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

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The views expressed in this report do not necessarily reflect the views of the European Commission

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1. FOREWORD

The Twinning project

In the first eight months of 2009 a Twinning Light Project has been carried out on the topic of “*Estimation of concentrations of radionuclides in Estonian ground waters and related health risks*”. Cooperation between Estonia and Italy has been established within the framework of Estonian Transition Facility 2006 sponsored by the European Union.

The not negligible amount of natural radionuclides, especially radium isotopes, contained in some ground waters in Estonia is due to geological reasons. The overall objective of the project was a deeper knowledge of the situation and the identification of proper strategies for the minimization of health risks.

The Italian team included: four experts of Regional Environment Protection Agencies (ARPA Lombardia and ARPA Veneto) in charge of data analysis, measure methods and sampling plans, two experts of water treatment plants and one expert of health effects of radiations (from the National Institute of Health in Rome) in charge of radioprotection studies and health evaluations.

The project has been carried out in tight cooperation with Estonian Health Protection Inspectorate (HPI). Other relevant institutions responsible for risk assessment and environmental control (as Geological Survey and Radiation Protection Centre), research centres (as Tallinn University and Tartu Polytechnic) and private stakeholders (as waterworks societies) were also involved.

The background

The project results are addressed to the full implementation of the requirements under the following EU Directives:

- Directive 2006/118/EC of the European Parliament and of the Council of 12 December 2006 on the protection of groundwater against pollution and deterioration;

- Directive 2000/60/EC of the European Parliament and of the Council establishing a framework for the Community action in the field of water policy;
- Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption.

The “Requirements for the quality and control of drinking water and analysis methods” are defined by contents in the Decree of the Minister of Social Affairs No. 82 from 31.07.2001. This was amended with the Decree of the Minister of Social Affairs No. 94 from 28.06.2002.

Concentrations of radionuclides were analysed and reported by laboratories in Estonia and abroad in the frames of different studies. Radiometric data are available only as reports, compiled by OÜ Geoloogiakeskus (the Geological Survey of Estonia LLC) and they show that the calculated total indicative doses (T.I.D.) of radionuclides in Cambrian-Vendian water bearing complex often exceeded the indicator value of 0.1 mSv/year. Such waters are widely used in Northern Estonia.

Moreover in 1994-1998 with the purpose of finding the most appropriate methods for decreasing indicative dose levels in water, EC initiated project “TENAWA” (Treatment Techniques for Removing Natural Radio-nuclides from Drinking Water) including experts from Finland, Austria, Germany and Sweden.

Development of the project

In the first phase of the project, a preliminary analysis of Estonian radiological database, mainly by geographic criteria, was accomplished in order to evaluate its representativeness. A screening on available technologies for radium removal from water resources was performed too, followed by a brief discussion of problems related to the management of wastes generated from treatment plants. Analytical methods for the measurement of gross alpha/beta activity, radium-226 and radium-228, currently used by ARPA Lombardia, were also given. Results of the first phase (Component 1) were summarized in the Technical Report 1, issued in April 2009.

In the second phase of the project (Component 2) a deeper analysis of data was performed. This allowed getting a clearer picture of the exposure of the population,

to define the most useful analytical parameters and to gather key information for an effective planning of a future representative monitoring. Possible health consequences were assessed taking into account the different age classes. Remedial actions were proposed and a first evaluation of their economic sustainability was done. Analyses were performed on five selected pumping stations provided by treatment plants in order to ascertain the effectiveness of the radium removal. An overview of international guidelines and regulations was then made, with the aim of providing support to any possible modification of Estonian regulation. All these topics are fully discussed in the Technical Report 2, issued in September 2009. Moreover, the above summarized results have been presented in three different meetings (September 2009) to all involved stakeholders.

In the third phase (Component 3) all data and information gathered in the whole project were re-examined and their interpretation refined. An extensive bibliographic research was accomplished to support our findings. The present Final Technical Report summarizes all the work done by the Italian team on the subject of Estonian waters from January to September 2009.

The following topics are presented and discussed:

- General behaviour of groundwaters and mechanisms of radioactivity transfer.
- Water supply zone sizes and their distribution in connection to radiological risk.
- Relationship between the radioactivity of water and related doses with the geological layer of aquifers.
- Analysis of radiological database on Estonian waters and assessment of dose to population and related health risks.
- Relationship between radiometric parameters and chemical-physical characteristics of waters
- Strategies and methods for an effective monitoring of radioactivity in waters
- General requirements for drawing up a representative sampling plan (guidelines for surveillance and analysis).
- Considerations on effectiveness of treatment plants presently operating in Estonian waterworks and possibility of their upgrading.
- Examination of possible alternative solutions and cost analysis.

- Radioactivity content of effluents and wastes from Estonian existing treatment plants and comparison with International Standards.
- Review of international regulation and recommendation.

In order to allow an easier understanding of this Technical Report, all these topics will be discussed in following chapters. Further information and useful documents will be reported in final Annexes.

Outcomes of the project

As a result of the project, Estonian authorities have been now provided with the appropriate knowledge and tools in order to successfully face the problem of radioactivity in drinking water through different and complementary pathways.

During the project time span, efforts were made to involve all the concerned Estonian authorities in order to present the results of the ongoing activities several technical meetings have been organized involving different institutions and a full report of the outcomes was shown in the kickout meeting.

Furthermore the aims and the results of the project were presented to the Ministry of Social Affairs, in charge of establishing national regulations, and to the Conference of Estonian Society of Health Protection, at Tartu University.

Since the problem of drinking waters has technical, health, economic and social implications, a wide and deep involvement of different public institutions certainly was a key point for the successful outcome of the project.

The outcomes of the project will allow the Estonian authorities to:

- establish a proper regulation tailored on national situation;
- carry on a complete and representative monitoring;
- set up local scale remedial actions according to their sustainability.

The improvement of drinking water monitoring related to radionuclides presence and all corrective actions could be quickly undertaken in a gradual way according to the current economic sustainability.

The cooperative attitude of both public institutions and private enterprises (aqueducts operators) gave the possibility of having a fruitful discussion that allowed considering the problem from several point of views and taking into account different requirements. Besides, the proper execution and organization of the project allowed the full achievement of mandatory results in the terms defined by the twinning contract. During all the stages of the Project implementation, satisfactory cooperation between Estonian and Italian work-teams has been reached ensuring the utter attainment of the Twinning Light goals.

A high commitment of short term experts as well as a valuable cooperation with qualified Estonian specialists permitted to avoid any failures in the achievement of the mandatory results. Moreover, the twinning partnership allowed all relevant knowledge and expertise to be utilized in the most appropriate way through the whole project cycle. A strict follow-up of the project is recommended to Estonian authorities in charge since the simultaneous action of public and private stakeholders is requested for a successful result.

2. RADIOACTIVITY IN GROUND WATERS

2.1. Origins of water radioactivity

Whilst groundwater is usually protected from manmade radioactivity, radionuclides of natural origin are normally present in water in different amounts. They are supplied from rocks and minerals which form the aquifer in the same way as other cations and anions: processes of erosion and dissolution bring radioactive elements from rocks into the water (Ivanovich, 1992). The most common natural radioelements are the following:

- Potassium-40. It is a beta-gamma emitter of primordial origin and goes along stable potassium in a fixed ratio (31.3 Bq per gram of potassium).
- Uranium-238 series. It is a natural radioactive series of many radionuclides one descending from the other. The most relevant are uranium-238 (^{238}U), uranium-234 (^{234}U), radium-226 (^{226}Ra) and radon-222 (^{222}Rn). ^{238}U is relatively abundant in the earth crust (around 3 mg/kg).
- Thorium-232 series. Thorium is 3-4 times more abundant than uranium. Radium-228 (^{228}Ra) belongs to this series.

In the rock lattice all elements belonging to the same series are in secular equilibrium (their radiological activity is the same). Nevertheless the radionuclide sorption from the rock and the following stability as solutes is ruled by complex chemical-physical mechanisms and by the individual characteristics of radio elements. Thus the secular equilibrium is lost for dissolved radionuclides and their concentrations are rather independent one from each other. Some general behaviour rule for radioactive elements is given here, mainly gathered from extensive studies run in U.S. (Cothorn, 1990):

- 1) radionuclides produced by an alpha decay are more readily driven out from the rock since the alpha decay causes a recoil of the atom that lowers its stability in the lattice.
- 2) Highly porous rocks (e.g. basaltic rocks) can easily reabsorb radionuclides from water. On the opposite, sandstones have a very low capability of absorption.

- 3) Radium has a relatively low solubility and does not form soluble complexes. Usually the most abundant radionuclides in water are uranium isotopes (^{234}U and ^{238}U).
- 4) In oxidizing conditions, uranium forms stable soluble complexes (e.g. carbonates) and can move for long distances.
- 5) In reducing conditions (absence of air) uranium precipitates thus forming concentrated secondary deposits. Levels of the descendant ^{226}Ra can be very high in sites of uranium enrichment.
- 6) The solubility of thorium is very low, thus it is scarcely mobilized from water. The transport of the descendant ^{228}Ra is limited by its short half-life (5.7 years). This means that levels of ^{228}Ra are directly controlled by the concentration of thorium in the aquifer solids. Due to the higher abundance of Th over U, ^{228}Ra can be the dominant radium isotope if no secondary enrichment of uranium is present.
- 7) Highest values of ^{228}Ra can be found in waters originated from granitic rocks, arkosic sand and sandstones (geometric mean 50-80 mBq/L, 95° percentile 450-650 mBq/L), quartzose sandstone aquifers. No relevant changes over time in radium concentrations are usually found.
- 8) The dependence of ^{226}Ra from the local geology is not so sharp due to the higher mobility of the radionuclide and of the parent ^{238}U .
- 9) In roughly geologically homogeneous areas, a linear dependence between radium isotopes concentration and total dissolved solids can often be found.
- 10) The radionuclide concentration is higher in ground water than in surface water.

2.2. Behaviour of radium in ground water systems

The mechanisms of radium transfer to waters and its transportation inside the aquifers are complex and influenced by many factors. Extensive reviews of these studies are available (IAEA 1990; Cothorn 1990, Ivanovich 1992). The variability of radium concentrations in ground water is higher than uranium one and its correlation with the radioactivity of aquifer rocks is more difficult to be established since many factors play a role in the transfer mechanism from rocks to water, nevertheless an

attempt to connect the geology of the aquifer and the radium concentration was made in order to set a provisional model (Hess, 1985).

The radium dissolution by weathering can be strongly helped by the alpha recoil process (see former paragraph). The radium is transferred from the lattice to the pores of the rock and equilibrium is established between the radium in solution and the radium absorbed on the mineral surface. The absorption/desorption mechanism depends both on the kind of the rock and on the chemical composition of the water. For many minerals the re-absorption mechanism can be very effective, so most of the weathered radium is retained on the particle surface in the absorbed state. On the opposite, the increase of water salinity favours the dissolution of radium; alkaline metals are especially effective in supporting the release of radium from mineral particles and rocks pores. Important exceptions can be found (Vengosh, 2009) and in those cases the explanation was the low availability of the absorption sites of the aquifer rocks (mainly sandstones).

Likewise, the stability of radium in solution depends on the solution chemistry. Radium belongs to the alkaline earths elements (II A) like calcium, strontium and barium, which do not form stable complexes and are precipitated as carbonates, sulphates etc. Though radium itself can hardly achieve the solubility limit (even in presence e.g. of high concentration of sulphate anion), it can co-precipitate with similar cations and especially with barium: secondary deposit of radiobarite can be found close to uranium deposits. Co-precipitation can occur even in the presence of iron and manganese when insoluble oxyhydroxides are formed.

Despite the chemical similarity with barium, correlations between radium and barium concentration are not always found. A possible explanation concerns the prevailing mechanism for radium release: if the recoil mechanism is dominating, no correlation with barium is expected; if weathering mechanism is dominating, high concentrations of barium are found in radium rich waters (Chalupnik, 2008).

Although definitive data are not available, the wideness of radium transportation within the aquifer is supposed to be small (IAEA, 1990 – Chapter 4-2, Chalupnik, 2008), in the order of magnitude of meters during an half life of ^{226}Ra (1602 y) and proportionally less for the short living ^{228}Ra (6.7 y)

^{226}Ra and ^{228}Ra come from two different series, the first is originated from the relatively soluble uranium, and the latter is a descendant of the insoluble thorium. The study of the highly variable $^{226}\text{Ra}/^{228}\text{Ra}$ ratio, extensively reviewed by Dickson (IAEA, 1990 – Chapter 4-2), can give information on the nature of the aquifer, the transfer mechanism and the radium shifting in the water table, although obtainable conclusions can only be referred to specific cases.

Despite the complexity of the problem, some conclusion on the radium behaviour in groundwater systems can be drawn:

- The concentration of radium isotopes in water depends on complex and competing mechanisms. It can be generally stated that both concentrations of radium isotopes and their ratio are not related to the concentrations of parent uranium and thorium in solution, which are not thus the source of the dissolved radium.
- The most important transfer processes are related to the chemical composition of water, by consequence the radium concentration in water usually depends more on the groundwater chemistry than on the concentration of radium in the host rock. The nature of the solid part of the aquifer mainly plays a role in the re-absorption of radium.
- Studies available up to now show that the temporal variability of radium concentration in aquifers is low (Hess, 1985).

2.3. Occurrence of radium in groundwater

The presence of radium isotopes in groundwater is highly variable; concentrations of ^{226}Ra and sometimes those of ^{228}Ra , can exceed 1 Bq/L, but most cases they are related to thermal springs, mineral water or drilled wells. Few examples of aqueducts contaminated by radium are available (IAEA 1990-pagg. 72-74).

In Finland many investigations have been carried out even in recent times (Vesterbacka, 2005 and 2006). The problem of radium concerns only private wells and in limited amount: 4% of examined wells exceeded 500 mBq/L of ^{226}Ra and 1% exceeded 200 mBq/L of ^{228}Ra concentration.

In Sweden (Salih, 2002) a considerable amount of private drilled wells (47%) exceed the national limit which is much lower yet (185 mBq/L for total radium).

Measurements carried out in western Spain (Fernandez, 1992) showed that in 13% drilled wells ^{226}Ra exceeded 1 Bq/L, with a maximum value of 9.3 Bq/L, but dug wells exhibited much lower radium levels. In Extremadura, close to Portugal, small aqueducts employ water with ^{226}Ra content up to 720 mBq/L and mitigation processes have been considered (Baeza, 2008).

Radium monitoring in water resources has been accomplished in many other countries (Ulbak 1984, Zhuo 2001, De Oliveira 2001, Ahmed 2004, Forte 2004, Bronzovic 2006, Wallner 2007, IRSN 2008, Kleinschmidt 2008).

Due to the increasing consumption of mineral waters, many papers have been published in recent year on radiological survey of bottled waters (Duenas 1999, Rusconi 2004, Somlai 2002, Obrikat 2004, Bronzovic 2005, Bituh 2009, Schonhofer 2008).

As far as United States are concerned, a huge amount of data is available from the early '80s (Hess, 1985). Nearly 80% of the 60,000 water supplies in the U.S. use ground water sources and 90% of them serves less than 3,300 people. In general radium was found to be a problem for some small aqueducts. A screening level of 185 mBq/L for gross alpha and of 110 mBq/L for ^{226}Ra was used: non-compliances were found mainly in south-east coastal regions (New Jersey, North Carolina, South Carolina, Georgia) and in the north-central regions (Minnesota, Iowa, Illinois, Missouri and Wisconsin). Those violations were mainly due to ^{226}Ra and average radium isotopes concentration was 370 mBq/L. In South Carolina approximately 3% of the ground water supplies exceeded the 185 mBq/L value, reaching the values of 980 and 440 mBq/L for respectively ^{226}Ra and ^{228}Ra . In Iowa radium concentrations up to 1.8 Bq/L (before treatment) are reported for a public water supply (Reid 1985, IAEA 1990). As a consequence, a number of aqueducts adopted specific treatments for radium mitigation in drinking waters.

Interesting cases are found in Middle East. In some dry areas in the south of Israel (Koch, 2008), the wide Nubian sandstone aquifer produces water with radium activities exceeding national regulations (which are in substantial agreement with WHO criteria). In this area water is a critical resource and few alternative sources are often available. In southern Jordan, nearby the cited area, the possibility of exploitation of this fossil and non renewable aquifer has been studied (Vengosh, 2009). Concentrations up to 1.3 Bq/L and 3.1 Bq/L for respectively for ^{226}Ra and ^{228}Ra have been found with very variable ratios.

As these literature data are considered, worrying radium levels ($^{226}\text{Ra} > 500$ mBq/L or $^{228}\text{Ra} > 200$ mBq/L) are seldom found in public water resources. Most cases are related to private drilled wells (especially in Scandinavia) and to mineral or thermal water, whose consumption is optional. In some areas (e.g. Australia) local uranium mining may cause water contamination but it does not concern drinking water resources. Aqueducts based on ground water normally exhibit low radium levels even in areas whose prevailing geology is supposed to be favourable to a radiological contamination of waters. Situations in which radium levels are high and a water treatment for its removal is necessary are spatially limited exceptions.

2.4. The radioactivity in Estonian aquifers

In Estonia more superposed water tables are present. They are called Quaternary, Silurian, Ordovician and Cambrian-Vendian (Cm-V) aquifer, according to their geological origin (Perens, 1997). High radioactivity concentrations are found especially in the last one, which is the deeper too. Cambrian-Vendian aquifer is large and quite deep (200-400 m), but near the coastal area it becomes shallower (90-100 m depth). At the same time in the Northern Estonia the other aquifers are too superficial and polluted to be used and mostly Cambrian Vendian aquifer supplies aqueducts: since the Cambrian Vendian aquifer is a confined system, the input of pollutants is not likely.

From a geological point of view Cm-V lays on a basement of crystalline rocks (granite, gneiss) approximately 10 m deep. The basement is covered by Cambrian-

Vendian water bearing siltstones and sandstones. The Cm-V aquifer is protected upward by the Lontova aquitard and it is divided into two sub-aquifers: Gdov (the deeper) and Voronka, the first mainly made of mixed grain sandstones and siltstones, the latter of quartzose sandstone and siltstone.

Isotopical analyses, mainly of ^{18}O (Raidla 2009), suggest that Cm-V water is extremely old and the aquifer is not recharged by meteoric water since Lontova aquitard protect it very effectively. Only in areas where ancient buried valleys are present, some infiltration of more superficial waters take place. In these areas lower concentrations of radionuclides are found. This fossil water is a mixing of water of Scandinavian continental melt ice of the last glaciation (Weichelian Ice Age) with much older brines rich in sodium and chlorine. In northern Estonia glacial water predominates while in central and southern Estonia brines give the higher contribution. In central and southern Estonia Cm-V aquifer is more deep and salty and it is not suitable as a drinking water but rather it is used for thermal baths. The residence time of water in the aquifer has been calculated in several tents of thousands years.

The concentration of Na and Cl is generally high, and in some areas even less common anions and cations (F and Ba) have unusually high concentrations. The so called “barium anomaly” poses concerns in some part of the country (Karro 2004, Marandi 2007). Some studies afforded the topic of correlation of radioactivity with dissolved salts (Na, Cl and especially Ba) but conclusive evidence has not been found yet (Mokrik 2009).

Some of the characteristics of Cm-V aquifer remind the above cited Nubian aquifer in Southern Jordan (Vengosh 2009), where the concentration of radium is very high too (see 2.3).

Available data (Tab. 2.1) show that average concentrations of both radium isotopes in the two Cambrian-Vendian sub-aquifers (Gdov and Voronka) are not significantly different.

The two sub-aquifers are claimed to be generally well separated in the North-eastern Estonia by the Kotlin clayey formation, but their intensive exploitation caused a difference in hydraulic pressure thus favouring the mixing of the water.

Table 2.1 *Radium isotope concentration in Cambrian-Vendian (Cm-V) sub-aquifers*

Cambrian-Vendian sub-aquifer	²²⁶Ra (Bq/L)	Std. dev.	²²⁸Ra (Bq/L)	Std. dev.
Not defined	0.38	0.20	0.34	0.21
Cm-V Gdov	0.45	0.23	0.36	0.21
Cm-V Voronka	0.48	0.32	0.42	0.29
Mixed	0.41	0.14	0.35	0.15

Moreover, recent studies (Karro 2004) showed that in last decades the composition of water in Kopli peninsula, close to Tallinn, changed. The increase of total dissolved solid indicates that the intense drainage of water caused the drawing up of deep water contained in the fractured crystalline basement (Karro 2004, Marandi 2007). Barium content raised too and this suggests an interesting question related to the change of radioactivity content in a time scale.

Barium is chemically similar to radium and a correlation between them has been observed (Mokrik 2009) and reported in present work as well (5.2). A possible explanation of the high radium concentration lies on its prolonged contact with the crystalline rocks of the basement with rather high radioactivity content, the reducing conditions of the aquifer which support the radium solubility and the presence of smectite-illite clays in which a mechanism of absorption-desorption of radium can take place (Mokrik 2009; see also 2.2). Nonetheless a thorough explanation of the phenomenon is not available yet.

3. AVAILABLE INFORMATION ON ESTONIAN AQUEDUCTS AND WATERS

3.1. Analysis of available information - Water Supply Zones

In Estonia 912 different water supply zones (WSZs, regions producing water with the same physical and chemical characteristics) can be identified, each of them supplying water to one or more aqueducts and serving, overall, 1110215 people.

The distribution of WSZs by served population is shown in Table 3.1 and in Figure 3.1. WSZs employing fresh water are included, while private water resources (wells) are not.

Table 3.1 *Distribution of WSZs in Estonia by size*

Class (served population)	Frequency	% Cumulative Frequency
Up to 50	146	16.0%
50 - 100	162	33.8%
100 - 250	269	63.3%
250 - 500	177	82.7%
500 - 1000	75	90.9%
1000 - 2500	40	95.3%
2500 - 5000	20	97.5%
5000 - 10000	9	98.5%
More than 10000	14	100.0%
Tot. people: 1110215	Tot. WSZs: 912	

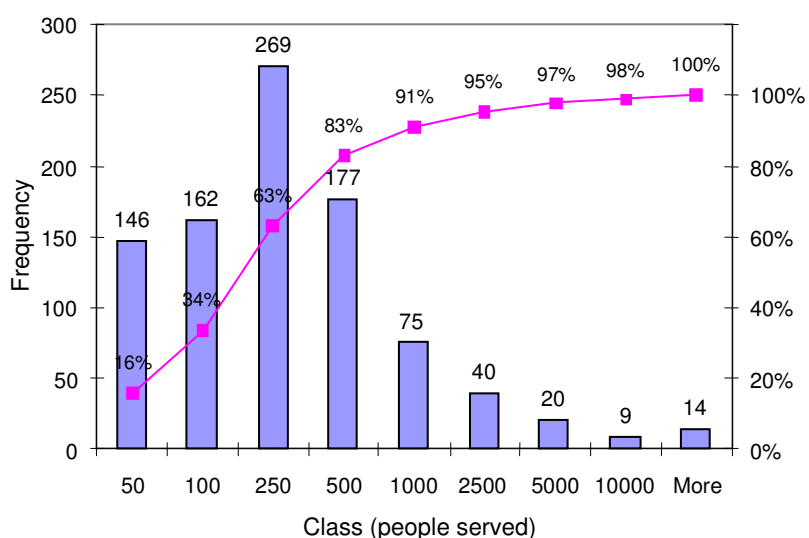


Figure 3.1 *Distribution of WSZs in Estonia by size*

Table 3.2 *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
Valgamaa	20314	48	44	92
Polvamaa	15186	42	39	93
Vorumaa	19726	47	44	94
Estonia	1100215	912	754	83

The distribution graph shows that 83% of all WSZs (754) serve 500 people or less. Most Estonian WSZs are small size ones. As can be seen from the data reported in Tab. 3.2, in every county a similar percentage of small WSZs is found.

3.2. Analysis of available information - Radiometric data

Estonian Authorities carried out, over many years, a monitoring program aimed at assessing the radioactivity content of drinking water. As it was known that water radioactivity was mainly due to natural radionuclides, namely radium isotopes (uranium is a minor contributor to Estonian water radioactivity), monitoring programs focused on the measurement of ^{226}Ra and ^{228}Ra ; in some cases gross alpha and

beta activities were measured as well. The available data are summarized and commented in the following paragraphs.

Table 3.3 resumes the overall number of available radiometric data (gross alpha and beta activity and radium isotopes), grouped according to the county; Figure 3.2 shows the percentage of population using, in each county, water of known radioactivity content.

Table 3.3 *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

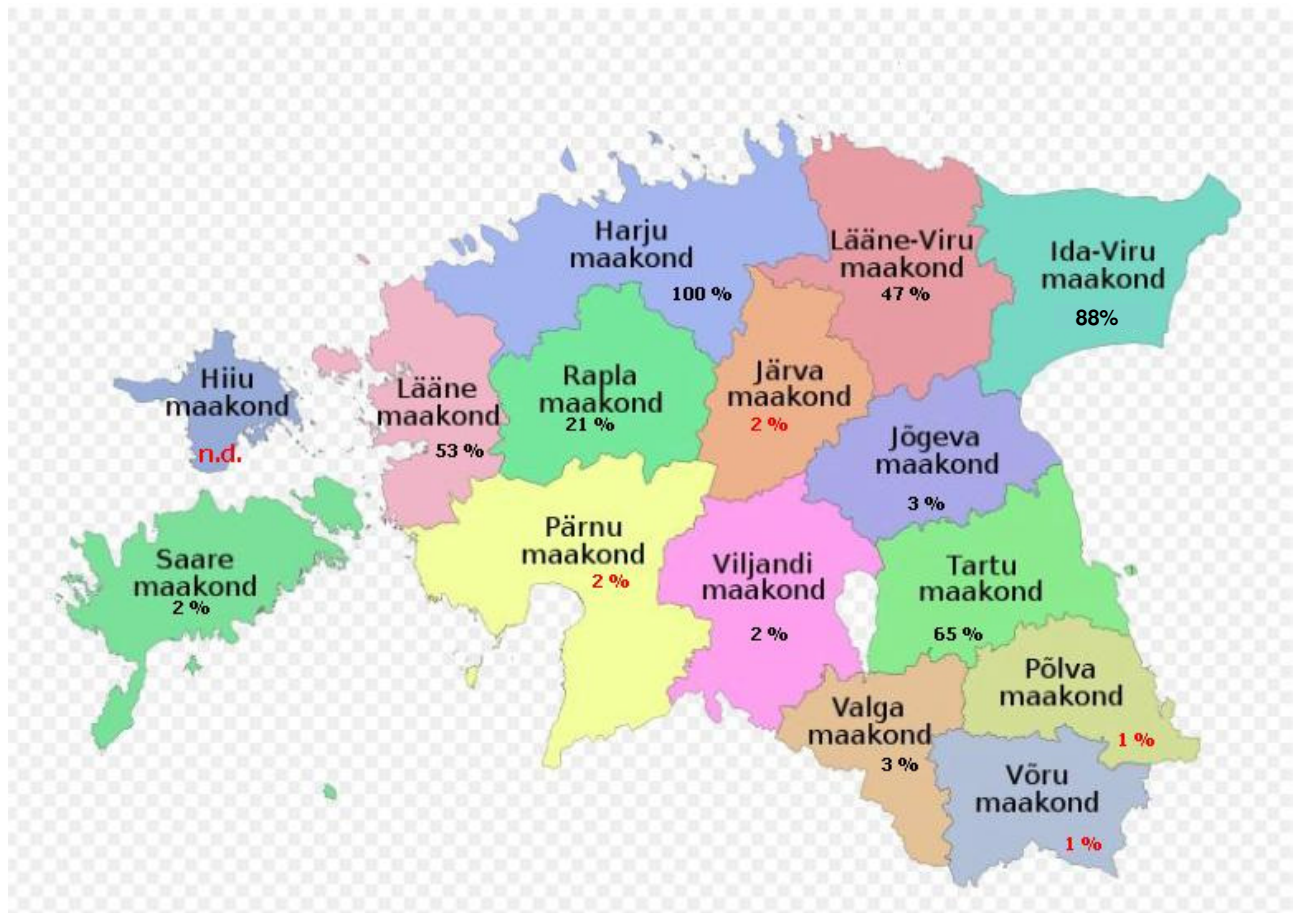


Figure 3.2 Percentage of population using water of known radioactivity content

In most counties (excluding the Northern ones of Harjumaa and Ida-Virumaa) few analytical data are available. In many cases (mostly in Southern counties) only gross activity values were measured, that cannot be used to deduce reliable information on radium isotopes concentration.

The following analysis, which aims at identifying areas interested by the problem and assessing the radiation dose due to the use of drinking water, is therefore based only on ^{226}Ra and ^{228}Ra values. Two main points are addressed in the following:

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.

3.3. Occurrence of high radioactivity values in Cambrian-Vendian aquifers

A preliminary analysis of the full data set showed that waters from Cambrian-Vendian aquifer and from its sub-layers (Gdov (V2gd) and Voronka (V2vr) sub-aquifers) have similar radionuclide concentrations; furthermore, 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. “Cm-V” group, including data for Cambrian-Vendian aquifers (Cm-V, V2gd and V2vr);
2. “non Cm-V” group, including data for the remaining aquifers (O-C, O, D, Q, S, S-O).

The following graphs (Figures 3.3, 3.4, 3.5 and 3.6) 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 that class added to the percentages in preceding size classes.

The Derived Water Concentration (DWC), that is the activity concentration in drinking water of each isotope corresponding to an adult ingestion dose of 0.1 mSv/y, is also shown (for more details on DWC see Addendum 4.1).

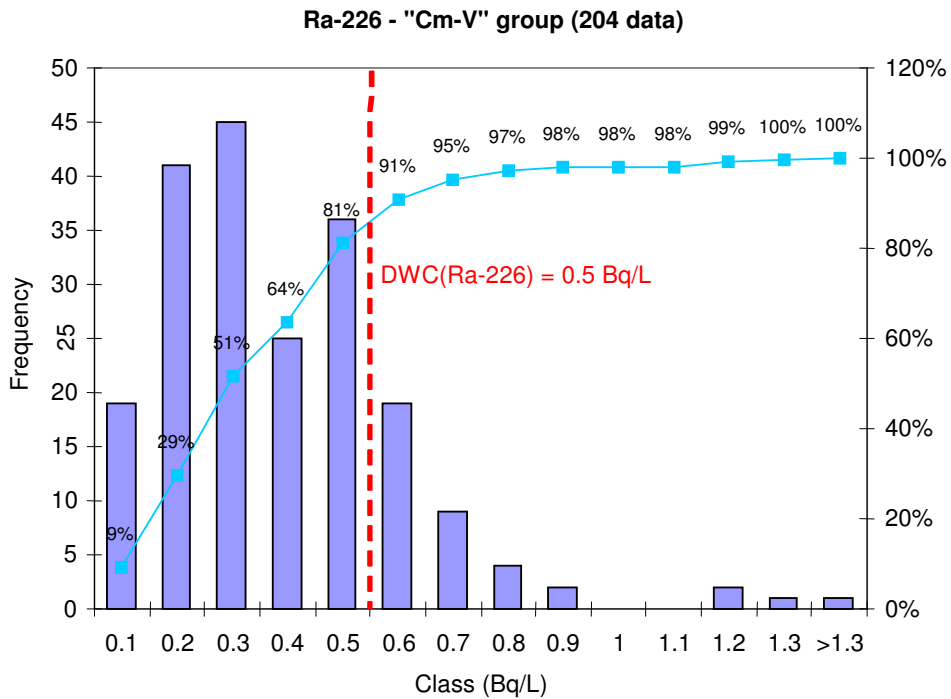


Figure 3.3 Frequency histogram of ^{226}Ra concentration in "Cm-V" aquifers

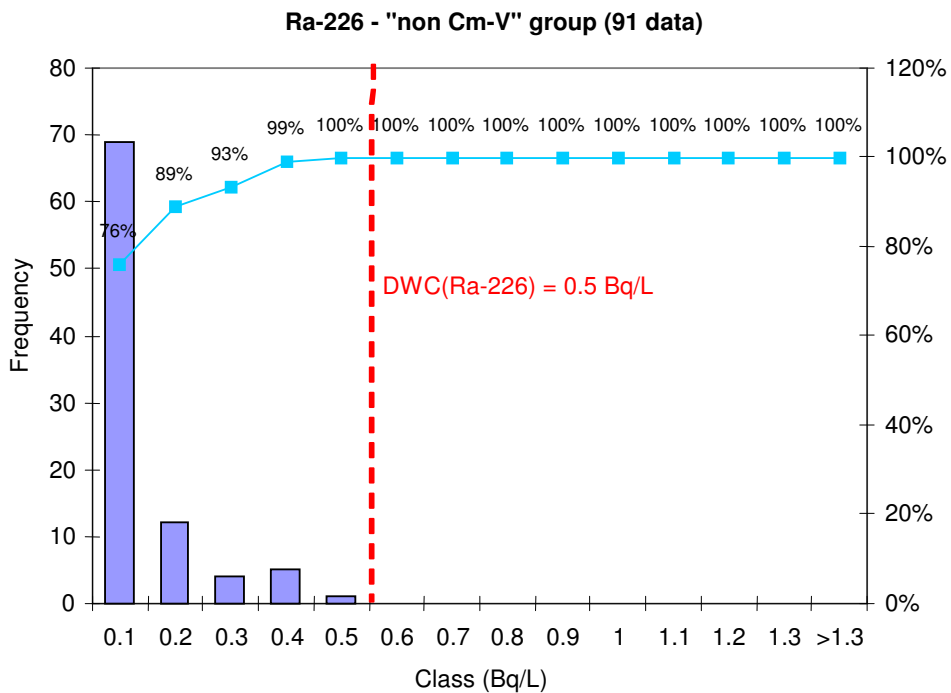


Figure 3.4 Frequency histogram of ^{226}Ra concentration in "non Cm-V" aquifers

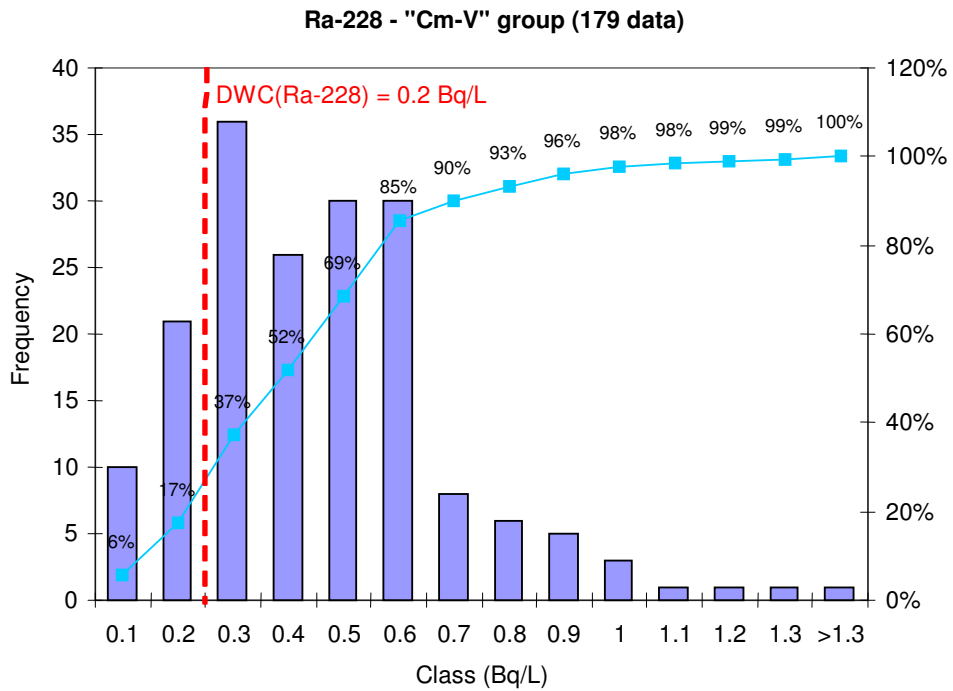


Figure 3.5 Frequency histogram of ^{228}Ra concentration in "Cm-V" aquifers

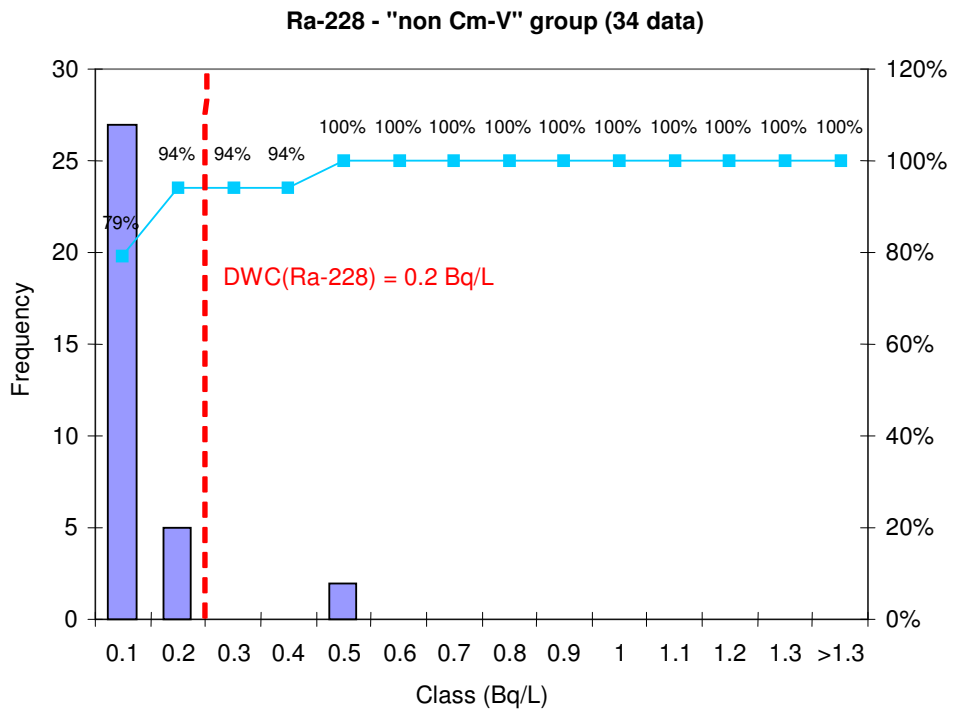


Figure 3.6 Frequency histogram of ^{228}Ra concentration in "non Cm-V" aquifers

The most relevant data are summarised in Table 3.4:

Table 3.4 *Summary of available radium data*

Group	²²⁶ Ra		²²⁸ Ra	
	No. of data	% of data > DWC (DWC = 0.5 Bq/L)	No. of data	% of data > DWC (DWC = 0.2 Bq/L)
Cm-V	204	19%	179	93%
non Cm-V	91	0%	34	6%

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

Waters from “non Cm-V” aquifers look ‘safer’, but only a more detailed monitoring could exclude the radiological concern as the number of available data is low and not all the values are lower than the DWCs.

3.4. Occurrence of high radioactivity values in Estonian counties

In the whole Estonia 912 different water supply zones can be identified; as most of them take water from a single aquifer, the same criteria used for grouping radiometric data (waters from “Cm-V” and “non Cm-V” aquifers) can be used for WSZs (the few zones taking water from different aquifers are classified according to the prevalent water source). Results are shown in Table 3.5 and in Figure 3.7.

Table 3.5 *Distribution of “Cm-V” and “non Cm-V” WSZs in Estonian counties*

County	No. of “non Cm-V” WSZs	No. of “Cm-V” WSZs	% of “Cm-V” WSZs on the total	Population served by “Cm-V” WSZs
Harjumaa	88	84	48.8%	134771
Ida-Virumaa	41	44	51.8%	76013
Laane-Virumaa	80	9	10.1%	23451
Laanemaa	32	3	8.6%	13365

County	No. of “non Cm-V” WSZs	No. of “Cm-V” WSZs	% of “Cm-V” WSZs on the total	Population served by “Cm-V” WSZs
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
	Tot: 772	Tot: 140		

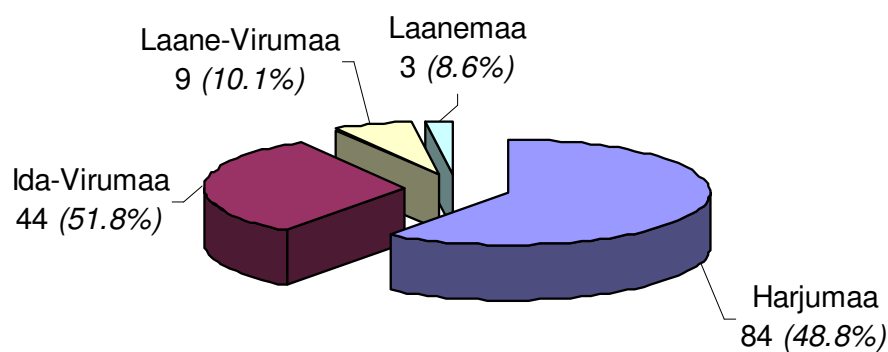


Figure 3.7 Distribution of “Cm-V” water supply zones (tot. 140) in Estonian counties

WSZs taking water from Cm-V aquifers 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 about 10% in Laane-Virumaa and Laanemaa.

If we assume that high levels of radioactivity can be found mainly in the “Cm-V” group water supply zones, we can conclude that the problem is mostly limited to 140 water supply zones, corresponding to about 250,000 people (22% of Estonian

population). Actually, 12 WSZs out of 140 (the biggest ones, serving more than 5000 people) feed water to about 152,000 people (Table 3.6).

Table 3.6 *Biggest (serving more than 5000 people) “Cm-V” water supply zones*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/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, Täkumetsa 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 (Figures 3.8 and 3.9) provide more information about the “Cm-V” group supply zones: 83 supply zones serve less than 500 people; the 12 biggest zones altogether serve about 152000 people, 10 out of them producing more than 1000 m³/day of water. The 71 supply zones producing less than 50 m³/day of water altogether serve about 12400 people. Furthermore, in 16 out of 140 zones, water from “Cm-V group” aquifers is mixed with water from “non Cm-V” aquifers or surface water.

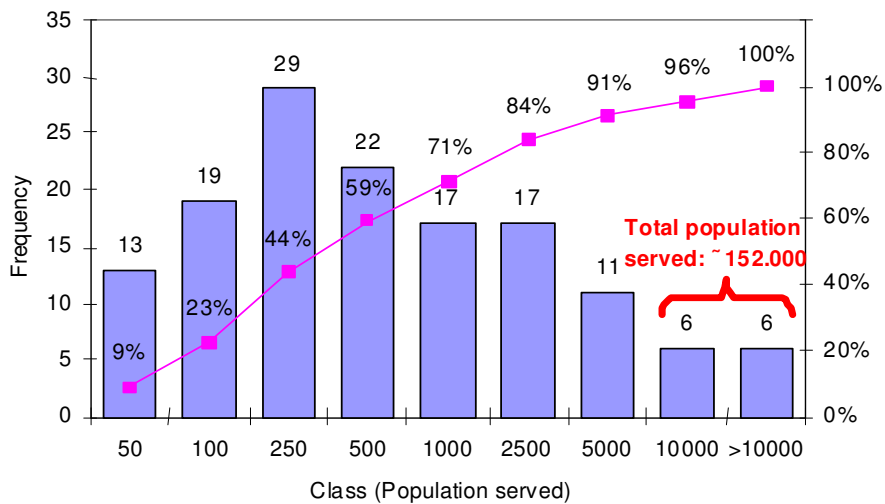


Figure 3.8 Distribution of “Cm-V” water supply zones by population served

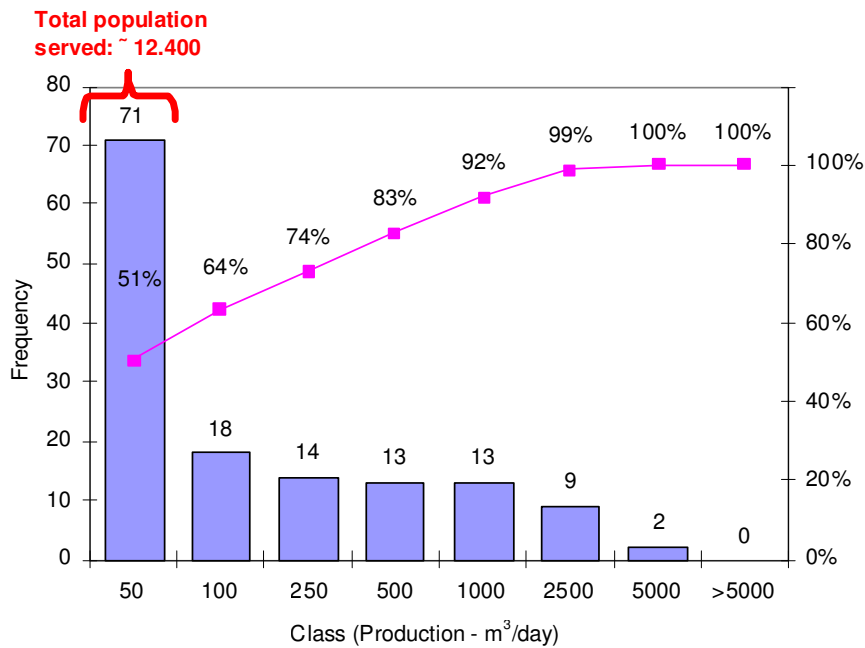


Figure 3.9 Distribution of “Cm-V” water supply zones by daily water production

4. REGULATIONS AND RISK ASSESSMENT

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, on request of the European Parliament, also included requirements for radioactivity. This does not mean that previously there was no concern on radioactivity in drinking water, but its safety from a radiological point of view was considered in any case guaranteed by the general system of radiation protection (see EC, 1996).

4.1. 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* (EC, 1998) was published in the Official Journal of the European Union. It was a revision of the Council Directive of 1980 (EC, 1980) 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).

As reported above for the first time 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 4.1.

Table 4.1. *Indicator parameters for radioactivity in drinking water (EC, 1998)*

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

From the Total Indicative Dose (TID) tritium, potassium 40², radon and radon decay products³ are excluded explicitly. For the assessment of the TID the effective 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 TID. Indeed, it corresponds to a committed effective dose for adults of 1 µSv/y. This value was requested by the amendment of the European Parliament (see ACES, 1994) and it

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 of intake.

³ For radon and its decay products, some years later the European Commission issued a Recommendation (EC 2001) (see Addendum 4.4).

was judged that the adoption of such a 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, implies 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 (EC, 1998). These requirements, not yet

published, should have suggested to Member States how to guarantee the required level of protection.

4.2. Total indicative dose (TID)

In 1999, a working party of the Article 31 Group of Experts of the EURATOM Treaty was set up in order to draw up a proposal for an environmental monitoring plan that would guarantee the level of radiological protection required by the Directive (see Addendum 4.1).

In order to draw up this plan, an estimate of water intake for different age classes was essential and after an overview of different intake values used by different organisations (EPA, ICRP, UNSCEAR, WHO) those reported in Table 4.2 were chosen.

Table 4.2. Annual drinking water intake assumed for different age groups

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

These intake values are needed for calculating the Total Indicative Dose, which is given by the following equation

$$TID(Sv / y) = \text{conc.}(Bq/L) \times \text{committed eff. dose per unit intake (Sv/Bq)} \times \text{ann. intake (L/y)}$$

The parameters *committed effective dose per unit intake* are the dose coefficients for different age classes of *members of the public*, calculated and published by the International Commission for Radiological Protection (ICRP, see Addendum 4.2), which were adopted by the EURATOM Basic Safety Standards (EC, 1996). For ingestion of ²²⁶Ra and ²²⁸Ra they are reported in Table 4.3.

Table 4.3 *Ingestion dose coefficients for members of the public*

Nuclide	Committed effective dose per unit intake (Sv/Bq)					
	Age class (y)					
	≤ 1	1 - 2	2 - 7	7 - 12	12-17	> 17
²²⁶ Ra	4.7 10 ⁻⁶	9.6 10 ⁻⁷	6.2 10 ⁻⁷	8.0 10 ⁻⁷	1.5 10 ⁻⁶	2.8 10 ⁻⁷
²²⁸ Ra	3.0 10 ⁻⁵	5.7 10 ⁻⁶	3.4 10 ⁻⁶	3.9 10 ⁻⁶	5.3 10 ⁻⁶	6.9 10 ⁻⁷

The above equation applied to a single radionuclide concentration estimates actually the committed effective dose by ingestion, which is only coincident with the TID if one radionuclide is only contained in the water. Otherwise, the committed effective doses relevant to each nuclide should be summed up to assess the TID.

On the basis of the intake values of Table 4.2, the ²²⁶Ra and ²²⁸Ra doses (mSv/y) for unit activity concentration (Bq/L) were evaluated. For the age class (10-17)y, being no agreed data about annual consumption, the calculations were made for the two classes (10-12)y and (12-17)y⁴ with the annual intake of both the lower and the higher age class. The results are reported in Table 4.4.

Table 4.4 *Committed effective dose per year and unit activity concentration by ingestion for different age classes*

Nuclide	Committed effective doses per year and unit activity concentration by ingestion (mSv y ⁻¹ /Bq L ⁻¹)						
	Age class (y)						
	≤ 1	1 – 2	2 - 7	7 - 10	10-12	12-17	> 17
²²⁶ Ra	1.2	0.3	0.2	0.3	0.3 – 0.6	0.5 – 1.1	0.2
²²⁸ Ra	7.5	2.0	1.2	1.4	1.4 – 2.9	1.9 – 3.9	0.5

It can be noticed that both nuclides are critical for lower age classes, in particular for infants (< 1 year) and small children. Moreover, the age class (10 – 17) y is also critical - even if not the most at risk - due to the high radium metabolic absorption in

⁴ The two classes cannot be joined due to the different dose coefficients.

bones during the growing age, having radium the same chemical behaviour of calcium.

In order to give a clear and easy picture of the higher doses that could be received by lower age classes, in Table 4.5 the ratios of the doses per unit intake for different age classes to the doses per unit intake for adults are reported.

Table 4.5 *Multiplicative factors which show the higher committed effective doses that can be received by younger people when consuming drinking water rich in radium*

Nuclide	Different age class dose per unit activity concentration/ adult dose per unit activity concentration						
	Age class (y)						
	≤ 1	1 - 2	2 – 7	7 - 10	10-12	12-17	> 17
²²⁶ Ra	6.0	1.50	1.0	1.5	1.5 – 3.0	2.5 – 5.5	1.0
²²⁸ Ra	15.0	4.0	2.4	2.8	2.8 – 5.8	3.8 – 7.8	1.0

4.3. Total indicative doses for Estonian drinking water

The Total Indicative Dose for Estonian drinking water due to the radium isotopes was calculated – with the equation and the parameters reported in the previous paragraph – using radium values reported by Estonian authorities and assuming a continuous and exclusive use of contaminated water.

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

The results for “adult” age class are summarised in the following graphs (Fig. 4.1 and 4.2):

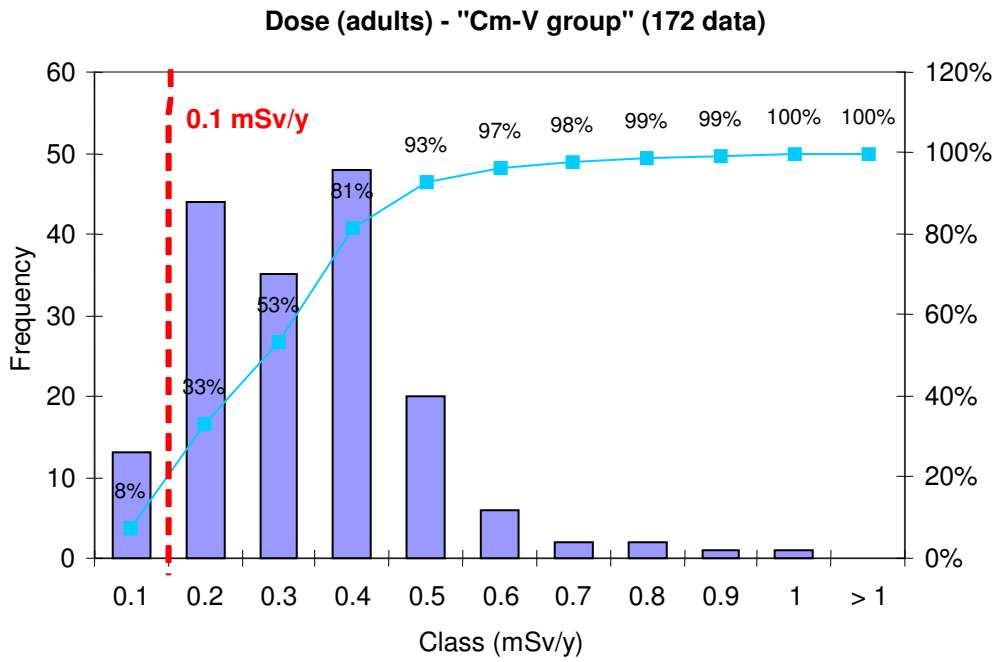


Figure 4.1 Distribution of adult dose - "Cm-V" group waters

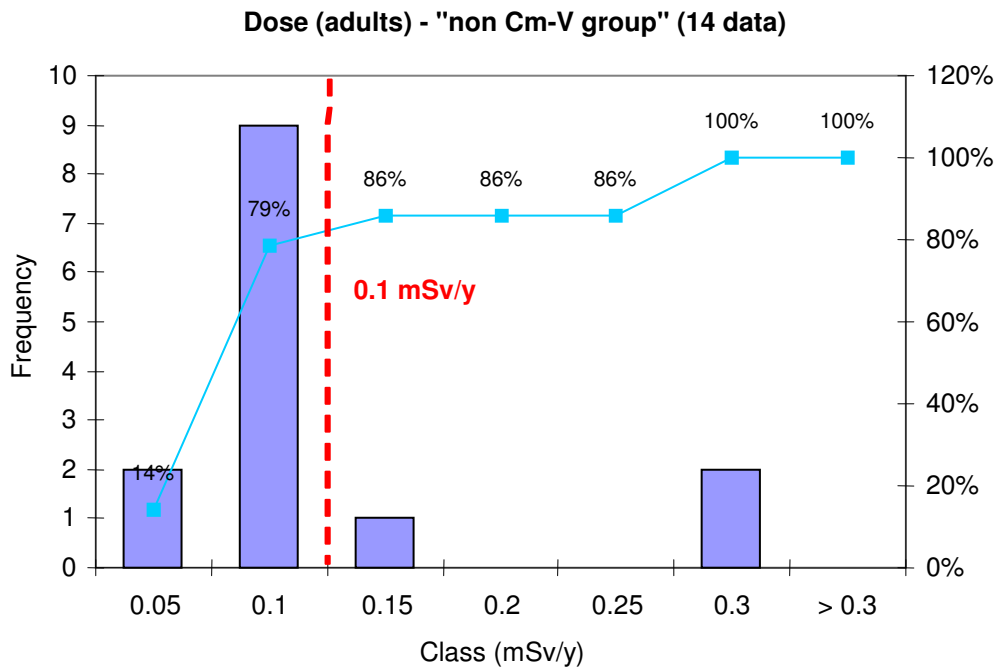


Figure 4.2 Distribution of adult dose - "non Cm-V" group waters

TIDs due to waters from Cm-V aquifers exceed the parametric value of 0.1 mSv/y in 92% of the cases, as expected on the basis of radium data analysis.

Available data for waters from “non Cm-V” aquifers are few, nonetheless it is worth pointing out that not all TID values are lower than 0.1 mSv/y in this case, too. This is even more heavy if starting from the same measured data the dose is calculated for the “infant” (≤ 1 y) age class, as shown in the Fig. 4.3 and 4.4.

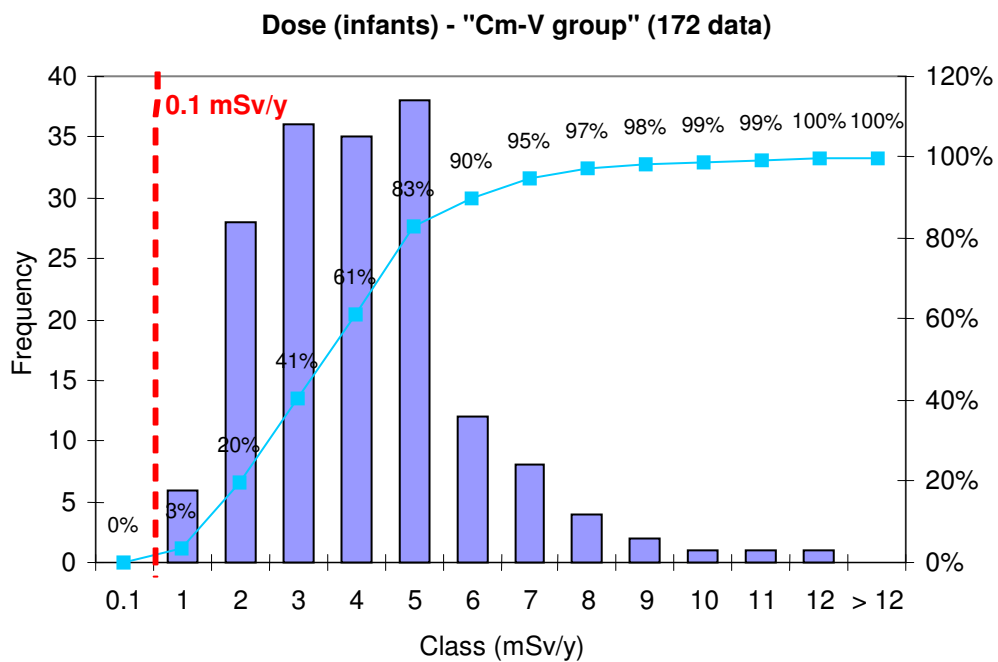


Figure 4.3 Distribution of infants (≤ 1 y) dose - “Cm-V” group waters

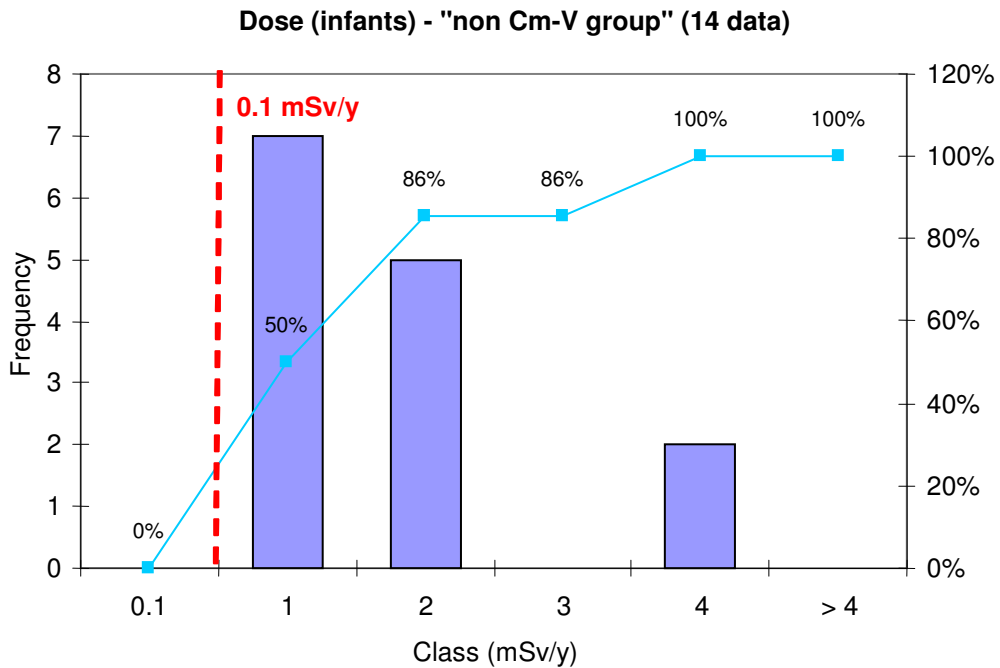


Figure 4.4 Distribution of infants (≤ 1 y) dose - "non Cm-V" group waters

It is worth pointing out that, despite the similar activity concentration of the two radium isotopes, the contribution to total dose is higher for ^{228}Ra , especially in the case of infants (≤ 1 y) (Fig. 4.5 and 4.6) due to higher ingestion coefficient (as outlined in paragraph 4.2).

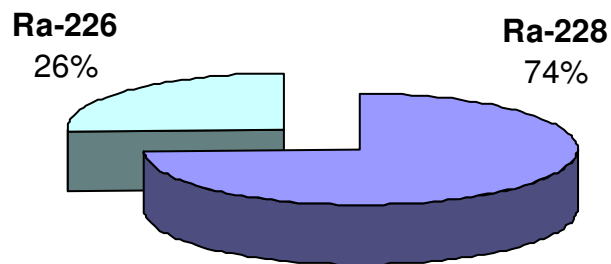


Figure 4.5 Average relative contributions to total dose of the two radium isotopes - Adults

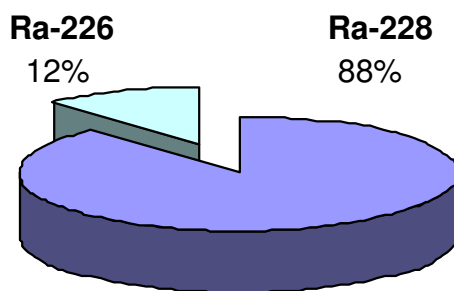


Figure 4.6 Average relative contributions to total dose of the two radium isotopes - Infants

In the previous graphs adults and infants were only considered. However, dose values are always lower for adult age class than for any other age class and this is especially true for infants, small children and teenagers, as shown in Tab. 4.6 and in Fig. 4.7 for waters belonging to “Cm-V” group.

Table 4.6 Committed effective dose for Estonian population drinking “Cm-V” group water

Dose values ¹ mSv/y	Age class						
	≤ 1	1 - 2	2 – 7	7 - 10	10-12 ²	12-17 ³	> 17
Average	3.6	1.0	0.6	0.7	0.9	1.7	0.3
Standard deviation	1.9	0.5	0.3	0.4	0.5	0.9	0.2
Median	3.5	0.9	0.6	0.7	0.8	1.6	0.3
95 th percentile	7.0	1.9	1.1	1.3	1.7	3.3	0.6

¹:Dose values calculated as outlined in paragraph 4.2

²:Assumed annual water intake: 450 L/y

³:Assumed annual water intake: 600 L/y

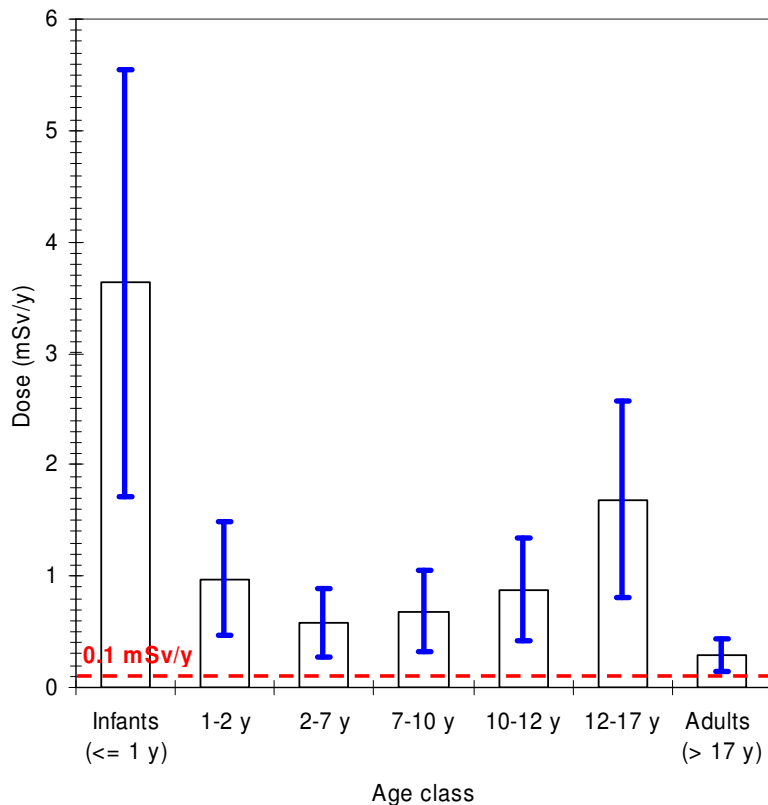


Figure 4.7 Dose to Estonian population per age class (average and standard deviation) - “Cm-V” group waters

4.4. Radiological risk for Estonian drinking water

The health risks related to ionising radiation exposure are described very clearly and effectively in the last ICRP Publication (see Addendum 4.2).

“Radiological protection deals with two types of harmful effect. High doses will cause deterministic effects, often of an acute nature, which only appear if the dose exceeds a threshold value. Both high and low doses may cause stochastic effects (cancer or heritable effects), which may be observed as a statistically detectable increase in the incidences of these effects occurring long after exposure

The Commission’s system of radiological protection aims primarily to protect human health. Its health objectives are relatively straightforward: to manage and control exposures to ionising radiation so that deterministic effects are prevented and the risks of stochastic effects are reduced to the extent reasonably achievable.” (ICRP, 2008).

The EURATOM Directive “Basic Safety Standards for the protection of health of workers and the general public against the dangers arising from ionizing radiation” (EC, 1996), issued in 1996, still in force, and transposed in the national legislation of Member States, was based on the *1990 Recommendation of the International Commission on Radiological Protection* (ICRP, 1991). Therefore, health risks due to a radiation exposure should be based on this ICRP document.

ICRP (ICRP, 1991) uses the concept of *detriment* to estimate the stochastic effects of exposure to radiation doses well below the thresholds for deterministic effects. Detriment is the total harm to health that would eventually be experienced by an exposed group and its descendants as a result of the group’s exposure to a radiation source. It includes the risk of fatal cancer in all relevant organs, non fatal cancer, severe hereditary diseases and length of life lost if the harm occurs (see Addendum 4.2).

In the 1990 Recommendations (ICRP, 1991), the detriment for the whole population was assessed to be $7.3 \cdot 10^{-2} \text{ Sv}^{-1}$ (ICRP, 1991). This means $7.3 \cdot 10^{-5}$ per mSv. With this factor the detriment of a person drinking water from “Cm-V group” aquifers in all his/her lifetime was calculated, starting from the average dose value for each age class (see Fig. 4.7). The detriment for each age class is shown in Table 4.7, with a total of $2.6 \cdot 10^{-3}$ on the whole life. The percentage contribution to whole life detriment due to consumption of water in each age class is also reported in Table 4.7:

Table 4.7 *Detriment for each age class and its percentage contribution to whole life detriment due to consumption of water in each age class*

	Age class						
	≤ 1	1 – 2	2 – 7	7 – 10	10-12	12-17	> 17
detriment for 1 year exposure	$265 \cdot 10^{-6}$	$71 \cdot 10^{-6}$	$43 \cdot 10^{-6}$	$50 \cdot 10^{-6}$	$64 \cdot 10^{-6}$	$123 \cdot 10^{-6}$	$21 \cdot 10^{-6}$
Supposed length of exposure (years)	1	1	5	3	2	5	53
% contribution to whole life detriment	10	3	8	6	5	24	44

4.5. Radiological protection conclusions

Some general comments can be drawn from the above discussion:

- the annual dose level of waters from Cambrian-Vendian aquifers hardly complies with 0.1 mSv/y value recommended by the European Union; for non Cambrian Vendian waters few data are available, and not always compliant with EU reference level. In both cases the dose is mainly due to the contribution of ^{228}Ra ;
- non compliance with the 0.1 mSv/y TID is more striking for infants and small children that are the population group most at risk. This also means that the derived activity concentration in drinking water calculated for adults might not be conservative enough for these age classes; great caution should therefore 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 (BU, 2006) and in Italy some recommendations suggested by the Istituto Superiore di Sanità (Nuccetelli et al., 2004) were adopted by the Italian Minister of Health (but for mineral and spa water);
- in terms of health risk, estimated using the whole life detriment, the main contribution is from consumption of drinking water in adult age as for this age class dose values are lower but the exposure is supposed to last for many (53) years; on the other hand, an important (and feasible) reduction in whole life detriment could be achieved limiting the use of contaminated water for infants and teenagers. This would be effective considering the higher radiosensitivity of the younger age classes.

As regards the implementation of the European Directive (EC, 1998) two general criteria should be kept in mind: the fact that TID 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 paragraph: "*Intervention*" in Addendum 4.2).

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.

Addendum 4.1 - Calculation of the derived reference levels for drinking water

As already referred in paragraph 4.2, 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 (EC, 1996), 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 (Risica, 2000) 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 (EC, 1996), and the water intake values chosen by the Article 31 working party (see Table 4.2 in paragraph 4.2).

Calculations were carried out for the age classes $\leq 1y$, (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 $1 \times 10^{-4} \text{ Sv/y} = 0.1 \text{ mSv/y}$ is the set Total Indicative Dose.

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

In the database (Risica, 2000) 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 A.4.1 for main radionuclides in a nuclear emergency as reported in table VIII of (IAEA, 1986) and in Table A.4.2 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 A.4.1. *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 A.4.2. *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 4.4)

° For uranium in drinking water see Addendum 4.5

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

Observing the two tables, it can be noted that, caesium 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 (Risica, 2000) 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 caesium isotopes) to 17.3 for ^{228}Th . In the complete database (Risica, 2000) 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 compliance with the DWC for adults could expose infants and small children to a dose much higher than the TID.

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.

Addendum 4.2 - 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 (ICRP, 2009).

“The advice of the Commission is aimed principally at regulatory authorities, organisation and individuals that have responsibility for radiological protection... The Commission provides guidance on the fundamental principles on which appropriate radiological protection can be based.” (ICRP, 2008).

ICRP and natural radioactivity

ICRP starting from its 1977 Recommendation (ICRP, 1977) 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” (ICRP, 1984).

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.

Estimate of “Detriment”

As already reported in paragraph 4.4, detriment assesses “the total harm that would eventually be experienced by an exposed group and its descendants as a result of the group’s exposure to a radiation source” (ICRP, 1991). It includes four main components: “the risk of fatal cancer in all relevant organs, a specific allowance for differences in latency which result in different values of expected life lost for fatal cancer in different organs, an allowance for the morbidity resulting from induced non fatal cancers an allowance for the risk of serious hereditary disease in all future generations descended from the irradiated individual”. (ICRP, 1991).

The corresponding value for adult workers and whole population are reported in Table A.4.3.

Table A.4.3. *Nominal probability coefficients for stochastic effects*

	Detriment (10^{-2} Sv^{-1})*			
Exposed population	Fatal cancer	Non-fatal cancer	Severity hereditary effects	Total
Adult workers	4.0	0.8	0.8	5.6
Whole population	5.0	1.0	1.3	7.3

* Rounded values

Recently ICRP decided to adopt a revised set of Recommendations (ICRP, 2008), where new probability coefficients are given for stochastic effects. Main differences come from the much lower coefficients for probability of heritable effects, because calculation was stopped at the second generation. The total detriment for the whole population is $5.7 \cdot 10^{-2} \text{ Sv}^{-1}$.

The intervention

“In situation where the sources of exposure and the exposure pathways are already present the only type of action available is intervention.

The system of radiological protection recommended by the Commission for intervention is based on the following general principles:

- a) "The proposed intervention should do more good than harm, i.e. the reduction in detriment resulting from the reduction in dose should be sufficient to justify the harm and the costs, including social costs, of the intervention.
- b) The form, scale, and duration of the intervention should be optimised so that the net benefit of the reduction of dose, i.e. the benefit of the reduction in radiation detriment, less the detriment associated with the intervention, should be maximised" (ICRP, 1991).

Addendum 4.3 - Comparison of the European Directive with WHO and EPA regulations for drinking water

1. 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* (WHO, 2004), 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 (ICRP, 1996), which were adopted by the EU Basic Safety Standards (EC, 1996). 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 (WHO, 1993), which were used as a basis both in drafting the 1998 Drinking Water Directive (EC, 1998) and by the *Article 31 working party* 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 A.4.4. and A.4.5. the results of these calculations are

reported for main radionuclides in a nuclear emergency and for main radionuclides of natural origin, respectively.

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 (Risica, 2000). In order to stress the point, in Tables A.4.6. and A.4.7. WHO guidance levels are compared with those obtained from the European DWCs for the critical age, rounded 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, when e. g. radium isotopes are the prevailing radioactive components, the suggested screening levels both for gross alpha and for gross beta activity may be not enough protective, not guaranteeing the compliance with the *recommended reference dose level* of 0.1 mSv/y, as discussed in paragraph 6.1.

Table A.4.4. *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 A.4.5. *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 (WHO, 2004).

Table A.4.6. 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 A.4.7. 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 (WHO, 2004).

2. 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 (EPA, 2000) 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 (EPA, 1999) 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 A.4.8. in SI units. No specific value of activity concentration for other radionuclides is given.

Table A.4.8. EPA maximum contaminant levels

Parameter	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.

3. 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 (EC, 1998), nor *guidance levels* that, if exceeded, "...should be regarded as an indication that further investigation... is needed" (WHO, 2004), 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 (EC, 1998) or *the recommended reference dose level* (WHO, 2004)
- the MCL for the sum of ^{226}Ra and ^{228}Ra is much lower than those of the single radionuclides calculated starting from the TID of the European Directive (DWC for adults 0.5 Bq/L and 0.2 Bq/L, respectively) and than that of WHO
- a specific MCL for ^{210}Po is not provided for, because ^{210}Po is included in gross alpha; however, ^{210}Po is excluded by the European Directive (but, as reported above, a proposal for its limitation was issued in the 2001 Euratom Recommendation (EC, 2001)), but included in the WHO guidance levels (with a value of 0.1 Bq/L)

- ^{210}Pb is explicitly not regulated by EPA, excluded by the European Directive (but, as for Po-210, included in (EC, 2001)), 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.

Addendum 4.4 - 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 (EC, 1998), 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 (EC, 1998), the 2001 Recommendation (EC, 2001) 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 undertaking 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 considerations 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 (EC, 1996).

Addendum 4.5 - Uranium in drinking water and its limitation

1. The European Directive Approach

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 TID (see Table A.2) 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 (corresponding to 24.8 mBq/L of U-238) (WHO, 1998a), 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" (WHO, 1998b).

In the last edition of the Guidelines (WHO, 2004) 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 (EPA, 2000), as already shown in Table A.8, EPA set a maximum contaminant level of 30 µg/L for uranium, that is “protective of both kidney toxicity and 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. Discussion

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. CORRELATIONS BETWEEN ANALYTICAL PARAMETERS

5.1. Purposes

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

In literature straightforward connections between chemical and radiochemical parameters or between groups of different isotopes can hardly be found (IAEA 1990, Cothorn 1990), nevertheless the peculiarity of Estonian water stimulated such an attempt.

5.2. Radium vs. chemical parameters

Statistical analysis was affected by the limited amount of samples with both radium isotopes data and complete chemical analyses. The group of Cambrian-Vendian water was examined first (56 data).

In Tab. 5.1 correlation coefficients R between ^{226}Ra or ^{228}Ra activity concentrations and some chemical parameters are reported. The strongest correlation ($R=0.802$) is found between ^{228}Ra activity concentrations and water mineralization (the sum of chemical macro-components, approximately the total salt content). A weaker relation ($R=0.590$) is between mineralization and ^{226}Ra . Scatterplot and linear regression between ^{228}Ra and mineralization is shown in Fig. 5.1. Cambrian-Vendian water are not more mineralised than more superficial waters (Ordovician-Cambrian, Silurian-Ordovician, Quaternary Devonian) as shown in Tab. 5.2, so the correlation between total salts and radium is a specific feature of this group of waters actually.

Other chemical parameters as total residue, sodium, magnesium, calcium and chloride concentrations, show similar high correlations with radium isotopes (Tab. 5.1): this is reasonable since both mineralization and total residue are proportional to the amount of dissolved salts, sodium magnesium calcium and chloride are a major component of them. Total residue, sodium, magnesium, calcium and chloride

concentrations exhibit strong correlations (not reported here) with mineralization (R= 0.80-0.99).

Table 5.1 *Correlations between chemical parameters and radium isotope activity concentrations (expressed as correlation coefficient R)*

	²²⁶ Ra	²²⁸ Ra
Mineralization	0.590	0.802
Residue	0.554	0.788
Na	0.500	0.785
K	0.445	0.625
NH₄	0.118	0.154
Ca	0.555	0.755
Mg	0.558	0.628
Fe²⁺	0.169	0.324
Fe³⁺	0.004	-0.087
Al	0.131	-0.040
Mn	0.156	-0.011
Ba	0.204	0.347
Cl	0.446	0.621
SO₄	-0.347	-0.211
PO₄	0.229	0.041
HCO₃	0.388	0.218
CO₂	0.300	0.176
SiO₂	-0.083	0.258
Oxidability	0.170	0.245
pH	-0.069	-0.160

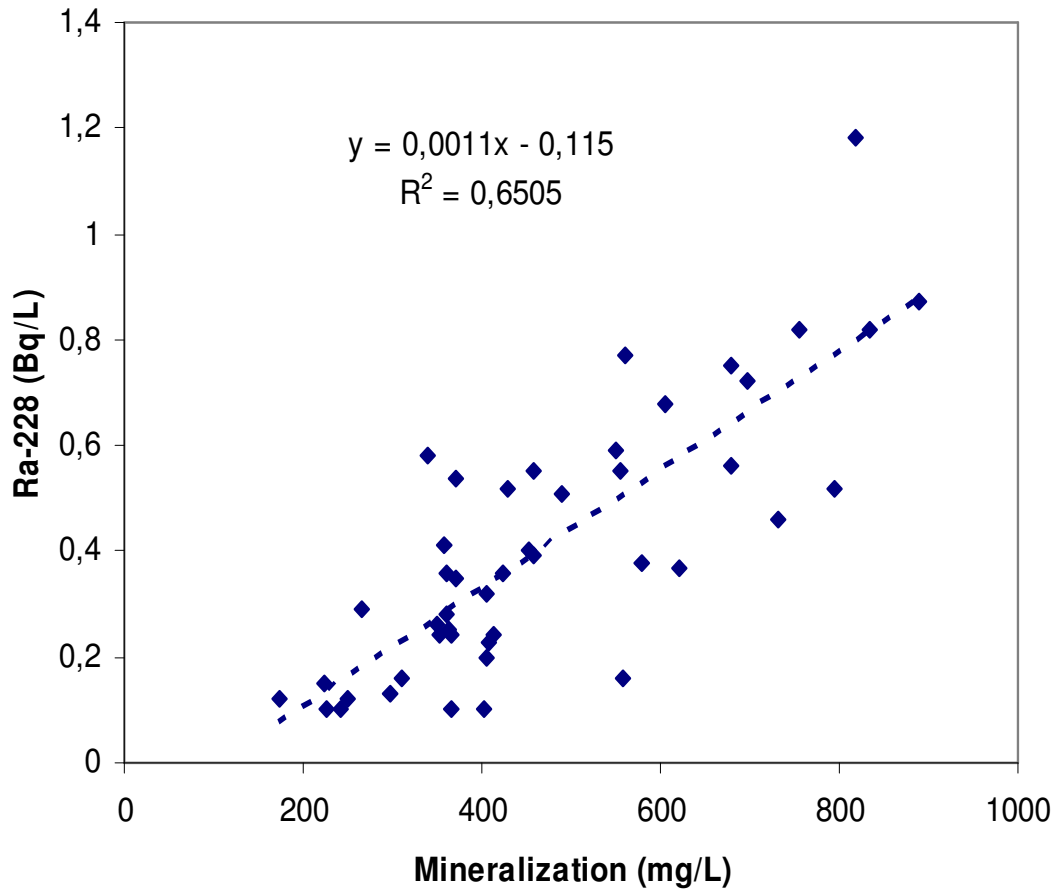


Figure 5.1 Scatterplot (square points) and linear regression (dashed curve) between ²²⁸Ra and mineralization in Cambrian Vendian waters

Table 5.2 Distribution of mineralization (total salt content) in different waters

Water/ aquifer	N° of samples	Mean (mg/L)	25° percentile (mg/L)	Median (mg/L)	95° percentile (mg/L)
Quaternary Devonian	18	421	362	451	556
Silurian Ordovician	18	510	452	487	680
Ordovician Cambrian	56	524	367	435	1017
Cambrian Vendian	207	482	340	390	819

It is interesting to note that the actually weak correlation with sulphates and pH (Tab. 5.1), is a reverse one. This is reasonable since coprecipitation of radium should increase with higher pH or higher sulphate concentration.

Since other parameters, as barium, exhibit potentially interesting correlation a multiparametric analysis was attempted. In Tab. 5.3 we report the percentages of explained variance, of both radium isotopes (${}^i\text{Ra}$, $i=226, 228$), through the following linear model:

$${}^i\text{Ra} = \alpha_1 + \alpha_2 \text{Chemical1} + \alpha_3 \text{Chemical2} + \alpha_4 \text{Chemical3} \quad (1)$$

Table 5.3 Percentages of explained variance of radium isotopes vs. combined chemical parameters-model (1)

Chemical 1	Chemical 2	Chemical 3	${}^{226}\text{Ra}$	${}^{228}\text{Ra}$	Number of observations
Mineral.	-	-	35%	64%	46
Mineral.	Ba	-	41%	71%	34
Mineral.	Fe^{2+}	-	40%	66%	46
Mineral.	Cl	-	38%	64%	46
Cl	Ba	-	27%	57%	38
Fe^{2+}	Cl	-	21%	43%	56
Fe^{2+}	Ba	-	43%	17%	38
Mineral.	Fe^{2+}	Ba	42%	72%	34
Mineral.	Cl	Ba	54%	72%	34
Mineral.	Fe^{2+}	Cl	43%	67%	46
Fe^{2+}	Cl	Ba	28%	57%	38

A slight increase of the explained variability is obtained if barium is considered together with the mineralization, though the increase is partly due to the further reduction of the data set (e.g. the mineralization plus barium concentration was

available for only 34 samples). The future application of this statistical approach will be likely meaningful on a wider set of data.

It should be finally noted that the parameters α_i ($i=1, 2, 3\dots$) in equation (1) of the linear regressions (not reported here) between all chosen group of chemical parameters and respectively ^{226}Ra and ^{228}Ra are very similar. This interesting feature allows us to conclude that chemical parameters affect the radium isotopes in the same way.

5.3. ^{226}Ra vs. ^{228}Ra

Literature clearly shows that no relationship between ^{226}Ra and ^{228}Ra should be *a priori* expected (IAEA 1990, Cothorn 1990, Ivanovich 1992) since they belong to two different natural series which can have very different abundance in different rocks; moreover their parent radionuclides have a very different chemical behaviour.

Nevertheless results reported in the previous chapter stimulated us to investigate the relationship between ^{226}Ra and ^{228}Ra . 191 couples of ^{226}Ra and ^{228}Ra activity concentration values were used as input parameters.

The first step was a nonparametric analysis in order to identify a possible parametric dependence structure between the two variables. As shown in Fig. 5.2, the nonparametric curve (Kernel regression, red line), is quite linear, so a linear regression was chosen. Statistical methods have thus been performed in order to verify the significance of the linear regression. In particular, the usual test statistics (t-values and p-values, not reported here) support the use of the linear model at the usual significance level of 95%. Parameters of the regression model are reported in Tab. 5.4.

Table 5.4 *Parameters of linear regression (^{226}Ra vs. ^{228}Ra)*

Parameter	Value	Std. err.
Intercept	0.07399	0.01890
Angular coeff.	0.65322	0.04422
R^2	0.536	-

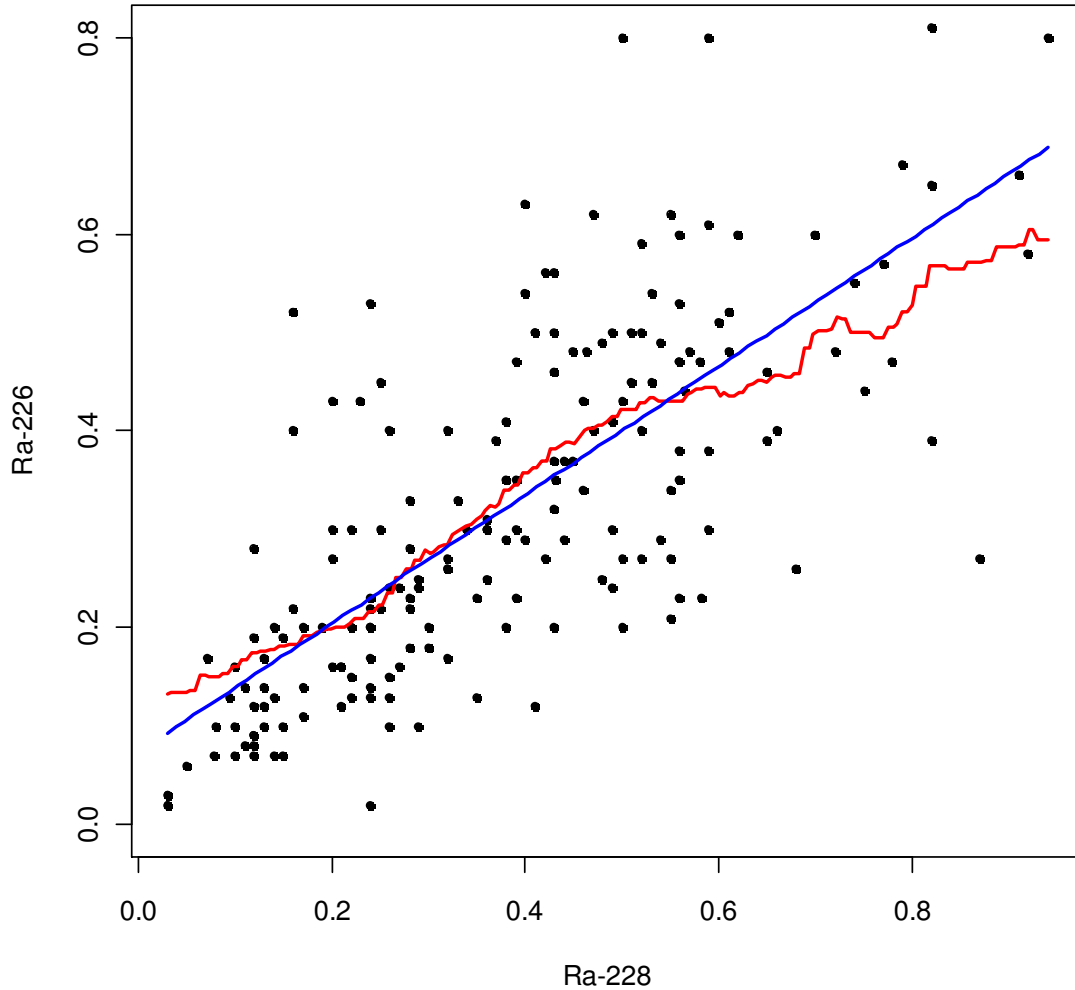


Figure 5.2 Scatterplot (black points), nonparametric curve obtained using Kernel regression (red line) and linear regression (blue line) for ^{226}Ra vs. ^{228}Ra (Bq/L).

Although further refinements as confidence intervals calculations should still be done, this correlation has the advantage of allowing the rough estimate of ^{226}Ra concentration from the experimentally measured activity concentration of ^{228}Ra . If the estimated linear regression in Tab 5.4 is used, the ^{226}Ra concentration can be calculated by the following equation:

$$\hat{C}_{\text{Ra}226} = 0.65322 * C_{\text{Ra}228} + 0.07399 \quad (2)$$

where \hat{C}_{Ra226} and C_{Ra228} are, respectively, the estimated ^{226}Ra and the measured ^{228}Ra activity concentrations in Bq/L. From these values, the dose for the two radium isotopes and then the total dose can be estimated. Applying the ingestion dose coefficients reported in Tab 4.3, the following equation can be obtained:

$$D_{\text{adults}} = 0.6372 * C_{Ra228} + 0.0151 \quad (3)$$

where D_{adults} is the total ingestion dose for adult age class in mSv/y. In Fig. 5.3 a graphic representation is given. Despite the similar activity concentration of the two radium isotopes, the contribution to the total dose is higher for ^{228}Ra (see 4.3). Since the relative contribution of ^{226}Ra is small, even a rough estimation of it will not heavily affect the total dose evaluation.

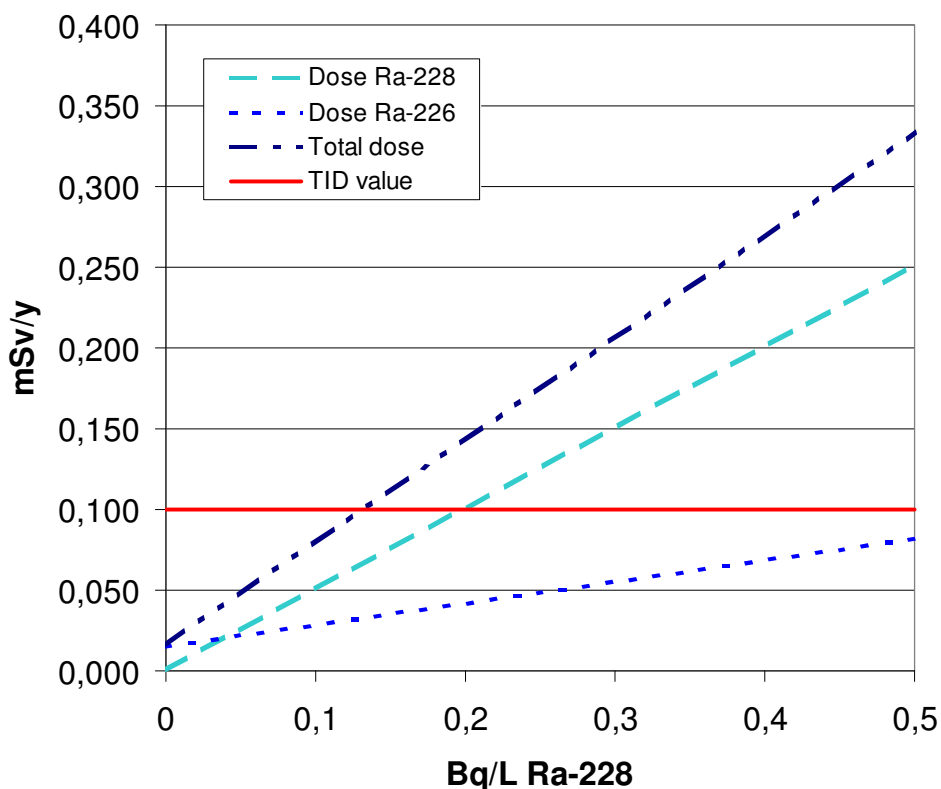


Figure. 5.3 Dependence of dose from ^{228}Ra activity concentrations: relative contributions of ^{226}Ra and ^{228}Ra

In this simplified treatment some specific assumptions related to modelling relationship between variables with uncertainty have been made (e.g. constant uncertainties of experimental parameters) (Wald 1940 pag. 298).

As an example in Fig. 5.4 and 5.5 the graph of total dose calculated from ^{228}Ra concentration are reported together with confidence intervals. The width of confidence interval depends on both linear regression and uncertainty of ^{228}Ra concentrations. The blue dashed lines provide confidence intervals (at 95% level) related to linear regression (Formula 2), the red dashed lines represent the overall confidence intervals assuming a fixed uncertainty of ± 0.01 Bq/L for ^{228}Ra concentrations.

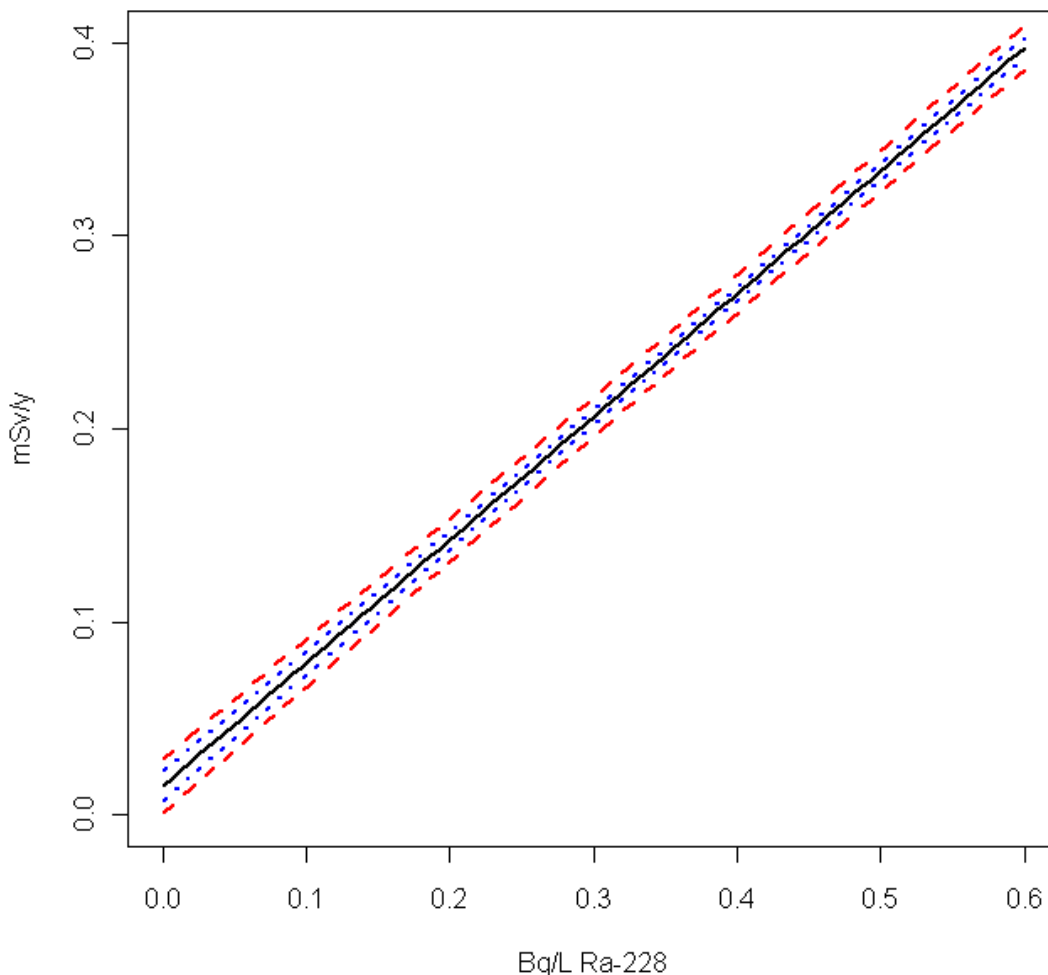


Figure 5.4 Total dose as a function of ^{228}Ra (black line) and confidence intervals (95% level) considering linear regression between ^{228}Ra and ^{226}Ra (blue lines) and with ^{228}Ra measurement uncertainty (0.01 Bq/L) contribution added (red line)

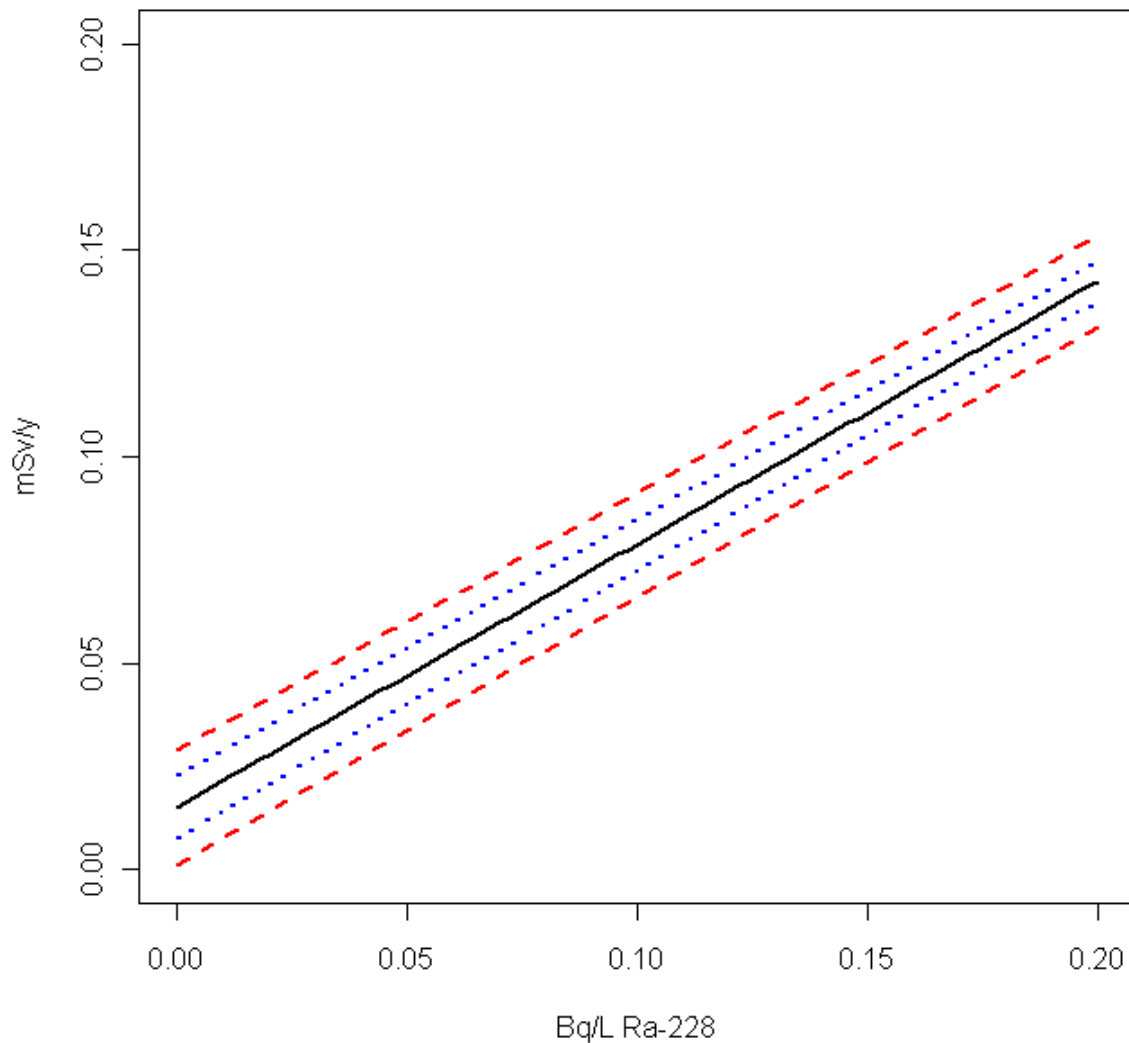


Figure 5.5 *Expansion of graph in Fig. 5.4.*

5.4. Final considerations

Some preliminary results can be drawn from the analysis of correlations between chemical and radiometric parameters.

- Within the Cambrian-Vendian water group the variability of radium isotopes concentration can be explained by the water mineralization degree and, at a lower extent, by barium concentration. This correlation could be useful as a preliminary screening and in addressing water monitoring programs.

- A statistically significant correlation between ^{226}Ra and ^{228}Ra is found. This partially contradicts literature data but it may be justified if the Cambrian-Vendian aquifer can be considered relatively homogeneous in the studied area. As both rock characteristics and chemical-physical feature of the aquifer are relatively similar only quantitative differences in dissolved solids may occur.
- A rough total dose evaluation can be done using only ^{228}Ra concentration, since the contribution of ^{226}Ra is smaller and moreover it can be calculated from the concentration of ^{228}Ra by the estimated linear model. A first evaluation, shows that a total indicative dose value of 0.1 mSv/y should be reached when a 0.12 – 0.15 Bq/L concentration of ^{228}Ra occurs.

All these findings should be considered provisional since the data set, especially for chemical correlation, was small and they should be confirmed with the analysis of a wider one. Statistical analysis and correlation studies are yet a useful tool to be developed especially to provide suitable estimator for anticipatory purposes.

6. CHOICE OF RELEVANT PARAMETERS AND ANALYTICAL TECHNIQUES

6.1. Gross alpha and beta activities

WHO recommends the measurement of gross alpha and beta activities as a 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 ^{226}Ra will lead to a 0.1 mSv/y total indicative dose (TID) by itself and 1 Bq/L of the beta emitter ^{228}Ra will lead to much higher doses.

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

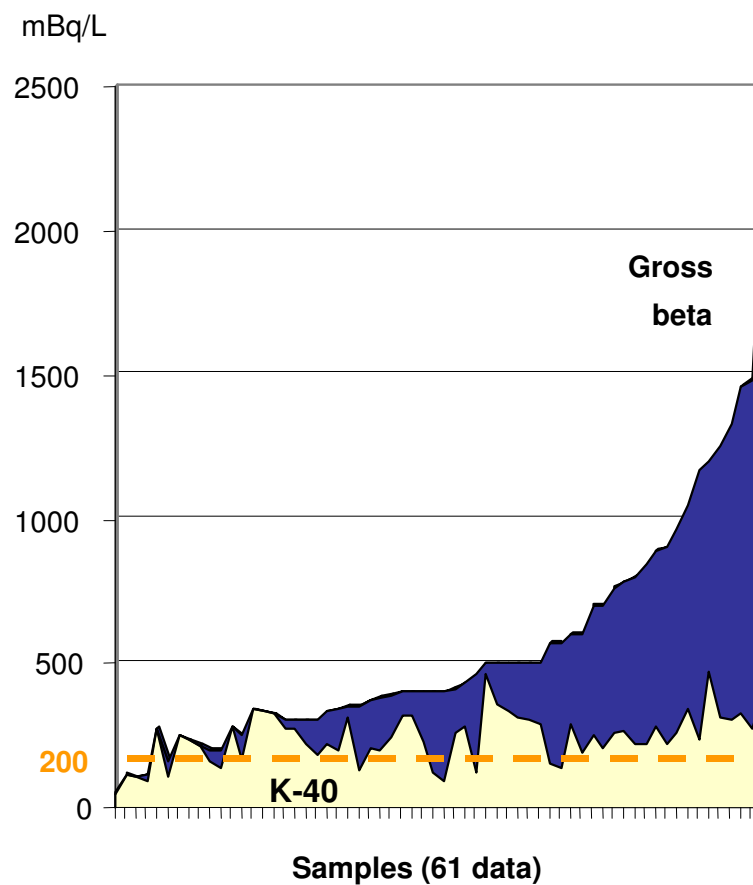


Figure 6.1 ^{40}K contribution to gross beta activity concentration (mBq/L)

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

By examining the database results, the average contribution of ^{40}K to the beta activity is 0.24 Bq/L (Fig. 6.1). ^{40}K is the prevailing contributor to gross beta activity in more than 50% of the samples (Fig. 6.2)

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

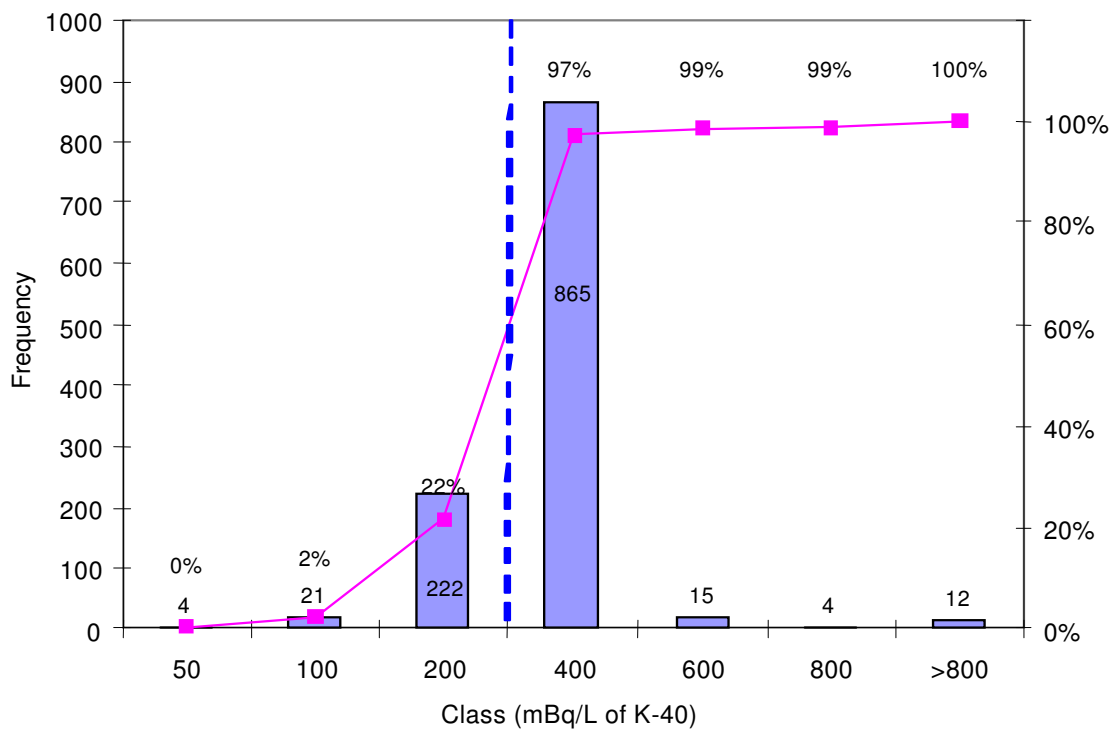


Figure 6.2 *Frequency histogram of ^{40}K activity concentration based on chemical measurement of potassium (1143 input data)*

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

6.2. Radium isotopes analysis

Radium-228

The ^{228}Ra concentration is the most relevant radiometric parameter in Estonian waters since it provides a major contribution to the total dose and thus it should be always measured.

Many analytical methods are available in literature or as standard methods. Some are based on the measure of beta emission by low background beta counter of ^{228}Ac (decay product of ^{228}Ra) after radiochemical separation of ^{228}Ra and 15 days actinium ingrowth. (EPA 1986-1).

Gamma spectrometry measurement of the daughter ^{228}Ac provides a lower sensitivity but it is a quicker and less cumbersome technique. The water sample has to be concentrated, transferred into an appropriate container and counted without any chemical treatment. As an example, if a 30% efficiency hyperpure germanium detector (HPGe) is used and a Minimum Detectable Activity (MDA) of 50 mBq/L is desired, a water sample of 8 L should be used (4000 min counting).

The concentration of such an amount of water can be performed:

- by evaporation of the water sample to dryness. Neither special equipment nor operator skill are required, nevertheless both the final volume of obtained solid and its chemical composition depends on water features. The calibration should take into account the possible variations of geometry and autoabsorption.
- by elution on ion-exchange resin. Both cation and mixed bed resin can be used in amount depending on the salinity of the sample (approximately some hundreds of millilitres of resin are needed for a 20 L sample). The retaining efficiency is very high (often 100% can be assumed) and resins with a saturation indicator are available. The resin is then dried and counted. This method is routinely applied by ARPA Lombardia, the detailed procedure was given in Technical Report 1.

Radium-226

The analysis of ^{226}Ra concentrations is often performed indirectly by the measurement of the daughter ^{222}Rn (and descendants) at isotopic equilibrium. The advantage is the ease of purification and extraction of the radon since it is a noble gas. The disadvantages are the time required for achieving the equilibrium and the difficulty of avoiding radon losses. Widely employed techniques are reported below.

- Emanometric technique: the water sample is sealed in a tight container till the equilibrium with the daughter ^{222}Rn is established. Then the radon is removed by flushing an inert gas in the container, transferred in a scintillation cell (Lucas cell) and then counted. The water sample can be previously concentrated by thermal evaporation, by sulphate precipitation or by ion exchange resins elution (EPA 1986-2, ASTM 1997-1).
- Liquid scintillation counting (LSC): double phase method can be applied: a small sample (around 10 mL) is transferred in a scintillation vial. A scintillation cocktail not miscible with water is added. The ^{222}Rn produced by the decay of ^{226}Ra is selectively adsorbed by the scintillation cocktail and is counted as the isotopic equilibrium is attained. (Forte 2004), The LSC procedure used by ARPA Lombardia was provided in previous Technical Reports, but care should be paid since a small interference by ^{228}Ra may occur. Single phase method can also be used, in this case a scintillation cocktail miscible with water is employed. Radium is previously purified by sulphate precipitation and counted applying alpha beta discrimination and choosing suitable windows (Yong-Jae Kim 2001, Repinc 2002).
- Gamma spectrometry: if ^{226}Ra is measured directly the gamma line 186 keV is used and the possible interference by ^{235}U should be taken into account. If the measurement of radium through the gamma emission of its progeny is desired, radon tight containers should be employed (e.g. a steel container with o-ring lid is suitable). The simple sealing of plastic Marinelli beakers does not ensure the complete radon retention.
- Alpha spectrometry: radium is chemically purified, electrodeposited and counted by high resolution alpha spectrometry. The sensitivity of this method is high and ^{224}Ra can be determined as well. Nevertheless it is rather cumbersome and the evaluation of chemical yield is difficult (Martin 2004). An interesting alternative is

the use of manganese dioxide coated filters which can selectively absorb radium from water. Disks are then counted by alpha spectrometry (Eikenberg 2001).

Radium-226 and Radium-228 simultaneous measurement

In last years some procedures for the simultaneous determination of ^{226}Ra and ^{228}Ra have been published, based either on gamma spectrometry or liquid scintillation counting. A previous concentration and purification of radium is always required. Since no convenient tracer or stable carrier is available, the determination of chemical recovery is not easy. In some cases it is assumed quantitative (100%), in other cases barium is employed as a carrier since its chemical behaviour is close to the radium one. The gamma emitter ^{133}Ba can be also employed, under this hypothesis.

Liquid scintillation counting. The previously described method of radium purification through sulphate precipitation followed by LSC counting can be applied to the simultaneous determination of ^{226}Ra and ^{228}Ra if proper counting windows are chosen. This procedure is currently being tested by IAEA as recommended procedure (Chang-Kyu Kim, 2009).

An interesting alternative for radium concentration and purification is filtration on special filters (Radium Rad Empore disks) which selectively retain radium. Radium or decays products are then eluted and counted by liquid scintillation. Procedures have been published for the quick determination of radium isotopes (Schonhofer 2001, Mobius 2006).

Gamma spectrometry: the previously cited Radium Rad disks have been used for concentrating radium before gamma spectrometry measurement (Rihs 2002). Radium can be also coprecipitated with Pb or Ba as sulphate and measured. An attractive preconcentration method employs coprecipitation with manganese dioxide which can quantitatively absorb radium from large volumes of water (Salih 2002). The precipitated radium is sealed and measured at isotopic equilibrium with descendants. For ^{226}Ra determination gamma lines at 609 and 295, 352 keV of respectively ^{214}Bi and ^{214}Pb were used.

6.3. Other radiometric parameters

Uranium

The measurement of uranium isotopes (^{234}U , ^{235}U and ^{238}U) seems to be not crucial for Estonian water. Available data show that concentrations are not exceptionally high (up to 0.54 Bq/L of ^{238}U). Since the dose coefficient for uranium isotopes is much lower than radium ones, the dose contribution due to uranium is not relevant (usually less than 1%). Nevertheless, since international guidelines suggest a reference value of 15 ppb (corresponding to 0.187 Bq/L of ^{238}U) for uranium due to its chemical toxicity (WHO 2004), its determination can be useful in selected cases. The uranium measurement by ICP-MS is more and more applied due to its ease and quickness (ASTM 1997-2). A rather convenient procedure employs liquid scintillation counting: uranium is extracted with a selective scintillation cocktail and measured by LSC (Forte 2004). In this way the total uranium activity and $^{238}\text{U}/^{234}\text{U}$ ratio can be determined but ^{235}U cannot, due to the low resolution of the technique. The three isotopes can be measured with high sensitivity if high resolution alpha spectrometry is applied on radiochemically purified and electrodeposited samples (ASTM 1997-3). Unfortunately this technique is too cumbersome to be applied to wide range monitoring.

^{222}Rn , ^{210}Pb and ^{210}Po

These radionuclides are not included in the scope of the European Directive 98/83 (EC 1998), on the opposite they are included in that of a recent European recommendation (EC 2001).

or the measurement of radon many quick and simple procedures are available. The key point is the water sampling since the radon gas can be easily desorbed from the sample.

Attention should be paid to lead-210 and polonium-210 because of their high dose coefficient. The few available measurements of Estonian water show that their concentration is not always negligible, thus the measurement of both radionuclides should be advisable at least in selected cases.

Polonium-210 is usually measured by alpha spectrometry on deposited samples (autodeposition on metal disk can be achieved in controlled conditions) (Matthews 2007).

Lead can be determined along with radium with some of the above described procedures or by specific methods (radiochemical separation followed by gross beta counting or liquid scintillation counting) (Desideri 1995, Forte 2009).

7. SAMPLING PLAN

In planning a monitoring campaign for the control of radioactivity it should be always kept in mind that radiometric analyses are lengthy and more cumbersome than chemical ones, hence a special care has to be paid in maximizing results with a sustainable amount of measurements. Representativeness of data is another key point as complex situations (like a nation-wide survey) are considered. Finally a choice has to be made in connection to the aim of the study since criteria may be different if a deep knowledge of phenomenon rather than the evaluation of dose (and risk) to population is desired.

The main outcomes of the previous analysis of the Estonian situation are resumed in the following as they can help to define the best suited sampling plans:

- Estonian waters belong to two major groups, one of them taking water from Cambrian-Vendian (mainly in Northern Estonia) aquifers and the other from non Cambrian-Vendian aquifers (Central and Southern Estonia);
- almost all waters from Cambrian-Vendian aquifers (140 out of 912), which supply most of the water in Northern counties, show high radioactivity concentrations, being ^{228}Ra the main contributor;
- waters from non Cambrian-Vendian aquifers (772 out of 912) look 'safer' but the number of available data is low and not all the values comply with relevant reference values (i.e. DWCs); the number of available data in counties others than the Northern ones is very limited and not sufficient;
- most Estonian water supply zones are small sized ones (83% of WSZs serves 500 people or less) and this percentage is almost the same in any county; on the other hand, the 15 largest zones (see Table 7.1) serve altogether about 740,000 people (67% of Estonian population); the 12 biggest water supply zones serve about 150,000 people (see Table 7.2).
- within the Cambrian-Vendian water group a correlation between chemical and radium parameters has been found: higher radium concentrations correspond to higher water mineralization degree and, at a lower extent, to higher barium concentration (see par. 5.2).
- a statistically significant correlation between ^{226}Ra and ^{228}Ra is generally found;

- gross alpha and beta activities are not a good radiological descriptors for Estonian waters due to high ^{40}K concentration and its contribution to gross beta activity;
- water treatment plants can lower the radioactivity of water but the effectiveness seems to be highly variable;
- most available data are related to water radioactivity at the well (before any treatment).

On the basis of the data and information reported above, some hints for a general outline of a control plan can be given.

Choice of sampling points

- Since the aim of the campaign should be the assessment of actual radiation dose to Estonian population, water should be measured as delivered to consumers. Thus the sampling should be carried out at the distribution network (downstream pumping stations) in order to take into account any variation in water radioactivity due to mixing of different wells or to purification treatments. In case of particularly complex aqueducts with several distribution hubs, sampling in one point of each hub is suggested. Samples can only be drawn directly at wells if the water is neither mixed nor processed before delivery.
- Within Cm-V group WSZs (Northern Estonia), a further monitoring should be carried out in order to collect data for safe dose assessment; on this basis, the need for countermeasures can be evaluated reliably. As the total number of Cm-V aquifers is quite high (140), it is advisable to start the monitoring from those distributing water to the largest part of the population; the list of the 12 biggest WSZs (supplying water to 60% of people using Cm-V waters) is reported in Tab. 7.2. The possibility of using chemical parameters like mineralization degree and barium concentration (see par. 5.2) as indicator for most radioactive waters from Cm-V aquifer could be considered.
- The representativeness of water monitoring should be increased for Central and Southern Estonia (and for non Cm-V aquifers in general).

- Largest supply zones in different counties should therefore be selected and monitored. Table 7.3 shows the percentage of coverage (in terms of monitored population) of the proposed plan under the following two hypotheses:
 - a. the 3 biggest WSZs in each county are monitored: in this case the monitoring plan would enable to assess the dose for 72% of Estonian population (minimum and maximum values in different counties being 44% and 86%, respectively);
 - b. the 10 biggest WSZs in each county are monitored: in this case the monitoring plan would enable to assess the dose for 85% of Estonian population (minimum and maximum values in different counties being 65% and 98%, respectively). Under this hypothesis the increase in percentage of monitored population is limited as most WSZs are very small.

As a further refinement, the choice of monitored WSZs could also take into account their location in order to guarantee an even distribution over the whole territory; the list of the 10 biggest WSZs in each county is reported in Tables from 7.4 to 7.18.

Choice of the analytical approach

- The analytical parameters to be determined are radium isotopes, mainly ^{228}Ra as it is the main contributor to dose and it is fairly correlated to ^{226}Ra as well (see 4.3 and 5.3).
- As far as reference levels for radium isotopes are concerned, the DWL (Derived Water Level) proposed in the database commissioned by the European Union (Risica 2000), i.e. 0.2 Bq/L for ^{228}Ra and 0.5 Bq/L for ^{226}Ra , are recommended. They ensure compliance with the parametric value for the committed dose of 0.1 mSv/year calculated in the hypothesis that the ingested water contains only the interested isotope, the age individual is an adult and that the water consumption is 730 L/year. Taking into account that both radium isotopes (^{228}Ra and ^{226}Ra) are present, and on the basis of the analysis discussed in paragraph 5.3, we suggest to adopt the following rule of thumb:
 - waters with $^{228}\text{Ra} < 0.1$ Bq/L reasonably produce TID values lower than 0.1 mSv/y;

- waters with $^{228}\text{Ra} > 0.15 \text{ Bq/L}$ reasonably produce TID values higher than 0.1 mSv/y ;
 - waters with: $0.1 \text{ Bq/L} < ^{228}\text{Ra} < 0.15 \text{ Bq/L}$ reasonably need further investigation and direct measurement of ^{226}Ra concentration.
- A careful evaluation of existing analytical resources should be carried on. The choice of analytical methods and priorities should take into account the availability of them: if gamma spectrometry analyses were for instance readily available, an extensive measurement campaign based firstly on measurement of ^{228}Ra by gamma spectrometry on concentrated water samples would be recommended.
 - It is worth reminding the (future) need to monitor other radionuclides content (e.g. ^{210}Pb and ^{210}Po) in Estonian waters, as they are possibly present in significant amount and can give an important contribution to radiation dose.

Monitoring frequency

- The repetition of the sampling and measurement campaign (at least two times) is advisable, as significant variations of the content of natural radioactivity are sometimes observed in aqueducts fed by several supply wells when mixing rates are modified; furthermore, the use of treatment plants before water delivery can affect water final radioactivity at a variable extent. Controls should be repeated on the basis of a program established at a National level.
- It has been shown that overexploitation of water layers in Cm-V aquifer is causing a variation in chemical composition (increase of total residue and barium) due to the drawing up of deep water contained in the fractured crystalline basement (Karro 2004, Marandi 2007). In these cases the follow up of temporal variations of radioactivity in selected points could be useful. Sampling should be performed at the well before any treatment and highly exploited Cambrian-Vendian aquifers should be chosen.

Table 7.1 *Biggest Estonian water supply zones (serving more than 10000 people)*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
1	Harjumaa	Tallinn	Tallinn ground water WSZ	AS Tallinna Vesi	130000	350000	non Cm-V (surface water)
519	Tartumaa	Tartu town	Tartu town and Kõrveküla village WSZ	Tartu Veevärk AS	14500	99000	non Cm-V (Q, D2tr, D2pr-S, O-C)
852	Ida-Virumaa	Narva town	Narva town WSZ	Narva Vesi AS	8994	67497	non Cm-V (surface water)
250	Pärnumaa	Pärnu town	Pärnu town	Pärnu Vesi AS	7000	34350	non Cm-V (S, D2-1-S)
6	Harjumaa	Tallinn	Nõmme wells WSZ no 7	AS Tallinna Vesi	4800	33300	Cm-V (Cm-V, O-C)
362	Viljandimaa	Viljandi town and Viiratsi settlement	Viljandi town and Viiratsi settlement WSZ	AS Viljandi Veevärk	2410	20328	non Cm-V (S)
835	Ida-Virumaa	Kohtla-Järve town	Kohtla-Järve town Järve district, Tākumetsa and Peeri villages WSZ	Järve Biopuhastus OÜ	2364	19689	Cm-V (Cm-V)
838	Ida-Virumaa	Kohtla-Järve town	Kohtla-Järve town Ahtme district WSZ	Järve Biopuhastus OÜ	2813	19300	non Cm-V (Q)
742	Lääne-Virumaa	Rakvere town	Rakvere town WSZ	Rakvere Vesi AS	1500	16000	Cm-V (Cm-V)
288	Saaremaa	Kuressaare town	Kuressaare WSZ	Kuressaare Veevärk AS	2800	15950	non Cm-V (S)

- Continues -

- Continues -

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/ day	Population served	Aquifer group
851	Ida-Virumaa	Sillamäe town	Sillamäe town WSZ	Sillamäe Veevärk AS	2053	15680	Cm-V (V2vr)
194	Läänemaa	Haapsalu town	Haapsalu	Haapsalu Veevärk AS	1600	13000	Cm-V (Cm-V, O-C)
846	Ida-Virumaa	Jõhvi town	Jõhvi town WSZ	Jõhvi Veemajandus OÜ	Q: 712,5 V2gd: 1877,6 Total: 2590	12400	Cm-V (Q, V2gd)
424	Valgamaa	Valga town	Valga town WSZ	Valga Vesi AS	9600	10800	non Cm-V (D2)
350	Võrumaa	Võru town	Võru town	Võru Vesi AS	1300	10000	non Cm-V (D2)
Tot. population: (67% of Estonian population)						737294	

Table 7.2 *Biggest Cambrian-Vendian water supply zones (serving more than 5000 people)*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer
6	Harjumaa	Tallinn	Nõmme wells WSZ no 7	AS Tallinna Vesi	4800	33300	Cm-V, O-C
835	Ida-Virumaa	Kohtla-Järve town	Kohtla-Järve town Järve district, Tākumetsa and Peeri villages WSZ	Järve Biopuhastus OÜ	2364	19689	Cm-V
742	Lääne-Virumaa	Rakvere town	Rakvere town WSZ	Rakvere Vesi AS	1500	16000	Cm-V
851	Ida-Virumaa	Sillamäe town	Sillamäe town WSZ	Sillamäe Veevärk AS	2053	15680	V2vr
194	Läänemaa	Haapsalu town	Haapsalu	Haapsalu Veevärk AS	1600	13000	Cm-V, O-C
846	Ida-Virumaa	Jõhvi town	Jõhvi town WSZ	Jõhvi Veemajandus OÜ	2590	12400	Q, V2gd
9	Harjumaa	Maardu town	Kallavere south district (old)	AS Maardu Vesi	1500	9700	V2gd
16	Harjumaa	Keila town	Keila town WSZ	AS Keila Vesi	1100	9258	Cm-V
849	Ida-Virumaa	Kiviõli town	Kiviõli town WSZ	Kiviõli Vesi OÜ	653	6734	Cm-V, V2gd
10	Harjumaa	Maardu town	Kallavere north district	AS Maardu Vesi	650	5700	V2gd
2	Harjumaa	Saue town	Saue wells WSZ no 8	AS Tallinna Vesi	1800	5200	Cm-V, O-C
98	Harjumaa	Viimsi municipality	Lääneranniku WSZ	AS Viimsi Vesi	1400	5012	Cm-V x 12
Tot. population: (about 60% of people using Cm-V waters)						151673	

Table 7.3 *Percentage of people served by the 3 (and 10) biggest WSZs*

County	Population	Total No. of WSZs	Pop. served by the 3 biggest WSZs	% Pop. served by the 3 biggest WSZs	Pop. served by the 10 biggest WSZs	% Pop. served by the 10 biggest WSZs
Hiiumaa	6588	14	4820	73,2	6428	97,6
Laanemaa	21870	35	15700	71,8	18760	85,8
Harjumaa	510201	172	393000	77,0	432270	84,7
Laane-Virumaa	49044	89	25495	52,0	35792	73,0
Ida-Virumaa	167833	85	106486	63,4	149031	88,8
Raplamaa	22437	51	10065	44,9	15807	70,5
Jarvamaa	22527	59	13475	59,8	17040	75,6
Jogevamaa	20403	48	8900	43,6	13207	64,7
Saaremaa	23260	27	17250	74,2	20615	88,6
Parnumaa	55042	59	39150	71,1	46177	83,9
Viljandimaa	35782	62	22840	63,8	27250	76,2
Tartumaa	120002	74	103300	86,1	108661	90,5
Valgamaa	20314	48	12750	62,8	15645	77,0
Polvamaa	15186	42	7530	49,6	10490	69,1
Vorumaa	19726	47	11380	57,7	14228	72,1
Estonia	1100215	912	792141	72,0	931401	84,7

Table 7.4 *Biggest water supply zones in Estonian Counties - Hiiumaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
193	Hiiumaa	Kärdla town, Palade village	Kärdla-Palade	Kärdla Veevärk AS	250	3100	non Cm-V (O)
192	Hiiumaa	Käina settlement	Käina settlement	Kärdla Veevärk AS	140	1200	non Cm-V (O-C)
184	Hiiumaa	Kõrgessaare municipality, Lehtma village	Lehtma sadam (port)	Direct.Consulding AS	30	520	non Cm-V (O-C)
187	Hiiumaa	Kõrgessaare settlement, Sadama road	Kõrgessaare settlement	Kõrgessaare Local municipality	80	500	non Cm-V (O-C)
186	Hiiumaa	Kõrgessaare municipality, Lauka village	Lauka village	Kõrgessaare Local municipality	15	280	non Cm-V (O)
189	Hiiumaa	Pühalepa municipality, Suuremõisa village	Suuremõisa	Kärdla Veevärk AS	37	260	non Cm-V (S-O)
183	Hiiumaa	Emmaste municipality, Tärkma village	Tärkma houses	Emmaste municipality EMKO	23,9	200	non Cm-V (S)
181	Hiiumaa	Emmaste municipality, Emmaste village	Emmaste Põhikool (school)	Emmaste municipality EMKO	6,8	180	non Cm-V (S)
191	Hiiumaa	Käina municipality, Männamaa village	Männamaa	Kärdla Veevärk AS	10	118	non Cm-V (O)
182	Hiiumaa	Emmaste municipality, Hindu village	Tohvri hooldekodu (nursing home)	Emmaste municipality EMKO	13	70	non Cm-V (S)

Table 7.5 *Biggest water supply zones in Estonian Counties - Laanemaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
194	Läänemaa	Haapsalu town	Haapsalu	Haapsalu Veevärk AS	1600	13000	Cm-V (Cm-V, O-C)
201	Läänemaa	Lihula town	Lihula centre	Lihula Vesi OÜ	80	1400	non Cm-V (S)
223	Läänemaa	Taebla	Taebla settlement	AS Taebla Kodu	123	1300	non Cm-V (O-C)
211	Läänemaa	Palivere settlement	Palivere	AS Taebla Kodu	72	900	non Cm-V (O-C)
224	Läänemaa	Virtsu municipality	Kaare village	Virtsu Vesi MTÜ.	32	500	non Cm-V (S)
205	Läänemaa	Linnamäe settlement	Linnamäe centre	Linnamäe Kodu	40	400	non Cm-V (O)
197	Läänemaa	Kullamaa municipality	Kullamaa village	Kullamaa Local municipality	55	360	non Cm-V (O)
196	Läänemaa	Koluvere settlement	Koluvere WSZ (nursing home)	Koluvere Hooldekodu	70	350	non Cm-V (S-O)
215	Läänemaa	Pürksi municipality	Pürksi 1	Noarootsi Soojus OÜ	77	300	non Cm-V (O-C)
213	Läänemaa	Panga village	Panga	Pangavesi AS	17	250	non Cm-V (S)

Table 7.6 *Biggest water supply zones in Estonian Counties - Harjumaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/ day	Population served	Aquifer group
1	Harjumaa	Tallinn	Tallinn ground water WSZ	AS Tallinna Vesi	130000	350000	non Cm-V (surface water)
6	Harjumaa	Tallinn	Nõmme wells WSZ no 7	AS Tallinna Vesi	4800	33300	Cm-V (Cm-V, O-C)
9	Harjumaa	Maardu town	Kallavere south district (old)	AS Maardu Vesi	1500	9700	Cm-V (V2gd)
16	Harjumaa	Keila town	Keila town WSZ	AS Keila Vesi	1100	9258	Cm-V (Cm-V)
10	Harjumaa	Maardu town	Kallavere north district	AS Maardu Vesi	650	5700	Cm-V (V2gd)
2	Harjumaa	Saue town	Saue wells WSZ no 8	AS Tallinna Vesi	1800	5200	Cm-V (Cm-V, O-C)
98	Harjumaa	Viimsi municipality	Lääneranniku WSZ	AS Viimsi Vesi	1400	5012	Cm-V (Cm-V x 12)
11	Harjumaa	Paldiski town	Paldiski town WSZ	Paldiski Linnahoolduse OÜ	1388	4800	Cm-V (Cm-V, V2gd)
13	Harjumaa	Saku municipality	Saku settlement WSZ	AS Saku Maja	680	4800	Cm-V (V2gd)
84	Harjumaa	Saue municipality	Laagri settlement WSZ	AS Kovek	1000	4500	Cm-V (Cm-Vx4)

Table 7.7 Biggest water supply zones in Estonian Counties – Laane-Virumaa

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
742	Lääne-Virumaa	Rakvere town	Rakvere town WSZ	Rakvere Vesi AS	1500	16000	Cm-V (Cm-V)
756	Lääne-Virumaa	Tapa	Tapa town WSZ	Tapa Vesi AS	980	4995	non Cm-V (O)
719	Lääne-Virumaa	Kunda town	Kunda town WSZ	Kunda Vesi AS	478	4500	Cm-V (Cm-V)
755	Lääne-Virumaa	Tamsalu	Tamsalu town WSZ	Tamsalu Vesi AS	290	2630	non Cm-V (O)
711	Lääne-Virumaa	Kadrina settlement	Kadrina WSZ	Kadrina Soojus AS	260	2055	non Cm-V (O-C)
781	Lääne-Virumaa	Väike-Maarja	Väike-Maarja Tammi WSZ	Pandivere Vesi AS	80	1500	non Cm-V (S-O)
751	Lääne-Virumaa	Sõmeru	Sõmeru settlement WSZ	Rakvere Vesi AS	200	1362	Cm-V (Cm-V)
705	Lääne-Virumaa	Haljala settlement	Haljala Naaritsa WSZ	Haljala Soojus AS	240	1000	non Cm-V (O)
773	Lääne-Virumaa	Vinni	Vinni settlement WSZ	Askoterm OÜ	75	970	Cm-V (Cm-V)
744	Lääne-Virumaa	Roodevälja village	Rakvere Lihakombinaat WSZ (food factory)	Rakvere Lihakombinaat AS	n.a.	780	non Cm-V (O-C)

Table 7.8 *Biggest water supply zones in Estonian Counties – Ida-Virumaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
852	Ida-Virumaa	Narva town	Narva town WSZ	Narva Vesi AS	8994	67497	non Cm-V (surface water)
835	Ida-Virumaa	Kohtla-Järve town	Kohtla-Järve town Järve district, Täkumetsa and Peeri villages WSZ	Järve Biopuhastus OÜ	2364	19689	Cm-V (Cm-V)
838	Ida-Virumaa	Kohtla-Järve town	Kohtla-Järve town Ahtme district WSZ	Järve Biopuhastus OÜ	2813	19300	non Cm-V (Q)
851	Ida-Virumaa	Sillamäe town	Sillamäe town WSZ	Sillamäe Veevark AS	2053	15680	Cm-V (V2vr)
846	Ida-Virumaa	Jõhvi town	Jõhvi town WSZ	Jõhvi Veemajandus OÜ	2590	12400	Cm-V (Q, V2gd)
849	Ida-Virumaa	Kiviõli town	Kiviõli town WSZ	Kiviõli Vesi OÜ	653	6734	Cm-V (Cm-V, V2gd)
855	Ida-Virumaa	Narva-Jõesuu town	Narva-Jõesuu town WSZ	Narva Vesi AS	750	3019	Cm-V (V2vr)
850	Ida-Virumaa	Püssi town	Püssi town WSZ	OÜ Järve Biopuhastus	190	1600	Cm-V (V2gd)
861	Ida-Virumaa	Aseri municipality	Aseri settlement WSZ	Aseri Kommunaal OÜ	300	1570	Cm-V (V2gd)
841	Ida-Virumaa	Kohtla-Järve town	Oru settlement WSZ	Oru Kodu OÜ	170	1542	Cm-V (V2vr)

Table 7.9 *Biggest water supply zones in Estonian Counties – Raplamaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
784	Raplamaa	Rapla town	Rapla town WSZ	Rapla Vesi AS	656.	4885	non Cm-V (O)
786	Raplamaa	Märjamaa	Märjamaa settlement WSZ	Marjamaa Vesi OÜ	485	3000	non Cm-V (O)
792	Raplamaa	Kohila settlement	Kohila settlement WSZ	OÜ Kohila Maja	266	2180	non Cm-V (O-Cm)
805	Raplamaa	Kehtna settlement	Kehtna WSZ	Kehroada Elamu OÜ	206,7	1400	non Cm-V (O)
818	Raplamaa	Järvakandi settlement	Järvakandi settlement WSZ	Järvakandi Kommunaal OÜ	105	1350	non Cm-V (O)
820	Raplamaa	Alu	Alu WSZ	Tepek Kinnisvara OÜ	74	900	non Cm-V (O)
797	Raplamaa	Kaiu settlement	Kaiu WSZ	OÜ Kaiu Revival	66	556	non Cm-V (O)
826	Raplamaa	Orgita settlement	Orgita centree WSZ	OÜ Orgita Elamu	65	550	non Cm-V (S)
827	Raplamaa	Orgita settlement	Orgita Masinakeskuse WSZ	OÜ Orgita Elamu	59	550	non Cm-V (O)
793	Raplamaa	Prillimäe settlement	Prillimäe WSZ	OÜ Kohila Maja	131	436	non Cm-V (O)

Table 7.10 *Biggest water supply zones in Estonian Counties – Järvamaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
666	Järvamaa	Paide town	Paide town WSZ	AS Paide Vesi	1100	7600	non Cm-V (S)
676	Järvamaa	Türi town	Türi town WSZ	Türi Vesi OÜ	450	4950	non Cm-V (S-O, O)
659	Järvamaa	Koeru municipality Koeru settlement	Koeru settlement WSZ	Koeru Kommunaal AS	170	925	non Cm-V (O)
651	Järvamaa	Järva-Jaani municipality Järva- Jaani settlement	Järva-Jaani settlement WSZ	Järva-Jaani Teenus OÜ	87,7	635	non Cm-V (S-O)
674	Järvamaa	Roosna-Alliku municipality Roosna-Alliku settlement	Roosna-Alliku settlement WSZ	AS Paide Vesi	40	600	non Cm-V (O)
640	Järvamaa	Ambla municipality Aravete settlement	Aravete settlement WSZ	Avoterm Aqua OÜ	43	500	non Cm-V (S-O)
647	Järvamaa	Imavere municipality Imavere village	Imavere village WSZ	Imavere Soojus OÜ	45	500	non Cm-V (S1)
684	Järvamaa	Türi municipality Särevere settlement	Järvamaa Kutsehariduskeskus Särevere WSZ (centre of vocational education)	Järvamaa Kutsehariduskeskus	100	500	non Cm-V (S)
689	Järvamaa	Väätsa municipality Väätsa settlement	Väätsa settlement WSZ	Väätsa Soojus OÜ	60	480	non Cm-V (S-O, O3)
662	Järvamaa	Koigi municipality Koigi village	Koigi village WSZ	AS Paide Vesi	40,5	350	non Cm-V (O3)

Table 7.11 *Biggest water supply zones in Estonian Counties – Jõgevamaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
627	Jõgevamaa	Jõgeva town	Jõgeva town, WSZ on the left bank of river Pedja	Jõgeva Vesi OÜ	490	4500	non Cm-V (Ox3)
606	Jõgevamaa	Põltsamaa town	Põltsamaa town WSZ	Melior OÜ	412	3500	non Cm-V (S)
593	Jõgevamaa	Tabivere settlement	Tabivere WSZ	Veemajanduse OÜ	80	900	non Cm-V (S)
599	Jõgevamaa	Siimusti settlement	Siimusti settlement WSZ	Kuremaa Enveko AS	62	750	non Cm-V (S)
607	Jõgevamaa	Mustvee town	Mustvee town WSZ	Mustvee LHA Mustvee Teenus	150	700	non Cm-V (O)
619	Jõgevamaa	Adavere settlement	Adavere elamute WSZ	Põltsamaa municipalityvara OÜ	65	662	non Cm-V (S)
595	Jõgevamaa	Võisiku settlement	Võisiku Hooldekodu WSZ	Võisiku Hooldekodu	69	605	non Cm-V (O-C)
604	Jõgevamaa	Jõgeva town	Jõgeva settlement WSZ	Kuremaa Enveko AS	50	550	non Cm-V (S)
629	Jõgevamaa	Luuu settlement	Luuu Metsanduskooli WSZ	Luuu Metsanduskool	70	540	non Cm-V (S)
634	Jõgevamaa	Elistvere village	Elistvere WSZ	Tempos Grupp OÜ	4,2	500	non Cm-V (Q)

Table 7.12 *Biggest water supply zones in Estonian Counties – Saaremaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
288	Saaremaa	Kuressaare town	Kuressaare WSZ	Kuressaare Veevärk AS	2800	15950	non Cm-V (S)
295	Saaremaa	Sõmera village	Sõmera village Hooldekodu WSZ (nursing home)	Sõmera Hooldekodu	80	700	non Cm-V (S)
290	Saaremaa	Aste	Aste settlement WSZ	Arco Ehitus OÜ	60	600	non Cm-V (S)
305	Saaremaa	Orissaare	Orissaare settlement WSZ	Orissaare Soojus OÜ	80	600	non Cm-V (S)
307	Saaremaa	Kaali	Kaali-Kõljala villagede WSZ	Pihtla Local municipality	50	520	non Cm-V (S)
312	Saaremaa	Salme	Salme settlement WSZ	SVK Salme OÜ	35	515	non Cm-V (S)
314	Saaremaa	Valjala settlement	Valjala WSZ	Valjala Local municipality	30	500	non Cm-V (S)
301	Saaremaa	Liiva	Liiva village WSZ	Muhu Local municipality	25	450	non Cm-V (S)
298	Saaremaa	Leisi	Leisi settlement WSZ	Leisi VV Kommunaaalosakond	24	400	non Cm-V (S)
300	Saaremaa	Lümända	Lümända village WSZ	Lümända Local municipality	28	380	non Cm-V (S)

Table 7.13 *Biggest water supply zones in Estonian Counties – Parnumaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
250	Pärnumaa	Pärnu town	Pärnu town	Pärnu Vesi AS	7000	34350	non Cm-V (S,D2-1-S)
266	Pärnumaa	Sindi	Sindi town (Pärnu -Sindi water network)	Sindi Vesi OÜ	150	2500	non Cm-V (S,D2-1-S)
245	Pärnumaa	Paikuse settlement	Paikuse settlement (Reiu - Paikuse-Sindi network)	Pärnu Vesi AS	165	2300	non Cm-V (S, D2-1-S)
280	Pärnumaa	Vändra	Vändra settlement	Vändra MP OÜ	275	2287	non Cm-V (S)
243	Pärnumaa	Pärnu-Jaagupi	Pärnu-Jaagupi settlement	Mako AS	85	1350	non Cm-V (S)
272	Pärnumaa	Tootsi	Tootsi settlement	Tootsi Kommunaal OÜ	240	930	non Cm-V (S)
262	Pärnumaa	Sauga	Sauga settlement	Sauga Varahaldus AS	60	800	non Cm-V (S)
257	Pärnumaa	Tihemetsa settlement	Tihemetsa centre	Saarde Kommunaal OÜ	97	620	non Cm-V (D2-1)
241	Pärnumaa	Lavassaare	Lavassaare settlement	Lavassaare Kommunaal OÜ	160	540	non Cm-V (S)
268	Pärnumaa	Tõstamaa	Tõstamaa settlement	SuFe OÜ	55	500	non Cm-V (S)

Table 7.14 *Biggest water supply zones in Estonian Counties – Viljandimaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
362	Viljandimaa	Viljandi town and Viiratsi settlement	Viljandi town and Viiratsi settlement WSZ	AS Viljandi Veevärk	2410	20328	non Cm-V (S)
363	Viljandimaa	Karksi-Nuia town	Veetorni well WSZ	livakivi AS	211	1412	non Cm-V (S)
368	Viljandimaa	Võhma town	Võhma town WSZ	Võhma Elko AS	108	1100	non Cm-V (O)
370	Viljandimaa	Abja-Paluoja town	Töökoja well WSZ	Abja Elamu OÜ	95	995	non Cm-V (D2-1)
405	Viljandimaa	Pärsti municipality, Ramsi settlement	Ramsi settlement WSZ	Ramsi VK OÜ	75	700	non Cm-V (S)
419	Viljandimaa	Viiratsi municipality, Vana-Võidu	Vana-Võidu WSZ	Viljandi WSZ AS	50	640	non Cm-V (S)
384	Viljandimaa	Suure-Jaani municipality, Olustvere	Olustvere settlement WSZ	Suure-Jaani Haldus AS	55	550	non Cm-V (S)
411	Viljandimaa	Pärsti municipality, Jämejala village	Viljandi Haigla SA peahoone WSZ (hospital)	Viljandi Haigla SA	69	540	non Cm-V (S)
374	Viljandimaa	Mõisaküla town	Mõisaküla village town WSZ	Mõisavillage Linnahooldus	90	495	non Cm-V (D2-1)
372	Viljandimaa	Suure-Jaani town	Suure-Jaani town WSZ	Suure-Jaani Haldus AS	47	490	non Cm-V (S, D2-1)

Table 7.15 *Biggest water supply zones in Estonian Counties – Tartumaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
519	Tartumaa	Tartu town	Tartu town and Kõrveküla village WSZ	Tartu Veevärk AS	14500	99000	non Cm-V (Q, D2tr, D2pr-S, O- C)
473	Tartumaa	Elva town	Elva town WSZ	Aqua & Waste Services OÜ	460	2800	non Cm-V (D2-1-S)
543	Tartumaa	Tõrvandi	Tõrvandi settlement WSZ	Olme OÜ	120	1500	non Cm-V (D2-1-S)
511	Tartumaa	Puhja	Puhja settlement WSZ	Sangla Turvas AS	87	1100	non Cm-V (D2-1-S)
544	Tartumaa	Ülenurme	Ülenurme settlement WSZ	Olme OÜ	150	1100	non Cm-V (D2-1)
504	Tartumaa	Nõo settlement	Nõo settlement, Lao ja Voika road WSZ	Nõo WSZ OÜ	95	740	non Cm-V (D2-1)
534	Tartumaa	Märja	Märja settlement WSZ	Märja Soojus OÜ	70	675	non Cm-V (D2-1, S)
520	Tartumaa	Lähte	Lähte Ühisgümnaasiumi WSZ (school)	Lähte Ühisgümnaasium	16	600	non Cm-V (Q)
480	Tartumaa	Kambja	Kambja settlement WSZ	CAMBI OÜ	147	596	non Cm-V (D2)
513	Tartumaa	Rannu	Rannu settlement WSZ	Emajõe Veevärk AS	70	550	non Cm-V (D2-1)

Table 7.16 *Biggest water supply zones in Estonian Counties – Valgamaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
424	Valgamaa	Valga town	Valga town WSZ	Valga Vesi AS	9600	10800	non Cm-V (D2)
429	Valgamaa	Otepää town	Uus veehaare WSZ	OTEPÄÄ Veevärk AS	170	1000	non Cm-V (D2)
428	Valgamaa	Tõrva town	Riiska housesistricti WSZ	Tõrva Linnahooduse Asutus	150	950	non Cm-V (D2-1)
449	Valgamaa	Puka municipality Puka settlement	Puka settlement	Puka Local municipality	45	700	non Cm-V (D2)
450	Valgamaa	Hummuli municipality Hummuli settlement	Hummuli settlement WSZ	Hummuli Local municipality	40	400	non Cm-V (D2)
455	Valgamaa	Tõlliste municipality Tsirguliina settlement	Tsirguliina settlement WSZ	Ühismajandamise OÜ	22	400	non Cm-V (D2)
435	Valgamaa	Palupera municipality Hellenurme village	Hellenurme village WSZ	Palu-Teenus OÜ	27	390	non Cm-V (D2)
440	Valgamaa	Helme municipality	Helme town WSZ	Helme Teenus OÜ	33	350	non Cm-V (D2-1)
444	Valgamaa	Helme municipality Helme settlement	Helme settlement WSZ	Valgamaa Kutseõppekeskus	42	350	non Cm-V (D2)
452	Valgamaa	Tõlliste municipality Laatre settlement	Laatre settlement WSZ	Ühismajandamise OÜ	30	305	non Cm-V (D2)

Table 7.17 *Biggest water supply zones in Estonian Counties – Põlvamaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
584	Põlvamaa	Põlva town	Põlva town WSZ	Põlva Vesi AS	600	5800	non Cm-V (D2, D2-1)
555	Põlvamaa	Räpina town	Kastani road WSZ	Revekor AS	150	1200	non Cm-V (D2)
586	Põlvamaa	Värskla settlement	Värskla settlement WSZ	Värskla Calor OÜ	30	530	non Cm-V (D2)
582	Põlvamaa	Vastse - Kuuste village	Vastse - Kuuste village WSZ	Vastse -Kuuste Soojus OÜ	55	500	non Cm-V (D2)
552	Põlvamaa	Kanepi settlement	Kanepi settlement and school WSZ	Kanepi Local municipality MA Karjäär	78	460	non Cm-V (D2)
549	Põlvamaa	Himmaste village	Himmaste village WSZ	Põlva Vesi AS	50	450	non Cm-V (D2)
563	Põlvamaa	Mammaste village	Mammaste village WSZ	Põlva Vesi AS	50	450	non Cm-V (D2)
570	Põlvamaa	Põlgaste village	Põlgaste settlement WSZ	Kanepi Local municipality MA Karjäär	80	380	non Cm-V (D2)
566	Põlvamaa	Mooste settlement	Mooste settlement WSZ	Mooste Olme OÜ	40	370	non Cm-V (D2)
548	Põlvamaa	Erastvere village	Erastvere Hooldekodu WSZ (nursing home)	Hooldekandeteenused AS	45	350	non Cm-V (D2)

Table 7.18 *Biggest water supply zones in Estonian Counties – Vorumaa*

Id. No.	County	Municipality	Name of water supply zone	Water producer	Production m³/day	Population served	Aquifer group
350	Võrumaa	Võru town	Võru town	Võru Vesi AS	1300	10000	non Cm-V (D2)
320	Võrumaa	Võru municipality, Parksepa village	Parksepa	Väimela Soojus OÜ	60	800	non Cm-V (D2)
348	Võrumaa	Vastseliina municipality, Vastseliina settlement	Vastseliina settlement	Vaks OÜ	100	580	non Cm-V (D2)
359	Võrumaa	Võru town	Kuperjanovi sõjaväeosa (army houses)	Kuperjanovi Üksikjalaväe pataljon	100	500	non Cm-V (D2)
338	Võrumaa	Lasva municipality, Kääpa village	Kääpa	Lasva Local municipality	38	428	non Cm-V (D2)
326	Võrumaa	Antsla municipality, Kobela settlement	Kobela	Antsla Local municipality	60	420	non Cm-V (D2)
324	Võrumaa	Antsla town	Antsla town	Antsla Local municipality	41	400	non Cm-V (D2)
328	Võrumaa	Antsla municipality, Lusti village	Lusti village	Antsla Local municipality	44	400	non Cm-V (D2)
317	Võrumaa	Võru municipality, Väimela settlement	Väimela Tehnikumi	Väimela Soojus OÜ	68	350	non Cm-V (D2)
354	Võrumaa	Sõmerpalu municipality, Sõmerpalu	Sõmerpalu settlement	Sõmerpalu Local municipality	11	350	non Cm-V (D2)

8. REMEDIAL ACTIONS

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 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.

8.1. 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. 8.1 – 8.2 the distribution of the radium removal percentages are shown as a function of the analyzed wells.

As an example, if the adult age class is concerned (Fig. 8.1), the following removal rates are needed to lower the TID to 0.1 mSv/y:

- with a radium removal of 70% compliant cases (referred to measured wells) 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.

When the infant age class is considered (Fig. 8.2), 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 demineralised water, not best suited as drinking water and dangerous for the distribution system (problems of pipe corrosion), therefore this result is hardly achievable.

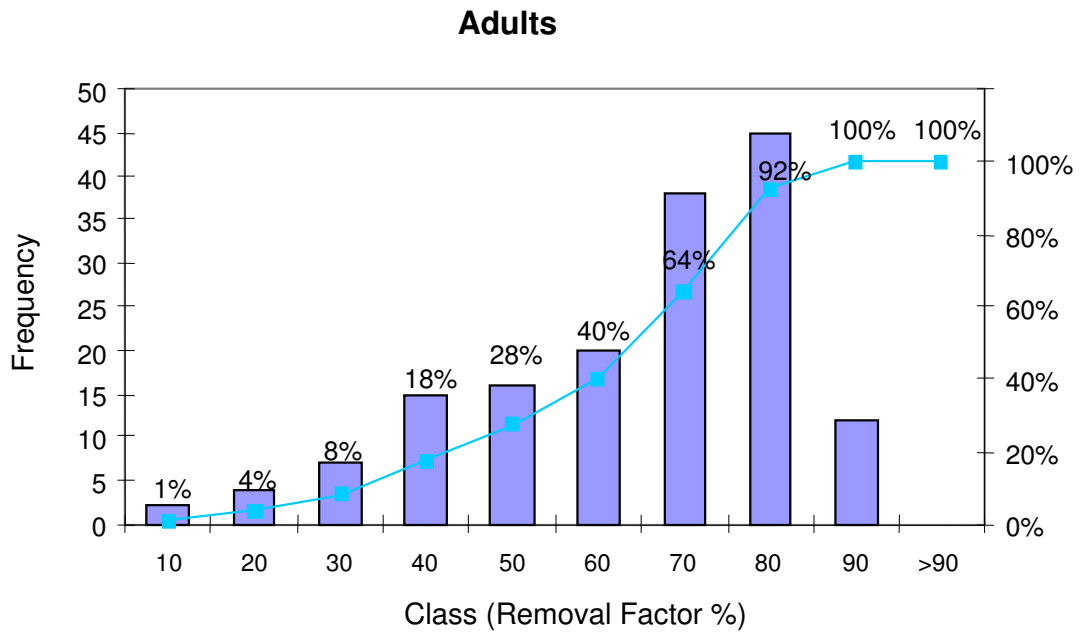


Figure 8.1 *Distribution of the radium removal level to achieve the 0.1 mSv/y TID for the adult age class. On Y axis: measured wells.*

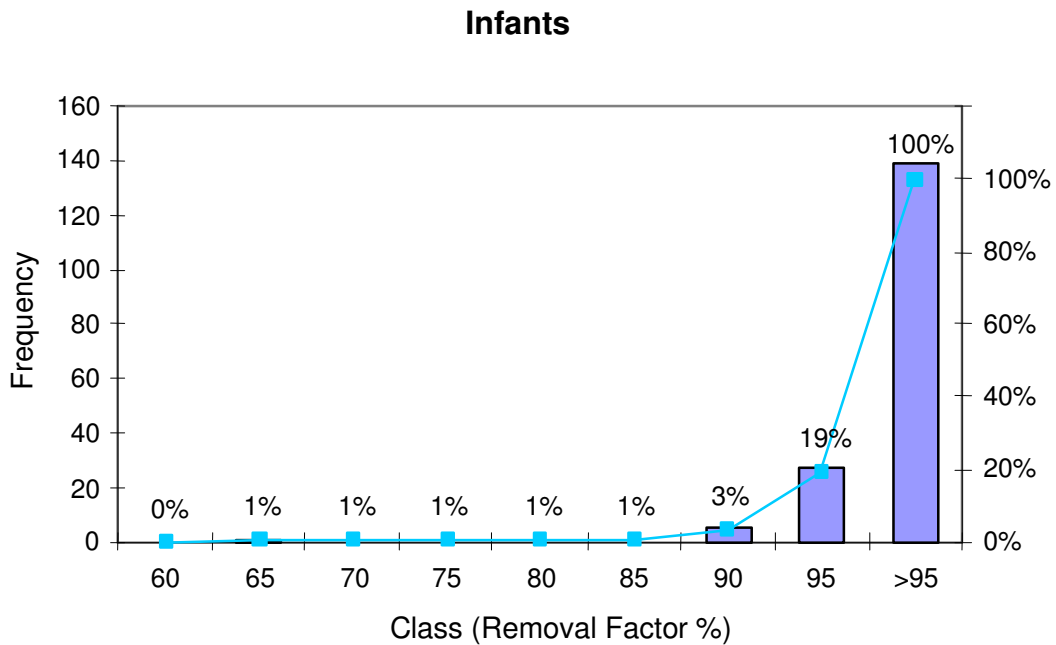


Figure 8.2 *Distribution of the radium removal level to achieve the 0.1 mSv/y TID for the infant age class. On Y axis: measured wells.*

8.2. 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 NH_4 .

Keila waterworks supplies water to about 9700 people. Groundwater drawn from 4 wells, is only delivered. Water is mixed before being collected in treatment tanks. Fe and Mn are then removed in a similar way as in the 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 8.1.

Table 8.1 Analyses of ^{226}Ra and ^{228}Ra in water before and after treatment for existing systems

Waterworks	Station/Well	Sampling	^{226}Ra (Bq/L)		^{228}Ra (Bq/L)		TID mSv/y
			Conc.	Unc. (2σ)	Conc	Unc. (2σ)	
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	Haabneme/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 8.1 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 8.2 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 8.2 *Decontamination factors for existing treatment plants*

Waterworks	Treatment station	Laboratory	²²⁶Ra D[%]	²²⁸Ra D[%]	TID D[%]
Tallinna Vesi	Jugapuu	Finland	24	36	34
	Tiskre	EE - Radiation Protection Centre	22	11	14
	Toomse-oitsu	EE - Radiation Protection Centre	n.r. *	n.r.	n.r.
Keila Vesi	Keila	EE - Radiation Protection Centre	19	26	15
Rakvere Vesi	Rakvere	EE - University of Tartu	n.r.	n.r.	n.r.
Average **			22	24	21
Standard deviation **			3	13	11
Viimsi Vesi	well 412 ***	EE - Radiation Protection Centre	82	71	73

(*) n.r.: not relevant

(**) average and standard deviation values calculated on the basis of the 3 positive values only

(***) additional cleaning after column I-II

Decontamination factors (in terms of TID reduction) 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.

To compare the treatment performances, a brief analysis of data belonging to another European area, Bavaria, has been carried out (BayLfU, 2006). Several treatment devices operate in that zone to ensure waters are fit for human consumption. A large amount of analysis of natural radionuclides activity concentrations has been executed on waters (raw and treated) and on residues of treatment plants. Single regions average ²²⁶Ra contents of raw water range from 7 to 107 mBq/l; the interval is 9 ÷ 154 mBq/l as far as average ²²⁸Ra levels are concerned. Common purifying devices provide iron, manganese (and often arsenic) removal by preaerated sand filtration; these systems are of the same kind as those

encountered in surveyed Estonian waterworks. Deacidification technique (limestone filtration) is also used, combined with Fe, Mn, (As) removal stage or on its own. In Fig. 8.3, for each of these techniques, figures for ^{226}Ra and ^{228}Ra decontamination factors are given. As for Estonian plants, data variability is elevated. Nevertheless, on average, radium removal effectiveness seems to be better: decontamination factor in Fe, Mn, (As) plants is 31 % for ^{226}Ra and 37 % for ^{228}Ra .

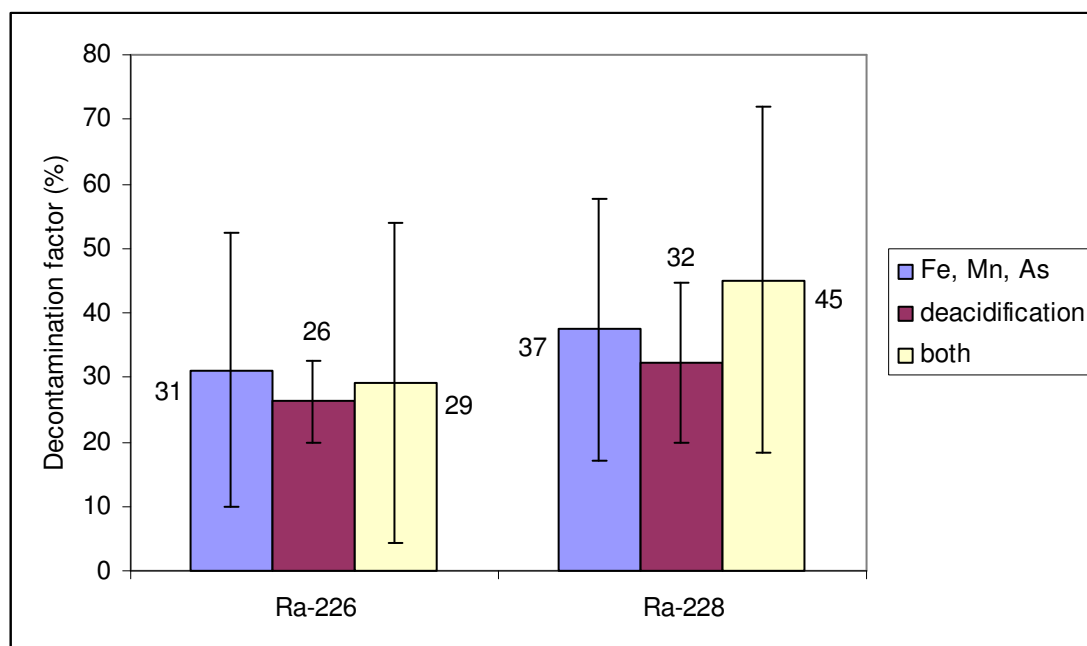


Fig. 8.3 Average decontamination factors for three treatment techniques applied in Bavarian Water Treatment Plants: a) preaerated Fe, Mn, As filtering (24 and 22 values for ^{226}Ra and ^{228}Ra respectively), b) deacidification (4 and 3 values for ^{226}Ra and ^{228}Ra respectively), c) combination of two techniques (7 and 6 values for ^{226}Ra and ^{228}Ra respectively). Standard deviations of data are also reported

8.3. Summary of the information about removal techniques

Radium removal from drinking waters has not to be regarded as a technical problem. Actually, many radium removal methods are available, with different costs, effectiveness and management problems.

Most of these processes are applied in United States where radium decontamination is needed in some areas as:

- Midwest (Iowa, Illinois, Wisconsin, Missouri)
- California
- Pennsylvania
- North and South Carolina
- Georgia
- New Jersey
- Florida
- Minnesota
- Oklahoma
- New York
- Maine.

Information on radium levels in U.S. water can be easily found in literature and in the web. An overview has been reported in 2.3.

In Table 8.3 we summarize the most used techniques. In brackets the number of the technical sheet (reported in 8.5) is given; 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 a US-EPA documents (USEPA 2007). 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.

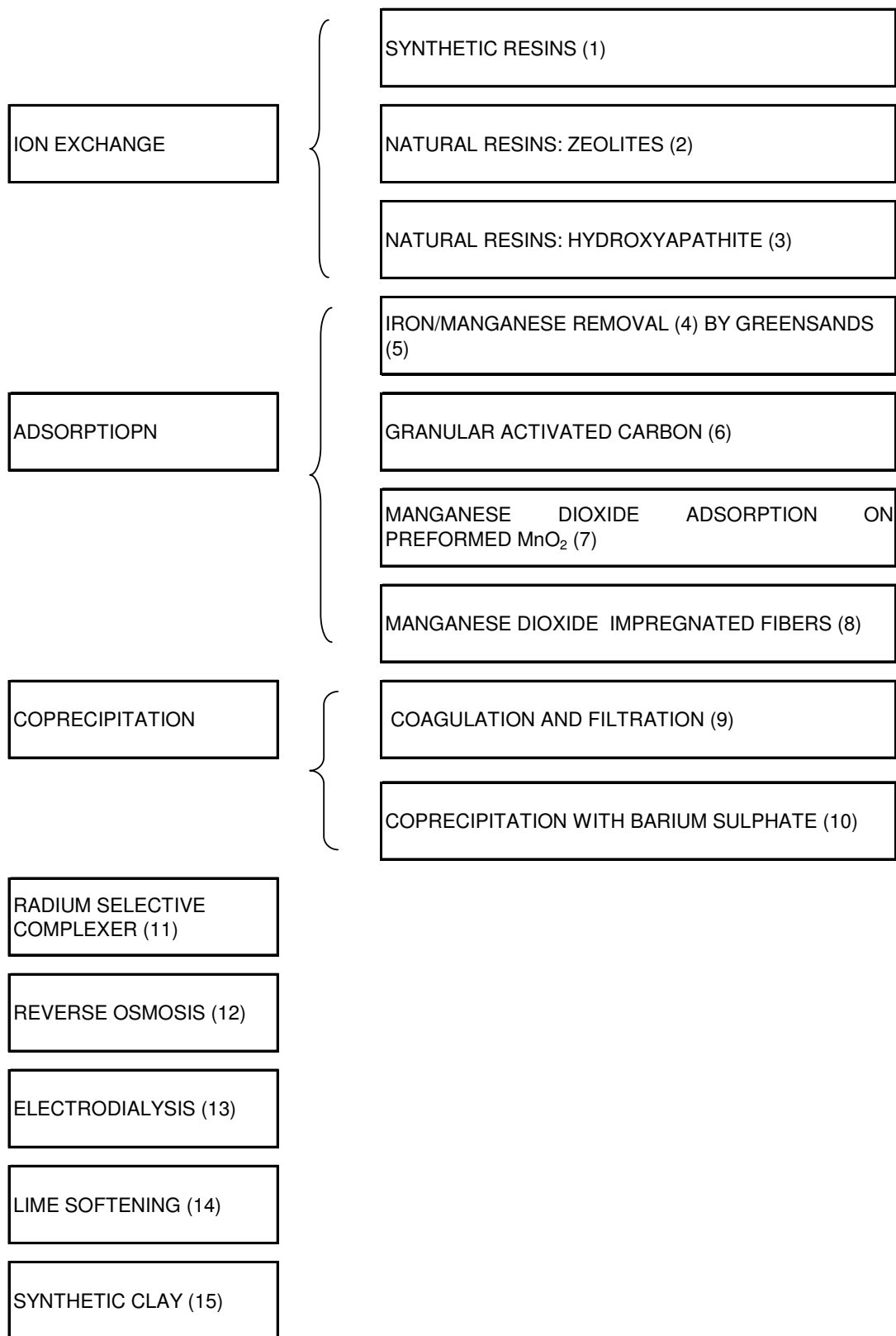


Fig. 8.3 Techniques for radium removal

Table 8.3 *Removal techniques: general overview*

Unit Technologies	Limitations	Skill level required	Effectiveness %
Ion exchange: synthetic resins (1)	a, b	Basic-intermediate	Up to 95 - 99 %
Ion exchange: natural resins -zeolites (2)	a, b	Basic-intermediate	90 – 97 %
Ion exchange: natural resin - hydroxyapatite (3)	a, b	Basic-intermediate	not available
Iron-manganese removal (4)	b, d, f, h	Intermediate	14 – 58 %
Iron-manganese removal by greensands (5)	d, f, h	Basic-intermediate	50 - 90 %
Granular activated carbon (6)	a, f, h	Basic	34 - 97 %
Pre-formed hydrous manganese oxide (7)	a, d, h	Intermediate	19 – 95 %
Manganese dioxide impregnated fibres (8)	a, d, h	Advanced	not available
Coagulation and filtration (9)	a, b, c	Advanced	32 - 70 %
Coprecipitation with barium (10)	a, e	Intermediate to advanced	40 – 98 %
Radium selective complexer (11)	a, h	Basic	50 – 98 %
Reverse Osmosis (12)	a, h	Advanced	90 – 99 %
Electrodialysis (13)	a, h	Basic-intermediate	40 – 98 %
Lime Softening (14)	a, b, c	Advanced	50 – 90 %
Synthetic clay (15)	a, g	not available	not available

- 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 sulphate 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.

The previous cited STUK-A 169 report (STUK 2000) provides an evaluation of the removal efficiency limited to the main available techniques (Table 8.4).

Table 8.4 *Removal techniques: TENAWA Report*

Technique	Effectiveness %
Ion exchangers - cation resins (1) (2) (3)	69 - 99
Aeration – filtration (4) (5)	3 – 93
Manganese based methods (7) (8)	56 – 97

8.4. 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 (New Jersey, 2002) 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 (8.5) summarizes the annualized cost, as a function of plant dimensions / population served.

Table 8.5 *Annualized cost estimates – an example from USA*

Size of Plant (m ³ /day)	Population served (N. people)	Annualized cost* (€)	Annualized cost per person* (€)
26.5	500	56,000 – 84,000	450 – 670
132	2,500	101,400 – 153,800	160 – 246
264	5,000	171,300 – 255,000	137 – 204
> 264	10,000	> 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.

8.5. Radium removal techniques: technical features of available methods

1) CATION EXCHANGE WITH SYNTHETIC RESINS

Principles

Water is eluted through a cation resin containing exchangeable ions. Cation exchange resins generally exchange sodium or potassium with other cations as calcium, magnesium, radium etc

Advantages

- suitable for small treatment plants
- commonly available
- relatively simple and easy to operate
- little space required
- it removes hardness along with the Ra if used in H⁺ or Na or K form
- it does not remove hardness along with the Ra if used in Ca or Mg form
- cation exchange reactors can be automated to a high degree meaning less operator attention required
- cation exchange can also be used in point-of-entry (POE), generally for small systems, or in point-of-use (POU).

Disadvantages

- if used in Na form an increase in the Na content of the treated water is produced (potential problem for people with restricted diets and hypertension)
- if used in the H⁺ form, a strong acid (for instance H₂SO₄) is used to regenerate it. Chemical hazard is associated with this operation
- the softened water can be corrosive to the distribution systems due to the very low mineral content
- a pretreatment (such as filtration) before ion exchange could be necessary
- disposal of spent brines, backwash water, rinse water and ultimately exhausted cartridges can be a serious problem
- it requires careful long-term operations, maintenance and monitoring plans to ensure proper performance.

Remarks

- a chosen amount of raw water could be blended to softened water in order to reduce its corrosiveness

References:

Reid 1985; STUK 2000; USEPA 2007; Havener 2007; Moore 1975; Lucas 1987; Clifford (in Cothorn 1990).

2) CATION EXCHANGE WITH NATURAL RESINS: ZEOLITES

Principles

Radium-contaminated water flows through a fluidized ion-exchange bed of zeolite. This treatment is not selective for radium, other cations are removed in variable amounts.

Advantages

- suitable for small treatment plants
- relatively simple and easy to operate
- little space required
- it does not remove hardness along with the Ra: Ca, Fe, Mg and Na content of the water remain essentially unchanged,
- cation exchange reactors can be automated to a high degree: lower operator surveillance is required
- cation exchange can also be used in point-of-entry (POE), generally for small systems, or in point-of-use (POU).

Disadvantages

- pretreatment such as filtration before ion exchange could be necessary
- disposal of spent brines, backwash water, rinse water and ultimately exhausted cartridges can be a serious problem
- it requires careful long-term operations, maintenance and monitoring plans to ensure proper performance.

Remarks

- the technique is finding increasing applications in USA.

References

STUK 2000; Richland 2004.

3) CATION EXCHANGE WITH NATURAL RESINS: HYDROXYAPATITE

Principles

Hydroxyapatite has a good radium absorption capability, if small flow rates are used. It works as a selective ion exchanger.

Advantages

- easy to use
- selective

Disadvantages

- need of low flow rate.

Remarks

- at present is not a widespread technique yet. Little literature is available.

References

STUK 2000.

4) IRON - MANGANESE REMOVAL BY SANDS

Principles

Water is firstly aerated to oxidize iron and manganese and then filtered on sand. Radium is probably adsorbed onto the iron (III) oxide particles and removed by filtration.

Advantages

- low costs (where the treatment is already in use) maintenance
- easy to operate
- the backwashing waters are not heavily contaminated by radium
- long life of the media.

Disadvantages

- the radium removal is low; removal rates are highly variable (see also Estonian data in this chapter)
- the technique is best suited in cases of low radium contamination.

Remarks

- Radium removal efficiency is widely variable and depends on the chemical composition of waters, the sand granulometry, the retention time etc.
- best results are obtained if greensands are often regenerated

References

STUK 2000; Clifford 1990; Bronzović 2006; Baeza 2008.

5) IRON - MANGANESE REMOVAL BY GREENSAND

Principle of the technique

Manganese greensand is a naturally occurring glauconite material which catalytically oxidize Mn to hydrous manganese oxide (HMO) in the presence of potassium permanganate (or another oxidizing agent). Radium has a natural affinity for manganese dioxide HMO.

Advantages

- cost effective at small systems
- simple and operator friendly, above all if Mn is naturally occurring
- also remove iron, manganese and arsenic (if present)
- minimal maintenance requirements
- no Na is added to the water as a result of the treatment.

Disadvantages

- high iron to manganese ratio may lower radium adsorption on greensand filters
- production of wastes (sludge, backwash waters, spent filter media)
- waters with low manganese content should be added by manganese salts

Remarks

- radium removal improves as water pH increases
- in order to improve the efficiency of the radium removal it is needed to optimize the pre-oxidation process, the amount of added reagents, the detention time etc.

References

Reid 1985; Oureshi 2003.

6) FILTRATION ON GRANULAR ACTIVATED CARBON (GAC)

Principles

Radium is selectively absorbed on GAC

Advantages

- technique widely used in drinking water treatment
- easy to operate
- minimal maintenance required
- possibility of multiple filters (sand, greensand, fixed-leaf) to enhance radium removal
- no sodium is added to the water as a result of the treatment
- it can be applied to small systems.

Disadvantages

- cost of the GAC device and its disposal
- the results presented in the literature are uncertain; however a work of optimization for every situation is necessary (see also remarks).

Remarks

The factors influencing the efficiency of radium removal by GAC are

- granular size
- water hardness (the decrease of water hardness produce increase in Ra removal).

References

STUK 2000; Bronzović 2006; Jimenez 2002; Salas García 2005.

7) MANGANESE DIOXIDE ADSORPTION BY PREFORMED MnO₂

Principles

Radium has a natural affinity for manganese dioxide, thus it is selectively absorbed on the surface of the hydrous manganese oxide (HMO), which will be then filtered away

Advantages

- technique suitable for small and medium sized systems
- can rely on existing treatment facilities
- no sodium is added to the water as a result of the treatment
- radium accumulation does not occur because the HMO is backwashed out of the filter media
- cost effective

Disadvantages

- the HMO must be dosed carefully, since it could clog the filtration process; an insufficient amount will lead to ineffective removal. As a result, intermediate operator skill is required
- high iron levels can compete with radium for adsorption sites and lower radium removal; iron removal may be required before the addition of the HMO

Remarks

Two different options are possible:

- processes using manganese greensand or manganese oxide coated filtration media: the radium fraction which it is not absorbed on HMO particles, will be trapped on the filtration apparatus and thus will accumulate there;
- processes using silica sand or silica sand/anthracite dual filtering media: in this case filtration media will not trap radium and no accumulation will occur; from a waste management point of view this solution is recommended.

References

Pedersen 2001; Havener 2007; Valentine, 1988; Valentine 2002; Oureshi 2003; Docstoc 2008; Hungerford 2006; New Jersey 2002.

8) ABSORPTION BY MnO₂ IMPREGNATED FIBERS

Principles

Radium is adsorbed onto acrylic fibers impregnated with MnO₂. Exhausted fibers will be replaced with new ones.

Advantages

- effective for radium removal
- very simple to use
- suitable for small systems
- very high capacity of radium adsorbing
- no backwashing or regenerating of the system is required, thus eliminating the need of disposing the wastewater discharge
- no sodium is added to the water as a result of the treatment.

Disadvantages

- the preparation of the fibers is not simple, thus operational problems can occur in small systems application; qualified operators are necessary
- the practical performance of the units may be adversely affected by the washout of loosely held MnO₂ from the fibers
- there is the problem of the disposal of exhausted units

References

Moore 1973; Reid 1985; Clifford 1990.

9) COAGULATION AND FILTRATION

Principles

Radium is coprecipitated with $\text{Fe}(\text{OH})_3$ at pH 10 and filtered

Advantages

- technique well known in drinking water treatment

Disadvantages

- not suitable for small systems
- need of chemical reagents (operational problems)
- production of sludges to dispose
- reagent cost

Remarks

- calcium, magnesium and bicarbonates influence positively the radium removal

References

- Salas García 2005.

10) COPRECIPITATION WITH BARIUM SULPHATE

Principles

Coprecipitation of Ra-Ba sulphates by adding soluble barium (as chloride, carbonate, nitrate); the precipitate is separated by settling.

Advantages

- relatively cheap
- no sodium is added to the water as a result of the treatment.

Disadvantages

- effectiveness of removal not constant
- high level operator skill required
- requires high sulfate concentrations in raw water
- bulky plants / sedimentation basins are needed
- the technique does require careful control of Ba concentrations added to water and routine analysis of the treated water to assure achievement of target concentrations (high concentration of Ba are toxic)

Remarks

- the process is not widely employed in USA.

References

Reid 1985.

11) RADIUM SELECTIVE COMPLEXER (RSC)

Principles

Radium is adsorbed onto BaSO₄ crystals (>8% wt/wt BaSO₄ content) supported on a standard polystyrene-divinylbenzene strong-acid cation resin in the sodium form.

Advantages

- effective in radium removal
- high absorbing capacity (~6,000 Bq/g)
- minimized volume of waste
- confined and controlled absorbing media
- other ions (water hardness) are not removed
- simple process, suitable to both small and large water supply systems
- no operational requirements for backwashing and regeneration
- no sodium is added to the water as a result of the treatment.

Disadvantages

- RSC is an irreversible chemical adsorption process and concentration factors can be high; spent media may produce radon gas in large amounts (up to 6000 Bq/g)
- spent adsorbents need to be disposed as low radioactive solid wastes in specific sites
- may need iron removing before RSC process by prefiltering.

Remarks

- the RSC is a Dow Chemical patent
- a quite similar process can be carried out by adsorption onto BaSO₄ impregnated alumina
- the technique has been limited in the diffusion of the application, probably due to the disposal problems.

References

Reid 1985; Clifford 1990; Melis 1985; Shuguang Deng 2005; Mangelson 1990.

12) REVERSE OSMOSIS (RO)

Dissolved salts are removed by water by flowing feed-water through a semi-permeable membrane at a pressure greater than the osmotic pressure caused by the dissolved salts. Low pressure systems (nanofiltration) are also available.

Advantages

- high effective technique
- the process is very suitable for automated plant operation and use in small plants
- requires relatively small space
- relatively simple and easy to operate for small or domestic systems
- it removes hardness along with the Ra
- no sodium is added to the water as a result of the treatment
- arsenic, fluoride, microbes, nitrate, uranium, TOC, most metals, sulphate, calcium, magnesium, potassium, phosphorous are also removed
- RO can also be used in point-of-use (POU) devices

Disadvantages

- RO membranes easily foul, thus pretreatment to remove particulates and organics may be necessary (Ca and Mg can cause membrane fouling).
- cellulose acetate membranes can be damaged by biological fouling; free chlorine can be beneficial up to 1 mg/l.
- polyamide membranes can be damaged by chlorine, high iron and chloramines.
- depending on operating parameters from 20 to 40 % of raw water can be lost in the reject stream.
- advanced operator skill level required for complex systems.
- treatment costs are higher than other treatment options.
- concentrated waste stream and spent membranes are produced, which may contain elevated amounts of radionuclides
- testing is needed to ensure spent media is not hazardous or radioactive
- RO can significantly lower the alkalinity of the finished water and make it more corrosive. Treated water may need corrosion inhibitors added or pH raised to avoid corrosion of the aqueduct pipelines, which will cause at the same time an increase of lead and copper concentration in water
- the high pressure required for RO leads to higher energy costs, and capital costs for the membrane units can be significant compared to other technologies, making RO one of the more expensive treatment options
- lack of essential minerals for human health in produced water

Remarks

- the rapid and continuous advances in membrane technology, requiring much lower feed water pressure, are lowering costs and making RO more competitive

References

Havener 2007; Sengupta 1995; Reid 1985; USEPA 2007; STUK 2000; Clifford 1990.

13) ELECTRODIALYSIS / ELECTRODIALYSIS REVERSAL

Principles

Electrodialysis/Electrodialysis Reversal (ED/EDR) employs an ion exchange membrane to separate ionic contaminants. In particular The ED system operates in the same way as an RO system but selective cation and anion membranes are combined with a direct current (DC) electric field to demineralize or deionize water flowing through the device.

Advantages

- suitable for small systems
- effective in radium removal
- the polarity of the dc current is reversed periodically to reverse in EDR the direction of the ion movement and provide automatic flushing of the scale forming material on the membrane surfaces
- also removes uranium, arsenic, nitrates
- no sodium is added to the water as a result of the treatment.

Disadvantages

- membrane failure is a concern as is membrane fouling and scaling. EDR has the advantage over ED of reducing scaling potential and reducing membrane cleaning.
- depending on the ED design, 10-30% of the incoming water stream may be lost to waste
- it frequently requires pre-treatment for the aluminum and silica removal.

Remarks

- the technique is not widely diffused in USA at present.

References

New Jersey 2002; Sengupta 1995.

14) LIME SOFTENING

Principles

Hydrated lime or quicklime or soda ash is added to the raw water to raise pH and precipitate out calcium. In enhanced softening the pH is further raised in a second stage to at least pH 10.6 to remove also magnesium.

By the aid of coagulation, mixing, flocculation and sedimentation, metals and organic contaminants are removed.

Lime softening is typically practiced with waters that have natural high alkalinity and high hardness.

Advantages

- proven effectiveness in radium removal; package plants are available
- removes As, U, Fe, Mg and more.

Disadvantages

- relatively complicated process carried out in large reactors-clarifiers; not suitable for small waterworks; advanced operator skill level required
- it may be necessary to increase alkalinity after the treatment for corrosion control
- large amounts of lime sludge, and backwash waters are produced as wastes. Since concentration factors are high, testing is needed to ensure spent media is neither hazardous nor radioactive
- a large space is required for treatment plant

Remarks

- this process is the choice for large treatment plants; best for plants over 35,000 ÷ 40,000 m³/day.

References

Reid 1985; USEPA 2007; Clifford 1990; Sengupta 1995; New Jersey 2002; Degremont Handbook 1978.

15) SYNTHETIC CLAY

Principles

Synthetic clay (namely “sodium-4 mica”) is prepared by thermal treatment of kaolinite; during the treatment the layers of mica are displaced, thus creating a space in which radium is captured; when the mica is filled with radium, a shift in the layers occurs and the atoms of radium are trapped between the layered structure.

Advantages

- low cost
- the technique could be used in conventional ion exchange columns, after their pelletization.

Disadvantages

- the technique is innovative; no experiences about
- the problem of spent media disposal remains.

Remarks

- The technique needs to be focused by other experiences.

References

Komarneni 2001.

9. EFFLUENTS AND RESIDUES FROM EXISTING TREATMENT PLANTS

9.1. 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 9.1, 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 9.1.

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 9.2: it is worthwhile observing that, in filter material, Th-228 (followed by its decay products) also occurs, presumably due to growth during ²²⁸Ra 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 9.1. Waters are conveyed to sewage and the produced sludge is not used in agriculture.

Two samples of filter material (sand) were analysed to assess ^{226}Ra , ^{228}Ra , ^{228}Th (same considerations as above) and the results are shown in Table 9.2.

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 9.1.

As far as solid material is concerned, analysis results (^{226}Ra , ^{228}Ra , ^{228}Th) for filter material (sand) and for washing sediment are displayed in Table 9.2.

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

Previous information from stakeholders refers that: in one of the wells (n. 170) a sand filtering treatment plant operates (see Table 9.1), 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 9.1 (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. ^{226}Ra and ^{228}Ra analysis data on waters used to wash the two columns are presented in Table 9.1, together with information about flow and fate. Backwash water is conveyed to the same sewer as that of Tallinn waterworks.

Table 9.1: Information about effluents from treatment plants

Waterworks	Treatment station	Treatment type	backwash water (m ³ /y)	²²⁶ Ra	²²⁸ Ra	b.water fate	²²⁶ Ra	²²⁸ Ra	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, NH4 removal (preaerated)	1708	3100	8700	sewer	5.29E+06	1.49E+07	Finland
	Merivälja	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1770			sewer			
	Norma	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	268			sewer			
	Pirita	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1115			sewer			
	Laagri	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	267			sewer			
	Sae	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	207			sewer			
	Pääsküla	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1278			sewer			
	Raba	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	507	8730	14710	sewer	4.43E+06	7.46E+06	Rad.Prot.Centre
	Toome - Seedri	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	742			sewer			
	Hiiu	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	2183			sewer			
	Kagu - Võidu	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1482			sewer			
	Kandle	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1545			sewer			
	Pika - Voolu	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1413			sewer			
	Toome - Õitse	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1018	9350	13650	sewer	9.52E+06	1.39E+07	Rad.Prot.Centre
	Tiskre	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	581	5320	6040	sewer	3.09E+06	3.51E+06	Rad.Prot.Centre
	Piiri	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1118			sewer			
	Mahla	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1213			sewer			
	Tule (Saue)	sand/gravel filters for Fe, Mn, NH4 removal (preaerated)	1769			sewer			
	Segu (Saue)	sand/gravel filters for Fe, Mn, NH4 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	5,58E+03	5,36E+03	Rad.Prot.Centre
	well 412	II column (in parallel with I)	4.5	2370	2910		1,07E+04	1,31E+04	
		TOTAL well 412		9.0	1805	2050		1,62E+04	1,85E+04

* from gross α and β activity

Table 9.2 *Information about residues from treatment plants*

Waterworks	Treatment station	Material	²²⁶ Ra (Bq/kg)	²²⁸ Ra (Bq/kg)	²²⁸ Th (Bq/kg)	⁴⁰ K (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.

9.2. 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 9.2). 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 9.2), so, again, clearance levels could help.

RP 122 Part II (EC 2002) derives general clearance levels (GCLs) 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. GCLs are set to ensure compliance with 300 $\mu\text{Sv}/\text{y}$ individual effective dose to the public.

Discharge limits

Backwash water from treatment plants will contain a certain amount of natural radioactivity (namely, ^{226}Ra and ^{228}Ra). 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, IAEA 19 (IAEA 2001), NRPB 13/2 (National Radiological Protection Board 2002) and RP 135 (European Commission 2003), have been consulted. It should be stressed that IAEA and EC documents represent references at international level while NRPB is a national standard.

All documents give discharge limits values based on conservative scenarios. Limits are referred to the dose criterion of 0.3 mSv/y (individual effective dose), explicitly adopted by RP 135 and NRPB 13/2 (whilst IAEA 19 gives screening levels in terms of individual annual dose for single unit radionuclide discharge amount). The EU BSS (Basic Safety Standard) directive proposal (European Commission, 2009) considers 0.3 mSv/y the exemption level for public exposure for NORM work activities; actually, the directive proposal expressly includes the “ground water treatment” in the positive list of work activities potentially affected by NORM.

IAEA 19 and NRPB 13/2 present reference levels for liquid discharge into small rivers and sewers; critical scenario for discharge into sewer is sewing plant workers exposure for IAEA 19, the farmland use of formed sludges for NRPB 13/2. Screening levels of RP 135 concern release into rivers of various size and to coastal sea. Detailed description of cited documents is given in Addendum 9.1.

As Estonian national legislation (Radiation Act, 2004) does not define reference values for NORM wastes or discharges, reference to international documents is necessary.

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

Effluent comparison

Radium discharge rate from backwash water of Tallinn, Keila, Rakvere and Viimsi treatment systems (see Table 9.1) are compared to appropriate limits in Table 9.3.

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 limit 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 A.9.2 Addendum 9.1), 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 General Derived Constraint (GDC), more restrictive, has then been used (Table A.9.3 Addendum 9.1), that takes this scenario as the most critical for discharge into sewer. For Tallinn, discharge limits have been scaled with respect to Table A.9.2 Addendum 9.1 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 A.9.4 Addendum 9.1).

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 A.9.4 Addendum 9.1).

Table 9.3 *Discharge limits used to assess compliance of backwash water from existing treatment plants*

Reference	Compartment	²²⁶ Ra (Bq/y)	²²⁸ Ra (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

Actually, Tables 9.3, 9.4 and 9.5 summarize, to simplify reading, information about discharge limits (Tables A.9.2, A.9.3, A.9.4 Addendum 9.1) and discharge rates (Table 9.1), respectively; in the Table 9.5 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 9.4 *Discharge rates from existing treatment plants*

Effluent	Waterworks	²²⁶ Ra (Bq/y)	²²⁸ Ra (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 9.5 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 ²²⁶Ra only, NRPB 13 n. 2 document ²²⁸Ra GDC being not available.

Residues comparison

²²⁶Ra, ²²⁸Ra, ²²⁸Th activity concentrations of solid residues associated with filtration systems (Table 9.2) have been compared to General Clearance Levels of RP 122 part 2 (Table 9.6) to check whether conditions for unrestricted clearance (whatever being the residues fate) do apply.

Table 9.6 General Clearance Levels (Bq/kg) from RP 122 part II

	²²⁶ Ra ^(*)	²²⁸ Ra ^(*)	Th-228 ^(*)
All materials	500	1000	500

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

Comparison is carried out through the “sum index” estimate (see text above) and is shown in Table 9.7. For compliance, sum index must be less than 1.

Table 9.7 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 worker 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.

9.4. Literature data review

A short review on international literature data concerning management of effluents and residues from drinking water treatment plants is reported in Addendum 9.2. Analysing other world regions situation is, in fact, important to better understand specificity and common aspects of the Estonian treatment plants one.

Addendum 9.1- Description of reference documents for residues clearance levels and effluents discharge limits setting

The RP 122 Part II (EC 2002)

It derives general 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 $\mu\text{Sv/y}$ individual effective dose) achievement is itself the clearance level (called “general” as an effect of its derivation). Table A.9.1 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 A.9.1 *Rounded General Clearance Levels (Bq/kg) from RP122 Part II (European Commission, 2002)*

MATERIAL	²³⁸ Usec (*)	Unat (**)	²³⁰ Th	²²⁶ Ra (***)	²¹⁰ Pb (***)	²¹⁰ Po	²³² Thsec (*)	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 A.9.1 are “general” clearance levels, meaning that their compliance warrants dose criterion compliance independently 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.

The IAEA Report (IAEA 2001)

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 assessment 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 environmental factors are only 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 A.9.2, 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 as the same option is adopted in other technical reports related to similar topics (see, for instance, NRPB 13/2 and RP 135 documents described below).

Table A.9.2 “Generic environmental” screening factors and liquid discharge limits from IAEA (IAEA, 2001)

Scenario	²²⁶ Ra screening factor	²²⁶ Ra discharge limit (Bq/y) *	²²⁸ Ra screening factor	²²⁸ Ra 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 ²²⁶Ra only (not for ²²⁸Ra). To obtain ²²⁸Ra 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 (EPA 1993), and they have been calculated for each radionuclide taking into account the contribution of progeny grown up in a 30 y period of time: ²²⁶Ra, for instance, is considered to be in equilibrium with its decay products up to ²¹⁰Po, and ²³²Th is assumed in equilibrium with its progeny (²¹²Po included). For ²³²Th 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 96/29 Directive inhalation dose coefficient² for ²²⁸Ra (adults, most conservative absorption factor).

The NRPB document (NRPB, 2002)

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 individuals belonging to three age groups: infants (assumed to be 1 year old), children (assumed to be 10 years old) and adults (assumed to be 20 years old); 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 to irradiation and inhalation at river bank, ingestion of river water and fish, and 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.

Table A.9.3 *Generalised Derived Constraints for release to rivers and sewers from NRPB (NRPB, 2002)*

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

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

In Table A.9.3 GDCs for release to rivers and sewer are shown. Beside numerical values, limiting age group is reported.

When comparing data of tables a.9.2 and a.9.3 for river discharges, some differences are revealed. The level set for sewer by NRPB appears much more restrictive compared 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 ²²⁸Ra are lacking (as evident from table A.9.3).

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 RP 135 (EC 2003)

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 aspect description and detailed indications for air and water (rivers, sea) compartment 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 individual 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 A.9.4.

Screening levels are calculated under the following hypotheses:

- continuous release over 50 years
- secular equilibrium of short-lived daughters with long-lived parents within chain segments
- adult age group only considered

- exposure scenarios for river discharges include consumption of fish and water and external irradiation from bank sediment (PC Cream model was used)
- exposure pathways for marine discharges include ingestion of sea food (fish, molluscs, crustaceans) and external irradiation from beach sediments (Poseidon software, incorporating MARINA II modelling, was used).

The report recalls to apply 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).

A.9.4 Screening levels for discharge into rivers and sea from RP 135 (European Commission, 2003)

Scenario	²²⁶ Ra screening level (Bq/y) ⁽⁺⁾	²²⁸ Ra 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 segments from ²²⁶Ra to ²¹⁴Po

(-) equilibrium between ²²⁸Ra and ²²⁸Ac

Levels in Table A.9.4 can be compared to analogous figures in Tables A.9.2 and A.9.3 for river discharge. Table A.9.4 levels are also greater than the other ones 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 ²²⁶Ra and ²²⁸Ra, and 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, has to be mentioned that “Radiation Protection” series represents a sort of primary technical standard for EU Member States.

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

Addendum 9.2 - Literature data review

An important study to be cited is the book “Radon, Radium and Uranium in Drinking Water” (Cothorn, 1990), specifically the chapter titled “Disposal of Radium from Drinking Water treatment”, where the topic of management of effluents and residues deriving from effective well water radium removal techniques in USA is dealt with. These techniques are ion exchange resins, reverse osmosis, lime and lime-soda softening and iron filtration.

Backwash water values for iron filtration (sand, anthracite, aluminosilicate media) range from 0.4 to 17 Bq/l (radium combined, peak values), low ^{226}Ra levels are reported for washing water of lime-soda softening plants (0.7 to 1.4 Bq/l), whilst some relevant radium levels can occur in ion exchange and reverse osmosis waste waters: particularly, ion exchange brine peak combined radium ranges from 5 to 130 Bq/l. Typically, backwash water can be discharged to storm sewer⁵ or surface water (but sometimes recourse to sanitary sewer is due); brine, on the contrary, has to be treated in sanitary sewer systems. Generally, effluents from wastewater treatment plants show low radium content, contamination being removed by digested sludge.

Both water treatment plant sludges (lime, settled Fe-Mn backwash water) and wastewater treatment plant sludges are capable of containing not negligible radium concentrations: levels vary from some 40 Bq/kg (^{226}Ra , dry weight) up to 1300 Bq/kg both for ^{226}Ra and ^{228}Ra (dry weight). Similar values are found in other solid residues (filtration and ion exchange media), ranging from 50 Bq/kg (^{226}Ra) and 17 Bq/kg (^{228}Ra) to 1180 Bq/kg (^{226}Ra) and 440 Bq/kg (^{228}Ra). Practices with specific radium removal media (radium selective resins, ...) are discouraged due to elevated activity concentration of exhausted media, exceeding thresholds (74000 Bq/kg) for wastes to be managed as conventional radioactive ones.

As far as fate of solid residues from water treatment (sludges and ordinary spent media) is concerned, the authors refer to landfilling and landspreading (sludges only) options; landspreading is intended for agricultural purposes. Regulations for both

⁵ Sewer dedicated to rain water collection

clearance solutions are provided by Illinois and Wisconsin States. A graded approach for restrictions in material disposal and spread is established for increasing contamination levels of residues. Landfilling option is substantially neglected because landfill owners largely refuse to accept such wastes due to restrictions placed on disposal, site operation, final closure. Landspreading requirements are intended to limit final activity concentration of mixed soil (maximum ^{226}Ra activity concentration of 74 Bq/kg in Wisconsin, maximum combined radium activity concentration increase with respect to background of 4 Bq/kg in Illinois).

A comprehensive study on Ra, U, Rn removal techniques from drinking waters in USA was previously published by Reid (Reid, 1985). With regard to Ra, these techniques coincide with those described above: exchange resins (fit for small plants), reverse osmosis (fit for small plants), lime and lime-soda softening (fit for large plants) and iron filtration (through greensands only). One relevant assertion is that, with these conventional treatment systems, settled wastewater sludges would hardly exceed Ra activity concentrations of 3700 Bq/kg so they can be disposed in sanitary landfills (mixed with wastes of different nature) and they are not bound for designated hazardous or radioactive disposal sites. Figures similar to those in (Cothorn, 1990) are shown about Ra activity concentrations in brine from ion exchange treatment devices; however, this time, the authors believe that brine can be discharged to surface streams if there is sufficient dilution to handle salinity.

A recent research has been developed by some Australian scientists (Kleinschmidt, 2008) focused on assessing natural radionuclides content of urban raw and treated water from three waterworks and of respective effluents and residues. Water is mainly of surface origin and this represents an original aspect of the study. Surface water is treated mainly by filtration and disinfection (in addition, sometimes, to flocculation), groundwater through ion exchange resins and reverse osmosis. All potable treated waters appear to meet regulatory requirements, ranging, as far as ^{226}Ra is concerned, from 1.2 to 3 mBq/l (but also raw waters show low radioactivity content). Backwash water from surface water treatment plants are typically recycled, whilst liquid effluent from ground water treatment plants are discharged to sewer,

again compliant with regulatory standards (due to low activity concentrations of radionuclides - 3 to 14 mBq/l for ^{226}Ra). In the matter of solid residues formed both in surface water and groundwater treatment plants, ^{226}Ra levels in sediment/sludges are included between 37 and 85 Bq/kg, ^{226}Ra levels in spent treatment media (cationic resins, coal filters, alum concentrate) range from 6 to 120 Bq/kg. Sediment and sludges are stocked on site or cleared for land use application (including soil conditioning), spent media are disposed of by controlled landfill. Effective doses to individuals of the reference group of population due to the presence of various natural radionuclides in solid residues are assessed by RESRAD model (ANL, 2007) for three scenarios: workers of water treatment plants stockpiling formed sludge (max value: 58 $\mu\text{Sv/y}$), workers of landfill undertaking expired ion exchange resins (max value: $< .1\mu\text{Sv/y}$), suburban resident using sludge as a soil conditioner for home cultivation of vegetables and fruits (value: 205 $\mu\text{Sv/y}$). All estimates have to be considered conservative.

10. CONCLUSIONS

The problem of water radioactivity in Estonia

In a considerable part of Estonian groundwaters, a non negligible amount of natural radionuclides, especially radium isotopes, is found. Notwithstanding its natural origin - that is the geological structure of the aquifer – it causes a radiation protection issue, because the parameter value of 0.1 mSv/year of European Directive 98/83 (EC 1998), which is also the limit for water radioactivity in Estonian national regulation, is often exceeded.

The presence of small amounts of radionuclides in drinking water is a common, natural phenomenon. Both ground and fresh waters contain variable amount of elements (Ca, Mg, Na and so on) coming from erosion and dissolution of rocks; radionuclides of natural series (U and Th) are transferred into water in a similar way. Usually uranium isotopes, for chemical reasons, are the most abundant dissolved radionuclides. High concentrations of radium isotopes are relatively uncommon, especially as extended areas are concerned. Since radium isotopes are more radiotoxic than uranium ones, high doses values may occur when their concentration is relatively high. This is observed in Northern Estonia, in some counties of United States and in smaller areas of other countries (Spain, Sweden, Israel etc.). In Estonia radium isotopes concentration is considerably higher in the Cambrian-Vendian (Cm-V) aquifer, which is the deepest one and lies in contact with the crystalline geologic basement, but the detailed transfer mechanism and origin of radium have not been fully explained yet.

The Cm-V aquifer feeds a relevant part of aqueducts in Northern Estonia: around 50% of population in Harjumaa and Ida Virumaa counties and 10% in Ida Virumaa and Laanemaa counties drink this kind of water. This problem is well known since many years and a wide monitoring of water resources has been carried out in last decades. However, a careful examination of collected data shows that they suffer of some problems.

- Samples have been collected at wells, before any mixing or treatment thus they are not fully representative of the water actually delivered to population.
- Monitoring has been carried out mainly in Northern Estonia where the radioactivity problem is known to be more relevant. Population coverage from 50% to 100% was accomplished. On the opposite, the population coverage in Centre and Southern Estonia is very low (from 0 to 20%) even considering that radioactivity problem is lower though not absent.
- Data have been collected over many years, they come from different laboratories, the procedures employed are not always known, measure uncertainties are often lacking. They are quite unhomogeneous too: gross alpha and beta activities, radium and uranium isotopes concentrations are available, but only few samples are completely characterized.

Despite these limitations, Estonian data base is quite large and allows drawing some preliminary conclusions.

- As the adult age class is considered, the total indicative dose (TID) of 0.1 mSv/y is exceeded mainly in Northern Estonia and in areas provided with Cambrian-Vendian waters (91% of available data). In few cases it is exceeded in Central and Southern Estonia too (21% of (the few) available data).
- If the TID is calculated for non-adult age classes, the value of 0.1 mSv/y would almost always be exceeded in whole Estonia (in 100 % cases of lactation age population).
- The individual whole life detriment due to the consumption of Estonian Cm-V waters, all life long, is $2.6 \cdot 10^{-3}$.
The largest annual contribution (i.e. 10%) to whole life detriment is due to dose received in the first year (lactation age).
- The dose is mainly attributable to ^{228}Ra (74 % for adult age class and 88 % for infant age class)
- Some considerations about the size of Estonian waterworks are possible too: most aqueducts are small sized ones (83 % serves less than 500 people). A

uniform size distribution is found in the whole country. Nevertheless the 15 biggest waterworks provide water to about 740,000 people (67 % of Estonian population). These observations are potentially useful for both monitoring and remediation purposes.

Statistical elaboration of data gave the following interesting indications (to be confirmed with a wider set of data).

- Within the Cambrian-Vendian waters a correlation has been observed between some chemical parameter (e.g. mineralization, barium concentration) and radium activity concentration. This should allow identifying the potentially more hazardous situations on the basis of chemical analyses, normally available for every waterworks.
- An empirical correlation between ^{226}Ra and ^{228}Ra has also been found. This would allow a rough preliminary dose estimate based on the measurement of ^{228}Ra only. Resulting uncertainties could be reasonably low since its contribution to dose is considerably higher than ^{226}Ra one.

The regulatory system

The EC Council Directive on drinking water 98/83/EC includes 0.1 mSv/y of Total Indicative Dose (TID) among indicator parameters (not mandatory).

- When this value 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, applying the so called ALARA – As Low As Reasonably Achievable - principle.
- A non-mandatory Total Indicative Dose for drinking water should be preferred, being the use of strict limit values not recommended in situations involving wide exposure to natural radiation sources, and where a primary good is involved.
- In deciding requirements for monitoring and remedial actions, doses to lower age classes should not be neglected. In particular, a consumption limitation for high radium concentration water by babies in lactation age could be recommended.

- Being water for drinks and foodstuff preparation also included in the scope of the EC Directive 98/83, a specific regulation could be similarly envisaged for soft drinks production, both from the point of view of justification and to limit exposure to teenagers, which is an age class at risk (see Figure 4.7).

Waste management

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

Though neither a 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 (belonging to Tallinn, Rakvere, Keila and Vimsi waterworks), the following conclusion can be drawn.

- Backwash water of treatment systems generally comply with reference levels; Rakvere aqueduct is an exception, as its backwash waters is sent to the town sewer, whose sludges are employed in agriculture.
- Solid residues of treatment systems (filter material and sludges) exceed general clearance levels. Specific assessment is therefore required to search for specific clearance levels.

Literature data confirm that effluents and residues from water filtering devices can sometimes pose management problems, also in other parts of the world too (e.g. USA).

The improvement of water quality

An extension of the monitoring of Estonian water resources is needed as the first step. Some general requirements for a national campaign are given.

- The samples should be drawn after any mixing and treatment, they should represent the water as delivered to population
- The geographic representativeness should be increased (Central and Southern Estonia should be monitored too, even if to a lesser extent than Northern Estonia)

Some of the above discussed observations on existing data could be useful for the planning of the campaign. A proper choice of analytical parameters is of paramount importance since radiometric and radiochemical analyses are lengthy and cumbersome:

- Gross alpha and beta activities, which are normally used as indicator parameters, are quite useless in the case of Estonian waters.
- Uranium isotopes give a minor contribution to dose and their determination could be useful only to ascertain if their chemical toxicity has to be considered (U concentration > 15 ppb, corresponding to 186 mBq/L of ^{238}U).
- Other natural radionuclides as ^{210}Po and ^{210}Pb will be interesting for further refinements.
- Radium isotopes, and namely ^{228}Ra should be always measured.
- Many analytical procedures for radium isotopes are available. The chosen method should have a sufficient sensitivity (e.g. Minimum Detectable Activity ≤ 20 mBq/L) and it should employ widely available techniques and relatively quick procedure. For these reasons gamma spectrometry on preconcentrated samples is recommended for ^{228}Ra measurements. ^{226}Ra can be effectively determined by the same technique after progeny ingrowths or by liquid scintillation counting.
- Quality control procedures should be always adopted; if more laboratories are involved in measurements, periodic intercomparisons between them are highly recommended.

Once a reliable characterization of waters has been carried out, strategies for dose reduction can be adopted:

- Large waterworks fed by Cambrian-Vendian waters should be considered first, as the 12 biggest ones (out of 140) provide water to 60% of population using “risky” Cm-V waters.
- In some pumping stations treatment plants designed for iron and manganese softening are presently operating, but they have a variable and not sufficient effectiveness in radium removal. Their upgrade employing specific radium absorber (as manganese dioxide) could be studied.
- The performances of the experimental plant which is being tested in Viimsi aqueduct are not completely ascertained, especially if large scale treatment is concerned.
- Wastes from treatment plants can pose serious problems, due to high radium concentration even if the actual removal efficiency of existing equipments is quite low. More problems will arise with more effective treatment plants.
- Many commercial methods are available on the market. Most of them are currently employed in United States. Their cost are highly variable, depending on the technique and the size of aqueduct.
- For small sized aqueduct a sustainable removal technique can hardly be found. A rationalization of waterworks (reduction of their number and increase of their size) could allow facing effectively the problem.
- In many cases the use of alternative water resources (mixing water from shallower aquifer or freshwater) seems to be possible and should be encouraged.

11. AUTHORS' REFERENCES AND ACKNOWLEDGMENTS

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