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Disposal of Radioactive Waste Arising from Water Treatment: Recommen- dations for the EC

Final Report of the WP 8 of the TENAWA project

M. Annanmäki, T. Turtiainen, H. Jungclas, Ch. Rauße

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ABSTRACT

Ground water, especially bedrock water, may contain high amounts of natural radioactivity. Elevated levels of natural radionuclides in ground water are mainly associated with uranium and thorium rich soil and rocks. Various processes based on different principles can be applied to the removal of radioactivity from water. Aeration and granular activated carbon (GAC) filtration are used to remove radon from household water. Ion exchangers are applied to the removal of uranium and radium. Lead and polonium may sometimes be removed by ion exchange technology as well. Membrane techniques are applied to the removal of uranium, radium, lead and polonium. Radionuclide removal can also be carried out using adsorptive materials.

When different kinds of treatment methods are used to remove natural radioactivity from drinking water, wastes containing natural radioactivity will be produced. The wastes are in liquid or solid form. Liquid wastes are produced when materials used to accumulate radioactivity are regenerated or backwashed. Solid wastes are formed in cases where regeneration or backwashing are not used or cannot be used, and when the materials are taken out of service. GAC filters emit gamma radiation when they are in service.

To gather information on existing national regulations and guidelines on the treatment and disposal of radioactive wastes produced by various water treatment methods, a questionnaire was sent to all the Member Countries of the European Union.

It is recommended that if aeration is used to remove radon from drinking water the aeration system should be fitted in such a way that the radon released from water does not enter the rooms occupied by the residents but is ventilated into the outside air. It is recommended that the annual effective dose to inhabitants from external gamma radiation of a GAC filter should not exceed 0.1 mSv. It is also recommended that the dose rate at a distance of 1 m from the GAC filter should not exceed 1 μ Sv/h. To achieve these aims the GAC filter should be equipped with special shielding to attenuate gamma radiation. It is also recommended that the wastes containing natural radioactivity in solid form could be discharged into communal dumps and wastes containing natural radioactivity in liquid form could be discharged into the sewer.

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Avainsanat luonnon radioaktiivisuus, juomavesi, vedenkäsittely, radioaktiivinen jäte, säteilysuojus

TIIVISTELMÄ

Pohjavesi – varsinkin kalliopohjavesi – voi sisältää korkeita pitoisuuksia luonnon radioaktiivisia aineita. Korkeita luonnon radionuklidipitoisuuksia tavataan pääasiassa pohjavesissä, jotka esiintyvät uraani- ja toriumpitoisissa kivi- tai maalajeissa. Eri toimintaperiaatteisiin pohjautuvia menetelmiä voidaan käyttää juomaveden radioaktiivisuuden poistoon. Ilmastusta ja aktiivihiihiisuodatusta käytetään radonin poistoon talousvedestä. Ioninvaihtoa käytetään uraanin ja radiumin poistoon. Myös lyijyä ja poloniumia voidaan poistaa ioninvaihtotekniikoilla. Radionuklidien poisto voidaan toteuttaa myös kalvotekniikoilla, kuten käänteisosmoosilla ja nanosuodatuksella. Näitä kalvotekniikoita sovelletaan uraanin, radiumin, lyijyn ja poloniumin samanaikaiseen poistoon. Myös adsorptiivisia materiaaleja käytetään luonnon radioaktiivisuuden poistoon.

Erlaisia vedenkäsittelymenetelmiä käytettäessä syntyy jätteitä, jotka sisältävät luonnon radioaktiivisuutta. Jätteet voivat olla kiinteitä tai nestemäisiä. Nestemäisiä jätteitä muodostuu, kun radionuklideja kerääviä materiaaleja regeneroidaan tai niille tehdään vastavirtahuuhtelu. Kiinteitä jätteitä syntyy silloin, kun regenerointia tai vastavirtahuuhtelua ei voida tehdä tai kun materiaalit otetaan pois käytöstä. Aktiivihiihiisuodattimet lähettävät käytössä ollessaan gammasäteilyä.

Kaikille EU:n jäsenvaltioille lähetettiin kyselylomake, jolla tiedusteltiin voimassaolevia kansallisia säädöksiä ja ohjeita, jotka koskevat vedenkäsittelyssä syntyvien radioaktiivisten jätteiden käsittelyä ja hävittämistä.

Suosittelaa, että radonin poistoon käytetyissä ilmastimissa poistoilmaputki on kiinnitetty siten, ettei käsittelyssä syntyvä radonpitoinen ilma pääse huonetiloihin vaan se johdetaan tehokkaasti ulkoilmaan. Suositellaan, ettei aktiivihiiisuodattimista tulevan ulkoisen gammasäteilyn aiheuttama efektiivinen annos ylitä arvoa 0,1 mSv vuodessa. Lisäksi suositellaan, ettei annosnopeus yhden metrin päässä suodattimesta ylitä arvoa 1 µSv/h. Näiden tavoitteiden saavuttamiseksi aktiivihiiisuodattimet tulee varustaa säteilysuojilla, joilla vaimennetaan gammasäteilyä. Lisäksi suositellaan, että luonnon radioaktiivisuutta sisältävät kiinteät jätteet voidaan hävittää kunnallisille kaatopaikoille ja että nestemäiset jätteet voidaan laskea viemäriin.

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PREFACE

The shared-cost research project “Treatment Techniques for Removing Natural Radionuclides from Drinking Water” (TENAWA) was carried out in the fourth Framework Programme 1994–98 of research and training funded by the European Commission in the sector of Nuclear Fission Safety. The aim of the TENAWA project was the evaluation of treatment techniques for removing natural radionuclides from drinking water. It was carried out by the following partners:

1. STUK—Radiation and Nuclear Safety Authority, Finland
2. BALUF—Federal Institute for Food Control and Research, Austria
3. PUMA—Philipps University Marburg, Nuclear Chemistry, Germany
4. IWGA—Control University of Agriculture, Department for Water and Wastewater Engineering, Industrial Waste Management and Water Pollution, Austria
5. SSI—Swedish Radiation Protection Institute, Sweden
6. ESWE—Institute for Water Research and Water Technology, Germany
7. HYRL—University of Helsinki, Laboratory of Radiochemistry, Finland

The TENAWA project was divided into 13 work packages:

- WP 1.1: General Considerations: Literature Survey on Natural Radioactivity in Drinking Water and Treatment Methods in European Countries
- WP 1.2: General Considerations: Intercomparison of Analysis Methods
- WP 1.3: General Considerations: Definition and Classification of Different Water Types and Experimental Conditions
- WP 2.1: Removal of Radon by Aeration: Testing of Commercially Available Equipment for Domestic Use
- WP 2.2: Removal of Radon by Aeration: Testing of Various Aeration Techniques for Small Waterworks
- WP 3.1: Removal of Radionuclides from Private Well Water with Granular Activated Carbon (GAC): Removal of Radon

- WP 3.2: Removal of Radionuclides from Private Well Water with Granular Activated Carbon (GAC): Removal of U, Ra, Pb and Po**
- WP 4: Removal of Radioactivity by Methods Used for Fe- and Mn-removal from Private Wells**
- WP 5.1: Removal of U, Ra, Pb and Po by Ion Exchange Methods. Removal of U and Po from Private Ground Water Wells using Anion Exchange Resins**
- WP 5.2: Removal of U, Ra, Pb and Po by Ion Exchange Methods. Removal of Ra and Pb from Private Ground Water Wells using Cation Exchange Resins**
- WP 6: Removal of U, Ra, Pb, and Po with Adsorptive or Membrane Filters**
- WP 7: Speciation of U, Ra, Pb and Po in Water**
- WP 8: Disposal of Radioactive Wastes by Water Treatment Methods: Recommendations for the EC.**

Many discussions have taken place among our colleagues in and beyond the TENAWA project and we gratefully acknowledge these. We also wish to thank all the companies and the house owners for the good co-operation during the project.

1 INTRODUCTION

Groundwater, especially bedrock water, may contain high amounts of natural radioactivity. Contrary to the case of ground water the amount of natural radioactivity in surface water is low. Elevated levels of natural radionuclides in ground water are mainly associated with uranium and thorium rich soil and rocks. The naturally occurring radionuclides which may be present in drinking water are mainly radon (^{222}Rn) and its short-lived daughters, but also uranium (^{238}U , ^{234}U), radium (^{226}Ra), polonium (^{210}Po) and lead (^{210}Pb). The groundwater may also contain tritium (^3H), radioactive carbon (^{14}C) and potassium (^{40}K).

At the beginning of the TENAWA project a literature survey on the occurrence of natural radioactivity in drinking water and on the treatment methods used in European countries for removing natural radionuclides from drinking water was made. The report presents data on the radionuclide contents of drinking, mineral, ground and surface water from 17 European countries. The highest values were found in the crystalline areas (especially in granite areas) of Finland, Sweden, Spain and Ukraine.¹

If the amount of radioactivity in drinking water is too high some water treatment method to decrease the amount of radioactivity should be used. The water treatment methods normally used are aeration, GAC filtration (Granular Activated Carbon), ion exchange and reverse osmosis.

Aeration is a method that can be applied to removing high levels of radon from drinking water. GAC filtration is normally used when the radon concentration of water is less than 5,000 Bq/L, because GAC filters emit gamma radiation.

Fe- and Mn-removal equipment also remove natural radioactivity from water. Ion exchangers are applied to the removal of uranium and radium. Lead and polonium may sometimes be removed by ion exchange technology as well. Radionuclide removal can be carried out using reverse osmosis or

¹ Haberer K, Raff O, Akkermann-Kubillus A. Natural Radionuclides in Drinking Water in Europe and Treatment Methods for their Removal. Elsevier Science, Oxford (in press).

nanofiltration. These membrane techniques are applied to the removal of uranium, radium, lead and polonium. Adsorptive materials are also used to remove natural radioactivity.

When treatment methods for reducing natural radioactivity from drinking water are used, radioactive wastes in liquid or solid form are produced. In some cases the dose rate² in the vicinity of the treatment unit may also increase. When GAC filtration is used to remove radon the dose rate near the filter may vary from 1 to 100 $\mu\text{Sv/h}$. The amounts of radium and lead may also increase in the filter. Typical values for radium may be from 100 to 10,000 Bq (10–1,000 Bq/kg) and for lead from 10,000 to 1,000,000 Bq (1,000–100,000 Bq/kg).

In the case that the water contains uranium and anion exchangers are used to remove it, the amount of uranium collected may vary from 1 to 100 g. When anion exchangers are regenerated, the regeneration liquid may contain uranium from 0.1 to 10 g.

Some EU Member Countries have limits on the amount of natural radioactivity that a private person may have in his or her possession or that can be discharged either into the environment or into communal dumps. To find out these limits or other regulations concerned a questionnaire aiming at gathering information on existing national regulations and guides on the treatment and disposal of radioactive wastes produced by various water treatment methods, was sent to all the Member Countries (Appendix 1).

² 'Dose rate' is used throughout the text. Unless defined otherwise, the 'dose rate' stands for 'ambient dose equivalent rate'.

2 SCOPE

In the TENAWA project the capability of various water treatment methods to remove natural radioactivity from drinking water was studied. The amounts of wastes containing radioactivity produced by different water treatment methods were also estimated.

Some water treatment methods may also act as a source of radiation in the place of use. Radioactive wastes produced by various water treatment methods may increase radiation exposure to the people if released into the environment.

This report aims at recommending guidelines for the treatment and disposal of radioactive wastes produced by various water treatment methods. The Member Countries of the European Union can use the recommendations presented in this report as a basis for their own regulations or the European Commission may recommend a common approach for all the Member Countries. In order to achieve these aims:

- a questionnaire aiming at gathering information on existing national regulations and guides on the treatment and disposal of radioactive wastes produced by various water treatment methods, was sent to all the Member Countries
- the wastes produced by different water treatment methods were monitored and information was gathered during the whole of the TENAWA project
- the dose rate close to the GAC filters and other treatment units was measured
- the expected doses in handling different kinds of treatment units loaded with naturally occurring radionuclides were estimated.

3 NATURAL RADIOACTIVITY IN DRINKING WATER

3.1 Recommendations and guidelines for natural radioactivity in drinking water

3.1.1 EU directive

In Council Directive 98/83/EC on the Quality of Water Intended for Human Consumption, the European Union has laid down a total indicative dose expressed as an annual effective dose of 0.1 mSv. Radon and its daughters (short-lived and long-lived) as well as tritium and potassium are excluded. The limit is not compulsory but when it is exceeded, the respective Member Country should assess the health risk due to the use of water, and if needed, take actions to protect the health of the population.³

3.1.2 WHO recommendations

The World Health Organisation (WHO) has issued recommendations for natural and man-made radionuclides in drinking water. The limits for drinking water are:

- Gross alpha activity (without radon) 0.1 Bq/L
- Gross beta activity (without tritium) 1 Bq/L.

If either gross alpha or gross beta activity exceeds the limit, an individual radionuclide concentration has to be determined and a total annual dose has to be calculated. In the case this dose exceeds a value of 0.1 mSv, remedial actions should be taken.⁴

³ Council Directive 98/83/EC on the Quality of Water Intended for Human Consumption.

⁴ WHO. Guidelines for drinking-water quality, 2nd ed., Vol. 2. Geneva: World Health Organization, 1996.

3.1.3 Recommendations of the Nordic countries

The authorities responsible for radiation protection in the Nordic countries (Denmark, Finland, Iceland, Norway, Sweden) will issue recommendations (under preparation) on naturally occurring radiation. The recommendations also include recommendations on natural radioactivity in drinking water. It will be recommended that the exemption level for radon in drinking water be 100 Bq/L. The recommended upper level for radon in drinking water is 1,000 Bq/L. The recommended upper level, expressed as an annual effective dose, for exposure to long-lived radionuclides in drinking water is 1 mSv.

3.2 Natural radioactivity

The main primordial natural radionuclides as sources of human exposure to radiation are uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K). The heavy nuclides ^{238}U and ^{232}Th decay through long decay-chains which include several radioactive elements and end in the stable isotopes of lead. Uranium and thorium and nuclides of their decay-chains are present in the ground in all rocks and soils, the amount depending on the rock type. In the decay-chain of ^{238}U are radon (^{222}Rn) and its short-lived and long-lived daughter products. The long-lived daughter products of radon are ^{210}Po and ^{210}Pb . Table I sets out all the nuclides present in the uranium decay chain.

Nuclide ^{40}K will not be further discussed because variation in the amount of ^{40}K in the water has no substantial effect on the dose.

Table I. Radionuclides of the uranium decay chain.

Nuclide	Half-life	Radiation type*
U-238	4.5 billion years	alpha
Th-234	24 days	beta
Pa-234	1.2 minutes	beta
U-234	250,000 years	alpha
Th-230	75,000 years	alpha
Ra-226	1,600 years	alpha
Rn-222	3.8 days	alpha
Po-218	3.1 minutes	alpha
Pb-214	27 minutes	beta
Bi-214	20 minutes	beta
Po-214	160 microseconds	alpha
Pb-210	23 years	beta
Bi-210	5 days	beta
Po-210	138 days	alpha
Pb-206	stable	none

*most of these isotopes also emit gamma radiation.

Radiation exposure due to natural radioactivity can be divided into external and internal exposure. External exposure is caused by gamma radiation emitted by the nuclides in the decay chains and by potassium. Internal exposure is caused by the alpha radiation of the short-lived decay products of radon when radon-laden air is inhaled and by the alpha and beta activity of the long-lived decay products of radon when e.g. water containing these nuclides is ingested. In special cases also the short-lived decay products of radon may be a cause of internal exposure when ingested.

The exposure to radiation from indoor radon is usually greater than that from all the other components of natural radiation. The main cause of high concentrations of radon indoors is the influx of radon gas from the subjacent soil and bedrock.

All building materials contain varying amounts of naturally occurring radionuclides. Naturally occurring radionuclides in building materials contribute to the exposure to radiation through emitted gamma radiation and indirectly through the exhalation of radon from the material into the indoor air.

Drinking water is in some regions a remarkable source of radiation exposure. Elevated concentrations of natural radionuclides in drinking water have been reported e.g. in Finland, France, Germany, Spain and Sweden (UNSCEAR 1993). Internal exposure due to household water is caused by ingestion of radionuclides and by inhalation of radon gas released from water.

In underground uranium and other mines and in spas and tourist caves elevated levels of radon occur. Especially in the Nordic countries, indoor radon may also be high at normal, above-ground workplaces.

3.3 Occurrence of natural radioactivity in drinking water

The distribution of accumulations of natural radionuclides in Europe can be estimated—as a first approximation—on the basis of the occurrence of the main European uranium deposits. These are set out in Figure 1. Most of the deposits are related to the Moldanubian zone of the Herzynian (Variscan) orogen or to the directly adjacent zone. The Moldanubian Zone is the inner zone of the Herzynian Orogen, corresponding to the area of maximum orogenic, metamorphic and plutonic activity. It extends from the north-western part of the Iberian peninsula, through Brittany, central France, and southern Germany to former Czechoslovakia. Moldanubian outcrops mainly form the crystalline complexes of

- the Central Iberian Zone,
- the Vendée of the Armorican Massif (Brittany),
- the French Central Massif,
- the Vosges and the Black Forest (western France / southern Germany) and
- the western Bohemian Massif including the Erzgebirge (former Czechoslovakia and the southern part of eastern Germany).

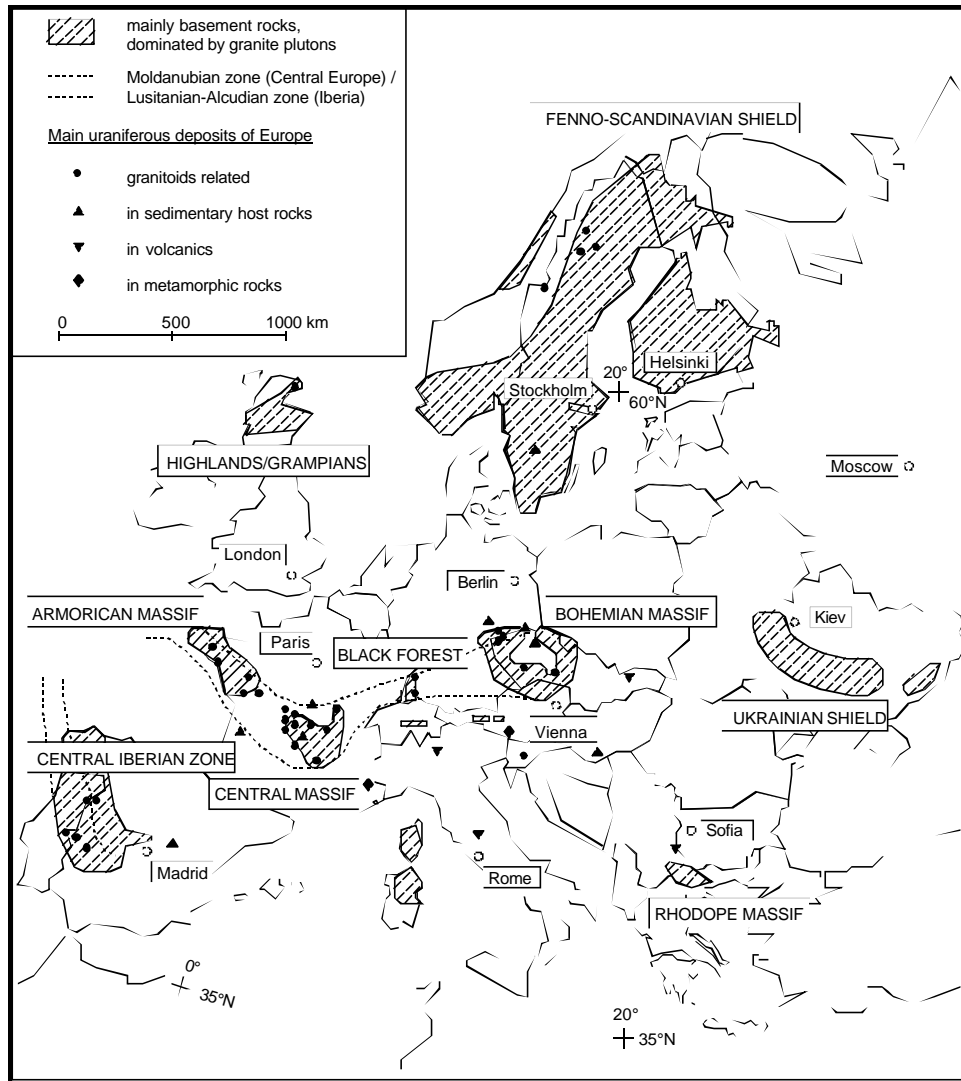


Figure 1. The main uraniumiferous deposits of Europe and zones dominated by basement rocks as areas with potentially elevated contents of natural radionuclides in ground water.

Besides the Moldanubian Zone there are some other European areas where uraniumiferous deposits occur. The largest and most important is Scandinavia. Most of Scandinavia is part of the proterozoic Baltic Shield (except for the Caledonian orogen in the north-west). The Baltic Shield mainly consists of high-grade metamorphic rock and large intruded igneous rock bodies

(mainly granitoids). So most of Finland, Sweden and the southern part of Norway form a huge crystalline complex (though there are many different tectonical and lithological units, of course). Not many major uraniferous deposits exist in Scandinavia (only a few in Sweden), but this is due to the economic point of view. Wide areas of the crystalline Scandinavian bedrock are known to have considerably elevated contents of uranium and other natural radionuclides. Most of them are related to granitoids.

A few other major uraniferous deposits exist in southern Europe (The Alps, Apennines, Carpathians, and Balkans) and in northern Britain. Most of them are also related to granitoids or to silicic volcanics.

The data concerning the radionuclide contents of European ground water and the distribution of uraniferous deposits in Europe show that there is a close relation between high radionuclide values and crystalline complexes, especially granitoids. In addition to the above-mentioned, such areas are:

- the Ukrainian crystalline complex (Ukrainian shield),
- the southern crystalline Balkans,
- the granted massifs of the Central Alps,
- the granitoid massifs of Corsica and Sardinia and
- the crystalline area of the Scottish Highlands.

Surely, there are many other, smaller-potential risk areas concerning high natural radionuclide contents in ground water in Europe. For example, areas with small uraniferous accumulations of only local importance. Many of these exist typically in smaller silicic volcanic rock bodies or in their sedimentary surroundings. Even some of the major European uraniferous deposits are hosted in sedimentary rock.

Generally speaking every geological uranium/thorium accumulation can cause high natural radionuclide contents of ground water in their surroundings. So far the highest values in Europe have been found in Finland, Sweden and western Spain. The degree of concentration and the extension of enriched ground water depend on lithological, hydraulical, and hydrochemical conditions.⁵

⁵ Haberer K, Raff O, Akkermann-Kubillus A. Natural Radionuclides in Drinking Water in Europe and Treatment Methods for their Removal. Elsevier Science, Oxford (in press).

4 METHODS FOR REMOVING NATURAL RADIOACTIVITY FROM DRINKING WATER

4.1 Introduction

Various processes based on different principles can be applied to remove radioactivity from drinking water. When assessing the potential of a treatment technique for producing radioactive wastes the following points should be considered:

1. *The operating principle of the equipment.* Some of the treatment methods retain radionuclides in a fixed matrix. Depending on the regeneration interval of the matrix, the activity of the influent and the rate of water use, large amounts of radionuclides may be accumulated in the removal unit. There are also matrices that cannot be regenerated and thus the activity will not be diluted into a larger volume or mass. Some of the treatment methods do not accumulate radionuclides and they operate in such a way that the radionuclides occurring in water are continuously rejected from the purified water and released into the sewer or into the open air.
2. *The half-life of the radionuclides.* The radionuclides that need to be removed from drinking water or household water mainly originate from the uranium series. The half-lives of the radionuclides found in the wastes determine how long the waste remains radioactive. The most long-lived are ^{238}U , ^{234}U , ^{230}Th , ^{226}Ra and ^{210}Pb . Therefore, the main stress is laid on these when estimating the amounts of radioactive wastes produced in water treatment.

When water treatment systems are set up, inhabitants' exposure to gamma radiation should be considered. Although most of the radionuclides set out in Table I emit gamma radiation, the most significant ones are ^{214}Pb (352 keV with a gamma intensity of 37% and 295 keV with a gamma intensity of 19%) and ^{214}Bi (609 keV with a gamma intensity of 46%, 1,760 keV with a gamma intensity of 16% and 1,120 keV with a gamma intensity of 15%). Therefore, if they are accumulated in a treatment unit, high dose rates can be found in the vicinity of the unit.

In this chapter, the removal principles of different water treatment units will be described. Based on the removal principles and the half-lives of the radionuclides, potentials for accumulation of radioactive wastes and for causing elevated dose rates will be discussed.

4.2 Aeration

Aeration is a method that is applied to remove radon from all household water (point-of-entry). Depending on the design of the treatment system, aeration can be used to remove even extremely high levels of radon or to treat large amounts of water. Therefore, aeration is the main technique for radon removal in systems that supply water for more than one household, or when the radon concentration exceeds 5,000 Bq/L.

Aeration is based on the mass transfer of radon from the aqueous phase to the gaseous phase. The transfer can be accomplished based upon different principles. Most commercial aerators combine these principles in order to achieve the best performance.

1. *Spray aeration.* Radon-laden water is forced into a ventilated tank through a spray nozzle. The water is transformed into small droplets and radon is released from the interfacial surface into the surrounding air. The radon-rich air is exhausted into the open air through a ventilation channel by a fan. The aerated water is fed into the household water line.
2. *Diffused bubble aeration.* Radon-laden water is fed into a tank where it is brought into contact with small air bubbles. The bubbles can be created with an air compressor and a plate diffuser fixed to the bottom of the tank, or by circulating the water through an ejector, which mixes clean air into the water. The bubbles rise to the surface of the water where they are exhausted into the open air through a ventilation channel.
3. *Packed tower aeration.* Water is directed into the top of a vertically positioned tower. The tower is filled with an inert packing material (e.g. nets or plastic balls). As the water passes through the packing material, a large surface area between the air and the water is created from which radon can be released. A small blower fan forces air up through the packing, which carries the radon gas out of the unit.

4. *Bubble plate or shallow tray aeration.* These techniques are similar to the diffused bubble aeration but the air bubbles are larger and more air is usually compressed through the water.

Aeration is capable of removing only radon, which is directly funnelled into the open air. Therefore, no accumulation of radioactivity occurs in the aeration process. The concentration of radon in the exhaust air depends on the radon concentration in raw water and the air-to-water (A/W) ratio. Mostly, A/W ratio of about 10 is sufficient to remove 95% of radon, though much higher ratios are often applied. Table II sets out the concentrations and volumes of radon-rich exhaust air produced by systems with different radon concentrations in raw water and different rates of water use.

Table II. The total activity and the activity concentration of radon and the volume of exhaust air generated daily by systems that apply A/W ratio 11 and treat different amounts of water exhibiting different concentrations of radon.

Rn in water (Bq/L)	Total Rn released daily (MBq), when the water flow rate is					Rn in exhaust air (Bq/m ³)
	0.3 m ³ /d	0.5 m ³ /d	1.0 m ³ /d	5.0 m ³ /d	10 m ³ /d	
1,000	0.3	0.5	1	5	10	91,000
3,000	0.9	1.5	3	15	30	270,000
5,000	1.5	2.5	5	25	50	450,000
10,000	3.0	5.0	10	50	100	910,000
Volume of exhaust air (m ³)	3.3	5.5	11	55	110	

The only radioactive substance extracted from water by aeration is the radon-rich exhaust air. Table II shows that radon concentration in exhaust air is usually very high. Therefore, the ventilation channel must be designed in such a way that there is no risk of letting the radon bearing exhaust air back into the house. The outlet of the exhaust channel must be as far as possible from the replacement air ventilators of the dwelling. Since the exhaust air contains water vapour, the outlet of the ventilation channel must be designed so that it does not become blocked due to freezing in winter. If the radon-laden air is properly released into the open air, it rapidly becomes diluted by air currents and incurs no exposure to the residents.

Aerators do not significantly increase the dose rates in the dwellings. Since aeration does not accumulate radionuclides, the gamma radiation originating from the aerator is caused by the short-lived radon daughters that normally occur in the water. Water attenuates the gamma rays relatively well. The highest dose rate measured on the surface of an aeration tank was 4 $\mu\text{Sv/h}$ (radon concentration 40,000 Bq/L).

4.3 Granular activated carbon filtration

Granular activated carbon (GAC) filters are 20–100 L filters that are installed at the water line right after the pressure tank in such a way that they treat all water used by a household (point-of-entry). The removal efficiency for radon depends on the bed size of the GAC filter and can be close to 100%. GAC filtration can best be employed in a single household where the radon concentration of water does not exceed 5,000 Bq/L because GAC filters emit gamma radiation when they are in service. The higher the radon concentration and the larger the water use, the more intense the dose rate around the filter becomes. Furthermore, GAC filters accumulate ^{210}Pb that is formed by the radioactive decay of radon inside the filter. The rate of accumulation of ^{210}Pb also depends on the rate of daily water use and the radon concentration in the influent.

Radon is retained on GAC by physical adsorption. Although the adsorption process is reversible, the GAC beds are capable of retaining most of the radon long enough to allow its radioactive decay inside the filter. After three weeks in service the adsorption rate of radon equals the rate of its radioactive decay; thus there is a constant number of radon atoms (and its short-lived daughter products) adsorbed on the GAC bed. This state is called the adsorption-decay steady state and results from the relatively short half-life of radon.

The short-lived decay products, ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po , and the long-lived ^{210}Pb generated by radon are retained on the carbon bed. ^{214}Pb and ^{214}Bi cause most of the gamma rays but are negligible in the waste due to their short half-life. The long-lived ^{210}Pb , however, is accumulated during the whole time the GAC filter is in service. Depending on the water consumption and radon concentration in the influent, high levels of activity

of ^{210}Pb may be found in spent GAC beds. ^{210}Pb attains secular equilibrium with its progeny, ^{210}Bi and ^{210}Po ; thus the total activity of the bed is three times higher than for ^{210}Pb alone. Figure 2 sets out the activity of ^{210}Pb accumulated on a GAC bed.

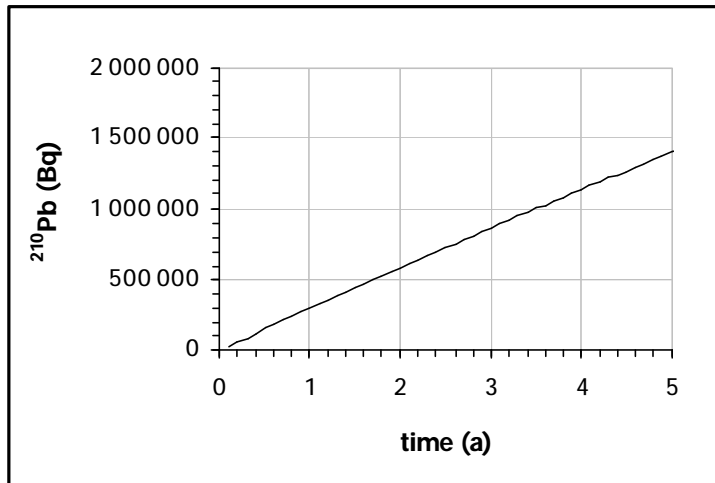


Figure 2. The total activity of ^{210}Pb accumulated on a GAC bed as a function of time. In this case, ^{222}Rn concentration in influent is 3,000 Bq/L, daily water use 600 litres and removal efficiency for radon 99%.

The activity of ^{210}Pb in spent GAC can be calculated if the radon concentration in influent and effluent, and the daily water use are known. The assumptions for calculating the ^{210}Pb accumulated on the GAC are:

1. Radon daughters that originate from the decay of ^{222}Rn are all completely retained on GAC.
2. All the ^{210}Pb originates from the decay of ^{222}Rn retained on GAC. The amounts of ^{210}Pb directly adsorbed or generated by ^{226}Ra are negligible.
3. The measured flow rates and radon concentrations in both influent and effluent for a given volume of GAC are constant over the sampling interval.

The equations that can be applied are:

$$(1) \quad A(\text{Rn}) = \frac{Q \cdot (C_i - C_e)}{I(\text{Rn})} \text{ and}$$

$$(2) \quad A(Pb) = A(Rn) \cdot (1 - e^{-I(Pb)t}), \text{ where}$$

$A(Rn)$	is the activity of ^{222}Rn (Bq) adsorbed on GAC in the steady-state,
$A(Pb)$	is the activity of ^{210}Pb (Bq) accumulated in GAC after time t (a) in service,
Q	is the daily flow rate of water (L/d),
C_i	is the radon concentration in influent (Bq/L),
C_e	is the radon concentration in effluent (Bq/L),
$I(Rn)$	= 0.181 d^{-1} the decay constant of ^{222}Rn ,
$I(Pb)$	= 0.0307 a^{-1} , the decay constant of ^{210}Pb and
t	the time the filter has been in service (a).

The specific activity of spent carbon can be calculated per volume or per mass provided that the volume of the GAC bed and the density of carbon are known.

In this study, the accumulation of ^{210}Pb on GAC was experimentally investigated. Because the concentration of radon in influent and effluent varied and the adsorption of ^{210}Pb directly from the influent was significant, the theoretical activity of ^{210}Pb accumulated in the carbon was calculated by converting the total activity of radon adsorbed on the GAC into its atomic quantity and then into the activity of lead. The results indicated that 100% of ^{210}Pb formed by the radioactive decay of radon was retained on GAC (Appendix 2).

GAC filters are capable of removing various amounts of other radionuclides of the uranium series, such as U and ^{226}Ra , ^{210}Pb and ^{210}Po . The removal efficiencies vary considerably and therefore it is difficult to estimate how large amounts of these radionuclides are accumulated in a GAC unit. The results, however, indicate that the activities of these nuclides are significantly less than that of ^{210}Pb .

4.4 Fe- and Mn-removal techniques

There are three main principles on which the commercial Fe- and Mn-removal equipment is based. All of these removal units are installed to treat all household water (point-of-entry) and are regenerated or backwashed at

certain intervals depending on the quality of the effluent and the daily water use. Table III sets out the removal efficiencies for different radionuclides, by the operation principle.

1. *Aeration-filtration technique.* The first step is to treat water with air or oxygen in order to oxidise iron from +II state (ferrous) to +III state (ferric). Ferric ions are readily hydrolysed and precipitate as ferric hydroxide, $\text{Fe}(\text{OH})_3$. Manganese is oxidised from +II state to +III or +IV state to achieve a subsequent precipitation of MnCO_3 , MnS , $\text{Mn}(\text{OH})_2$, MnOOH , MnO_2 or Mn_3O_4 . These metal precipitates may adsorb radionuclides such as uranium, radium, lead and polonium. In the second step, the precipitates are removed from water by filtration. Backwashing is applied to remove the precipitates from the filter and to drain the wastes into the sewer.
2. *Manganese greensand filtration.* In this technique, the oxidation and filtration steps are carried out applying one fixed bed, manganese greensand. Before commissioning, the greensand bed is conditioned with manganese to form MnO_2 sites on the greensand granules. These sites exhibit oxidising power that enables oxidation and subsequent precipitation of soluble species of iron and manganese. Therefore, radionuclides may also be retained. The bed is backwashed and regenerated with dilute KMnO_4 solution to restore the oxidising power at certain intervals.
3. *Ion exchange technology.* Strong acid cation exchangers can be applied to remove soluble species of iron and manganese as well as hardness from household water. The method is based on ion exchange between the soluble cations in water and the sodium ions located in the functional groups of the resin. Radionuclides occurring as cations or cationic compounds may be exchanged and thus removed by cation exchange. Regeneration is carried out with saturated sea-salt solution at certain intervals.

Table III. Removal efficiencies of Fe- and Mn- removal equipment for different radionuclides, by the operation principle.

Principle	Removal efficiency (%)				
	²²² Rn	U	²²⁶ Ra	²¹⁰ Pb	²¹⁰ Po
Aeration-filtration	10–90	0–90	0–90	20–70	30–80
Manganese greensand	0–40	10–60	60–100	10–60	40–90
Cation exchange	0	0–100	50–100	0–90	0–100

Table III shows that the removal efficiencies vary a great deal. Depending on the system, large amounts of radionuclides may be accumulated by the equipment. Backwashing or regeneration at regular intervals, however, enables safe daily use of these units—radionuclides are rinsed out of the fixed bed and drained into the sewer. The higher the concentration of radionuclides and the higher the daily water use, the more often backwashing or regeneration must be carried out. The regeneration interval is in most cases frequent enough to prevent this technique from causing a problem of waste disposal.

4.5 Ion exchange techniques

Ion exchangers are applied to the removal of uranium (²³⁸U, ²³⁴U) and radium (²²⁶Ra). Lead (²¹⁰Pb) and polonium (²¹⁰Po) may sometimes be removed (depends on water quality) by ion exchange technology as well. Generally speaking, uranium (and polonium) is removed with an anion exchanger while radium (and lead) are removed with a cation exchanger. These exchangers can also be combined in one unit to enable simultaneous removal. Different operation principles and exchange materials can be utilised. Organic ion exchangers (resins) can usually be regenerated, which means that the substances retained in the resin can be removed and the exchange properties restored. The properties of many inorganic exchangers (mineral based) cannot be restored by regeneration and therefore they must be discarded after exhaustion (see Chapter 4.6). The operation principles of ion exchange units are:

1. *Point-of-entry (POE) exchangers with automatic regeneration.* These units are the largest (bed volumes approx. 40 litres) and often make use of both anion and cation exchangers in one bed (mixed bed). They are

installed in the household water line in such a way that all household water is treated. An automatic feature starts the regeneration at regular intervals (time- or volume-related). Regeneration is mostly carried out with saturated sea salt solution.

2. *Point-of-entry (POE) exchangers without automatic regeneration.* The bed volumes of these units are usually 20–40 litres and they are mounted in the incoming water line of a household. An additional purpose of this installation is to remove uranium and humus before a GAC unit in order to prevent the GAC filter from fouling. After the estimated exhaustion, the filters are sent to a commercial company for regeneration or to be filled with a new batch of resin.
3. *Point-of-use (POU) exchangers.* These units are smaller; the bed volumes are mostly 5–20 litres. Since uranium, radium, lead and polonium are harmful only when ingested, only the drinking water needs to be treated. These units can be mounted in the kitchen sink cabinet or at the beginning of the cold water line to the kitchen because drinking water is mostly taken from the kitchen tap. When the capacity of the unit is near exhaustion, it is sent to a commercial company that regenerates the resin or replaces the resin with a fresh batch.
4. *Tap-mounted filters.* These units are attached directly to the kitchen tap and the installation needs no professional plumbing. The size of these filters is usually approx. one litre. Because the filters are relatively inexpensive, they are mostly discarded after a certain period of time and replaced with a new one. The resin can, however, be regenerated either by sending it to a commercial company or by performing it in a bucket filled with sea salt solution.
5. *Pour-through filters.* The bed volumes of these filters are very small; the volume of the resin is often 0.1–0.2 litres. The unit consists of two containers. The lower container (reservoir) is filled by pouring water into the upper basin and allowing the water to pass through the resin by exerting gravitational pressure (e.g. Brita™ can). The resin is discarded after a certain period (time- or volume-related) and replaced.

Strong basic anion exchangers (SBA resins) exhibit an extremely high affinity for uranium, that normally occurs as a negatively-charged carbonate complex in groundwater. After the exhaustion of the resin, substances that are more weakly bound in the resin are replaced by uranium and the removal of uranium remains undisturbed.

The largest volumes of water are treated with POE filters (1–2), since they are applied to treat all the household water. Some of these units, however, are equipped with an automatic regeneration feature and the regeneration interval can be adjusted so that the activity of the radionuclides accumulated is not high. Therefore, the amount of radioactive waste is the largest for those POE filters that are not regenerated automatically and are also designed to treat large volumes of water. Depending on the concentration of radionuclides in the influent, the daily water use and the regeneration (by the company) or decommissioning interval, high levels of radioactivity can be found in these units.

POU units (3–5) treat much smaller volumes of water. Only about 5% of the daily water use are due to preparation of food and drinks. The most convenient application is the 5–20 litre unit mounted in the cold water line to the kitchen. The capacity of the exchanger is usually sufficient to remove radionuclides even for some years before exhaustion. Therefore, maintenance of these units is easy. The larger the bed volume of a POU unit, the longer it can be used without regeneration and the more radioactivity is accumulated in the resin. Pour-through filters are usually small and often replaced and therefore the radioactivity accumulated in them is low.

4.6 Membrane techniques

Membrane techniques are mostly applied to simultaneous POE or POU removal of uranium, radium, lead and polonium. In principle, the feed water is forced into a membrane unit exerting relatively high pressure (5–50 bar). Inside the membrane unit there is a semi-permeable membrane. The pore radii of the membranes determine which ions are allowed to push through into the permeate (purified water) and which are rejected (concentrate). Radionuclide removal on either domestic or waterworks scale can best be carried out by reverse osmosis or nanofiltration:

1. *Reverse osmosis (RO)*. The yield of RO filtration is usually low, only 10–30%. This means that for every cubic metre of water there is 700–900 litres of concentrate. Therefore, these units are mostly applied as POU removal systems. Reverse osmosis removes most dissolved ions, thus the permeate is demineralized. The permeate, therefore, is corrosive and cannot usually be fed into the plumbing system without re-hardening.

2. *Nanofiltration.* The pore radii of nanofiltration membranes are larger, thus not all the ions are rejected. The yield is also higher, 30–80%, and the operational pressure lower. The permeate is not as corrosive as in RO filtration. Nanofiltration is a developing technique and can be expected to be utilised by private households for POE removal of radionuclides in the future.

The radioactive wastes produced by membrane techniques are not accumulated into fixed matrices. The concentrate contains radionuclides, which are constantly drained into the sewer as the unit operates. The concentrate may contain radioactivity about five-fold (80% yield) in excess of the feed water. Thus the concentrations of radionuclides in the concentrate are not significantly higher than in feed water. Membrane techniques, therefore, have no potential for producing radioactive wastes.

The pre-treatment components of a membrane unit, however, may accumulate radionuclides. A GAC filter is often installed as a pre-treatment step. As explained in Chapter 4.3, it is capable of accumulating ^{210}Pb as well as causing gamma radiation if radon is present in the feed water.

4.7 Adsorptive materials

A large variety of different materials are included in this group of removal techniques. The basic principle is that the radionuclides are retained on a fixed matrix (adsorbent), which may be a batch of i.e. activated aluminium oxide, barium sulphate crystals, manganese oxide, activated carbon or a salt of phosphoric acid, $\text{Ca}_3(\text{PO}_4)_2$, CaHPO_4 . Sometimes inorganic ion exchangers such as sodium titanate, mica and zeolite are considered as mineral adsorbents, too.

Usually, these materials cannot be regenerated. They can be applied in the same way as ion exchangers (see Chapter 4.5), POE, POU or pour-through. Depending on the volume of the adsorbent bed, different amounts of water may be treated with them. Table IV summarises the reductions of different radionuclides obtained by these materials. (The information is based on the literature and on laboratory experiments within WP 5 and WP 6.)

Table IV. Summary of the removal efficiencies and distribution coefficients (K_D) values for radionuclides attained by various adsorbents.

Adsorbent	Removal efficiency (%)			
	U	²²⁶ Ra	²¹⁰ Pb	²¹⁰ Po
Al ₂ O ₃ , activated ⁽¹⁾	good ⁽³⁾	60–80	–	–
BaSO ₄ ⁽²⁾	–	>99	–	–
MnO ₂	–	90	80–90	–
GAC	fair ⁽³⁾	fair	35–95	50–90
CaHPO ₄	70–80	>90	>90	good
K_D (ml/g)				
Sodium titanate	7 600	2 030 000	–	–
MnO ₂	650	5 400 000	–	–
Zeolite A	82	106 000	–	–
Na-4-mica	35 300	146 000	–	–

– no information

(1) easy to regenerate

(2) cannot be regenerated

(3) poor capacity

It seems that removal of radium can easily be carried out with different adsorbents. The best materials are barium sulphate and manganese oxide, which cannot be regenerated. It is important to recognise that radium generates radon, which is dissolved in the water passing through the bed. The best application for these materials is thereby pour-through units where no high concentration of radon can be formed in the treated water and the gamma radiation levels remain low.

Lead can best be removed by adsorption on MnO₂ or CaHPO₄. As was the case for radium, pour-through units are most suitable.

5 RADIOACTIVE WASTES PRODUCED BY WATER TREATMENT METHODS

5.1 Introduction

In order to produce estimates of the quantity of radioactive wastes the average water use by the consumers needs to be known. The estimates presented in this chapter are based on the water consumption and consumer habit of an average resident of Helsinki using water from Helsingin Vesi (Helsinki Waterworks). The amounts of water used for different purposes are set out in Table V.

Table V. The average water consumption and consumer habit in households per person according to Helsingin Vesi (Helsinki Waterworks).

Purpose of use	Usage (L/d)	Percentage (%)
Personal hygiene	72	39
Toilet	49	26
Dishes	25	13
Laundry	25	13
Food and drinking	9	5
Cleaning	6	3
Other	4	2
Total	190	100

An average household of three persons consumes 570 L/d or about 200 m³/a of water, which is the amount in POE applications. For POU installation the corresponding flow rates are 30 L/d and 10 m³/a. These values will be used here when estimating wastes produced by one household.

Secondly, an estimate of the number of wells where removal techniques are to be applied, is made. This information, however, is limited. Only in Finland and Sweden is the number of wells where removal techniques should be used, known.

Table VI sets out the treatment techniques and the corresponding concentration ranges of the radionuclides that are removed. It also indicates the nuclides that may be accumulated and therefore may cause problems when disposed of.

Table VI. Treatment techniques, the corresponding concentrations of the radionuclides that need to be removed, and the corresponding nuclides found in waste.

Technique	Radionuclide	Range of interest	Waste contains
Aeration	²²² Rn	> 1 000 Bq/L	no significant amounts
GAC filtration	²²² Rn	1 000–5 000 Bq/L	²¹⁰ Pb, (U, ²¹⁰ Po)
Fe-and Mn removal	–	–	no significant amounts
Ion exchange	U	>20 µg/L	U, ²²⁶ Ra, ²¹⁰ Pb, ²¹⁰ Po
	²²⁶ Ra	> 1 Bq/L	
Membrane technology	U	>20 µg/L	no significant amounts
	²²⁶ Ra	> 1 Bq/L	
	²¹⁰ Pb	> 1 Bq/L	
	²¹⁰ Po	> 1 Bq/L	
Adsorptive techniques	U	>20 µg/L	U, ²²⁶ Ra, ²¹⁰ Pb, ²¹⁰ Po
	²²⁶ Ra	> 1 Bq/L	
	²¹⁰ Pb	> 1 Bq/L	
	²¹⁰ Po	> 1 Bq/L	

5.2 Radionuclides accumulated in GAC filters

In Finland and Sweden, the approximate number of private wells is known (Table VII). A summary of the amount of radionuclides present in spent GAC after 3 years' use is given in Table VIII.

Table VII Number of the wells and users where ^{222}Rn concentration ranges from 1,000 to 5,000 Bq/L, the mean value of radon, uranium and ^{226}Ra in the range of interest (1,000–5,000 Bq/L).

Country	No. of wells (1–5 kBq/L)	No. of users (1–5 kBq/L)	Mean ^{222}Rn (Bq/L)	Mean U (mg/L)	Mean ^{226}Ra (Bq/L)
Sweden	10 000	30 000	–	–	–
Finland	5 800	17 000	2 150	0.12	0.25

Table VIII. The amount of radionuclides present in spent GAC. The minimum and maximum values of ^{210}Pb were calculated for radon concentrations of 1,000 and 5,000 Bq/L, respectively. The same mean concentrations of ^{222}Rn , U and ^{226}Ra were used for both Finland and Sweden (Table 6.3).

Users	Activity found in spent GAC batch (after 3 years' use)				
	^{210}Pb , min. (kBq)	^{210}Pb , max. (kBq)	^{210}Pb , mean (Bq/g)	U, mean (mg/g)	^{226}Ra , mean (Bq/g)
One person	93	470	8	0.3	0.5*
One household	300	1 500	24	0.3	1.6*
Country	Radionuclides accumulated in spent GAC material annually (if 5% of the radon wells employ GAC filtration)				
	^{210}Pb (MBq)	U (kg)		^{226}Ra (MBq)	
Sweden	100	1.1		11*	
Finland	60	0.64		20*	

Values marked with an asterisk (*) may contain very large errors

If 5% of households which have 1,000–5,000 Bq/L of radon in their household water were to apply GAC filtration to remove radon, about 60 MBq of ^{210}Pb would occur in the spent carbon that is annually disposed of at municipal dumps in Finland. In Sweden, the average ^{222}Rn concentration within the range of interest is not known, but by using the same value as for Finland, the corresponding activity would be 100 MBq. In Sweden, however, aeration is the main technology applied to mitigate high radon levels in drinking water. Therefore, it is not expected that GAC filtration would be applied on as large a scale as in Finland. Estimates of the quantity of the wastes cannot

be made for the other Member Countries since the information about the number of wells having high radon concentration is limited.

Although the GAC filters to some extent retain uranium, their capacity for uranium is usually poor. The breakthrough occurs when 1,000 BV (Bed Volumes) have been filtered. The reduction efficiency of a GAC filter for uranium decreases gradually, so it can be estimated that 50% retention occurs during the first 1,000 BV's. The estimate given in Table 6.4 is probably too high because anion exchangers are usually applied to reduce high levels of uranium before GAC filtration.

Radium is retained only fairly well by GAC filters. However, radium was removed from the waters that contained elevated concentrations of iron. The adsorption was probably due to the ferric hydroxide precipitate formed in the GAC units. Because such precipitates can be removed by backwashing, ^{226}Ra should not pose as large a problem as ^{210}Pb and uranium. The estimates presented in Table VIII are based on the assumption that the average ^{226}Ra concentration in influent is 0.25 Bq/L and the reduction of 25%. This reduction efficiency is probably too high in most cases.

^{210}Po as well as ^{210}Pb can be adsorbed on GAC directly from influent, if present. The retention is usually 30%–100%. However, their concentration is mostly much lower than that of the ^{210}Pb produced by the decay of radon inside a GAC unit.

5.3 Wastes produced by ion exchange techniques

Since an ion exchanger can be installed as POU or POE, two types of calculations will be applied. In POE installations, 570 litres of water is filtered daily in a household. In POU units, the amount is 27 L/d (Table V). For POE installation, the usual general regeneration interval is one week when automatic regeneration is carried out. The amount of brine (regeneration) and water (rinsing) used during one regeneration procedure is 350 litres on average. Provided that the removal efficiency is 100% the regeneration solution (brine and water) contains radionuclides about eleven-fold (concentration factor, Table IX) in excess of the influent. For POE units where no automatic regeneration is carried out, the regeneration interval is normally one year. Pour-through filters usually treat about 100 litres of water, after which they are discarded.

When the regeneration is carried out automatically the regeneration solution containing radionuclides is released into a sewer or a drainage pit. Manufacturers mostly drain the regeneration solutions into the municipal sewage system.

5.3.1 Uranium

The maximum contaminant level of uranium in drinking water in different countries is 20–160 µg/L. In this report it is assumed that uranium removal is generally applied when the concentration in drinking water exceeds 20 µg/L. The average concentration to be removed used in the calculations is 200 µg/L.

Table IX summarises the amount of uranium accumulated and the wastes produced by different anion exchange techniques. POE anion exchangers that are not regenerated automatically accumulate the largest amount of uranium, 40 grams per year on average. In POE units where the regeneration is carried out weekly, the amount is usually about 1 gram. POU and the tap-mounted filters contain about 2 grams of uranium after one year's use. The pour-through filters accumulate very small amounts of uranium. If a POE unit without automatic regeneration is applied to mitigate extreme levels of uranium (> 10 000 µg/L), the amount of uranium accumulated in the unit may be more than 2 kg per year.

Table IX. The amounts of uranium accumulated by one household (3 persons) annually, the volume of the regeneration solution and its uranium content and the concentration factors applying to different types of anion exchangers for removing uranium. The values are based on an average uranium concentration of 0.2 mg/L.

Type of exchanger	U accumulated in exchanger units	Volume of brine and water used for regeneration annually	Concentration of U in regeneration brine and rinsing water	Concentration factor of regeneration solution
POE, automatic reg.	40 g/year	20 m ³	2 mg/L	11
POE, no reg.	40 g/year	350 L	120 mg/L	600
POU, no reg.	2 g/year	350 L	6 mg/L	30
Pour-through	20 mg/100 L	none	none	none

5.3.2 ²²⁶Ra

Radium is normally removed from drinking water when its concentration exceeds 1 Bq/L. The average concentration to be removed is assumed to be 2 Bq/L. As was the case for uranium, different types of ion exchanger units can be used. The estimates of the amount of radium in the exchangers and the regeneration solutions are set out in Table X.

Table X. Activity of ^{226}Ra accumulated by one household (3 persons) , the volume of regeneration solution and its radium content produced annually, and the concentration factors applying to different types of cation exchangers for the removal of radium. The values are based on an average radium concentration of 2 Bq/L.

Type of exchanger	^{226}Ra accumulated in the units	Volume of brine and water used for regeneration annually	Concentration of U in regeneration solutions	Concentration factor of regeneration solution
POE, automatic reg.	8 kBq/week	18 m ³	20 Bq/L	11
POE, no reg.	420 kBq/year	350 L	1 200 Bq/L	600
POU, no reg.	20 kBq/year	350 L	50 Bq/L	30
Pour-through	0.2 kBq/100 L	none	none	none

5.3.3 ^{210}Pb and ^{210}Po

Ion exchange techniques cannot reliably be applied to the removal of ^{210}Pb and ^{210}Po . However, when ion exchange treatment is used for removing uranium and radium, various concentrations of these radionuclides may occur. The removal efficiencies for ^{210}Pb and ^{210}Po have ranged from 30% to 95%. Therefore, estimates of their contribution to the waste should be discussed.

It can be assumed that the average concentration of both lead and polonium in influent is 1 Bq/L and the retention is 50%. Similar calculations to those made for uranium and radium are set out in Tables XI and XII.

Table XI. Activity of ^{210}Pb accumulated by one household, the volume of the regeneration solution, its lead content and the concentration factors applying to different types of ion exchangers for the removal of lead. The values are based on an average lead concentration of 1 Bq/L and a removal efficiency of 50%.

Type of exchanger	^{210}Pb accumulated in the units	Volume of brine and water used for regeneration annually	Concentration of regeneration brine and rinsing water	Concentration factor of regeneration solution
POE, automatic reg.	2 kBq	18 m ³	6 Bq/L	6*
POE, no reg.	100 kBq	350 L	300 Bq/L	300*
POU, no reg.	5 kBq	350 L	14 Bq/L	14*
Pour-through	0.05 kBq	none	none	none

* concentration factor based on 50% removal

Table XII. Activity of ^{210}Po accumulated by one household, the volume of regeneration solution, its polonium content and the concentration factors applying to different types of ion exchangers for the removal of polonium. The values are based on an average polonium concentration of 1 Bq/L and a removal efficiency of 50%.

Type of exchanger	^{210}Po accumulated in the units	Volume of brine and water used for regeneration annually	Concentration of regeneration brine and rinsing water	Concentration factor of regeneration solution
POE, automatic reg.	2 kBq	18 m ³	6 Bq/L	6*
POE, no reg.	100 kBq	350 L	300 Bq/L	300*
POU, no reg.	5 kBq	350 L	14 Bq/L	14*
Pour-through	0.05 kBq	none	none	none

* concentration factor based on 50% removal

In Finland there are about 13,000 households consuming water from private wells where the uranium content exceeds 20 µg/L. If 5% of these households were to use POE units, the annual amount of uranium released into the sewers and drainage pits would be 28 kg. For POU units, the corresponding amount would be 1.3 kg. Therefore, the POU units should be favoured. Nevertheless, in some cases the use of a POE unit is essential. The radon removal efficiency of a GAC filter can decrease resulting from a high level of uranium or humus in the influent. In that case, a POE ion exchanger needs to be placed upstream of a GAC filter. A POE ion exchanger is also used when simultaneously removing high levels of iron, manganese and humus from household water.

5.4 Radionuclides accumulated in adsorptive matrices

Adsorptive materials (except GAC) are relatively rare among the water treatment techniques applied in private households. Because the adsorptive matrices cannot usually be regenerated, the bed volumes have to be as small as possible to optimise expenses. Therefore, POU units are the most common.

Various materials are commercially available. Their removal efficiencies for different radionuclides, however, vary and therefore simultaneous removal of all the long-lived radionuclides of the uranium series cannot mostly be attained. Table IV summarised the removal efficiencies and the K_D values of different matrices. Based on these removal efficiencies the amounts of radionuclides can be calculated (Table XIII). The average concentration in influent is assumed to be 2 Bq/L for ^{226}Ra , ^{210}Po and ^{210}Pb and 0.2 mg/L for uranium. All of these nuclides are fixed into a solid matrix.

Table XIII. The amount of different radionuclides accumulated in a POU unit employed by one household over a one-year period. The concentration in influent was 2 Bq/L for ^{226}Ra , ^{210}Pb and ^{210}Po , and 0.2 mg/L for uranium.

Adsorbent	U (g)	Radionuclides accumulated		
		^{226}Ra (kBq)	^{210}Pb (kBq)	^{210}Po (kBq)
Al ₂ O ₃ , activated	1.5	14	–	–
BaSO ₄	–	20	–	–
MnO ₂	–	18	17	–
GAC	1	5	10	14
CaHPO ₄	1.5	19	19	15
Sodium titanate	–	5	15	14
Zeolite A	–	14	15	12

6 ASSESSMENT OF DOSES DUE TO RADIOACTIVE WASTES

6.1 General

When water treatment systems are set up, the possibility of inhabitants being exposed to gamma radiation should be considered. Although most of the radionuclides included in Table I emit gamma radiation, the most significant ones are ^{214}Pb (352 keV gamma abundance 37% and 295 keV gamma abundance 19%) and ^{214}Bi (609 keV gamma abundance 46%, 1 760 keV gamma abundance 16% and 1120 keV gamma abundance 15%). Therefore, if they are accumulated in a treatment unit, high dose rates can be found in the vicinity of the unit.

Gamma rays originating from a GAC filter are subject to the amount of radon retained on GAC in steady-state operation. The short-lived daughter nuclides, ^{214}Pb and ^{214}Bi attain secular equilibrium with radon and hence contribute the most to the dose rate. The dose rate can be estimated when the water use, the radon concentration in raw water and the removal efficiency are known. A simple method for estimating the dose rate is to use *Carbdose* (A program freely downloadable at USEPA, United States Environmental Protection Agency homepage.) Figure 3 sets out the dose rates of different installations calculated with *Carbdose*.

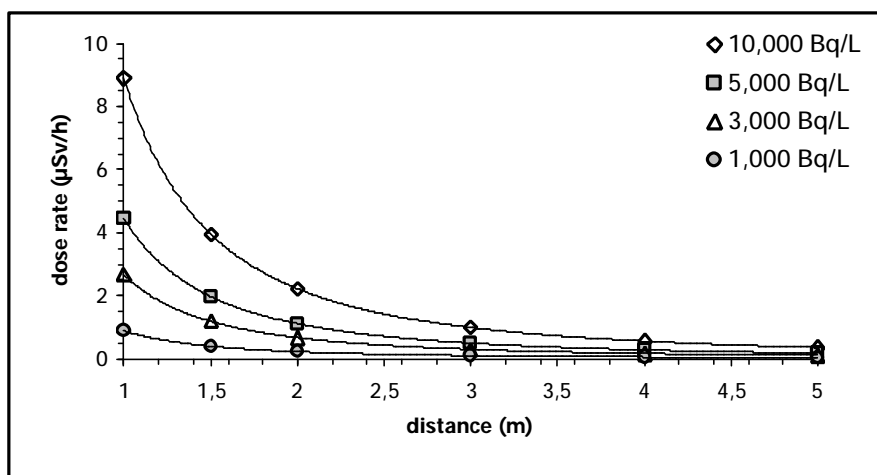


Figure 3. Dose rates at various distances from a GAC filter (daily water use 570 L/d and removal efficiency 100%), caused by removal of different radon concentrations in influent.

As can be seen in Figure 3, dose rates in the vicinity of the filter become a problem when radon concentration is high. In this study, the highest dose rate, 110 $\mu\text{Sv/h}$, was measured on the filter surface where ^{222}Rn concentration was 7,350 Bq/L and daily water use 320 L/d. In chapter 8.3 it is recommended that the annual dose to inhabitants from external gamma radiation from a GAC filter should not exceed 0.1 mSv. In this chapter the consequences of the action level 0.1 mSv/a are discussed.

The highest yield for radon removal is achieved by aeration techniques. This is not discussed in detail here, as the external radiation is hardly increased for people close to an aeration device, although inhalation can lead to a significant uptake of ^{222}Rn . The worst case in radiation protection for external sources is expected for GAC filtration due to its high efficiency for ^{222}Rn absorption, leading also to a high activity of the resulting daughter products. The general radiation protection problems are discussed in detail for GAC filtration.

6.2 Gamma radiation surrounding a GAC filter during operation

Assuming an efficiency of 100% for radon removal in a GAC filter (the worst case for the dose calculation) the steady state activity of radon collected in the filter is rewritten as

$$(3) \quad A(Rn) = \frac{Q \cdot C_i}{I(Rn)}$$

Taking a 3-person household with an average consumption of $Q = 200 \text{ m}^3/\text{a}$ into account plus factorising the radon concentration $C_i(Rn) = c_i \cdot 1 \text{ kBq/L}$ in the influent water results in the equation

$$A(Rn) = \frac{c_i \cdot 2 \cdot 10^5 \text{ kBq}}{66.2} \cong c_i \cdot 3 \text{ MBq}$$

Ignoring the attenuation of gamma radiation inside the GAC and in the air (worst case), the collected activity produces a dose rate at the distance r with the effective gamma dose rate coefficient $K_g = 0.2 \text{ } \mu\text{Sv/MBq} \cdot \text{m}^2/\text{h}$ for ^{222}Rn and its daughters (mainly ^{214}Bi):

$$(4) \quad \dot{H}_0(r) = \frac{K_g \cdot A(Rn)}{r^2}$$

At a distance $r = 1 \text{ m}$ this results in the equation

$$\dot{H}_0(1\text{m}) = 0.2 \cdot c_i \cdot 3 \mu\text{Sv/h} = c_i \cdot 0.6 \mu\text{Sv/h}$$

Considering $\dot{H}_1 = 1 \mu\text{Sv/h}$ as the maximum allowable dose rate at a distance of 1 m, no shielding is required, as long as the radon concentration in the influent is lower than 1.66 kBq/L ($c_i < 1.66$). Higher concentrations make it necessary to discuss shielding of the GAC filter.

$$(5) \quad \dot{H} = \frac{\dot{H}_0}{S}$$

In order to guarantee the limit $\dot{H}_1 = 1 \mu\text{Sv/h}$ also for higher radon concentrations ($c_i > 1.66$), the required reciprocal attenuation factor

$$\frac{1}{S} = e^{-\mu d}$$

must be smaller than the value $1.66/c_i$:

$$\frac{1}{S} \leq \frac{1.66}{c_i}$$

The shielding thickness d depends on the material i.e. its absorption coefficient μ and the value of c_i . For a worst case calculation one can assume a maximum radon concentration:

$$C_i(Rn) \leq 5 \text{ kBq/L} \quad \text{or} \quad c_i \leq 5$$

else aeration would be preferred over GAC. The corresponding reciprocal attenuation factor results in the equation

$$\frac{1}{S} = \frac{1.66}{5} = \frac{1}{3}$$

which can be achieved by a water layer with a thickness $d = 70$ mm. As a GAC filter in operation is already filled with water, this water causes significant self-absorption. The radiation emission is not at all isotropic due to a gradient of the collected radon activity along the flow direction inside the filter and due to the cylindrical shape of a GAC: the length and diameter of the filter influence the strength of self-absorption. The lowest dose rate is emitted in the direction of the water flow in the GAC material.

6.3 Annual exposure dose to residents using a GAC filter

Based on a given maximum dose rate of $\dot{H}_1 = 1 \mu\text{Sv/h}$ at a distance of 1 m, the annual dose to the residents of the house can be estimated. Table XIV lists typical rooms, distances from the filter, an approximate attenuation due to absorption in the building material, the remaining dose rate, estimated presence times and the resulting annual dose.

Table XIV. Annual dose to people living in a house at different locations

location	distance r (m)	reciprocal attenuation S^{-1}	$\dot{H}(r)$ ($\mu\text{Sv/h}$)	presence (h/a)	annual dose ($\mu\text{Sv/a}$)
service at filter	1	1.0	1.000	5	5
technical room	2	1.0	0.250	20	5
bedroom	5	0.2	0.008	7000	56
washing room	2	0.2	0.050	500	25
playground	5	0.3	0.012	500	6
					97

This example demonstrates that the annual dose is below the action level of 0.1 mSv/a for a house built by 13 cm-thick brick walls ($1/S = 0.3$), if the initial condition $\dot{H}_1(1\text{m}) \leq 1 \mu\text{Sv/h}$ is fulfilled by proper installation in the technical room. Self absorption inside the filter can be enforced by constructing new filter cartridges with an outer shell of water (for instance, two conical cylinders, only the inner one is filled with GAC material, the outer one—approximately 70 mm thick—solely with the influent water).

6.4 Transport of a used GAC filter

Once a GAC filter is taken out of operation, it should age about 4 weeks before being shipped to allow a decay of ^{222}Rn ($T_{1/2} = 3.8$ d) and its short-lived daughter products. After that period the main activity will be related solely to ^{210}Pb ($T_{1/2} = 22.3$ a). If t_s is the number of years that the filter has been in service (service time: $T_s = t_s \cdot 1\text{a}$), the remaining activity is

$$A(\text{Pb}) = t_s \cdot c_1 \cdot 92 \text{ kBq}$$

Again neglecting the attenuation of gamma radiation, the remaining lead activity produces a dose rate at distance r with $K_\gamma = 0.3 \mu\text{Sv/MBq} \cdot \text{m}^2/\text{h}$

$$\dot{H}_0 = K_\gamma \cdot A(\text{Pb}) / r^2$$

At a distance $r = 0.3$ m (practically on the surface of a shipping package) this results in the equation

$$\begin{aligned}\dot{H}_0(0.3\text{m}) &= 0.3 \cdot t_s \cdot c_1 \cdot 0.092 \cdot 0.3^{-2} \mu\text{Sv/h} \\ &= t_s \cdot c_1 \cdot 0.3 \mu\text{Sv/h}\end{aligned}$$

The consideration of $t_s = 3$ years' service time and a maximum radon concentration $C_i(\text{Rn}) \leq 5$ kBq/L ($c_1 \leq 5$) leads to a maximum dose rate of the unshielded filter with $4.5 \mu\text{Sv/h}$. Thus shipping is allowed without any additional shielding. Declaration and a label according to the category I-white (limit $5 \mu\text{Sv/h}$ in contact) is sufficient for most GAC filters to be shipped!

The same considerations lead to the unshielded dose rate at a distance of 1m

$$\dot{H}_0(1\text{m}) = t_s \cdot c_1 \cdot 0.028 \mu\text{Sv/h}$$

Again the case with $t_s = 3$ and $c_1 = 5$ results in the equation

$$\dot{H}_0(1\text{m}) = 0.417 \mu\text{Sv/h}$$

thus—after ageing—no transportation index will be needed!

Let's assume that a filter is replaced and it is shipped immediately without ageing. In this case it is strongly recommended to plug the filter with blind flanges and to keep it water-filled in order to maintain self-absorption. Taking care of this prevention, the remaining ^{222}Rn and its short-lived daughter ^{214}Bi can produce a dose rate at a distance of 1 m of at most $1 \mu\text{Sv/h}$. Thus the transportation index will be just 0.1 and the question whether the declaration is category II-yellow will depend on the dose rate on the surface of the package to be shipped.

7 EXISTING REGULATIONS ON WASTES CONTAINING NATURAL RADIOACTIVITY

7.1 European Union BSS Directive

The BSS Directive is applied to all practices⁶ which involve a risk from ionising radiation emanating from an artificial source or from a natural radiation source in cases where natural radionuclides are or have been processed in view of their radioactive, fissile or fertile properties.

In connection with natural radioactivity the BSS is also applied to work activities where the presence of natural radiation sources lead to a significant increase in the exposure of the workers or of the members of the public. The term 'work activities' is used instead of 'practices' to highlight the distinction that a natural radiation source is present but natural radionuclides are not or have not been processed in the view of their radioactive, fissile or fertile properties. The directive applies to these work activities in accordance with Title VII.

In Title III (Article 3) of the BSS special exemption levels are defined. The purpose of the exemption levels presented in Appendix 1 of the BSS, is to define levels of activities below which no reporting is required for practices. They have been derived by using scenarios reflecting small-scale use of radionuclides arising from practices.

The radioactive wastes arising from the treatment of water involve natural radionuclides which are not or have not been processed in view of their radioactive, fissile or fertile properties. Therefore, their production, processing, handling, use, holding, etc. cannot be considered practice and the exemption levels are not applicable.

⁶ Practice is defined as a human activity that can increase the exposure of individuals to radiation from an artificial source, or from a natural radiation source where natural radionuclides are processed for their radioactive, fissile or fertile properties, except in the case of emergency exposure.

However, wastes from water treatment methods involving natural radionuclides may lead to a significant increase in the exposure of members of the public (or workers who handle the waste). According to the principles set out in the BSS, the Member States may identify the production, processing, handling, use, etc. of such waste as a work activity in the sense of Title VII. In such case the Member States may apply the system of protection of practices to all or part, as they consider appropriate.

The Member States might decide that exemption levels should be applied to such work activities. However, this might not be a very reasonable action because the radiological criteria (10 μ Sv) and scenarios used in deriving the exemption levels are not necessary reasonable to be used for doses arising from natural radionuclides.

The exemption levels apply to natural radionuclides only in cases where they are, or have been, within a practice (processed in view of their radioactive, fissile or fertile properties). If a Member State decides to apply the exemption levels to work activities, it can do so, but a national decision to this effect is clearly needed. The BSS does not require such a decision to be made.

7.2 National regulations

To gather information on existing national regulations and guides on the treatment and disposal of radioactive wastes produced by various water treatment methods, a questionnaire was sent to all the Member Countries (Appendix 1). However, the questionnaire was not answered by all of them.

The questionnaire asked about national surveys on natural radioactivity in drinking water, about available estimates on population doses and about special regulations or recommendations on natural radioactivity in drinking water. The questionnaire also included questions about wastes produced by water treatment methods. The questionnaire also gave the possibility of free comment.

The Member States which have made nationwide or partly nationwide survey on natural radioactivity in drinking water are Austria, Denmark,

Finland, France, Germany, Ireland, Spain, Sweden and the UK. In Italy local surveys have been made, in Luxembourg a survey is in preparation.

The Member States which have made estimates on population doses due to natural radioactivity in drinking water are Austria, Finland, France, Germany, Ireland, Spain, Sweden and the UK. Different ingestion dose coefficients may have been applied in these studies and therefore these doses are not necessarily comparable.

A study made in **Austria** showed that in the province of Lower Austria, in areas having the highest amounts of natural radioactivity in drinking water the population receives from radon through inhalation a mean additional dose of between 0.26 and 1.27 mSv every year. The dose due to ingestion is less by an order of magnitude. The dose from ingested radium is negligible in most cases.⁷

According to the most recent estimates, there are 700,000 people in **Finland** who derive their household water from private wells. 200,000 of them use bedrock water from drilled wells. The number of people that use water in which the radon concentration exceeds 300 Bq/L, 1,000 Bq/L and 3,000 Bq/L are 70,000, 20,000 and 6,000, respectively. The annual doses from long-lived alpha emitters (^{238}U , ^{234}U and ^{226}Ra) exceeding 0.1 mSv, 0.5 mSv and 1 mSv are received by 10,000, 3,000 and 1,000 people, respectively. The long-lived radon daughters, ^{210}Pb and ^{210}Po cause higher doses. Doses of 0.1 mSv, 0.5 mSv and 1 mSv are received by 10,000, 8,000 and 4,000 people, respectively. In total, 300,000, 100,000 and 45,000 people using water from private wells receive annual doses of 0.1, 0.5 and 1 mSv caused by natural radionuclides in drinking water. For all the Finns (including those who use water supplied by water works) these figures are 1,000,000, 100,000 and 50,000, respectively.⁸

⁷ Schönhofer F. Measurement of ^{226}Ra in Water and ^{222}Rn in Water and Air by Liquid Scintillation Counting. *Radiation Protection Dosimetry* 1992; 45, ¼: 123—125.

⁸ Mäkeläinen I, Salonen L, Huikuri P, Arvela H. Dose from Drinking Water in Finland. In: Søgaard-Hansen J, Damkjær A (eds). *Proceedings of the Nordic Society for Radiation Protection 12th ordinary meeting 23–27 August 1999, Skagen, Denmark*. Roskilde: Risø National Laboratory, 1999: 117–120.

In **France** there are more than 1,400 thermal springs, out of which 750 had been studied in terms of radioactivity by the year 1990. Radioactivity in 26 of them, including the best-known mineral waters in France, had been the subject of a thorough survey regarding the amount of activity of ^{226}Ra , ^{238}U , ^{234}U , ^{232}Th , ^{230}Th , ^{228}Th and ^{40}K , and for some of them ^{222}Rn . The results of the study showed that radioactivity in French mineral waters was very low, sometimes non-measurable by usual technical means, and that the health hazards linked to the consumption of these waters were negligible.^{9, 10}

A more recent study concerning bottled mineral water was carried out in 1995. Out of 67 bottled mineral waters that were studied the gross alpha activity exceeded 0.1 Bq/L in 31 cases. A gross beta activity of 1Bq/L was exceeded in 26 cases. The concentration of ^{226}Ra exceeded 0.1 and 1 Bq/L in 26 and 5 cases, respectively. The maximum concentration measured was 2 Bq/L. The concentration of uranium exceeded 10, 30 and 50 $\mu\text{g/L}$ in 7, 5 and 2 cases, respectively. The highest concentration measured was 67 $\mu\text{g/L}$. One milligram of natural potassium contains 27.6 mBq of ^{40}K , which is a beta emitter. Potassium concentration exceeded 12, 50 and 100 mg/L in 15, 9 and 4 cases, respectively. The highest concentration measured was 190 mg/L (5.2 Bq/L of ^{40}K).¹¹

A nationwide survey of the radon concentration in drinking water and the resulting annual dose of the population has been carried out in **Germany** (Rühle, 1995). The median of more than 1,100 drinking water samples was 5.6 Bq/L and the maximum level 1,500 Bq/L. The annual effective dose due to the ingestion of radon-bearing drinking water was estimated to be 3 μSv for adults and 26 μSv for children on average. The annual effective dose due to the inhalation of radon released from drinking water in dwellings was estimated to be 8 μSv on average. Values in regions with higher uranium

⁹ Remy ML, Lemaitre N. Eaux minérales et radioactivité. *Hydrogéologie* 1990; 4: 267–278.

¹⁰ Remy ML, Pellerin P. Radioactivité naturelle de 250 sources hydrominérales Françaises. *Bulletin de l' I.N.S.E.R.M.T.* 1968; 23: 23–62.

¹¹ Minstre du travail et des affaires sociales, Secretariat d'etat a la santa et la securite sociale. Bilan sur la qualite des eaux conditionees –Annee 1995–. Paris: Direction Générale de la Santé, 1997.

contents in the bedrock, such as southern Saxony and the Fichtelgebirge, were substantially higher.¹²

A survey of natural radioactivity in drinking water was carried out in a granitic area in western **Spain** covering the so-called greywacke-schist complex. This region is known to be rich in uranium ores, so that natural radionuclides are to be expected in the groundwater. During 1988, 345 water samples were collected from the water supplies of 115 different villages. These samples were analysed for gross alpha, U, Th and Ra. The average concentrations of radionuclides were found to be 5–30 times higher in groundwater from bedrock than in groundwater from soil. The results indicate that radium makes the highest contribution to the effective dose.

Concentration levels of Ra-226 and Rn-222 have been analysed in most of the bottled waters commercially available in Spain. Concentrations up to about 0.6 Bq/L with a geometric mean of 0.012 Bq/L were observed for Ra-226. For Rn-222 a geometric mean of 1.2 Bq/L with values ranging from 0.014 to 52 Bq/L were measured. Doses resulting from the consumption of these waters were calculated. The effective doses due to intake of Ra-226 present in these waters are expected to range from about 102 to 2 μ Sv per year. Equivalent doses to the stomach due to Rn-222 intake through water consumption are estimated to reach values around 30 μ Sv per year.^{13, 14}

In **Sweden** about 800,000 permanent residents, nearly 10 percent of the population, use water from drilled wells. The number of wells in permanent use is about 200,000. Another 200,000 drilled wells are used in summer cottages. A survey of ²²²Rn and ²²⁶Ra from waterworks and a random sample of 500 private drilled wells in Sweden was conducted in the 1980s. The levels of ²²²Rn were low for water from large waterworks, the mean ²²²Rn

¹² Rühle H. Radongehalt des Trinkwassers in der Bundesrepublik Deutschland und Abschätzung der Strahlenexposition. Schriftenreihe Reaktorsicherheit und Strahlenschutz BMU-1995-415. Der Bundesminister für Umwelt, Naturschutz und Reaktorsicherheit, 1995.

¹³ Fernández F, Lozano JC, Gómez JMG. Natural Radionuclides in Ground Water in Western Spain. *Radiation Protection Dosimetry* 1992; 45, ¼: 227–229.

¹⁴ Dueñas C, Fernández MC, Carretero J, Liger E. Radon and Radium Concentrations in Bottled Waters: An Estimate of Ingestion Dose. Vienna: International Atomic Energy Agency. Report IAEA-CN-67/166.

concentration was about 20 Bq/L, while the mean concentration for drilled wells was 212 Bq/L. The highest concentration found so far in Sweden is 57,000 Bq/L. An estimated number of 20,000–30,000 permanently used wells, about 10 percent, have radon levels exceeding 500 Bq/L, and about 10,000 exceeding 1,000 Bq/L.

The levels of ^{226}Ra in the waterworks investigated were low, below 0.3 Bq/L with a mean of 0.01 Bq/L, while the concentrations for the drilled wells varied, between 0.002 and 2.5 Bq/L, with a mean of 0.05 Bq/L. The levels of uranium were measured in about 50 of the drilled wells. The mean concentration for ^{234}U was 0.5 Bq/L with a maximum of 6.9 Bq/L and the mean for ^{238}U was 0.2 Bq/L with a maximum of 2.8 Bq/L.

The levels of other natural radionuclides in drinking water in Sweden have not been investigated in a systematic manner.¹⁵

In the **UK** it has been recognised for many years that radon in drinking water is a possible exposure route. However, it is concluded that radon in public water supplies is not a significant public health issue but that a few private supplies might warrant further investigation.¹⁶

7.3 Limits and recommendations for reducing radioactivity in drinking water

Member Countries with regulations with reference to natural radioactivity in drinking water are Austria, Finland, France, Germany, Spain and Sweden (Table XV).

¹⁵ SSI—Swedish Radiation Protection Institute. Radon i vatten, SSI information series i98:03. Stockholm: Swedish Radiation Protection Institute, 1998.

¹⁶ Kendall G. Radon in Drinking Water. Radiological Protection Bulletin 1996; 181: 31–31.

Table XV: Limits on and recommendations for reducing radioactivity in drinking water (y = yes, n = no, ni = no information)

Member Country	Regulations on natural radioactivity in drinking water	Rn-222	Ra-226	Uranium	Other nuclides
Austria	y	n	y	y	y
Belgium	ni				
Denmark	n				
Finland	y	y	y	y	y
France	y	n	y	y	y (³ H, ²¹⁰ Pb)
Germany	y	n	n	n	n
Greece	ni				
Ireland	n				
Italy	n				
Luxembourg	n				
Netherlands	ni				
Portugal	ni				
Spain	y				
Sweden	y	y	n	n	n
U.K.	n				

Austria: In Austria the limits are laid down in the Austrian Radiation Protection Ordinance (BG.BL. 47/1972). The limit for radium is 0.123 Bq/L and for uranium 7.4 Bq/L. The limits for other nuclides are nuclide-specific.

Finland: In Finland the limit for natural radioactivity in drinking water is laid down in ST-guide 12.3: Radioactivity of drinking water. The ST-guide is applied only to water supplied by waterworks or used by professional producers of beverages and foods relying on their own well or water supply. According to the guide, the safety requirement for household water is that the effective dose due to radioactivity in the water shall not exceed 0.5 mSv per year. The radon released into indoor air is not taken into account in assessing the dose.

Presently under preparation is the decision of the Ministry of Social Affairs and Health on the radioactivity in drinking water. In the proposal for waterworks the limit for radon is 300 Bq/L and the dose limit for doses from

other radionuclides is 0.1 mSv per year. For privately-owned wells the limit for radon is 1,000 Bq/L. No special limit for other nuclides is proposed.

France: The limits are based on “LAI public”, more recently based on recommendations published by the WHO (1994). The limit for radium is 1 Bq/L and for uranium 160 µg/L. The limit for tritium is 100 Bq/L and for lead 0.1 Bq/L. The limits are based on an effective dose of 0.1 mSv per year. In the future Directive 98/83 will be applied.

Germany: Radiation protection criteria for the utilisation of water as drinking water from sources likely to have been influenced by uranium mining (Recommendation of the Commission on Radiological Protection, 10. December 1992, Bundesanzeiger vom 22 Mai 1993).

Spain: The Spanish legislation on radioactivity in drinking water (water supplies) is included in the “Real Decreto 1138/1990”. In this decree, guideline values are proposed for gross alpha activity (0.1 Bq/L) and gross beta activity (1 Bq/L). In these values the natural and artificial radioactivity contributions are included. The same values are also recommended for bottled drinking water.

In the Nuclear Safety Council Safety Guide (the Safety Guide 7.7: Rev 1. Drinking water radiological control) natural radionuclides are specifically considered. The concentration values included in the Safety Guide 7.7: Rev 1. are not limit values but reference levels (investigation, notification and action levels). These levels exist for the main natural radionuclides with the exception of radon. The reference levels are set out in Table XVI.

Table XVI. The reference levels included in the Safety Guide 7.7: Rev.1.

Nuclide	Reference level (Bq/L)		
	Investigation	Notification	Action
Ra-224	0.40	4.0	8.0
Ra-226	0.10	1.0	2.0
Ra-228	0.12	1.2	2.4
Th-230	0.10	1.3	2.6
Th-232	0.04	0.4	0.8
Pb-210	0.03	0.3	0.6
Po-210	0.10	1.3	2.6
U-234	0.50	5.5	11.0
U-238	0.70	6.8	13.6

Sweden: In Sweden the limit for radon is given in SLV FS 1997:32. If the amount of radon in water exceeds 1,000 Bq/L, the water is regarded unfit for human consumption. If the amount of radon in water exceeds 100 Bq/L, the water is serviceable with a warning concerning potential health risks (public water).

UK: In the UK, the limits prescribed by WHO are applied as recommended in "Guidelines for Drinking Water Quality. Vol. 1, Recommendations. 2nd Edition. World Health Organisation, Geneva, 1993."

7.4 Wastes produced by water treatment methods

In most of the Member Countries wastes produced by water treatment methods can be delivered to municipal dumps, and liquid wastes can be discharged into the communal pipeline, into private waste water systems or into the environment. However, in some cases this depends on the amount of radioactivity involved. In one country the exemption levels set in the BSS are applied.

Table XVII. Wastes produced by water treatment methods. Summary of the answers (y = yes, n = no, ni = no information)

Member Country	Regulations against a private person possessing material containing radioactivity	Can solid wastes due to water treatment methods be delivered to communal dumps?	Can liquid wastes due to water treatment methods be discharged into the environment?	Can liquid wastes due to water treatment methods be discharged into the communal pipeline?	Can liquid wastes due to water treatment methods be discharged into the private waste water system?
Austria	y	y	y	y	y
Belgium	ni				
Denmark	n	Depends on the circumstances	Depends on the circumstances	Depends on the circumstances	Depends on the circumstances
Finland	n			y	y
France	n				
Germany	n	y	y	y	y
Greece	ni				
Ireland	n	y	y	y	y
Italy	n				
Luxembourg	n	y	y	y	y
Netherlands	ni				
Portugal	ni				
Spain	n		no regulation	no regulation	no regulation
Sweden	y	y	y	y	y
U.K.	n				

Austria: In Austria solid wastes can be delivered to municipal dumps if the amounts are below the limit values laid down in the Austrian Radiation Protection Ordinance. For radium the limit value for notification to the authorities is 370 Bq and for licensing 3,700 Bq. As regards natural uranium any amount must be notified to the authorities, the limit value for licensing being 37,000 Bq. For liquid wastes to be discharged via the sewer the same limit values as for drinking water are applied.

Finland: Specific guidance is in preparation. Activated carbon may be delivered to a municipal dump after it has been regenerated or aged so that the dose rate has decreased sufficiently.

France: No official/authorised treatment method for the elimination of radionuclides from drinking water exists. In this respect no need has been expressed up to now either by water suppliers, local authorities or consumers. Consequently, no questions concerning the waste have arisen.

Water control and treatment directly at its production site make further treatment by consumers unnecessary except for problems associated with water hardness (a common problem in France). The distributed water meets the requirements/recommendations with respect to radioactivity.

Taking into account the waste emerging from such treatment (as used at an industrial level in very few places in France) and the general level of radioactivity in water fulfilling the hygienic requirements and supplied to the population, it seems to us that the most reasonable solution is a contra-indication of treatments that can, however, provide information on water properties and potential risks. Nonetheless, the problem of radon stripping, particularly at an industrial level, is a cause for concern.

Germany: Regulations concerning possession by private persons or concerning use: Radiation Protection Ordinance: 500 Bq/g of natural radioactivity.

Ireland: There are exemption limits specified in Irish law for the disposal of individual radionuclides, which are based on the EU Basic Safety Standards Directive. If the activities or activity concentrations are below the relevant limits then material may be disposed of as normal waste. It is clear, however, that if a water treatment method for removing natural radioactivity were to be employed on a medium to large scale, then in certain instances the quantities of waste produced would be likely to exceed these limits.

Italy: In Italy there is no experience of water treatment methods as regards radioactivity. This is due to the fact that up to now only some very limited Italian areas have been shown to have high natural radioactivity in drinking

water. Therefore no special regulation on natural radioactivity in drinking water, or on water treatment and connected wastes was ever issued.

Spain: There is no information about the existence of this type of radioactive solid wastes and there is no regulation on this topic.

Sweden: There is a limitation for gamma radiation in new buildings at $0.5\mu\text{Sv/h}$. This is, however, intended for reduction of the radiation from building materials. Questions concerning this type of waste products have so far been treated case by case. In some cases SSI gives recommendations on how to treat the waste. The situation will be different when the BSS is implemented and more attention is paid to natural radiation.

UK: Other questions not applicable, or no such wastes are produced from drinking water.

7.5 Comments of some countries

The questionnaire provided the possibility for the respective institute to make additional comments. The comments are presented as such.

Ireland: A common EU approach to water quality with regard to natural radioactivity is clearly very necessary. (Hopefully the new Drinking Water Directive will result in this.) Currently we in Ireland operate on the basis of the 1993 WHO recommendations. More detailed guidance on the implementation of these recommendations would be extremely useful. In particular, what is needed, I believe, is a standardised approach to the assessment of dose in the event that the total alpha 100 mBq/L investigation level is exceeded.

Spain: Spain is in favour of a possible common approach for inclusion in a EC regulation.

Sweden: If specific limits are included in the regulation, consideration has to be taken to the actual levels in the Nordic countries.

8 PROPOSAL FOR A EUROPEAN APPROACH

8.1 Introduction

When different kinds of treatment methods are used to remove natural radioactivity from drinking water, wastes containing natural radioactivity will be produced. The wastes are in liquid or solid form. Liquid wastes are produced when materials used to accumulate radioactivity are regenerated or backwashed. Solid wastes are formed in cases where regeneration or backwashing are not used or cannot be used, and when the materials are taken out of service.

The European Basic Safety Standards (96/29/EURATOM) BSS sets down a framework for controlling occupational and public exposure to natural radiation arising from work activities.

The radioactive wastes arising from the treatment of water involve natural radionuclides, but their production, processing, handling, use, holding, etc. can not be considered practice and the exemption levels laid down in Annex I of the BSS are not applicable. However, water treatment waste containing natural radionuclides **may lead to a significant increase in the exposure of members of the public (or of workers who are handling the waste).**

Aeration does not accumulate any radionuclides. It is capable of removing only radon, which should be directly funnelled into the open air.

GAC filters emit gamma radiation when they are in service. The higher the radon concentration and the larger the water use, the more intense the external gamma dose rate around the filter. Furthermore, GAC filters also accumulate ^{210}Pb . Depending on water consumption and radon concentration in influent, high activities of ^{210}Pb may be found in spent GAC beds.

Depending on the Fe- or Mn-removal system, large amounts of radionuclides may be accumulated by the equipment. Backwashing or regeneration at regular intervals, however, enables safe daily use of these

units—radionuclides are rinsed out of the fixed bed and drained into the sewer. The higher the concentration of radionuclides and the higher the daily water use, the more often backwashing or regeneration should be carried out. The regeneration interval is in most cases frequent enough to prevent this technique from causing a problem of waste disposal.

In connection with ion exchangers different operative principles and exchange materials can be utilised. Organic ion exchangers (resins) can usually be regenerated. The properties of many inorganic exchangers (mineral based) cannot be restored by regeneration and therefore they must be discarded after exhaustion.

The radioactive wastes produced by membrane techniques are not accumulated into fixed matrices. The concentrate containing radionuclides, is constantly drained into the sewer as the unit operates. The concentrations of radionuclides in concentrate, however, are low and do not create a waste problem.

These recommendations are meant to be applied in connection with water treatment methods used in private households and small waterworks. The Member Countries of the European Union can use the recommendations made in this report as a basis for their own regulations although it is preferable that the European Union recommend a common approach for all the Member Countries.

Taking into consideration

- that the exemption levels presented in the EU BSS should not be used in connection with the wastes containing natural radioactivity due to water treatment methods,
- that in most cases the effective doses to inhabitants or workers of the company selling and servicing the water treatment methods is small,
- and that to encourage people, by making the procedures needed simple, to start using different kinds of water treatment methods in the event that the water they are using contains high amounts of natural radioactivity,

the following is recommended:

8.2 Aeration

It is recommended that if aeration is used to remove radon from drinking water the aeration system should be fitted in such a way that the radon released from the water does not enter the rooms occupied by the residents but is ventilated into the outside air.

8.3 Gamma-radiation around the GAC filter

It is recommended that the annual dose to inhabitants from external gamma radiation of a GAC filter should not exceed 0.1 mSv. It is also recommended that the dose rate at a distance of 1 m from the GAC filter should not exceed 1 μ Sv/h. To achieve these aims the GAC filter should be equipped with special shielding to attenuate gamma radiation.

As a rule of thumb, if the radon concentration of water exceeds 2,000 Bq/L, special shielding is needed. Instead of constructing special shielding, the location of a GAC filter can be chosen in such a way that the distance from the filter to occupied rooms is long enough to attenuate the gamma radiation. If the radon concentration of water exceeds 5,000 Bq/L, the use of a GAC filter is not recommended.

Close to the GAC filter without shielding the external dose rate may be of the order of 100 μ Sv/h. During maintenance and transportation of a used filter, special attention should be paid to avoid an extra dose.

The best way to reduce the dose rate is to place the filter as far as possible from places where the residents spend time. If this is not possible, additional shielding material must be installed. The best materials are water (water jacket made of a tun), bricks (a case of bricks around the unit) and concrete (e.g. a concrete well ring split lengthwise, concrete tiles). Lead is not recommended because it is expensive and, as a heavy metal, harmful to human health. Assessment of the doses received by residents is discussed in more detail in Chapter 6.

8.4 Disposal of spent GAC

When the GAC filter is no longer used, the amount of radon in the filter decreases rapidly and is close to zero after four weeks. Simultaneously, the dose rate around the filter decreases.

Areas where GAC filters will possibly be used are located in the sparsely populated countryside, where public water supplies are far away. In these areas the municipal dumps are located far from the inhabitation. The densely populated areas where wastes are disposed of by incineration have public water supplies and no spent GAC containing ^{210}Pb will be produced. The authors believe that it is highly unlikely that high amounts of radionuclides can be accumulated in an individual communal dump. Therefore, it is recommended that spent GAC filter material be discharged into communal dumps after it has been "aged" after use for about four weeks.

8.5 Disposal of matrix of Fe- and Mn-removal equipment

It is recommended that if Fe- and Mn-removal methods are used and backwashing or regeneration is carried out about once a week, the liquid be discharged into the sewer and after use the spent matrix be discharged into communal dumps.

8.6 Regeneration of anion and cation exchange resins

It is recommended that if anion or cation exchange resins are used and regeneration is performed about once a week, the regeneration liquid be discharged into the sewer.

8.7 Disposal of adsorptive filters

It is recommended that the wastes produced be discharged into communal dumps.

8.8 Discussion

It was pointed out that the radioactive wastes arising from the treatment of water involve natural radionuclides, but their production, processing, handling, use, holding, etc. cannot be considered practice, and so the exemption levels of the Basic Safety Standards are not applicable. This is why there is the possibility of adopting a totally new approach when considering the attitude towards problems relating to radioactivity and to waste containing natural radioactivity in connection with different methods of removing radioactivity from drinking water.

The authors propose that the recommended annual dose to inhabitants from external gamma radiation of a GAC filter should not exceed 0.1 mSv. When methods for removing natural radioactivity from drinking water are applied, normally the doses which are to be avoided are of the order of 1mSv per annum. This is why a removal method which simultaneously acts as a gamma radiation source and might irradiate dwellers cannot be allowed to cause doses of the same order. The limit should be much smaller—one tenth—otherwise the whole procedure does not make sense. On the other hand, doses much smaller cannot be considered, because the doses from natural radioactivity normally encountered are quite high, from inhalation of radon alone about 1–2 mSv per annum and from gamma radiation of building materials about 0.1 mSv per annum.

The authors also propose that the recommended dose rate at a distance of 1 m from the GAC filter should not exceed 1 μ Sv/h. This limit means that one should stay close to the source 100 hours per year, to receive the 0.1 mSv dose referred to in the previous chapter. Normally this equipment is located in such parts of the dwellings that longer stays are not needed.

The recommendation was also proposed that possible solid and liquid wastes associated with removal methods be delivered to communal dumps or discharged into the sewer. The procedures with the radioactive wastes should be as simple as possible to deal with. This encourages private citizens to start using some radioactivity removal method in connection with their own water supplies, if needed. If too complicated procedures are applied the

dwellers keep on drinking the water containing high amounts of natural radioactivity.

The private companies selling removal equipment do not wish to bear responsibility for the possible wastes. Often the company selling the equipment has nothing to do with the fitting of the equipment in its place of use. The fitters are local private plumbers or small companies.

The fear of radioactivity operates in both directions. People are afraid of radioactivity and want to decrease the amount of natural radioactivity in their drinking water. On the other hand, when this is done there is a special problem with the wastes containing radioactivity. If it is difficult to get rid of them or it is expensive, this opposes the idea of lowering concentrations in drinking water.

RADIOACTIVE WASTES FROM WATER TREATMENT METHODS

Dear Sir/Madam,

The goal of the EU project Treatment Techniques for Removing Natural Radionuclides from Drinking Water (TENAWA) is to recommend methods for removal of natural radionuclides from drinking water. The main interest in the project lies in the methods used in private wells and small waterworks. The project is being carried out by several institutions from Finland, Sweden, Austria and Germany. The Finnish Radiation and Nuclear Safety Authority (STUK) is acting as the coordinator of the project. Some EU Member States already have limits on or recommendations for permissible levels of natural radioactivity in drinking water.

When treatment methods for reducing natural radioactivity from drinking water are used, radioactive wastes in liquid or solid form are produced. In some cases the dose rate near the treatment unit may also increase. When GAC filters (GAC = granular activated carbon) are used to remove radon (^{222}Rn) the dose rate near the filter may vary from 1 to 100 $\mu\text{Sv/h}$. The amounts of radium (^{226}Ra) and lead (^{210}Pb) may also increase in the filter. Typical values for radium could be from 0 to 100,000 Bq (0–10 Bq/g) and for lead from 100,000 to 1,000,000 Bq (10–100 Bq/g).

In the event that the water contains uranium (^{234}U , ^{238}U) and anion exchangers are used to remove it, the amount of uranium collected may vary from 1 to 100 g. When anion exchangers are regenerated; the regeneration liquid may contain from 0.1 to 10 g uranium. Some EU Member States have limits on the amount of natural radioactivity private persons can have in their possession or which can be discharged either into the environment or into communal dumps.

This questionnaire aims at gathering information on existing national regulations and guides, as well as providing the European Commission (CEC) with the information needed to recommend a common approach for possible inclusion in an EC regulation. To obtain all the information relevant to our study, we are collecting data from all the Member States. A detailed questionnaire is enclosed. I would greatly appreciate it if you would answer this questionnaire. An early reply would be accepted with gratitude. If you could send your reply before December 31 1998, I would be able to include your data and opinions in our report.

Sincerely yours,

Martti Annanmäki
TENAWA Project Coordinator

ENCLOSED Questionnaire
Executive summary of the TENAWA Project Mid-term report

QUESTIONNAIRE TO
THE MEMBER COUNTRIES

APPENDIX 1

Martti Annanmäki

27.10.1998

QUESTIONNAIRE

Radioactivity of Drinking Water

1 Limits on and recommendations for reducing radioactivity in drinking water

1.1 Is there any nationwide or partly nationwide survey on natural radioactivity in drinking water currently available in your country?
? Yes ? No

1.2 Do you have available any estimates on population doses due to natural radioactivity in drinking water?
? Yes ? No

If you have reports or other information about the natural radioactivity in drinking water in your country, I would greatly appreciate receiving copies of them.

1.3 Do you have special regulations or recommendations on natural radioactivity in drinking water currently in legislation?
? Yes ? No

1.4 If the answer was yes do you have a limit for

Limit

Radon?	? Yes	? No	_____
Radium?	? Yes	? No	_____
Uranium?	? Yes	? No	_____
Other nuclides?	? Yes	? No	_____

If there is a limit for one or all of these nuclides, would you please specify what is the limit and make reference to the specific regulation?

2. Wastes produced by water treatment methods

A short description of the wastes that might be due to the use of different water treatment methods was given in the covering letter. With reference to the figures presented, would you please answer the following questions. Please keep in mind that the higher ends of the intervals presented are normally encountered in the Nordic countries only.

2.1 When different water treatment methods are used by private households, minor amounts of natural radioactivity are collected and are in the possession of private persons in their home. Are there any regulations in your country against this?

? Yes ? No

If the answer was yes, would you please specify?

2.2 Can such solid wastes be delivered to communal dumps?

? Yes ? No

QUESTIONNAIRE TO
THE MEMBER COUNTRIES

APPENDIX 1

2.3 Can such liquid wastes be legally discharged into the environment?

? No

? Yes

communal waste water pipeline?

? Yes

? No

private waste water system

?

? Yes

? No

2.4 Are there any other reasons why these water treatment methods could not be used in your country?

? Yes

? No

If the answer was yes, would you please specify?

3. Comments

I would greatly appreciate receiving your comments or opinions on a possible common approach for inclusion in EC regulation or any other opinion you may have.

What is your national authority in questions related to natural radioactivity in drinking water? I would appreciate receiving the name and address of the authority and a possible contact person.

This information was given by:

Name:.....

....

Institute:.....

.....

Address:.....

.....

Phone:.....

....

Fax:.....

.

Please return this questionnaire to the following address:

Radiation and Nuclear Safety Authority (STUK)
Mr. Martti Annanmäki
P.O. Box 14
FIN-00881 HELSINKI
FINLAND

Thank you for your kind co-operation.

This questionnaire was sent to the following institutes:

Service de Protection contre les Radiations Ionisantes	BELGIQUE
Statens Institut for Straalehygiene	DENMARK
Bundesamt für Strahlenschutz	DEUTSCHLAND
Greek Atomic Energy Commission	GREECE
Consejo de Seguridad Nucleares	ESPAÑA
Office de Protection Contre les Rayonnements Ionisantes	FRANCE
Radiological Protection Institute of Ireland	IRELAND
Istituto Superiore di Sanita	ITALY
Division de la Radioprotection	LUXEMBOURG
Ministry of Housing, Spatial Planning and the Environment	NETHERLANDS
Departamento de Protecção e Segurança Radiologica	PORTUGAL

QUESTIONNAIRE TO
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APPENDIX 1

Swedish Radiation Protection Institute (SSI)
National Radiological Protection Board
Federal Institute for Food Control and Research

SWEDEN
UNITED KINGDOM
AUSTRIA

Experimental

A batch of activated carbon (39 l) was used for 407 days in order to remove radon from a water supply used by a single household. Water use and radon concentration in both influent and effluent were monitored at 1–3 month intervals. Table I shows the data collected. The concentrations of radon and lead in influent and effluent are set out in Figure 1.

Table I. Sampling dates, flow meter readings and the radon and lead concentrations in influent and effluent at the test location.

Date (d.m.y)	Time in service (d)	Consumed (m ³)	²²² Rn (Bq/L)		²¹⁰ Pb (Bq/L)	
			influent	effluent	influent	effluent
1.7.1997	0	0	–	–	–	–
9.10.1997	100	17	2,596	190	0.375	0.040
17.12.1997	169	50	2,746	180	0.640	0.045
25.3.1998	267	104	2,628	285	0.228	0.031
20.4.1998	293	115	–	290	–	–
25.5.1998	328	133	–	340	–	–
14.7.1998	378	158	2,660	170	0.132	0.015
12.8.1998	407	168	–	–	–	–

(–) not measured

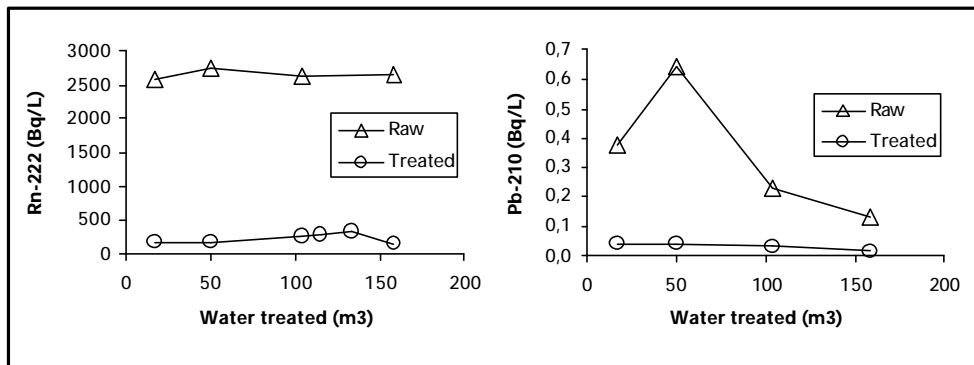


Figure 1. Concentration of radon and lead in influent and effluent as a function of filtered water volume. The radon chart is plotted on a logarithmic scale.

The graphical integrals of the curves set out in Figure 1 represent the total activity of radon in influent and effluent. From the data presented in Table I, the activity of radon retained in the filter, the average water usage, the average removal efficiency and finally the theoretical activity of ^{210}Pb accumulated in the filter can be calculated. The data are set out in Table II.

Table II. The total activity of radon in influent, effluent and retained in the GAC.

Mean values of	
Influent	2659 Bq/L
Effluent	231 Bq/L
Water use	414 L/d
Removal	91.3 %
^{222}Rn retained in GAC	409 MBq

Applying these data we can calculate the theoretical activity of ^{210}Pb in the spent GAC. Consequently:

$$A_{tot}(\text{Pb}) = \frac{3.8235 \text{ d}}{8140 \text{ d}} \cdot 408.9 \text{ MBq} = 192 \text{ kBq}, \text{ and}$$

$$A_f(\text{Pb}) = 192 \text{ kBq} \cdot e^{-8.515 \cdot 10^{-5} \text{ d}^{-1} \cdot \frac{407 \text{ d}}{2}} = 189 \text{ kBq} \quad (\text{ref. 13.8.1998}).$$

Based on the graphical integrals of the charts presented in Figure 2, the total activity of ^{210}Pb retained on GAC directly from the influent is

$$A_{dir}(Pb) = 52.1 \text{ kBq}$$

$$A_f(Pb) = 52.1 \text{ kBq} \cdot e^{-8.515 \cdot 10^{-5} \text{ d}^{-1} \cdot \frac{407 \text{ d}}{2}} = 51.3 \text{ kBq} \quad (\text{ref. 13.8.1998}).$$

Hence, the total theoretical activity of ^{210}Pb in the batch was 240 kBq on 13.8.1998.

The spent batch of GAC was homogenised and a part of it was dried with an infrared lamp. Four sub-samples were collected for gamma spectrometric determination of the radionuclides accumulated in the GAC. Two samples were counted in Marinelli geometry (500 mL) and two in cylindrical containers (diameter 43 mm, height 25 mm).

Results

The nuclides identified and their activities determined are presented in Table III.

Table III. The nuclides identified and the respective activities of four sub-samples collected from the spent batch of GAC. Two measurement geometry were applied.

Nuclide	Unit	#1	#2	#3	#4	Mean
		Marinelli	Marinelli	cylindrical	cylindrical	
Pb-210	Bq/g	10.45	11.57	10.74	12.06	11.20
Th-234	Bq/g	1.14	1.15	1.19	1.18	1.16
U-235	Bq/g	0.06	0.06	0.07	0.07	0.07

The results indicated that the carbon was homogenised well enough. The mean activity of ^{210}Pb calculated from the four sub-samples was 11.2 Bq/g. The density of the sub-samples was 0.6 kg/L. When the carbon was dried, its volume was compressed approximately 10%. Hence the activity in the batch on the day of determination was:

$$A_{tot}(Pb) = 0.6 \frac{\text{kg}}{\text{L}} \cdot (0.9 \cdot 39 \text{ L}) \cdot 11.2 \frac{\text{Bq}}{\text{g}} = 236 \text{ kBq} \quad (\text{ref. 15.3.1999})$$

This value is returned to the date when the GAC filter was taken out of service was:

$$A_f(Pb) = 236 \text{ kBq} \cdot e^{8.51510^{-5} \cdot 214} = 240 \text{ kBq} \quad (\text{ref. 13.8.1998})$$

^{238}U had attained secular equilibrium with ^{234}Th that was detected from the gamma spectrum. The total amount of uranium retained on GAC was thus

$$m(U) = 80.94 \mu\text{g Bq}^{-1} \cdot 1.16 \text{ Bqg}^{-1} \cdot 0.6 \text{ kgL}^{-1} \cdot 0.9 \cdot 39 \text{ L} = 2 \text{ g}$$

The activity of ^{235}U was low.

Conclusions

The measured activity of ^{210}Pb (240 kBq) was concordant with the calculated value (240 kBq). This experiment gave further evidence of the total retention of radon daughters on GAC filters. Additionally, ^{210}Pb was monitored in both influent and effluent during the experiment. An 89% mean efficiency for direct adsorption of ^{210}Pb from influent was recorded. The low concentration of ^{210}Pb in the effluent was another argument for the 100% retention of radon daughters. The radioactivity determinations would have revealed whether some ^{210}Pb was released from the GAC matrix.

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The full list of publications is available from

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