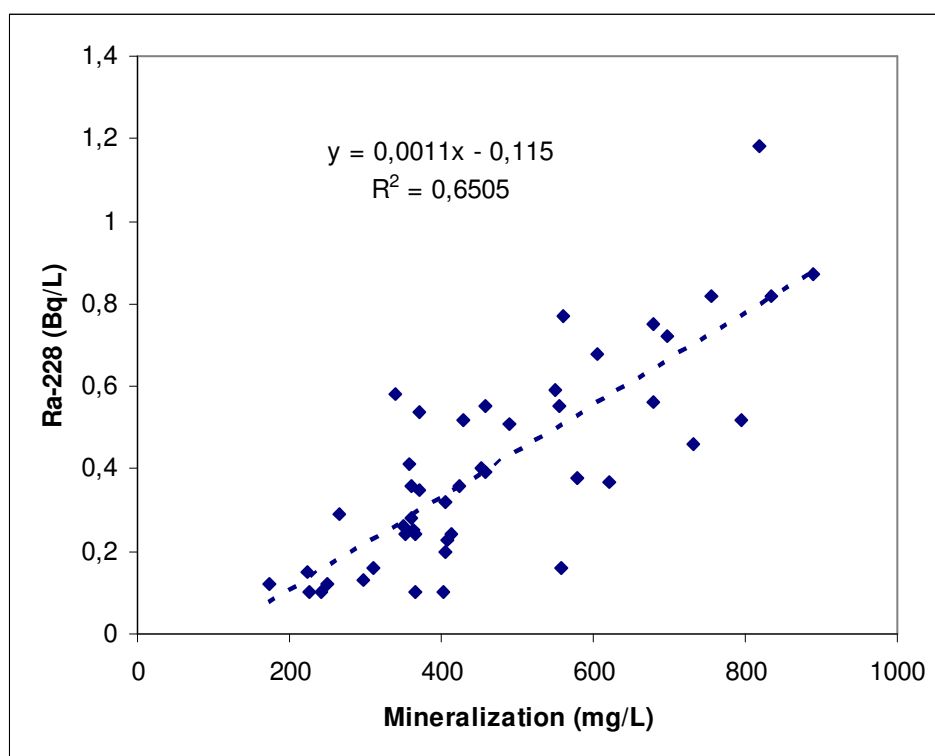


Figure 3.7 *Correlation graph between Ra-228 and water mineralization*

A further attempt to investigate relations between radium concentration and groups of chemical parameters (multiparametric analysis) gave poor results: no remarkable improvement with respect to single parameter correlations was observed. More sophisticated statistical techniques could be applied in the future.

3.2.2 Ra-226 vs Ra-228

The relationship between Ra-226 and Ra-228 was then investigated. 191 couples of Ra-226 and Ra-228 activity concentration values were used as input parameters.

The first step was a non parametric analysis in order to identify the real structure of the relationship. As shown in Fig. 3.8, both a linear and exponential model could be used. We chose the linear model for further elaborations since it is the simplest one.

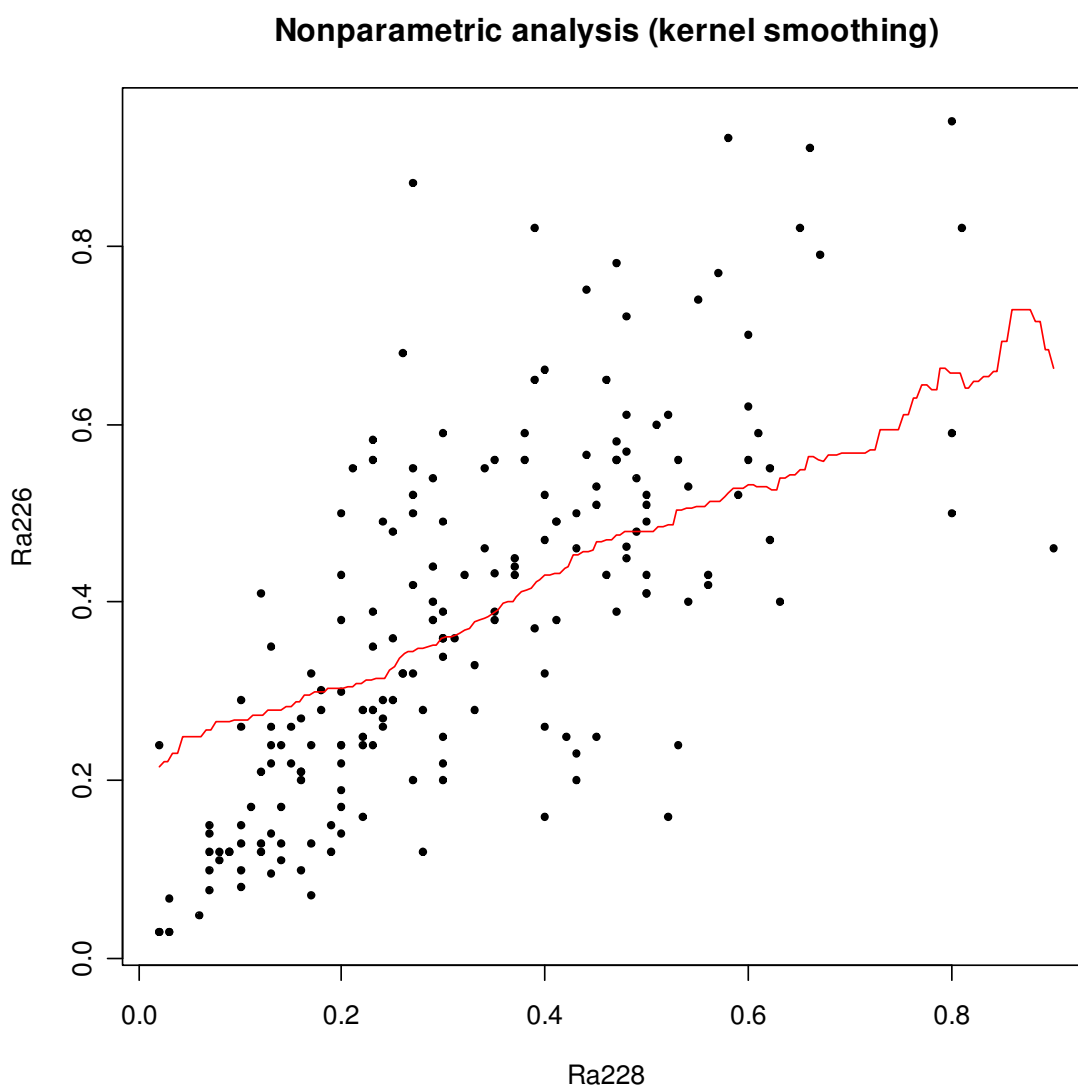


Figure 3.8 *First check of correlation between Ra-226 and Ra-228 (activity concentrations in Bq/L)*

The p-values reported in Table 3.1 support the use of the linear model at the usual significance level of 95%. All p-values show that both intercept and slope, are significantly different from 0, thus a linear model seems to be appropriate for our analysis.

Table 3.1 Parameters of linear regression (Ra-226 vs. Ra-228)

Parameter	Value	Std. err.	T value	P value
Intercept	0.07399	0.01890	3.914	$1,27 \cdot 10^{-4}$
Angular coeff.	0.65322	0.04422	14.771	$< 2 \cdot 10^{-16}$
R	0.732	-	-	-
R ²	0.536	-	-	-

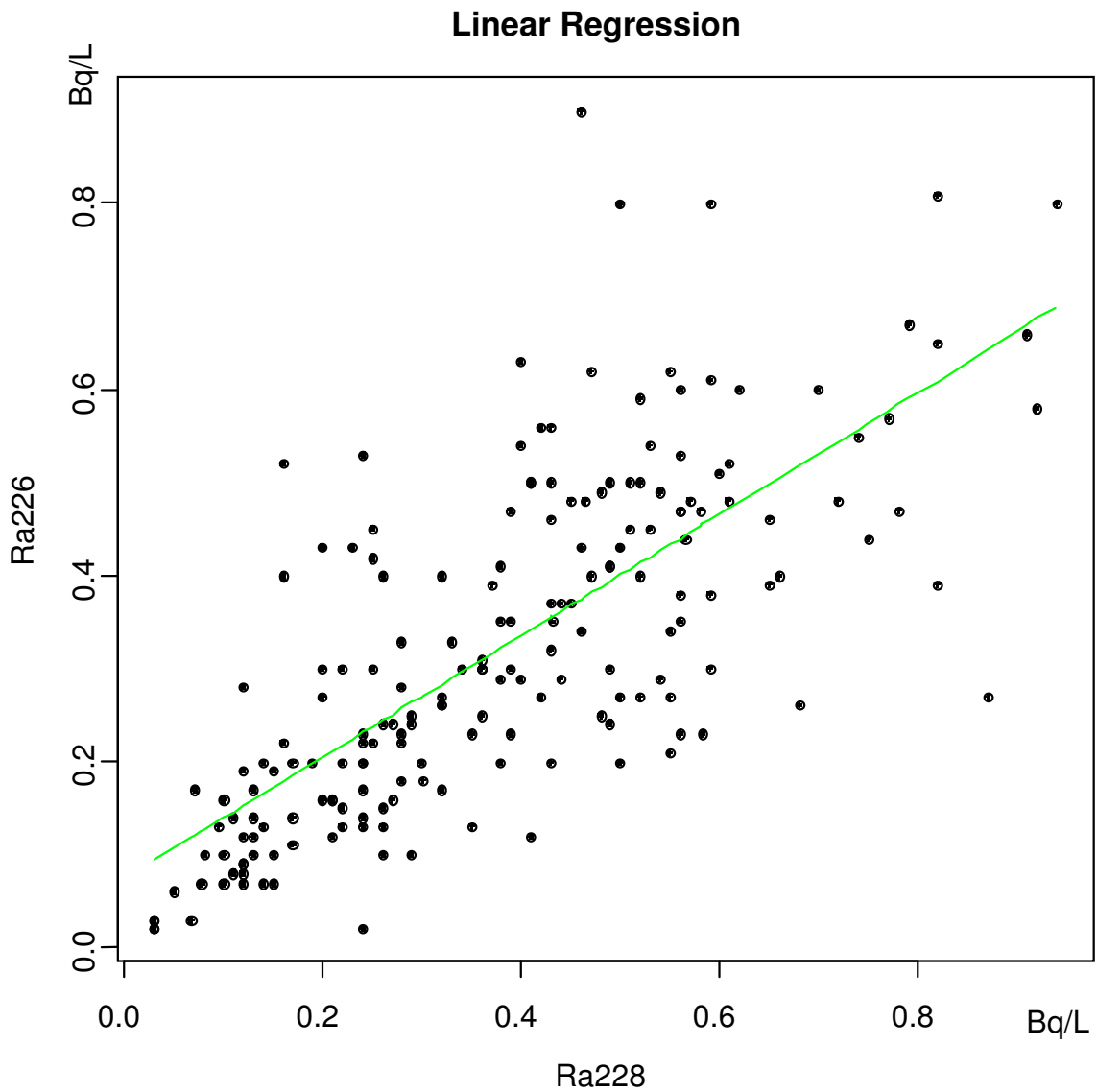


Figure 3.9 Linear regression graph (Ra-226 vs. Ra-228)

Although further refinements as confidence interval calculations should still be done, the correlation has the advantage of allowing the rough estimate of Ra-226 concentration from the experimentally measured activity concentration of Ra-228.

3.2.3 Preliminary remarks on total dose calculation

Ingestion dose coefficients for Ra-226 and Ra-228 are rather different, being the latter 2.5 times higher for adult class age and 6.5 times for infants (≤ 1 y) (Tab. 3.2)

Table 3.2 *Ingestion dose coefficients for Ra-226 and Ra-228*

Age class	Ra-226 (Sv/Bq)	Ra-228 (Sv/Bq)	Ra-228/Ra-226
Infants (≤ 1 y)	$4.70 \cdot 10^{-06}$	$3.05 \cdot 10^{-05}$	6.49
Adults	$2.80 \cdot 10^{-07}$	$6.90 \cdot 10^{-07}$	2.46

For this reason, despite the similar activity concentration of the two radium isotopes, the contribution to the total dose is higher for Ra-228 in the set of waters under examination especially in the case of infants (≤ 1 y) (Fig. 3.10 and 3.11),:

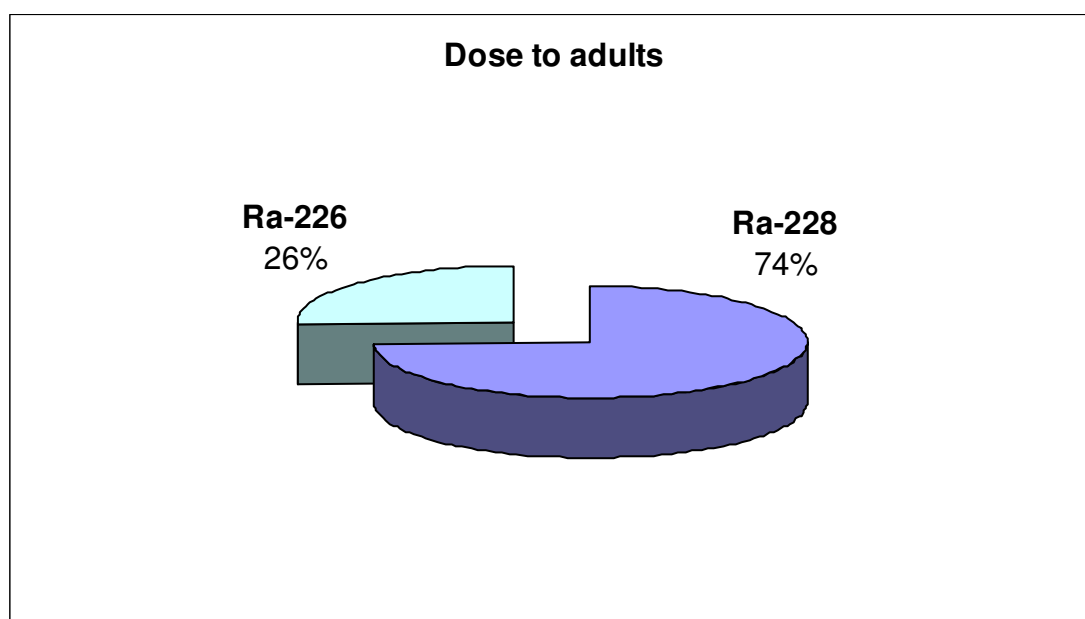


Figure 3.10 *Relative contribution to total dose for adults age class*

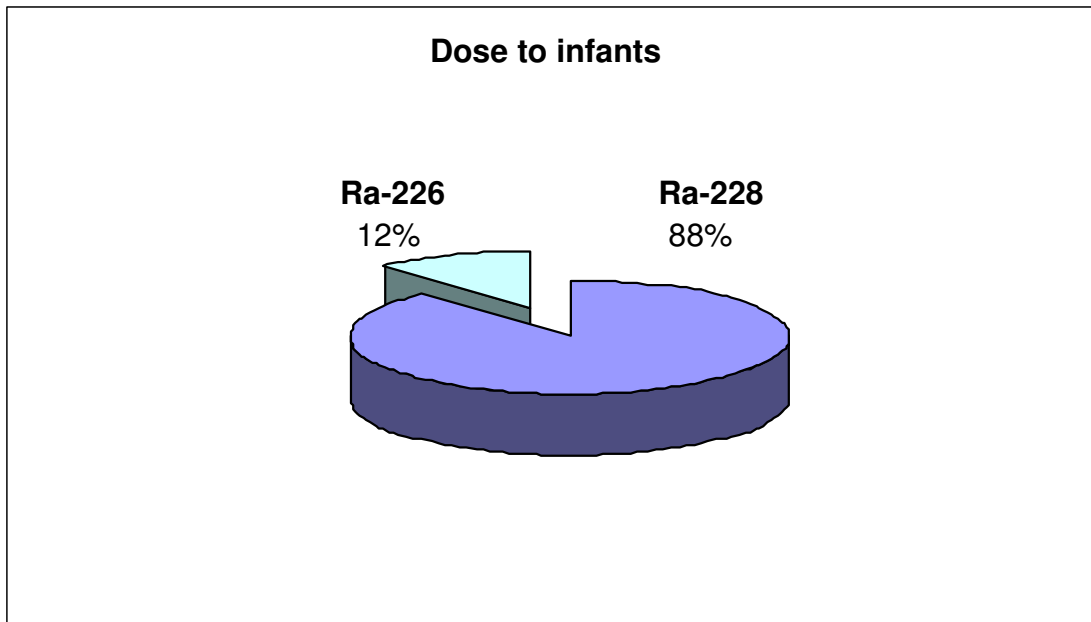


Figure 3.11 *Relative contribution to total dose for infants age class*

Since the relative contribution of Ra-226 is small, a rough estimation will not affect heavily the total dose calculation. Therefore, if only Ra-228 concentration has been experimentally measured, Ra-226 can be estimated on the basis of the linear regression previously shown (Fig. 3.9) by the use of parameters reported in Table 3.2 with the following equation:

$$C_{\text{Ra226}} = 0.65322 * C_{\text{Ra228}} + 0.07399 \quad (1)$$

where C_{Ra226} and C_{Ra228} are, respectively, Ra-226 and Ra-228 activity concentrations in Bq/L. From these values it is possible to calculate the dose for the two radium isotopes and then the total dose. Applying the ingestion dose factors, the following equation can be obtained:

$$D_{\text{adults}} = 0.6372 * C_{\text{Ra228}} + 0.0151 \quad (2)$$

Where D_{adults} is the ingestion dose for adult class age in mSv/y. In Fig. 3.12 a graphic representation is given.

These preliminary results should be considered only a useful tool to be developed. It may then be used as a screening method with the obvious advantage of calculating the total dose by the use of only one radiometric parameter. Moreover, activity concentrations of Ra-228 can be determined by gamma spectrometry on pre-concentrated samples (see Technical Report 1), with a faster procedure compared to the radiochemical determination of Ra-226.

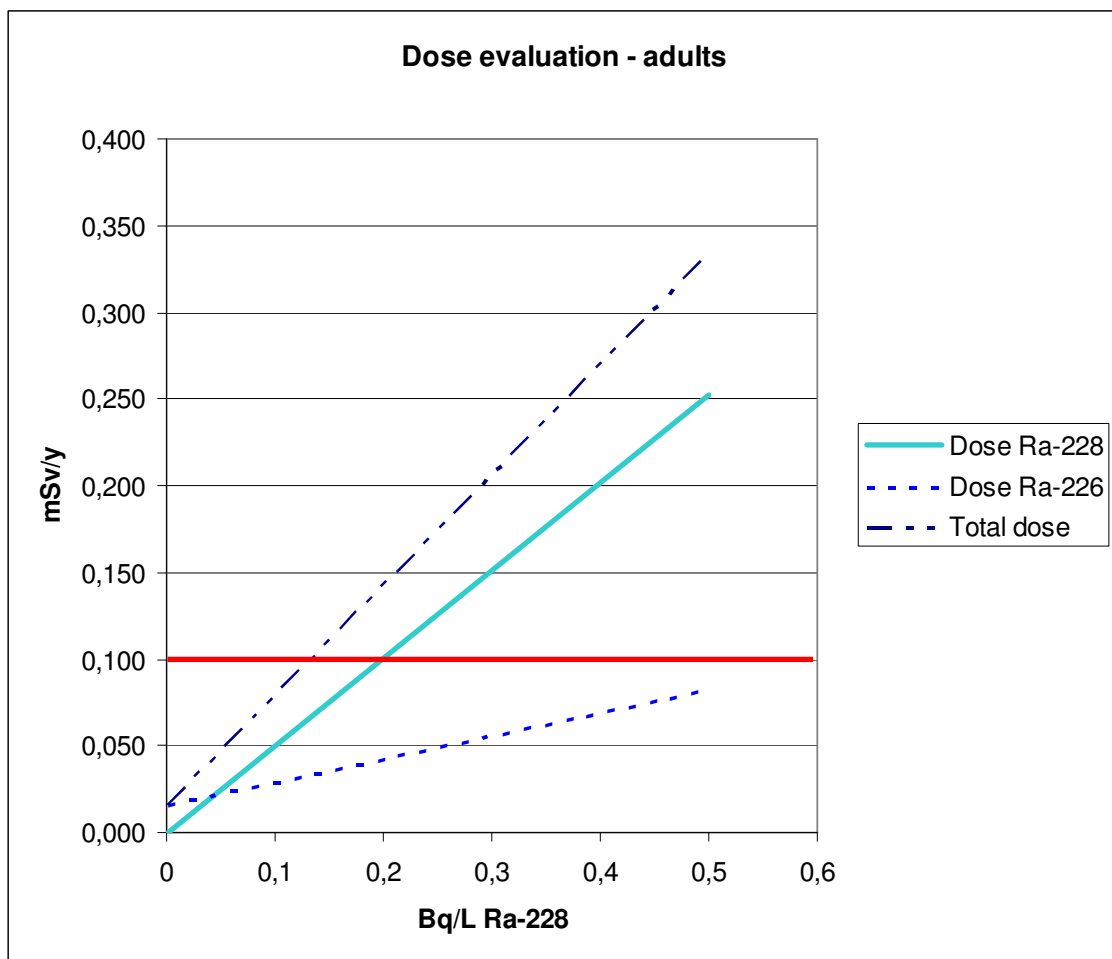


Fig. 3.12 Dependence of dose from Ra-228 concentrations

4. REMEDIAL ACTIONS

by Flavio Trotti and Franco Realini

The purposes of the present chapter are the following:

- To evaluate the necessary level of radium removal for the Estonian situation as can be inferred from the available data.
- To evaluate the radiological significance of effluents and residues from existing removal systems.
- To supplement the technical information exposed in chapter 2 of the previous report (Component 1 - April 2009).
- To supply information about the treatment costs.
- To underline some conclusions with regard to the possible applicable solutions to the Estonian situation.

4.1. The need of radium removal in Estonian water

In order to fix a goal for the required radium removal efficiency, a calculation of the removal percentage necessary to achieve the total indicative dose (TID) of 0.1 mSv/y was firstly performed. In Fig. 4.1 – 4.2 the distribution of the radium removal percentages are shown as a function of the analyzed wells.

As far as adult age class is concerned (Fig. 4.1), a rather high removal is needed to lower the TID to 0.1 mSv/y:

- With a radium removal of 70% compliant cases would be 64%.
- The removal percentage should raise up to 80% in order to achieve 92% compliant cases.

This gives a first indication on the requested performances of treatment plants since the chosen technique should assure a sufficient effectiveness (and related benefits) in order to justify the economic effort .

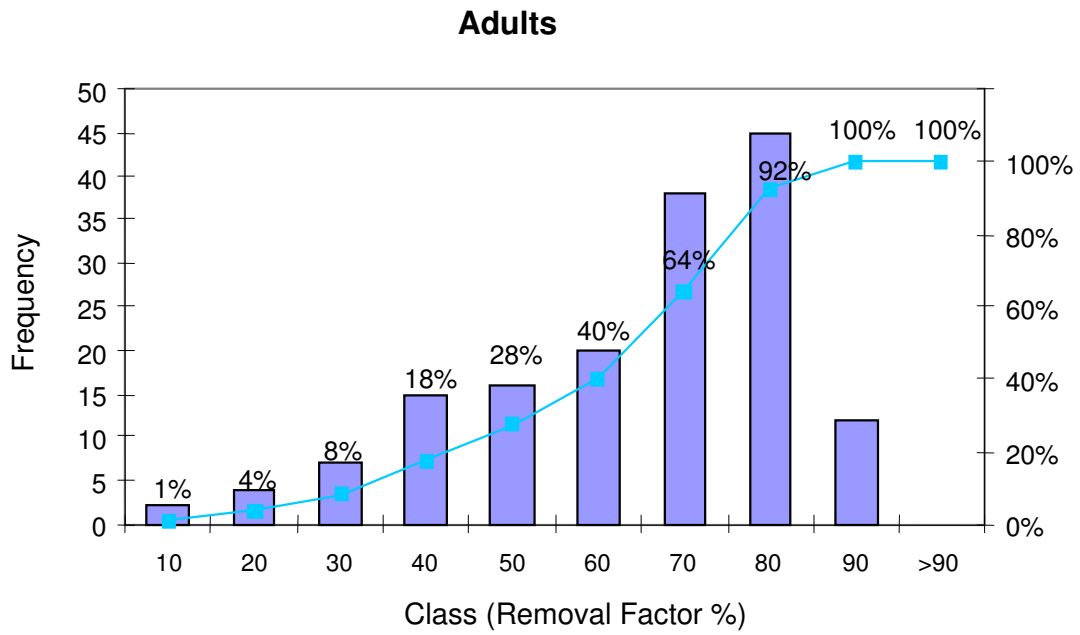


Fig. 4.1 *Distribution of the radium removal level to achieve the 0.1 mSv/y TID for the adults age class*

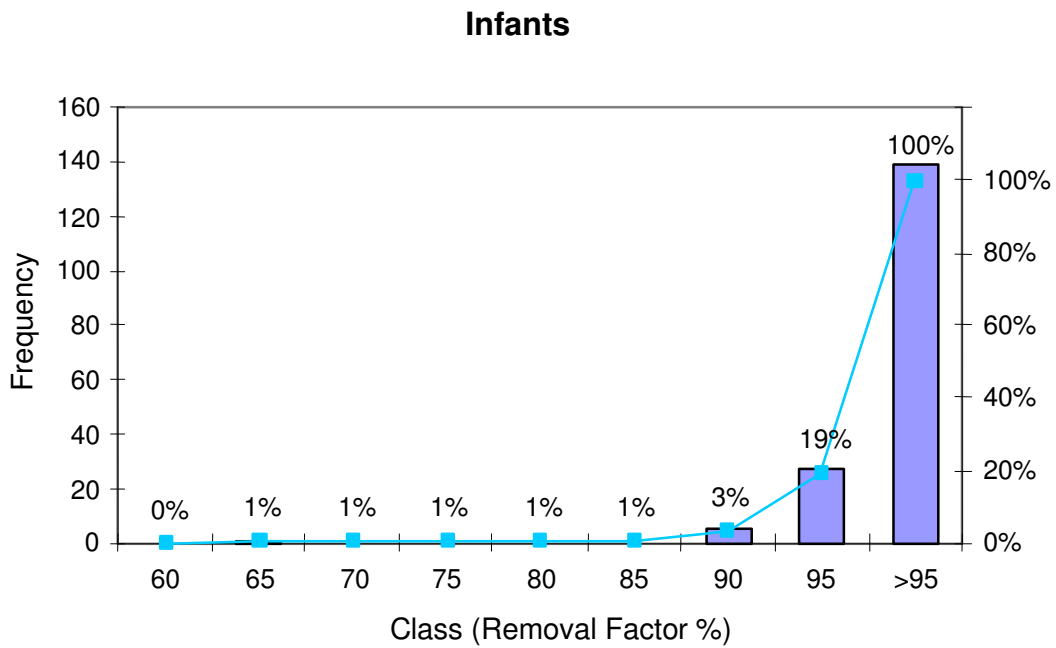


Fig. 4.2 *Distribution of the radium removal level to achieve the 0.1 mSv/y TID for the infants age class*

When the infant age class is considered (Fig. 4.2), the requirements are much higher. A rather complete radium removal (> 95%) would be needed to reach the fixed dose goal of 0.1 mSv/y as for adults. This narrows the choice of treatment technique to the most effective ones: reverse osmosis or ion exchange. Even though these techniques produce almost demineralized water, not best suited as drinking water and dangerous for the distribution system (problems of pipe corrosion), therefore this result in hardly achievable. Nevertheless we remind once more that the parameter value of 0.1 mSv/y is given only for adult age class in the EC Directive.

4.2. Removal efficiency for existing Estonian treatment plants

Some treatment plants are operating in Estonia at present although they are not devoted to radium removal. A brief description of Tallinn, Rakvere, Keila and Viimsi aqueducts which employ such plants is given:

Tallinn waterworks is the largest water company in Estonia and supplies water to about 400,000 people. Out of them, nearly 50,000 people get water from underground wells. As far as the groundwater network is concerned, 85 wells supply water to 56 pumping stations; 19 of them are supplied with treatment plants. Water is aerated and then filtered on sand and gravel to remove Fe, Mn and NH₄.

Keila waterworks supplies water to about 9700 people. Groundwater only, drawn from 4 wells, is delivered. Water is mixed before being collected in treatment tanks. Fe and Mn are then removed in a similar way as in Tallinn aqueduct.

Likewise Rakvere waterworks supplies 15,000 people by groundwater extracted from 5 wells.

Viimsi waterworks supplies 14,800 people by groundwater drawn from 35 independent wells and not mixed afterwards. All waters are extracted from Cambrian-Vendian layer (Vendian Voronka, Vendian Gdov or the mixture of both).

In some cases water are treated with the previously described sand filters (e.g. well n. 170, well n. 163), in other cases they are delivered without any treatment.

A small scale experimental device for high efficiency radium removal is working at Haabneeme pumping station. It is based on 1) aeration stage 2) filtration on sand and anthracite 3) filtration on zeolite.

In order to assess the effectiveness of radium removal by the above reported treatment plants, new radiochemical analyses, financed by the Twinning Project, have been carried out on the above described plants by Estonian Radiation Protection Centre and Tartu University. In only one case – Jugapuu Borehole, Tallinna Vesi analyses were performed by a Finnish laboratory. The results are shown in Table 4.1.

Table 4.1 Analysis of Ra-226 and Ra-228 in water before and after treatment for existing systems

Waterworks	Station/Well	Sampling	Ra-226 (Bq/L)		Ra-228 (Bq/L)		TID mSv/y
			Conc	Unc. (2 σ)	Conc	Unc. (2 σ)	
Tallinna Vesi	Jugapuu borehole	Before treatm.	0.34	n.a.	0.55	n.a.	0.35
		After treatm.	0.26	n.a.	0.35	n.a.	0.23
	Tiskre well, N°11031	Before treatm.	0.45	0.06	0.46	0.06	0.27
		After treatm.	0.35	0.05	0.41	0.05	0.23
	Toomse-Oitsu/well N° 223	Before treatm.	0.45	0.06	0.51	0.06	0.29
		After treatm.	0.47	0.07	0.51	0.03	0.29
	Raba/well N°225	Before treatm.	0.30	0.04	0.37	0.02	0.20
		After treatm.	0.62	0.09	0.62	0.09	0.36
Keila Vesi	Keila/well N° 552	Before treatm.	0.21	0.03	0.31	0.02	0.20
		After treatm.	0.17	0.03	0.23	0.03	0.17
Rakvere Vesi	Rakvere/well PK-2	Before treatm.	0.46	0.03	0.46	0.05	0.33
		After treatm.	0.49	0.04	0.45	0.04	0.33
Viimsi Vesi	Haabneeme/well 412	Before treatm.	0.34	0.05	0.45	0.07	0.30
		After treatm. (column 1)	0.05	0.01	0.09	0.04	0.06
		After treatm. (column 2)	1.95	0.29	2.70	0.09	1.76
		After treatm. (additional cleaning)	0.06	0.01	0.13	0.02	0.08

A decontamination factor, based on Table 4.1 data, has been computed for each station, given by:

$$DF (\%) = (Ra_b - Ra_a)/Ra_b \cdot 100$$

where Ra_b is the radium concentration before treatment and Ra_a is the radium concentration after treatment. Analogous relationship has been used for TID parameter.

In Table 4.2 all decontamination factors are reported. Raba pumping station (Tallinna Vesi) values were not considered because of anomalous conditions of the sample (filters had not been cleaned according to common frequency).

Viimsi values are reported separately since they refer to a small scale experimental technology.

Table 4.2 *Decontamination factors for existing treatment plants*

Waterworks	Treatment station	Laboratory	Ra-226 (Bq/l) D[%]	Ra-228 (Bq/l) D[%]	TID (mSv/y) D[%]
Tallinna Vesi	Jugapuu	Finland	24	36	34
	Tiskre	EE - Radiation Protection Centre	22	11	14
	Toomse-oitsu	EE - Radiation Protection Centre	-4	0	-1
Keila Vesi	Keila	EE - Radiation Protection Centre	19	26	15
Rakvere Vesi	Rakvere	EE - University of Tartu	-5	2	0
Average			11	15	12
Standard deviation			15	16	14
Viimsi Vesi	well 412 (*)	EE - Radiation Protection Centre	82	71	73

(*) additional cleaning after column I-II

Decontamination factors range from 0 to 30% with an average of 12 % removal. The high variability of results and the poor effectiveness of these plants in radium removal were expected (see Technical Report 1, 2.1). Viimsi plant, designed for radium remediation, exhibits promising performances but further confirmations on a larger scale plant are still needed.

4.3. Effluents and residues from existing treatment plants

4.3.1 Effluents and residues production data

Four of the waterworks involved in the project have treatment devices already working; data have been collected for a preliminary evaluation of the potential impact to the environment due to effluents and residues formed during treatment processes.

The ground water network of Tallinn waterworks consists of 85 wells afferent to 56 pumping stations; 19 of them are supplied with treatment systems. Water is filtered with sand and gravel to remove Fe, Mn and NH₄; an aeration stage precedes filtering. Periodically filters are cleaned by backwash water that is then channelized to sewage.

All backwash waters are conveyed to a single sewer (Tallinn city one) that serves about 400000 inhabitants; sludges from Tallin sewer are used as filling materials in landscape construction projects, purified water is released into sea. Radium content analysis results for such waters from four stations (Jugapuu, Raba, Toome-Oitse, Tiskre) are reported in Table 4.3, together with respective flows and discharge rates; total radium discharge rate from all Tallinn treatment stations based on average radium values were estimated and are also shown in Table 4.3.

Filtering material is replaced with low frequency (once in several years); this year Merivaljia pumping station filter has been removed and a radioactivity analysis on it has been carried out, the results of which are given in Table 4.4: it is worthwhile observing that, in filter material, Th-228 (followed by its decay products) also occurs, presumably due to growth during Ra-228 stay on filter.

In Keila waterworks, groundwater coming from 4 wells is mixed before being collected in treatment tanks where Fe, Mn removal techniques of the same type as those of Tallinn are applied.

Backwash water flow and radium activity concentrations, together with radium annual discharge rate, are presented in Table 4.3. Waters are conveyed to sewage and the produced sludge is not used in agriculture.

Two samples of filter material (sand) were analysed to assess Ra-226, Ra-228, Th-228 (same considerations as above) and the results are shown in Table 4.4.

In Rakvere waterworks, 5 groundwater wells deliver water to a treatment plant of the same kind as previous ones.

Backwash water is cleared to sewer; sewer sludge is used for farmland treatment; purified waters are released into a small river. Backwash water radioactivity concentrations, flow and radium discharge rate data are reported in Table 4.3.

As far as solid material is concerned, analysis results (Ra-226, Ra-228, Th-228) for filter material (sand) and for washing sediment are displayed in Table 4.4.

In Viimsi waterworks, water is drawn from 35 wells that are independent of each other.

Previous information from stakeholders refers that: one of the wells (n. 170) a sand filtering treatment plant operates (see Table 4.3), which is supposed to be of the same type as those described above, backwash water is channelized to sewage (no information on flow and radioactivity content is available); an experimental device for radium removal has been set at Haabneeme pumping station n. 2 (well n. 163), formed by two treatment stages, preaerated sand and anthracite filters followed by zeolite column, performed radioactivity analysis on backwash water are shown in Table 4.3 (although given as gross α and β activities), but no data on flow and fate have been diffused. This device has now stopped working.

From recent information there is evidence of a treatment device at well n. 412 (Katlamaja road), being formed by two parallel filtration columns followed by an additional common cleaning stage. Ra-226 and Ra-228 analysis data on waters used to wash the two column are presented in Table 4.3, together with information about flow and fate. Backwash water is conveyed to the same sewer as that of Tallinn waterworks.

Table 4.3 Information about effluents from treatment plants

Waterworks	Treatment station	Treatment type	backwash water (m ³ /y)	Ra-226	Ra-228	b.water fate	Ra-226	Ra-228	Laboratory
				b.water (Bq/m ³)	b.water (Bq/m ³)		b.water (Bq/y)	b.water (Bq/y)	
Tallinn	Jugapuu	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1708	3100	8700	sewer	5.29E+06	1.49E+07	Finland
	Merivälja	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1770			sewer			
	Norma	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	268			sewer			
	Pirita	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1115			sewer			
	Laagri	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	267			sewer			
	Sae	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	207			sewer			
	Pääsküla	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1278			sewer			
	Raba	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	507	8730	14710	sewer	4.43E+06	7.46E+06	Rad.Prot.Centre
	Toome - Seedri	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	742			sewer			
	Hiiu	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	2183			sewer			
	Kagu - Võidu	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1482			sewer			
	Kandle	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1545			sewer			
	Pika - Voolu	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1413			sewer			
	Toome - Õitse	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1018	9350	13650	sewer	9.52E+06	1.39E+07	Rad.Prot.Centre
	Tiskre	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	581	5320	6040	sewer	3.09E+06	3.51E+06	Rad.Prot.Centre
	Piiri	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1118			sewer			
	Mahla	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1213			sewer			
	Tule (Saue)	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1769			sewer			
	Segu (Saue)	sand/gravel filters for Fe, Mn, NH ₄ removal (preaerated)	1058			sewer			
		TOTAL		21242	6625	10775		1.41E+08	2.29E+08
Keila	all 4 wells	sand filters for Fe, Mn removal (preaerated)	1400	1380	1580	sewer	1.93E+06	2.21E+06	Rad.Prot.Centre
Rakvere	all 5 wells	sand filters for Fe removal (aerated)	36500	1774	1796	sewer	6.48E+07	6.56E+07	Tartu Univ.
Viimsi	well 170	sand filters				sewer			
	well 163	sand and anthracite filters (preaerated)+ zeolite column		1200 *	1200 *				
	well 412	I column (in parallel with II)	4.5	1240	1190	sewer	1.62E+04	1.85E+04	Rad.Prot.Centre
	well 412	II column (in parallel with I)	4.5	2370	2910				

* from gross α and β activity

Table 4.4 *Information about residues from treatment plants*

Waterworks	Treatm.station	Material	Ra-226 (Bq/kg)	Ra-228 (Bq/kg)	Th-228 (Bq/kg)	K-40 (Bq/kg)	Laboratory
Tallinn	Merivalja	Sand	8603	8681	5798		Tartu Univ.
Keila	All 4 wells	Sand filter 1	5524	5754	3817	40	Tartu Univ.
		Sand filter 2	5202	5618	3139	35	Tartu Univ.
Rakvere	All 5 wells	Sand filter	3788	3047	1768		Tartu Univ.
		Wash water sediment	20103	15034	7176	1054	Tartu Univ.

4.3.2 Clearance levels and discharge limits

Clearance levels

To check the adequacy of eliminating radioactive solid materials from the site of their production, clearance levels can be used. For the investigated cases (4 waterworks of previous paragraph), backwash water is conveyed straightaway to sewage without purification in sedimentation basins, thus apparently no sludge is formed in filter cleaning procedure (for waterworks in other regions such as Bavaria water sedimentation in basins is common); nevertheless one sample of backwash water sediment has been collected in Rakvere waterworks (see Table 4.4). Other waterworks in Estonia not involved in this project, could employ sedimentation processes after filter cleaning. Furthermore, filtering material in treatment devices is subject to replacement from time to time (see Table 4.4), so, again, clearance levels could help.

Radiation Protection 122 Part II (1) derives clearance levels for natural radionuclides in residues and waste from work activities involving NORM (in the present case, drinking water treatment is assumed to be such an activity). Residues with specific activity lower than the clearance levels can be reused, recycled, delivered for disposal with no constraint as far as their radiological aspects are concerned. Clearance levels are estimated considering different exposure scenarios (transport, storage, disposal, houses built with materials containing NORM residues,

...), involved persons (workers, population), exposure pathways (irradiation, inhalation, ...) and material type (waste rock, ash, ...): the most restrictive condition is selected and the specific nuclide concentration causing dose criterion (300 μ Sv/y individual effective dose) achievement is itself the clearance level (called “general” as an effect of its derivation). Table 4.5 shows these (rounded) levels. Where several nuclides are involved, single concentrations are normalized to respective clearance levels and the sum of all ratios (Sum Index) must be less than 1, for compliance.

Table 4.5 *Rounded General Clearance Levels (Bq/kg) from RP122 Part II (1)*

MATERIAL	²³⁸ Usec (*)	Unat (**)	²³⁰ Th	²²⁶ Ra (***)	²¹⁰ Pb (***)	²¹⁰ Po	²³² Thse c (*)	²³² Th	²²⁸ Ra (***)	²²⁸ Th (***)	⁴⁰ K
All materials	500	5000	10000	500	5000	5000	500	5000	1000	500	5000
Wet sludge from oil/gas industry	5000	100000	100000	5000	100000	100000	5000	100000	10000	5000	100000

(*) whole decay chain in secular equilibrium (²³⁵U series included in natural isotopic ratio to ²³⁸U);

(**) three uranium isotopes in fixed natural ratio with respective short half-life daughters;

(***) short half-life daughters in secular equilibrium

Values in table 4.5 are “general” clearance levels, meaning that their compliance warrants dose criterion respect independent of the specific situation. RP 122 itself allows less restrictive clearance levels, where computations based on specific (actual) scenarios demonstrate that these levels are compatible with dose criterion.

Discharge limits

Backwash water from treatment plants will contain a certain amount of natural radioactivity (namely, Ra-226 and Ra-228). Thus compliance with discharge limits should be checked when leaving waterworks. It is important to know fate of the water in order to select modeled scenarios and limits that suit best: in the present case, all 4 waterworks discharge purification water into sewage system.

Three documents by IAEA (2), NRPB (3) and European Commission (4) have been consulted. It should be stressed that IAEA and EC documents represent references at international level while NRPB is a national standard.

The IAEA Report (2)

The scope of the document is to give screening levels for radioactive substance discharges into air and water. A graded approach is suggested in which the complexity of dose assessments methods increases as subsequent reference levels are exceeded. The screening models contained in the report particularly fit situations involving discharges from small scale facilities (hospitals, research laboratories, ..) where the amount of radioactive effluent is expected to be low. Analytical models description are articulated in: estimation of radionuclides concentration in air and water (rivers, lakes, ocean coasts) following discharge, concentration in terrestrial and aquatic food, estimation of doses.

In the document (Annex I) a list of screening levels is shown: they are all calculated using methods and data presented in the body of the Safety report itself. Screening levels are given in terms of individual annual dose (critical group) for single unit radionuclide discharge amount, meaning that discharge limits are derived once individual dose criterion is established by the authority. Two sets of screening levels are proposed: the “no dilution” ones (extremely pessimistic hypothesis of individual exposure at the point of discharge) and the “generic environmental” factors (where standardized assumptions about discharge characteristics and location of the critical group are made).

In this report generic only environmental factors are being considered. Liquid discharge factors are presented for two conditions.

1. Liquid is discharged into a sewage system: plant worker exposure is accounted for due to external irradiation from radionuclides in the sludge and inhalation of radionuclides resuspended in air from sludge.

2. Liquid is discharged into a river: the modeled river is small ($0.1 \text{ m}^3/\text{s}$ flow), the critical group lives 500 m downstream from the discharge point and it is exposed through drinking water, freshwater fish ingestion and irradiation from shore/beach sediment.

In table 4.6, screening factors are shown for both scenarios; discharge limits are also inserted, based on the effective dose value of 0.3 mSv/y .

The choice of a dose criterion of 0.3 mSv/y is supported by the same option adopted in other technical reports of this branch (see, for instance, NRPB (3) and EC (4)

documents below described) and by the indication of EU BSS of such a figure as exemption level for public exposure for NORM work activities; actually, the new EU BSS proposal expressly includes in the positive list of work activities potentially affected by NORM the “ground water treatment”.

Table 4.6 “Generic environmental” screening factors and liquid discharge limits from IAEA (2)

Scenario	Ra-226 screening factor	Ra-226 discharge limit (Bq/y) *	Ra-228 screening factor	Ra-228 discharge limit (Bq/y) *
Discharge into sewage system	$1.6 \cdot 10^{-12}$ (Sv/y per Bq/y)	$1.9 \cdot 10^8$	$2.2 \cdot 10^{-12}$ (Sv/y per Bq/y)	$1.4 \cdot 10^8$
Discharge into river	$1.6 \cdot 10^{-5}$ (Sv/y per Bq/s)	$5.9 \cdot 10^8$		

* referred to 0.3 mSv/y

It should be remarked that actually in the IAEA Report, screening factors are explicitly given for Ra-226 only (not for Ra-228). To obtain Ra-228 factor in sewer exposure scenario, the following considerations have been done:

a) in the document, external irradiation dose coefficients are taken from EPA Federal Guidance (5), and has been calculated for each radionuclide taking into account the contribution of progeny grown up in a 30 y period of time: Ra-226, for instance, is considered to be in equilibrium with its decay products up to Po-210, and Th-232 is assumed in equilibrium with its progeny (Po-212 included). For Th-232 chain the radiation dose is due to gamma emitting daughters.

b) As far as worker inhalation pathway is concerned, the screening factor has been simply derived based on 29/96 Directive inhalation dose coefficient for Ra-228 (adults, most conservative absorption factor).

The NRPB document (3)

This document gives general derived constraints (GDCs) for discharge of Polonium, Lead, Radium and Uranium into atmosphere, river and sewage. GDCs refer to a single controlled source, they are set based on the critical group individual effective dose of 0.3 mSv/y and are particularly suitable for small (non-nuclear industry) users

discharging low levels of radioactivity in the environment. GDCs are intended for general application being based on a generic definition of discharge location and receiving environment. Actually, cautious assumptions are relied on for all modelled scenarios; nevertheless, site-specific assessments are encouraged in case of exceeding a significant fraction of GDC. For each radionuclide, GDCs are calculated for three age group individuals: infants (1 y), children (10 y) and adults (20 y); the most conservative of these situations is then taken as the adopted value. Where several radionuclides form the radioactive effluent, single discharge rates are normalized to respective GDC and the sum of all ratios (Sum Index) must be less than 1, for compliance.

Discharge to river. A small river has been considered ($1 \text{ m}^3/\text{s}$ flow), critical group individuals live 500 m downstream the discharge point and are exposed by irradiation and inhalation at river bank, by ingestion of river water and fish, by ingestion of vegetables grown on soil irrigated with river water. Short-lived progenies are assumed to be in secular equilibrium with parents for all GDCs; growth of long-lived progenies is accounted for in river bank and vegetable ingestion pathways. The river size selection is conservative because of moderate pollutants dilution.

Discharge to sewers. A sewer serving a small rural community (500 inhabitants) has been chosen, with a correspondent low raw effluent flow; this is a cautious assumption viewing at occurring dilution. Furthermore the document mentions studies indicating that doses from discharge of radionuclides to large urban works are lower than those from rural works. Three different scenarios are analyzed: release of treated effluent to river (having kept 100% radioactivity of raw effluent; same exposure pathways of point 1), the exposure of workers at treatment plant (100% radioactivity associated with sludge; irradiation from sludge, inhalation of resuspended sludge and inadvertent ingestion of it), use of sludges (having kept 100% radioactivity of raw effluent) in farm land; exposure pathways for this case are irradiation from treated soil, inhalation and inadvertent ingestion of soil, ingestion of animal food (milk and beef) from cattle grazing on treated land. The authors remark that land treatment is one of the most significant disposal route for sludge from small and medium rural sewage treatment works. Short-lived progenies are assumed to be in secular equilibrium with parents for all GDCs; ingrowth of long-lived progenies is accounted for river and land sludge use fate. The most conservative scenario (farm

land use of sludge) has been chosen for GDC setting; in this scenario, the critical exposure pathway is the ingestion of food coming from cattle grazing on treated land.

In Table 4.7 GDCs for release to rivers and sewer are shown. Beside numerical values, limiting age group is reported.

Table 4.7 *“Generalised Derived Constraints for release to rivers and sewers from NRPB (3)”*

Scenario	Ra-226 GDC (Bq/y)	Limiting age group
Discharge into sewage system	$1.0 \cdot 10^7$	First year ⁽⁺⁾
Discharge into river	$3.0 \cdot 10^9$	Adult

⁽⁺⁾ refers to infants in the first year of life on an all-milk diet

When comparing data of tables 4.6 and 4.7 for river discharges, some differences are revealed. Much more restrictive appears the level set for sewer by NRPB with respect to the analogue in IAEA report; the reason for that could be searched in the limiting scenario adopted by NRPB (farm land use of sludge) ignored by IAEA. Unfortunately, as in IAEA Report, data for isotope Ra-228 are lacking (as evident from table 4.7).

According to the document guideline, refinements of calculations could be carried out to reflect assumptions (less restrictive) more sticking to actual Estonian sewage management, in the event of waterworks discharge levels close to GDCs.

The European Commission (4)

This report is composed of three parts. The first concerns a review of NORM industries in the EU that produce radioactive effluents and wastes and the analysis of regulatory systems within EU Member States dedicated to the control of public exposure to such effluents and wastes (the analysis is based on questionnaires filled by MS). The second part aims at giving guidance for assessment of public exposure from NORM discharges (into atmosphere and water bodies). The last one proposes screening levels for discharges into air and water in order to identify situations requiring control.

The guidance for correct assessment of public exposure from NORM release is developed through general aspects description and detailed indications for air and water (rivers, sea) compartments presentation. Within general suggestions, the following items are remarked: instead of using single natural radionuclides data it is useful to consider natural decay chains as divided in segments, each one being composed by short-lived daughters following the long-lived nuclides; commonly, continuous release assumptions are acceptable; for NORM release, environmental monitoring cannot usually be used for public exposure assessments due to the presence of natural background, thus recourse to models is necessary in conjunction with empirical characterization of the source; attention is addressed to actual definition of critical exposure pathways, reference groups and environmental accumulation conditions.

Screening levels are calculated based on cautious assumptions such that compliance with them would certainly ensure compliance with the dose constraint. Basic step in deriving screening levels is the choice of the dose criterion: a dose criterion of 0.3 mSv/y is selected in the document for the individuals reference group, but a serious discussion is established about the possibility to refer to different (lower) values. Screening levels are given for the following compartments: atmosphere, rivers (small, medium and large size, with respective flow of 2.5 m³/s, 100 m³/s and 500 m³/s), marine (open sea and coastal sea) and they are presented in table 4.8.

Important features of screening levels derivation include: continuous release for 50 y, secular equilibrium of short-lived daughters with long-lived parents within chain segments, adult age group only considered, exposure pathways for river discharge being river water drinking together with fish ingestion and external irradiation from bank sediment, exposure pathways for marine discharge being ingestion of sea food (fish, molluscs, crustaceans) and external irradiation from beach sediment, PC Cream model used for river scenarios, Poseidon software (incorporating MARINA II modelling) for marine scenarios.

The report recalls the “summation index” procedure to be applied whenever several radionuclides are discharged simultaneously (single nuclides discharge rates are normalized to respective screening levels and the sum of all ratios must be less than 1 for compliance).

Table 4.8 *Screening levels for discharge into rivers and sea from RP 135 (4)*

Scenario	Ra-226 screening level (Bq/y) ⁽⁺⁾	Ra-228 screening level (Bq/y) ⁽⁻⁾	Critical pathway
Discharge into small river (2.5 m ³ /s)	7.5·10 ¹⁰	4.2·10 ¹⁰	Water ingestion
Discharge into medium river (100 m ³ /s)	1.4·10 ¹²	6.4·10 ¹¹	Fish ingestion
Discharge into large river (500 m ³ /s)	6.9·10 ¹²	3.2·10 ¹²	Fish ingestion
Discharge into coastal sea	2.2·10 ¹³	1.2·10 ¹³	Fish ingestion

⁽⁺⁾ all chain segment from Ra-226 to Po-214

⁽⁻⁾ equilibrium between Ra-228 and Ac-228

Levels in table 4.8 can be compared to analogous figures in tables 4.6 and 4.7 for river discharge. Table 4.8 levels are greater than the other ones also for the same reference situation (small river). NRPB document describes a more comprehensive scenario including pathways for ingestion of food grown on land irrigated with river water (not considered by other reports). Nevertheless, RP 135 data appear to be preferable for some reasons: levels are given both for Ra-226 and for Ra-228, levels are different for different river sizes to better fit actual conditions, refined calculations are based on sophisticated computer models, the document is the most recent one (2003); apart from those arguments, it is to be mentioned that “Radiation Protection” series represents a sort of primary technical standard for EU Member States.

For evident reasons RP 135 levels are to be selected as far as discharge to the sea is eventually of interest.

Reference to EU technical reports appears to be relevant for the study case as no waste/residues clearance levels nor discharge limits for natural radionuclides not

being used in practices are provided by the Estonian national legislation (Radiation Act, 2004).

4.3.3. Comparison of effluents and residues radioactivity content with international standards

Effluents comparison

Radium discharge rate from backwash water of Tallinn, Keila, Rakvere and Viimsi treatment systems (see Table 4.3) are compared to appropriate limits in Table 4.9.

For Tallinn, discharge rates referred to all 19 pumping stations are used (based on the average radium concentration values of available data).

Backwash waters from Tallinn and Viimsi treatment plants go to the same sewer (Tallinn city one), so their contribution has been added in order to make comparison with reference levels.

Some considerations about discharge limits selection. Waters from filter cleaning of Tallinn and Keila plants are bound to sewer, and sludge formed in process is not used in agriculture; recourse has been done in this case on limits of IAEA document for sewer (Table 4.6), that is coherent with the actual conditions. Rakvere backwash waters also go to sewage but the produced sludge is used in farmland treatment: sewer NRPB GDC (more restrictive) has then been used (Table 4.7), that takes this scenario as the most critical for discharge into sewer. For Tallinn, discharge limits have been scaled with respect to Table 4.6 figures to take into account the greater number of served inhabitants of Tallinn sewer than the one modelled in IAEA document (400000 vs 20000), corresponding to a larger amount of produced sludge and a proportional radioactivity dilution in it.

For Tallinn, being known that sewer purified waters are finally released to sea, the alternative conservative assumption, that all backwash water radium content is not associated with sludge and gets to sea, has been considered; comparison has then been carried out with RP 135 coastal sea discharge limits (Table 4.8).

Similarly for Rakvere, being known that sewer purified waters are finally released to small river, the alternative conservative assumption, that all backwash water radium content is not associated with sludge and gets to river, has been considered; comparison has then been carried out with RP 135 river discharge limits (Table 4.8).

Actually, Table 4.9 is composed of three tables: Table 4.9a and Table 4.9b summarize, to simplify reading, information about discharge limits (Table 4.8) and discharge rates (Table 4.3), respectively; in the Table 4.9c comparison in terms of the “sum index”, that is the sum of ratios of single nuclides activity to respective limit (it must be less than 1 for compliance) is shown.

Table 4.9a *Discharge limits used to assess compliance of backwash water from existing treatment plants*

Reference	Compartment	Ra-226 (Bq/y)	Ra-228 (Bq/y)	Critical pathway
IAEA 19	Sewer	$1.9 \cdot 10^8$	$1.4 \cdot 10^8$	Sewer workers
IAEA 19 (Tallinn) (*)	Sewer	$3.8 \cdot 10^9$	$2.7 \cdot 10^9$	Sewer workers
NRPB 13 n. 2	Sewer	$1 \cdot 10^7$		Sludge used for agriculture
RP 135	Coastal sea	$2.2 \cdot 10^{13}$	$1.2 \cdot 10^{13}$	Ingestion (fish)
RP135	Small river	$7.5 \cdot 10^{10}$	$4.2 \cdot 10^{10}$	

(*) Document levels scaled to account for Tallinn sewer served inhabitants number

Table 4.9b *Discharge rates from existing treatment plants*

Effluent	Waterworks	Ra-226 (Bq/y)	Ra-228 (Bq/y)
Backwash water	Tallinna Vesi	$1.5 \cdot 10^8$	$2.62 \cdot 10^8$
	Viimsi Vesi	$1.62 \cdot 10^4$	$1.85 \cdot 10^4$
	Keila Vesi	$1.93 \cdot 10^6$	$2.21 \cdot 10^6$
	Rakvere Vesi	$6.48 \cdot 10^7$	

Table 4.9c *Comparison between discharge rates and limits for washing waters of existing treatment plants*

Compartment	Waterworks	Sum index (*)	Reference
Sewer	Tallinna and Viimsi Vesi	1.37E-01	IAEA 19 (mod.)
Coastal sea	Tallinna and Viimsi Vesi	2.87E-05	RP 135
Sewer	Keila Vesi	2.60E-02	IAEA 19
Sewer	Rakvere Vesi	6.48	NRPB 13 n. 2
River (small)	Rakvere Vesi	2.42E-03	RP 135

(*) Sum Index is the sum of ratios of single nuclides activity to respective limit

Commenting results, compliance is revealed for backwash waters of Tallinn (although based, at the moment, on few analytical data) and Keila waterworks. For Rakvere, sum index threshold referred to sewer compartment is exceeded, due to both sewer sludge fate (farmland use) that requires more restrictive standards and elevated flow. Moreover Rakvere for sewer compartment compliance test has been carried out for Ra-226 only, NRPB 13 n. 2 document Ra-228 GDC being not available.

Residues comparison

Ra-226, Ra-228, Th-228 activity concentrations of solid residues associated with filtration systems (Table 4.4) have been compared to General Clearance Levels of RP 122 part 2 (Table 4.5) to check whether conditions for unrestricted clearance (whatever being the residues fate) do apply.

Comparison is carried out through the “sum index” estimate (see text above) and is shown in Table 4.10.

Table 4.10 *Comparison between activity concentrations of solid residues from existing treatment plants and GCLs of RP 122*

Waterworks	Treatment station	Material type	Sum index ^(*)
Tallinn	Merivalja	Sand	37
Keila	All 4 wells	Sand filter (sample 1)	24
		Sand filter (sample 2)	22
Rakvere	All 5 wells	Sand filter	14
		Wash water sediment	70

^(*) Sum Index is the sum of ratios of single nuclides activity to respective limit

All materials reveal not compliance. GCLs have been selected as the worst (most conservative) condition recorded in modelled exposure situations, as far as material type, reference scenario and population group are concerned: actually the adopted scenario, people living in a house made of building materials containing the radioactive residues, appears not reliable for the case we are discussing (solid residues from drinking water treatment plants); however, even if a different and more

suitable scenario is selected (such as workers exposure in road constructions with use of contaminated material or people living in a house close to the contaminated residues disposal site), compliance is not achieved.

As suggested by RP 122 part 2 itself, computations should then be performed based on specific (actual) scenarios to investigate whether less restrictive levels exist, compatible with dose criterion (specific clearance levels).

This preliminary analysis indicates, anyway, that a special care has to be taken in managing solid residues from drinking water treatment devices.

4.4. Summary of the information about removal techniques

Radium removal from drinking waters is not a technical problem. Actually, many radium removal methods are available, differing from each other for what complexity and removal effectiveness is concerned.

In Table 4.11 we summarize the most used techniques; for each of them we supply information about the removal effectiveness (if available), possible limitations and the main problems they may cause. Most of the data are taken from the US-EPA documents (6). Furthermore, the provided information especially refers to the so called Small System Compliance Technologies (SSCT), i.e. technologies that can be successfully applied in small plants.

Table 4.11 *Removal techniques: sounded methods*

Unit Technologies	Limitations	Operator Skill Level Required	Effectiveness %
Ion Exchange (IE)	a	Intermediate	Up to 95 – 99
Point of Use (POU) IE	b	Basic	-
Reverse Osmosis (RO)	c	Advanced	At least 90, up to 99
POU RO	b	Basic	-
Lime Softening	d	Advanced	40 - 80, up to 90
Green sand filtration	e	Basic	60 – 97
Co-precipitation with barium sulfate	f	Intermediate to advanced	40 – 90, up to 98
Electrodialysis/Electro dialysis reversal	N/A	Basic to intermediate	Up to 95
Pre-formed Hydrous Manganese Oxide filtration	g	intermediate	From 21 to 90 (h)

- a) Disposal options should be carefully considered before choosing this technology.
- b) Requires careful long-term operations, maintenance and monitoring plans to ensure proper performance.
- c) Reject water disposal options should be carefully considered before choosing this technology.
- d) Variable source water quality and complex water chemistry make this technology too complex for small water systems.
- e) Removal efficiencies can vary depending on water quality.
- f) This technology is most applicable to systems that have sufficiently high sulfate levels and that already have filtration in place.
- g) This technology is most applicable to small systems that already have filtration in place. The wide scattering in removal efficiency is due to difference in the experimental setup (optimum setup always assures high performances).
- h) Depending on the water quality.

In the following table (4.12) we provide information about more methods which have been described in the literature; some of them are quite new, while others are not extensively used.

Table 4.12 *Removal techniques: more methods*

Unit Technologies	Effectiveness %
Iron Based Technique (9, 10)	70
Zeolites (7, 11)	90 (in pilot plants up to 97%)
Granulated Active Carbon (12)	50-90, up to 97
Syntethic Clay (13)	N/A
Hydroxyapatite (7)	N/A

The previous cited STUK-A 169 report (7) provides an evaluation of the removal efficiency limited to the main available techniques (Table 4.13).

Table 4.13 *Removal techniques: TENAWA Report*

Equipment type	Effectiveness %
Aeration – filtration	3 – 93
Manganese based methods	56 – 97
Ion exchangers (cation resins)	69 - 99

4.5 Radium removal techniques: provisional cost evaluation

In the following some information is provided about the costs of different removal techniques.

In the report issued by the New Jersey Drinking Water Quality Institute (8) an evaluation of the costs for three of the most important radium removal technologies is given:

- Ion exchange
- Reverse osmosis
- Preformed hydrous manganese oxide.

The following table (4.14) summarizes the annualized cost, as a function of plant dimensions / population served.

Table 4.14 *Annualized cost estimates – An example from USA*

Size of Plant (m³/day) and Population served	Annualized cost* (Euro)
26.5 500 people	56,000 – 84,000 450 – 670 per person
132 2500 people	101,400 – 153,800 160 – 246
264 5,000 people	171,300 – 255,000 137 – 204
Greater than 264 10,000 people	> 245,000 17 – 25

*Annualized costs include debt service, capital recovery, maintenance and operation. The operating cost assumes that the radium may be disposed of as a soluble waste via sanitary sewers or septic system not requiring a radium selective complexer or resin.

According to this Report, one of the main factor affecting the choice of the most suitable technology is the production and management of radioactive wastes.

4.6 Conclusions

Five waterworks take part in this twinning project, Tallinna Vesi, Keila Vesi, Rakvere Vesi, Viimsi Vesi and Esmar Eithus. All these waterworks use groundwater (exclusively for Keila, Rakvere, Viimsi, Esmar Eithus, partially for Tallinn). Groundwater traditional treatment devices for the removal of iron and manganese are active in most of them (1 in Keila, 1 in Rakvere, 1 in Viimsi and 19 in Tallinn); for Viimsi an experimental system presumably dedicated to the mitigation of radium also operates.

Radiochemical analysis have been carried out on waters before and after treatment stage in several stations to check effectiveness in radium removal. Treatment techniques ordinarily used for Fe, Mn mitigation appear not so effective for radium: on average, the decontamination factor is slightly more than 10 % and the dispersion of data is elevated (of the same order of magnitude). Viimsi system shows an overall

good performance (decontamination factor of more than 70 %), but some critical stage within cleaning process seems to exist.

For Tallinn, Keila, Rakvere and Viimsi radium measurements have also been carried out on backwash water from some treatment stations, in order to preliminary assess the environmental compatibility of such effluents. Backwash waters from Tallinn, Rakvere and Keila waterworks are conveyed straightaway to sewers and sludge is produced which is used for agricultural purposes in Rakvere, but not in Tallinn and in Keila (specific information about Viimsi waterworks is not available thus the assessment has not been possible for it); after sewage processing, waters are released to sea in the case of Tallinn. Discharge data have been compared to reference levels taken from international technical standards. It comes out that compliance is revealed for backwash waters of Tallinn (although based, at the moment, on few analytical data) and Keila waterworks, but not for Rakvere due to both sewer sludge fate (farmland use) that requires more restrictive standards and elevated flow. Nevertheless, it should be pointed out that reference levels used for comparison are based on very conservative assumptions and calculations may be refined to better fit actual scenarios involved in backwash water fate.

For Tallinn, comparison has been developed providing both alternative assumptions that all backwash water radium content is binded in sewer sludge (as for Keila and Rakvere) and remains in water being released to sea.

Analysis of Ra-226, Ra-228 and Th-228 have also been carried out in solid materials sampled in some treatment plants of Tallinn, Keila, Rakvere waterworks: materials are filter sands and backwash water sediment. The purpose was, again, the estimate of the environmental compatibility once these residues are cleared from the plants. In this case, reference was to GCLs (general clearance levels) of RP 122 part 2 document. All materials reveal non compliance. It must be said that GCLs have been selected as the worst (most conservative) condition recorded in modelled exposure situations; as suggested by RP 122 part 2 itself, computations should then be performed based on specific (actual) scenarios to investigate whether less restrictive levels exist, compatible with dose criterion (specific clearance levels).

This preliminary analysis indicates, anyway, that a certain amount of care has to be taken in managing solid residues from drinking water treatment devices.

The above synthesized considerations about effluents and residues environmental compatibility concern ordinary iron-manganese treatment systems with low radium removal effectiveness; the situation could be somewhat worse for treatment plants with elevated radium mitigation effectiveness.

Radium removal from drinking waters is not a technical problem. Actually a number of methods for the radium removal are available, each of them with various complexity and different removal effectiveness.

Some aspects should be carefully considered:

- if the decision is to act with technological ways of radium removal (producing in any case some costs) then there is an inevitable charge on the water cost. This incidence is more or less important with regard to the selected technology.
- The plants for the radium removal involve problems of management and maintenance. In this case also the incidence is more or less important with regard to the selected technology. In a situation like in Estonia where there is a considerable presence of small plants scattered on the territory this aspect is very important.
- The radium removal produces some contaminated by-products with the consequent problems of disposal. These problems can be more or less complicated with regard to the chosen technology and to the concentration of radionuclides. Some techniques based on radium adsorption produce residues with high radioactivity content which should be disposed in suitable sites. On the opposite, techniques based on filtration and frequent backwashing are available: their drain exhibit low radioactivity content, and they might be acceptable also for common sewers.
- Not negligible also is the problem of the change in the quality of the produced water compared to that of the raw water. For instance the process based on Ion Exchange with resins and generally all the processes producing a

softened water are certainly very efficient from the radium removal point of view but the obtained water is practically a demineralized water, without useful minerals or poor of them. In similar situations it is possible to act with a “remineralization” of the water but with other costs and results in any case questionable from the organoleptic point of view.

Due to the above reported reasons and taking into account the Estonian situation it's our opinion that all the possible ways for the identification of the alternative sources of the drinking water supply have to be explored. Among them we can consider surface waters, groundwater from not contaminated layers and in particular and restricted situations rain water (as you can find in the south of Italy). In our opinion, facing the examined data, this is the first priority.

In this way we surely have a worsening of the organoleptic qualities of the supplied water, but the level of the radium contamination is not a negligible problem.

If we consider also the particular situation of the babies, pregnant women etc the above exposed assertions have to be intended in a drastic way.

4.7 References

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5. LEGISLATIVE ASPECTS AND PRINCIPLES FOR LIMITING RADIONUCLIDES IN DRINKING WATER

by Serena Risica

5.1. Introduction

Drinking water can contain significant concentration of radionuclides of natural origin, mainly uranium and thorium and their decay products. This is generally due to the geological features of the reservoirs and the solubility of the chemical compounds in which radionuclides are bound. However, radioactive pollution of water can also be due to discharges and/or byproducts released in the external environment by Naturally Occurring Radioactive Material industries, like oil and gas extraction, fertilizer production etc. Indeed, these discharges and byproducts generally contain high concentrations of uranium and thorium and their decay products, in high disequilibrium, as well as potassium 40.

Drinking water is an essential good for life in the world and for this reason it is of prime importance to guarantee, through careful monitoring, its quality. For this reason at the end of the 1990's the Council of the European Union issued a new Drinking Water Directive, which, for the first time, also included requirements for radioactivity.

5.2. The Council Directive 98/83/EC on the quality of water intended for human consumption

In December 1998, the *Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption* (1) was published in the Official Journal of the European Union. It was a revision of the Council Directive of 1980 (2) on the same issue, and had to be transposed by Member States into national laws within two years of its entry into force (i.e., December 2000).

It applies to (article 2)

- (a) "...all water, either in its original state or after treatment, intended for drinking, cooking, food preparation... whether it is supplied from a distribution network, from a tanker, or in bottles or containers..." and

- (b) “all water used in any food-production undertaking... unless the competent national authorities are satisfied that the quality of the water cannot affect the wholesomeness of the foodstuff in its finished form”.

It does not apply to (article 3)

- (a) natural mineral waters
- (b) waters which are medicinal products.

Therefore it does not apply to mineral water¹, but it applies not only to tap water, but also to bottled water and water used for food production.

Three types of parameters are set:

- (a) microbiological parameters (Annex I PART A)
- (b) chemical parameters (Annex I PART B)
- (c) indicator parameters (e.g. Cl, conductivity, odour, taste..) (Annex I PART C).

For the first time, on request of the European Parliament, the 1998 *Drinking Water Directive* also included requirements for radioactivity. But contrary to what was proposed by the Parliament, the Commission decided not to make these requirements mandatory (putting them in Annex I PART A or B), but only indicative (considering them among *indicator parameters* in Annex I PART C).

The indicator parameters for radioactivity are reported in table 5.1.

Table 5.1. *Indicator parameters for radioactivity in drinking water (1)*

Parameter	Parametric value	unit
Tritium	100	Bq/L
Total Indicative Dose	0.10	mSv/y

From the Total Indicative Dose (TDI) tritium, potassium 40², radon and radon decay products³ are excluded explicitly. For the assessment of the TDI the effective

¹ For radioactivity in mineral water up to now no regulation was issued by the European Commission.

² This is due to the well-known fact that K-40 does not accumulate in the body, but remains at a constant level independently on intake.

³ For radon and its decay products, some years later the European Commission issued a Recommendation (3) (see Addendum 1).

dose from a consumption of 1 year of drinking water should be considered, taking into account all natural and artificial radionuclides (except those listed above).

The indicator parametric value of 100 Bq/L for tritium activity concentration was not chosen with the same radiation protection base as the TDI. Indeed, it corresponds to a committed effective dose for adults of 1 μ Sv/y. This value was requested by the amendment of the European Parliament (see (7)) and it was judged that the adoption of a so low value was a cost-free action due to the absence of heavy water reactors in the European Union Member States at that time and therefore the very low possible tritium contamination of EU drinking water.

In article 5 the Directive prescribes that

1. "Member States shall set values applicable to water intended for human consumption for the parameters set out in Annex I...", and these values
2. "...shall not be less stringent than those set out in Annex I. As regards the parameters set out in Annex I, Part C, the values need be fixed only for monitoring purposes and for the fulfilment of the obligations" ...imposed in a successive article regarding remedial action and restrictions in use.
3. "a Member State shall set values for additional parameters not included in Annex I where the protection of human health within its national territory or part of it so requires..."

In article 8 "Remedial action and restrictions in use" it is stated that "In the event of non-compliance with the parametric values or with the specifications set out in Annex I, Part C, Member States shall consider whether that non-compliance poses any risk to human health. They shall take remedial action to restore the quality of the water where that is necessary to protect human health".

The reason of the different obligation for parameters of Part C is that *indicator parameters* (e.g. colour, smell, flavour, conductivity) do not imply a risk for human health in their own right, but were inserted in the Directive to be a timely signal of variations in water quality and of the possible need to adopt preventative actions to protect human health. Therefore, it may be concluded that both tritium concentration and TID should have a similar status, indicating a potential radiological problem when exceeded, and should not be regarded as limit values. Radioactivity, on the

other hand, imply a risk for human health and dose, in particular, is an expression of risk.

As regards radioactivity monitoring frequencies, monitoring methods and the most relevant locations for monitoring, they were to be set by the Commission within 18 months after the Directive's entry into force (1). These requirements, not yet published, should have suggested to Member States how to guarantee the required level of protection.

5.3. Calculation of the derived reference levels for drinking water (DWC)

In 1999, a working party of the Article 31 Group of Experts of the EURATOM Treaty was set up and worked in collaboration with a working party of the Article 36 Group of Experts of the EURATOM Treaty, in order to draw up a proposal for an environmental monitoring plan that would guarantee the level of radiological protection required by the Directive. The proposal was submitted to and approved by the Article 31 Group of Experts.

Since the beginning of the working party activities, it became clear that, in order to prepare a monitoring plan, a database was needed with derived activity concentrations for drinking water ingestion (DWC) for all radionuclides listed in Table A of Annex III of the EURATOM Basic Safety Standards (4), for different age groups of members of the public, on the basis of the appropriate ingestion dose coefficients. Under contract from the EU Commission, some experts of the *Istituto Superiore di Sanità* (Rome, Italy) performed the work and made the database available in a report (5) for possible use by health authorities and environmental radioactivity monitoring laboratories.

The DWC were evaluated using committed effective doses per unit intake via ingestion (Sv/Bq) for members of the public at different age groups, reported in the EURATOM Basic Safety Standards (4), and on the basis of the intake values chosen by the Article 31 working party. A decision on these values was taken after an overview of different intake values used by different organisations (EPA, ICRP, UNSCEAR, WHO). The chosen values are reported in table 5.2.

Table 5.2. Annual drinking water intake assumed for different age groups

Age group (years)	Annual intake (liter/year)
≤ 1	250
1-10	350
> 17	730

Calculations were carried out for the age classes ≤ 1y, (1 - 2)y, (2 - 7)y, (7 - 10)y, and >17y, whereas they were not for the age class (10 – 17)y, for there was no agreement on the related intake value.

The DWCs (Bq/L) in terms of activity concentration in drinking water were calculated using the following formula:

$$\text{DWC (Bq/L)} = \frac{1 \times 10^{-4} \text{ (Sv/y)}}{\text{Ann. intake value (L/y)} \times \text{Committed eff. dose per unit intake (Sv/Bq)}}$$

where 1×10^{-4} Sv/y = 0.1 mSv/y is the set Total Indicative Dose.

These DWCs were the base for suggesting a strategy for monitoring radioactivity in drinking water.

In the database (5) for each radionuclide, the committed effective doses per unit intake via ingestion and DWCs for the cited age classes are reported. For each nuclide the critical value of concentration (critical concentration, in Bq/L) and the corresponding critical age group were also identified and reported.

Some examples of DWCs and critical age groups are reported below, specifically in Table 5.3 for main radionuclides in a nuclear emergency (as reported in table VIII of (6)) and in Table 5.4 for the major radionuclides of natural origin which can cause a significant exposure of population. In the last column of the two tables the adult/critical age DWC ratio is also reported.

Table 5.3. *DWC for main radionuclides in a nuclear emergency for different age classes, critical concentration, critical age and adult/critical age DWC ratio*

Nuclide	Derived activity concentration in drinking water (Bq/L)					Critical concentr. (Bq/L)	Critical age (y)	Adult DWC/critical age DWC
	Age class (y)							
	≤ 1	1 - 2	2 - 7	7 - 10	> 17			
Sr-89	1.1E+01	1.6E+01	3.2E+01	4.9E+01	5.3E+01	1.1E+01	≤1	4.8
Sr-90	1.7E+00	3.9E+00	6.1E+00	4.8E+00	4.9E+00	1.7E+00	≤1	2.9
Zr-95	4.7E+01	5.1E+01	9.5E+01	1.5E+02	1.4E+02	4.7E+01	≤1	3.0
Ru-103	5.6E+01	6.2E+01	1.2E+02	1.9E+02	1.9E+02	5.6E+01	≤1	3.4
Ru-106	4.8E+00	5.8E+00	1.1E+01	1.9E+01	2.0E+01	4.8E+00	≤1	4.2
I-131	2.2E+00	1.6E+00	2.9E+00	5.5E+00	6.2E+00	1.6E+00	1 - 2	3.9
Te-132	8.3E+00	9.5E+00	1.8E+01	3.4E+01	3.6E+01	8.3E+00	≤1	4.3
Cs-134	1.5E+01	1.8E+01	2.2E+01	2.0E+01	7.2E+00	7.2E+00	>17	1.0
Cs-137	1.9E+01	2.4E+01	3.0E+01	2.9E+01	1.1E+01	1.1E+01	>17	1.0
Ce-144	6.1E+00	7.3E+00	1.5E+01	2.6E+01	2.6E+01	6.1E+00	≤1	4.3
Pu-238	1.0E-01	7.1E-01	9.2E-01	1.2E+00	6.0E-01	1.0E-01	≤1	6.0
Pu-239	9.5E-02	6.8E-01	8.7E-01	1.1E+00	5.5E-01	9.5E-02	≤1	5.8
Pu-240	9.5E-02	6.8E-01	8.7E-01	1.1E+00	5.5E-01	9.5E-02	≤1	5.8
Pu-241	7.1E+00	5.0E+01	5.2E+01	5.6E+01	2.9E+01	7.1E+00	≤1	4.1
Am-241	1.1E-01	7.7E-01	1.1E+00	1.3E+00	6.8E-01	1.1E-01	≤1	6.2

Table 5.4. *DWC for main radionuclides of natural origin for different age classes, critical concentration, critical age and adult/critical age DWC ratio*

Nuclide	Derived activity concentration in drinking water (Bq/L)					Critical concentr. (Bq/L)	Critical age (y)	Adult DWC/ critical age DWC
	Age class (y)							
	≤ 1	1 - 2	2 - 7	7 - 10	> 17			
Pb-210*	4.8E-02	7.9E-02	1.3E-01	1.5E-01	2.0E-01	4.8E-02	≤1	4.2
Po-210*	1.5E-02	3.2E-02	6.5E-02	1.1E-01	1.1E-01	1.5E-02	≤1	7.3
Ra-226	8.5E-02	3.0E-01	4.6E-01	3.6E-01	4.9E-01	8.5E-02	≤1	5.8
Ra-228	1.3E-02	5.0E-02	8.4E-02	7.3E-02	2.0E-01	1.3E-02	≤1	15.4
Th-228	1.1E-01	7.7E-01	1.3E+00	1.9E+00	1.9E+00	1.1E-01	≤1	17.3
Th-230	9.8E-02	7.0E-01	9.2E-01	1.2E+00	6.5E-01	9.8E-02	≤1	6.6
Th-232	8.7E-02	6.3E-01	8.2E-01	9.9E-01	6.0E-01	8.7E-02	≤1	6.9
U-234	1.1E+00	2.2E+00	3.2E+00	3.9E+00	2.8E+00	1.1E+00	≤1	2.5
U-235	1.1E+00	2.2E+00	3.4E+00	4.0E+00	2.9E+00	1.1E+00	≤1	2.6
U-238°	1.2E+00	2.4E+00	3.6E+00	4.2E+00	3.0E+00	1.2E+00	≤1	2.5

*The calculations were also carried out for Pb-210 and Po-210, which are excluded from the scope of the Directive, as specified above. On this issue the EU Commission issued at a later stage a Recommendation (see Addendum 1)

° For uranium in drinking water see Addendum 2

It must be stressed that the calculated DWCs have not the meaning of derived limits, for the choice, cited above, to set TDI as an indicator parameter of a potential radiological risk.

Observing the two tables, it can be noted that, Cs isotopes excluded, the population groups most at risk are infants (≤ 1 year) and small children (1 – 2) year, notwithstanding the lower annual intakes. Similarly, in the complete database (5) except for 5 radionuclides (Zr-93, I-129, Cs-134, Cs-135 and Cs-137), infants (≤ 1 year) and small children (1 - 2 year) are the population groups most at risk.

As regards the adult/critical age DWC ratio it ranges from 1 (for the Cs isotopes) to 17.3. In the complete database (5) it ranges from 1 to 24.6, mean value 4, standard deviation 2.3. It can be concluded that in case of a prolonged contamination of drinking water – particularly for some radionuclides – the respect of the DWC for adults could expose infants and small children to a dose much higher than the TID.

In particular, for Ra-226 and Ra-228, this calculation was made not only for the critical concentration (see table 5.4: the critical concentration corresponds to infants less than one year old), but for all age classes, to give a clear and easy picture of the higher doses that could be received by infants and children of different age classes, when the TID of 0.1 mSv/y is complied with.

Table 5.5 *Multiplicative factors which show the higher doses that can be received by infants and children when the TDI is complied with*

Nuclide	Adult DWC / different age class DWC				
	Age class (y)				
	≤ 1	1 - 2	2 - 7	7 - 10	> 17
Ra-226	5.8	1.6	1.1	1.4	1.0
Ra-228	15.4	4.0	2.4	2.7	1.0

Lastly, the age class (10 – 17) y, not considered in the database (5) for the reasons reported above, is also critical - even if not the most at risk - for some radionuclides (e.g. Ra-226 and Ra-228).

In any case the overcoming of one of the critical concentrations does not imply necessarily the overcoming of the Total Indicative Dose, because the critical concentration was calculated for an annual intake. On the other hand, if the drinking water contamination is due to more than one radionuclide, the TID could be overcome even without the overcoming of one of the critical concentrations, because the total committed effective dose would be the sum of the single doses.

5.4. Comparison of the European Directive with WHO and EPA regulations for drinking water

The Guidelines for Drinking water Quality of the World Health Organization

Unlike the European Union, the World Health Organization (WHO) issued over the years several legal provisions on radioactivity in drinking water. The most recent one, i.e. the third edition of *Guidelines for Drinking-water Quality* (8), was issued in 2004. The guidelines apply "...to routine ("normal") operational conditions of existing or new drinking water supplies. They do not apply to a water supply contaminated during an emergency involving the release of radionuclides into the environment", but they "... also apply to radionuclides released due to nuclear accidents occurred more than 1 year previously".

The approach taken for controlling radiological hazards is said to have two stages:

- "initial screening for gross alpha and/or beta activity to determine whether the activity concentrations (in Bq/L) are below levels at which no further action is required; and
- if these screening levels are exceeded, investigation of the concentration of individual radionuclides and comparison with specific guidance levels".

A *recommended reference dose level* (RDL) of the committed effective dose, from 1 year's consumption of drinking water, is stated equal to 0.1 mSv/y. K-40 is excluded and radon 222 is discussed separately.

Specific guidance levels are calculated based on the RDL, assuming that the annual volume of drinking water ingested by adults is 730 L/y and using dose coefficients for adults provided by ICRP (9), which were adopted by the EU Basic Safety Standards (4). The similarity of approach with the Directive 98/83 is not surprising, because the same approach had been already adopted by WHO in the

previous edition of the guidelines (10), which were used by the *Article 31 working party* as a basis for the elaboration of the EU proposal for the monitoring plan. However, in WHO's recent document guidance levels "...are rounded according to averaging the log scale values (to 10^n if the calculated value was below 3×10^n and above $3 \times 10^{n-1}$)". In Tables 5.6 and 5.7 the results of these calculations are reported for main radionuclides in a nuclear emergency and for main radionuclides of natural origin, respectively.

Table 5.6. *Guidance levels for main radionuclides in a nuclear emergency*

Nuclide	Guidance level (Bq/L)
Sr-89	100
Sr-90	10
Zr-95	100
Ru-103	100
Ru-106	10
I-131	10
Te-132	100
Cs-134	10
Cs-137	10
Ce-144	10
Pu-238	1
Pu-239	1
Pu-240	1
Pu-241	10
Am-241	1

Table 5.7. *Guidance levels for main radionuclides of natural origin*

Nuclide	Guidance level (Bq/L)
Pb-210	0.1
Po-210	0.1
Ra-226	1
Ra-228	0.1
Th-228	1
Th-230	1
Th-232	1
U-234	10
U-235	1
U-238*	10

* The provisional guideline value for uranium in drinking water is 15 µg/L (187 mBq/L), based on its chemical toxicity for the kidney (8).

The guidelines state that “the higher age-dependent dose coefficients calculated for children (accounting for the higher uptake and/or metabolic rates) do not lead to significantly higher doses due to the lower mean volume of drinking water consumed by infants and children”. This statement, as shown above, is not that convincing because the lower mean volume of drinking water consumed by these age classes does not compensate for the effect of higher dose coefficients (5). In order to stress the point, in Tables 5.8 and 5.9 WHO guidance levels are compared with those obtained from the DWCs for the critical age, rounded according to the same rule (according to averaging the log scale values). It can be noticed that for most radionuclides (highlighted in bold in the tables) the guidance value for the critical age (generally, as reported above, infants (≤ 1 year) and small children (1 – 2) year), is one order of magnitude lower.

Screening levels for drinking water, below which no further action is required, are set at: 0.5 Bq/L for gross alpha activity and 1 Bq/L for gross beta activity. The contribution of K-40 to beta activity should be subtracted following a separate determination of total potassium. The gross alpha activity screening level in the previous editions of the guidelines was 0.1 Bq/L and was increased to 0.5 because they claimed that “...this activity concentration reflects values nearer the radionuclide specific guidance RDL”.

However, as discussed above (see paragraph 3.1), when e. g. radium isotopes are the prevailing radioactive components, the suggested screening levels may be not enough protective, not guaranteeing the compliance with the *recommended reference dose level* of 0.1 mSv/y.

Table 5.8. WHO guidance levels and guidance levels calculated for the critical age for main radionuclides in a nuclear emergency

Nuclide	Guidance level (Bq/L)	Guidance level based on the critical conc. (Bq/L)
Sr-89	100	10
Sr-90	10	1
Zr-95	100	100
Ru-103	100	100
Ru-106	10	10
I-131	10	1
Te-132	100	10
Cs-134	10	--
Cs-137	10	---
Ce-144	10	10
Pu-238	1	0.1
Pu-239	1	0.1
Pu-240	1	0.1
Pu-241	10	10
Am-241	1	0.1

Table 5.9. WHO guidance levels and guidance levels calculated for the critical age for main radionuclides of natural origin

Nuclide	Guidance level (Bq/L)	Guidance level based on the critical conc. (Bq/L)
Pb-210	0.1	0.1
Po-210	0.1	0.01
Ra-226	1	0.1
Ra-228	0.1	0.01
Th-228	1	0.1
Th-230	1	0.1
Th-232	1	0.1
U-234	10	1
U-235	1	1
U-238*	10	1

*The provisional guideline value for uranium in drinking water is 15 µg/L (187 mBq/L), based on its chemical toxicity for the kidney (8).

The National Primary Drinking Water Regulations of the Environmental Protection Agency (EPA)

The Environmental Protection Agency (EPA) also issued some regulations in the past. The most recent one, the National Primary Drinking Water Regulations (11) was issued in December 2000 and became effective in December 2003. The previous rule had been promulgated in 1976, but in 1991 EPA initiated a wide consultation phase regarding a number of proposed changes and additions to the previous rule. The basis for the final regulatory decisions was the new information that became available as from the 1991 proposal, in particular a new study (12) providing the numerical factors used in estimating cancer risks from low-level exposures to radionuclides.

This regulation is only applicable to community water systems and it "...finalises *maximum contaminant level goals* (MCLGs), *maximum contaminant levels* (MCLs), and monitoring, reporting and public notification requirements for radionuclides". As regards the MCLGs, they are defined as "non-enforceable health-based target" and it is stated that "the final MCLGs are zero for all radionuclides, based on the no-threshold cancer risk model for ionizing radiation." The maximum contaminant levels are defined as "enforceable regulatory limits" and are reported in Table 5.10 in SI units. No specific value of activity concentration for other radionuclides is given.

Table 5.10. *EPA maximum contaminant levels*

	Maximum Contaminant Level
Gross alpha (excluding Rn and U)	555 mBq/L
Beta particle and photon radioactivity	40 μ Sv/y
Ra-226 + Ra-228	185 mBq/L
Po-210	included in gross alpha
Pb-210	explicitly not regulated
Uranium	30 μ g/L

As regards children exposure, EPA claims that "...the Agency does have reason to believe that radionuclides in drinking water present higher unit risks to children than to adults, since there is evidence that children are more sensitive to radiation than adults. Because of this, we have explicitly considered the risks to children in evaluating the lifetime risks associated with the current MCLs...In summary, today's decision to retain the current more stringent MCLs for radionuclides and to establish an MCL for uranium in drinking water is consistent with the protection of children's health."

May be this is the reason why EPA requirements are generally much stricter than EU and WHO requirements.

Discussion

The EPA approach is quite different from those of EU and WHO, because the suggested MCLs are neither *indicator parameters* aimed at assessing the quality of drinking water (1), nor *guidance levels* that, if exceeded, "...should be regarded as an indication that further investigation... is needed" (8), but actual limits. Taking into account this important difference, it can be noticed that:

- the gross alpha MCL is almost the same of the value suggested by WHO, but in the former case is a limit, in the latter one only a screening level "below which no further action is needed"
- the MCL for beta particles and photon radioactivity is less than half the TID (1) or *the recommended reference dose level* (8)
- the MCL for the sum of Ra-226 and Ra-228 is much lower or lower than those of the single radionuclides in the European Directive (DWC for adults 0.5 Bq/L and 0.2 Bq/L, respectively) and presumably than that of WHO (an additive formula of the two concentrations weighted by the two guidance levels, that is 1 Bq/L and 0.1 Bq/L, respectively)
- a specific MCL for Po-210 is not provided for, because Po-210 is included in gross alpha; however, Po-210 is excluded by the European Directive (but, as reported above, a proposal for its limitation was issued in the 2001 Euratom Recommendation (3)), but included in the WHO guidance levels (with a value of 0.1 Bq/L)

- Pb-210 is explicitly not regulated by EPA, excluded by the European Directive (but, as for Po-210, discussed in (3)), and included in the WHO guidance levels (with a value of 0.1 Bq/L).

In conclusion EU, WHO and EPA regulations on the content of radionuclides in drinking water look quite different, even if two of them seem to start from the same goal of limiting the population exposure to less than 0.1 mSv/y.

As reported above, the practical application of the European Directive calls for the issuing of "... detailed requirements on monitoring frequencies, methods and locations" (see notes to Annex I PART C). As soon as they will be available the comparison and discussion about the approaches chosen by these international authorities could be extended to these aspects.

5.5. Radiological protection conclusions

From the above discussion it is all too clear that in case of radioactive contamination of drinking water infants and small children are the population groups most at risk. This means that in case of prolonged contamination the derived activity concentration in drinking water calculated for adults might not be conservative enough for these age classes. This means that great caution should be used when implementing the derived reference levels in drinking water for these age classes. As far as we know, no recommendation or regulation specific for infants or children has ever been issued at the international level, whereas at the national level two examples of a specific attention to these age classes can be reported: in Germany a regulation concerning radium 226 and 228 in drinking and mineral water was issued in 2006 (13) and in Italy some recommendations suggested by the Istituto Superiore di Sanità (14) were adopted by the Italian Minister of Health (but for mineral and spa water).

However, it should be kept in mind that the total indicative dose of 0.1 mSv/y is only reached if the contamination lasts for one entire year.

As regards the implementation of the European Directive (1) two general criteria should be kept in mind: the fact that TDI is an indicator parameter, not a limit, and the obligation (article 8, see above) of taking remedial actions "...to restore the

quality of the water where that is necessary to protect human health” in case of non-compliance with the parametric values set out in Annex I, Part C.

Moreover, the feasibility of the possible remedial actions should also be carefully analysed (see Addendum 3).

In conclusion, being drinking water a primary need, taking into account the above cited conclusions, a thorough risk-benefit analysis should be done before deciding any limitation to its use.

5.6. Addendum 1 - The Commission Recommendation of 20 December 2001 on the protection of the public against exposure to radon in drinking water supplies

The Directive 98/83/EC, as specified above, excluded radon and its decay products. However, there are circumstances under which polonium 210 and lead 210 in drinking water pose a comparable or a higher radiation risk than some natural radionuclides monitored in accordance with the Directive. For this reason in year 2001 the European Commission issued a recommendation devoted to this issue.

Similarly to the Directive 98/83 (1), the 2001 Recommendation (3) applies to

- (a) all water intended for drinking, cooking, food preparation or other domestic purposes, regardless of its origin and whether it is supplied from a distribution network, from a tanker, or in bottles or containers
- (b) all water used in any food-production undertaking.

It does not apply to:

- (a) natural mineral water
- (b) water which are medical products.

It recommends to undertake representative surveys to determine the scale and nature of exposures caused by radon and long-lived radon decay products in domestic drinking water supplies originating from different types of ground water sources and wells in different geological areas. In particular, attention should be paid to drilled wells, especially those in crystalline rock areas, and waterworks using rock or soil aquifers.

The recommendations are different in case of water supplied as part of a commercial or public activity and an individual water supply.

In the former case, the following actions should be taken

- above 100 Bq/L Member States should set a reference level for radon
- a level > 100 Bq/L may be adopted if national surveys show it is necessary for implementing a practical radon programme
- above 1000 Bq/L remedial action is deemed to be justified on radiological protection grounds
- radon concentration measurements should be required if there is a specific reason to suspect that the reference level might be exceeded

- where significant concentration of polonium 210 and lead 210 are suspected monitoring should be arranged
- above a reference level of 0.1 Bq/L for polonium 210 and 0.2 Bq/L for lead 210 consideration should be given to whether remedial action is needed to protect human health.

On the other hand, for an individual water supply, the following action should be taken

- a level of 1000 Bq/L for radon should be used for consideration of remedial action
- the urgency of the remedial action should be commensurate with the extent to which the reference concentration. is exceeded
- where remedial action is considered necessary because of radon, the levels of other natural radionuclides should be screened and the consumers concerned should be informed of the radon levels and the remedies available.

Drinking water distributed in public premises such as residential homes, schools, and hospitals should comply with the principles for water supplied as part of a commercial or public activity and measurements should be made with appropriate methods and equipment which have undergone approved calibration and quality assurance programmes.

Member States are recommended to

- concentrate their attention on the highest exposures and on those areas where action is most likely to be effective
- provide guidance on different methods available for removing radon and long-lived radon decay products from water
- provide instructions on the handling and disposal of radioactive waste generated by the removal process
- provide adequate public information.

Lastly, possible exposure of workers in waterworks, spas and swimming pools should be controlled according to the Title VII of the Directive 96/29/EURATOM.

5.7. Addendum 2 - The Uranium in drinking water and its limitation

1. The Approach of the European Directive

In the European Directive 98/83 uranium, notwithstanding its chemical toxicity, is not considered among parameters in Annex I, Part B, but, due to its radioactivity, it should be taken into account in the TID assessment. However, the derived activity concentrations calculated starting from the TDI (see table 5.4) are meaningless, because, as it is well known, uranium is much more chemotoxic than radiotoxic.

2. The WHO Approach

A few years ago, WHO set a provisional guideline value for uranium in drinking water of 2 µg/L (15), obtained by means of the Tolerable Daily Intake (TDI) of 0.6 µg/kg of body weight, assuming a 60-kg adult consuming 2 litres of drinking water per day and a 10% allocation of the TDI to drinking water. The 10% allocation was justified by the fact that the greatest part of the daily intake of uranium was shown to come from food. The TDI was derived using the LOAEL (lowest-observed-adverse-effect level) of 60 µg/kg of body weight per day and an uncertainty factor of 100 (for intra-and interspecies variation).

It was specified that TDI "...is an estimate of the amount of a substance in food or drinking water, expressed on a body weight basis (mg/kg or µg/kg of body weight) that can be ingested daily over a lifetime without appreciable health risk TDIs ... are not so precise that they cannot be exceeded for short periods of time. Short-term exposure to levels exceeding the TDI is not a cause for concern, provided the individual's intake averaged over longer periods of time does not appreciably exceed the level set ... However, consideration should be given to any potential acute toxic effects that may occur if the TDI is substantially exceeded for short periods of time" (16).

In the last edition of the Guidelines (8) a new provisional guideline value for uranium in drinking water of 15 µg/L is suggested, again based on its chemical toxicity for the kidney. It is stressed that it is still a provisional guideline value, because of "outstanding uncertainties regarding the toxicology and epidemiology of uranium as well as difficulties concerning its technical achievability in smaller supplies".

Looking into the details of the relevant discussion, it can be noted that all the values of the parameters used in the former guidelines for this choice remained unchanged, except one. That is: a TDI of 0.6 µg/kg of body weight per day, based on the application of an uncertainty factor of 100 to a LOAEL of 60 µg/kg of uranium per kg of body weight per day and an adult of 60-kg, with a consumption of 2 litres/day. But the allocation to water is now set at 80% of TDI, maintaining that “the data on intake from food in most areas suggest that intake from food is low and support the higher allocation to drinking water”. Moreover, the old provisional value is called a “health-based guideline value” and it is said “This guideline value was designated as provisional, because it may be difficult to achieve in areas with high natural uranium levels with the treatment technology available and because of limitations in the key study. It was noted that several human studies are under way that may provide helpful additional data”.

3. The EPA Approach

As uranium is radioactive, EPA, which uses a non-threshold linear risk model for ionising radiation, set the MCLG (non-enforceable health-goal) for this contaminant at zero. The Safe Drinking Water Act requires EPA to set the MCL as close to the MCLG as is feasible. In 1999 EPA proposed a feasible level of 20 µg/L, determining that uranium may be treatable and quantifiable at levels below 20 µg/L, however levels below were not considered feasible under the Safe Drinking Water Act.

In its most recent regulation (11), as already shown in table 5.10, EPA set a maximum contaminant level of 30 µg/L for uranium, that is “protective of both kidney toxicity carcinogenicity”. EPA believes the feasible level be still 20 µg/L, but maintains that “EPA selected a less stringent MCL for uranium of 30 µg/L by invoking the discretionary authority for the Administrator to set an MCL less stringent than the feasible level if the benefits of an MCL set at feasible level would justify the costs. As a result, fewer water systems will be in violation of the uranium MCL, reducing the number of systems that may face radioactive waste disposal issues, and resulting in the ability of a higher percentage of water systems to use on-treatment options for achieving compliance (e:g. new wells, blending of water sources, modifying existing operations, etc.)”

This MCL is a factor of 2 higher than the one suggested by the most recent WHO guidelines.

4. Conclusions

All the cited International and National Organisation agree on the fact that uranium in drinking water should be limited more strictly for its chemical toxicity than for its radioactivity. However, there is no agreement on a unique value to be set due to both the still elevated uncertainties in the assessment of its toxicity and the feasibility of remedial actions.

5.8. Addendum 3 - ICRP and the limitation of natural radioactivity

The International Commission on Radiological Protection (ICRP) is “an independent non-governmental organisation”, established “to advance for the public benefit the science of radiological protection”. It “provides recommendations and guidance on protection against the risks associated with ionising radiation, from artificial sources widely used in medicine, general industry and nuclear enterprises, and from naturally occurring sources”. These reports and recommendations provide in-depth coverage of specific subject areas and are the base of both EURATOM Directive and IAEA Basic Safety Standards (17).

ICRP starting from its 1977 Recommendation (18) gave emphasis on the exposure of mankind to natural radioactivity, recognising that “there may be levels of natural radiation which might have to be controlled, to the extent practicable, in much the same way as for artificial sources”.

However, it is only in 1984 that ICRP began to give practical guidance on the principles for such control - starting from radon concentration in dwellings and at work – introducing the concept of *remedial action*. It maintained that “in deciding whether to take action, the hazard or social cost involved in any remedial measure must be justified by the reduction of risk that will result” and “the process of deciding how far to go with remedial actions... should involve a process similar to that of the optimization of protection” (19).

In the following 25 years several recommendations or statements on natural radioactivity were issued by ICRP and its philosophy in the field has been evolving, but the major concept of radiological protection in this field is still the feasibility of remedial actions.

5.9. References

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6. CONCLUSIONS

From the above report, some provisional conclusions can be drawn on following topics:

Representativeness and completeness of available data:

In Estonia a large majority of water supply zones is small (more than 80% serves less than 500 people) and consequently the number is relatively high. Obviously this makes it more difficult for complete monitoring of water resources.

Nevertheless, it can be observed that most of the data are collected in coastal counties and are referred to Cambrian-Vendian aquifer.

If it is true that generally higher values are found in Cambrian-Vendian waters, radiation protection problems cannot be completely excluded for other aquifers, thus a better knowledge of their radioactivity content is desirable. This is of paramount importance if age classes out of adults are considered.

When dose coefficients for infants are applied, even waters from “safer” aquifers (non Cambrian-Vendian) often give rise to rather high radiological exposures.

A proposal for a more complete monitoring will be discussed in the final report. A key point will certainly be the choice of sampling points: for a better knowledge of aquifers untreated water should be sampled directly at wells (as it has been done up to now), but if the estimate of the dose to population is the primary goal, samples should be drawn as ready for distribution and after any treatment.

Analytical techniques and choice of the parameters

Due to the extent of the analytical effort, it is important to take care in choosing the most significant analytical parameters.

Gross alpha and beta activity measurements give inconclusive or misleading information in the case of Estonian waters.

Radium isotopes concentrations are certainly the most relevant parameters. In particular Ra-228 is responsible for most of the committed dose. Determination of Ra-228 is quite easy and could be a priority objective.

The application of statistical analysis could be of some help both to improve sampling plans and to increase the significance of results.

Water quality improvement

Though only partial and preliminary data are available on the efficiency of radium removal by depuration plants which presently operate in Estonian aqueducts, it is arguable that their effectiveness is probably unsuited (15% provisional average).

Much higher removal efficiencies (> 70-80%) would be needed in most cases to attain an actual remedial action thus achieving the goal of 0.1 mSv/y.

No further data from experimental Viimsi plant were obtained thus no statement has been made here on this issue.

Many commercial methods are available but their cost is usually not negligible. Furthermore, only some of them can provide such high removal efficiency. A possible worsening of the water quality has also to be taken into account.

As international guidelines suggest, the first action should be made on the choice of aquifers to be exploited. The percent contribution of fresh water and non Cambrian-Vendian layers should be increased whenever possible. The use of Cambrian-Vendian waters should be avoided in any new plant/well.

Waste management

The currently operating sand filters produce both solid and liquid residues which contain the radium removed from water.

Though neither an European regulation nor a national one are presently in force, an assessment of radiological impact of the clearance should be done. Furthermore, the draft of the forthcoming new European Basic Safety Standard expressly requires radiological evaluations of groundwater treatment plants.

Some international technical documents issued by IAEA, NRPB and European Commission can help for a preliminary compliance test. When these criteria are applied to the selected treatment plants, the following conclusion can be drawn:

- a. Backwash waters of treatment systems generally comply with reference levels; Rakvere aqueduct is an exception, as its backwash waters are sent to the town sewer, whose sludges are employed in agriculture.

b. Solid residues from filters exceed general clearance levels. Specific assessment is therefore required to search for specific clearance levels.

A treatment plant more effective in radium removal than the present ones will obviously produce higher activity residues. Therefore, a special attention will have to be paid to the management of wastes as efficient treatment devices will be introduced.

Regulatory system

The EC Council Directive on drinking waters 98/83/EC includes Total Indicative Dose (TID) among indicator parameters (not mandatory). In a similar way World Health Organization reports 0.1 mSv/y as a recommended reference dose value. When it is exceeded, the balance between advantages and disadvantages of remedial actions should be considered, taking into account economic, social and health factors and any possible drawback of these actions (the so called ALARA – As Low As Reasonably Achievable - principle). The use of mandatory limit values is not recommended in situations involving wide exposure to natural radiation sources, and where a primary good is involved.

In spite of the fact that international regulations refer, implicitly or explicitly, to adult age class, in this case it would be advisable to take into account other age classes too. An advice to limit the consumption of waters with high radium concentrations in the case of babies in lactation age could be considered.

EC Directive 98/83 also concerns water used for drinks and foodstuff preparation, thus specific regulations can be similarly envisaged for soft drinks production, both from the point of view of justification and to limit exposure of another sensitive class of age as teenagers.