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Radon and radium in well water: Measurements and mitigation of exposure

Tuukka Turtiainen

Säteilyturvakeskus
Strålsäkerhetscentralen
Radiation and Nuclear Safety Authority

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Department of Environmental Science

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Abstract

More than 90% of Finns have access to water distribution services. The distributors are required to comply with statutory monitoring of water quality. About 500 000 people use private wells (most commonly dug wells or drilled wells) or have formed small local co-operatives for abstracting groundwater from communal wells. Monitoring of these wells, however, is often neglected. Presently, drilled wells account for about 40% of all wells in permanent use. Naturally occurring radionuclides, especially radon, can be found in drilled wells in such high concentrations that the use of water causes significant radiation doses and thereby increases the risk of cancer among the consumers. Connecting to the existing water distribution network is often not possible and the only remaining option is household water treatment. The aim of the present study was to assess the applicability of two water treatment methods, aeration and activated carbon filtration, for removing radon at homes. Special consideration was given to the proper installation method, testing of the performance, the quality of the treated water and wastes arising from the treatment. In addition, the impact of household water treatment on radiation doses among the users of drilled wells was estimated, and the occurrence of the isotopes of radium in water from drilled wells was assessed.

Most aerators and activated carbon filters were able to remove most radon from the water, and the methods were not observed to impair the water quality. Shortcomings relating to the reliability of operation and proper installation were observed among some aeration units. Due to these shortcomings, the radon removal efficiency was not always adequate, water leakages occurred and the noise from the units was found disturbing. By contrast, activated carbon units were very reliable. The short-lived radon daughters that accumulate in the carbon emit gamma rays, which necessitates the safe installation of these units. The activated carbon inside the units must be exchanged at 3- to 5-year intervals, applying safe handling methods.

The first units were installed in 1997. In 2007, more than 15% of households using water in which the guideline value set for radon, 1 000 Bq/L, is not met

have acquired water treatment units. Theoretically, a few cancer cases related to the consumption of water from drilled wells have already been averted due to the use of water treatment units. Companies specialising in water treatment have continued to improve the reliability of the units. At present, waterborne radon can be safely removed at homes.

According to the survey, isotopes of radium seldom cause any significant radiation dose to the consumers.

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Tiivistelmä

Yli 90 % suomalaisista saa juomavetensä vesihuoltolaitoksilta, joiden toimittama vesi kuuluu valvonnan piiriin. Noin puoli miljoonaa suomalaista ottaa vetensä yksityiskaivoista (tavallisesti rengaskaivo tai porakaivo) tai he ovat liittyneet pieniin vesiosuuskuntiin, joiden vedenhankinta tapahtuu yhteisistä kaivoista. Näissä tapauksissa veden laatua ei usein seurata. Nykyisin noin 40 % vakituisesti käytetyistä yksityiskaivoista on porakaivoja. Luonnon radioaktiivisia aineita, etenkin radonia, esiintyy osassa porakaivoja sellaisina pitoisuuksina, että veden käytöstä aiheutuu merkittävää säteilyannosta ja sitä kautta veden käyttäjien syöpäriski kasvaa. Liittyminen yhteiseen vedenjakeluun ei usein ole mahdollista, minkä vuoksi ainoaksi vaihtoehdoksi jää käsitellä talousvettä kotona. Tutkimuksen tavoitteena oli selvittää kahden käsittelymenetelmän, ilmastuksen ja aktiivihiilisuodatuksen, soveltuvuutta kotitalousmittakaavassa. Tutkimuksessa kiinnitettiin erityisesti huomiota laitteiden hyvään asennustapaan, laitteiden toimivuuden testaamiseen, käsitellyn veden laatuun ja syntyviin jätteisiin. Lisäksi arvioitiin laitteiden käytön vaikutusta porakaivovettä käyttävien suomalaisten säteilyannokseen ja selvitettiin radiumin isotooppien esiintymistä porakaivovedessä.

Useimmat ilmastimet ja aktiivihiilisuodattimet pystyivät puhdistamaan radonin vedestä lähes täysin eikä menetelmien havaittu heikentävän veden muuta laatua. Osassa ilmastimia havaittiin kuitenkin puutteita, jotka liittyivät toimintavarmuuteen ja vääränlaiseen asentamiseen. Puuteiden johdosta radonin puhdistuminen ei aina ollut riittävää, vesivuotoja esiintyi ja laitteen aiheuttama melu häiritsi asumista. Aktiivihiilisuodattimet sitä vastoin olivat erittäin toimintavarmoja. Aktiivihiileen kertyvän radonin lyhytikäiset hajoamistuotteet lähettävät läpitunkevaa gammasäteilyä, minkä vuoksi laitteen turvalliseen asentamiseen tulee kiinnittää erityistä huomiota. Suodattimessa oleva aktiivihiili on myös vaihdettava 3 – 5 vuoden välein ja jätteen käsittelyssä on noudatettava turvallisia työskentelytapoja.

Ensimmäiset laiteasennukset tehtiin vuonna 1997. Vuoteen 2007 mennessä noin 15 % porakaivotalouksista, joissa veden radonpitoisuus ylittää

enimmäisarvon 1 000 Bq/l, oli hankkinut vedenkäsittelylaitteen radonin poistoon. Laskennallisesti voidaan arvioida, että vedenkäsittelyn ansiosta on pystytty välttämään muutamia mahasyöpätapauksia. Laitevalmistajat ja -myyjät ovat kehittäneet laitteiden toimintavarmuutta ja alkuvaiheen asennusvirheistä on opittu. Nykyisin kaivovedessä esiintyvä radon on täysin ratkaistavissa oleva ongelma.

Otantatutkimuksen perusteella porakaivovedessä esiintyvät radiumin isotoopit aiheuttavat ainoastaan pienen säteilyannoksen veden käyttäjille.

List of original publications

Chapter 2 Tuukka Turtiainen. Radon removal by aeration: Observations on testing, installation and maintenance of domestic treatment units. Water Science and Technology: Water Supply 2009; 9 (4): 469–475.

Chapter 3 Tuukka Turtiainen, Laina Salonen, Pauliina Myllymäki. Radon removal from different types of groundwater applying granular activated carbon filtration. Journal of Radioanalytical and Nuclear Chemistry 2000; 243 (2): 423–432.

Chapter 4 Tuukka Turtiainen, Laina Salonen. Prevention measures against radiation exposure to radon in well waters: Analysis of the present situation in Finland. Journal of Water and Health 2010; 8 (3): 500–512.

Chapter 5 Pia Vesterbacka, Tuukka Turtiainen, Sirpa Heinävaara, Hannu Arvela. Activity concentrations of ^{226}Ra and ^{228}Ra in drilled well water in Finland. Radiation Protection Dosimetry 2006; 121 (4): 406–412.

The author independently conducted the work presented in Chapter 2. The sampling programme relating to work presented in Chapter 3 was designed together with Laina Salonen, Ph.Lic. The field work described in the chapter was carried out jointly with Pauliina Myllymäki, M.Sc.(tech.). The results were analysed and the manuscript written by the author. The manuscript for Chapter 4 was written by the author after discussions with and fruitful commenting by Laina Salonen. In Chapter 5, the author's role was to design and implement the measurements of the ^{228}Ra isotope. This work also included quality control and uncertainty assessments.

Contents

Foreword

Water is one of the primary nutrients in our diet. It serves several functions in the human body such as the transport of nutrients, maintenance of tissue structures, regulation of the body temperature and waste discharge. In order to sustain these functions we must drink water regularly. Continuous access to clean, potable water sources is therefore vital to us.

Unfortunately, adequate drinking water supplies are not a certainty. The World Health Organization has estimated that more than three million water-related deaths occur each year, many of them among children. Microbiological contamination is the largest problem – diarrhoeal diseases account for more than two million of these deaths.

Besides microbial contamination, drinking water supplies are exposed to chemical contamination, which can originate from human settlements, industrial activities and water treatment processes. Detrimental natural chemical contamination of drinking water sources can also occur. The most significant of these contaminants globally are fluoride and arsenic. Naturally occurring radionuclides additionally belong to this category. They originate from uranium and thorium, which have existed in the Earth's crust since its formation. Radon – the main topic of this thesis – is one of them.

The health risk posed by naturally occurring radionuclides is fortunately confined to certain rock aquifers and a limited number of people. According to our latest estimates, about 20 000 people in Finland abstract drinking water from private wells in which the health-based guideline value set for radon is not met. Of all naturally occurring radionuclides, radon is the main contributor to the public dose.

This thesis is a compilation of four articles that originated from different research projects during 1997–2009. They are all related to natural radioactivity encountered in well waters. Three of the articles are related to the prevention of radon exposure, which is the focus of this thesis.

Chapter 1 provides an overview of radon, its origin and occurrence in the environment. A very brief introduction to dosimetric quantities used in radiation protection is presented. This aims at assisting those readers who are not familiar with radiation protection to digest later chapters. The health effects related to radon exposure are also reviewed.

Chapters 2 and 3 present two different household water treatment methods for removing radon from private water supplies. The main focus is on the efficiency of the units as well as their applicability in homes.

Chapter 4 summarises the factors that have affected the magnitude of exposure to waterborne radon in Finland, concentrating on the reductive effect of household water treatment on doses to the public.

Chapter 5 introduces a survey in which the occurrence of radium in private wells was investigated. This survey examined two isotopes of radium (^{226}Ra and ^{228}Ra), and thus complemented a previous survey in which information on ^{228}Ra was lacking.

Finally, Chapter 6 provides a synthesis of previous chapters and discusses future challenges in protecting the public against exposure to waterborne radon.

1 Radon, its occurrence and health effects

This chapter provides an introduction to radon: its sources, transfer in the environment and routes of public exposure. In addition, the dosimetric quantities used in radiation protection are briefly described, together with a review of studies on the health effects of radon. The magnitude of exposure to waterborne radon in Finland is then described. The chapter serves as a basis for the research that is presented in the subsequent chapters.

1.1 Origin of radon

Naturally occurring radionuclides have existed in the Earth's crust since its formation. Uranium (^{238}U and ^{235}U) and thorium (^{232}Th) have half-lives that are of the same order as Earth's age, which is 4.6 billion years. As these isotopes decay, they turn into new radioactive elements, i.e. daughter isotopes. The decay and formation of new isotopes continues until a stable isotope is reached, producing what is referred to as a decay series. In terms of exposure to the public, the most important decay series are those of uranium and thorium (Figure 1).

The isotopes of the decay series reach the so-called secular equilibrium when all members of the decay chain have the same activity in the global inventory (except in the case of branching, e.g. ^{208}Tl). Equilibrium, however, does not occur everywhere in nature. Different elements of the decay series exhibit different chemical and physical properties, which affect their transport in the environment. Uranium, thorium, protactinium and actinium are all actinides, radium is an alkaline earth metal, polonium is a metalloid, while bismuth, lead and thallium are post-transition metals.

The chemistry of radon (^{222}Rn and ^{220}Rn) is radically different from the other elements, since it is a noble gas and virtually inert. In ambient conditions, radon is a monatomic gas and it can easily escape from matrices where it is formed. Radon does not form any chemical compounds in the environment, and hence is not fixed by microbes, plants or sediments. This allows radon to be readily transported in the environment.

Soils are mostly comprised of granular mineral matter and organic materials. Between the grains there are pores filled with air and water. As radon is formed in the mineral grains or organic matter, some of it escapes into the pores where, according to Henry's law, part of it dissolves in the soil water and part passes into the air volume. From the air space of the pores, radon gas is transported via diffusion or, in some cases convection, into outdoor air. The rate of transport depends on soil permeability. When radon reaches outdoor air it can travel hundreds of kilometres from its place of origin.

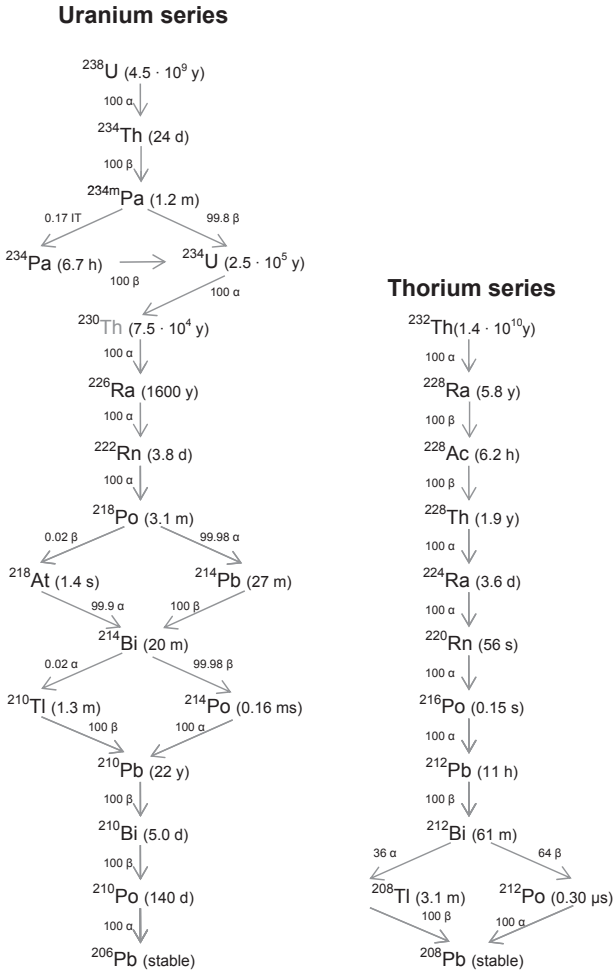

Figure 1. Uranium and thorium series. The half-lives are provided in parentheses and the mode of decay (α, β or IT) is indicated next to the arrows. The branching ratio is given before the decay mode as percentage (Bé *et al.*, 2008).

Radon is thus constantly produced in soils under buildings. There is generally slight underpressure inside buildings to prevent moisture diffusion from indoor air into the structures. This enhances radon entry into the building through concrete joints and lead-throughs found in the foundation, and may increase the indoor air radon concentration to a level hundreds of times higher than what is typically found outdoors. Underground workplaces such as mines are also potential sites for elevated radon exposure.

Groundwater is the zone where all soil pores are saturated with water. Since these pores contain no air, all radon that is released from the soil grains

dissolves in the groundwater. Bedrock is generally much less porous, especially crystalline rocks that are prevalent in Finland. There, groundwater saturates the joints, fractures and weathered areas of the rock. The radon concentration in soil groundwater is typically 1000 times higher than that in surface waters. In bedrock groundwater, radon concentrations are the highest, sometimes even ten thousand times higher than in soil groundwater. As groundwater is abstracted from groundwater aquifers, exposure to waterborne radon can occur through ingestion. Radon dissolved in water readily escapes from water during usage and hence may also cause exposure through inhalation.

The United Nations Scientific Committee on the Effects of Atomic Radiation has assessed the exposure to workers and the public from natural radiation. Radon is the main source of exposure in most underground mines, accounting for about 80% of the total natural radiation exposure of all workers. For the public, radon in indoor air contributes about 50% of the natural radiation exposure globally (UNSCEAR, 2008). In Finland, more than half of the total exposure to radiation (including natural radiation, medical use of radiation, nuclear weapons testing, Fukushima and Chernobyl fall-out) is caused by radon in indoor air (Muikku *et al.*, 2005). Radon in drinking water may cause elevated exposure to people who use drilled wells. Their proportion of the total population is, however, only 4% (Mäkeläinen *et al.*, 2001). Even though the number of the exposed may be relatively small, the individual exposure may be high. It is thus evident that protecting workers and the public against radon is one of the primary targets for the radiation protection authorities (WHO, 2009).

1.2 Radiation and health

The health detriment caused by ionizing radiation is based on its energy imparted to a tissue, for instance by alpha or beta particles, neutrons or photons. This energy can induce chemical reactions in the tissues, such as DNA breakage or the formation of free radicals that can further damage DNA strands. If the cells are not able to repair DNA damage properly, early senescence, apoptosis or mutations occur, the latter of which may lead to cancer. As alpha particles have the highest mass and energy, they release their energy in a very short distance (tens of micrometres), and hence incur more local damage to tissues than beta particles or gamma photons.

The hazard caused by radiation is governed by several factors such as the radiation type, energy and activity, the transfer of a radioactive element inside the body, the biological half-life, physical half-life and target tissue. The first quantity in assessing doses from radiation is the absorbed dose, D, which is the energy imparted to a mass unit. Its unit is the gray (Gy), which equals J kg^{-1}.

The equivalent dose, H_T, is defined as the average absorbed dose in a volume of a specified organ or a tissue due to radiation of type R.

$$H_T = \sum_R w_R D_{T,R}$$

where w_R is the radiation weighting factor. The unit of the equivalent dose is the sievert (Sv). The radiation weighting factor reflects the harmfulness of the radiation type. For photons (gamma rays) and beta particles it is 1 and for alpha particles 20. It must be noted that the equivalent dose is always specific for a tissue or an organ.

The effective dose, E, is defined by the weighted sum of tissue or organ equivalent doses.

$$E = \sum_T w_T H_T$$

where w_T is the tissue weighting factor. The unit of the effective dose is also the sievert (Sv). The tissue-weighting factor has been defined as 0.12 each for bone marrow, the colon, lungs, stomach, breasts and remainder tissues. For the gonads, the weighting factor is 0.08, and for the bladder, oesophagus, liver and thyroid 0.04. For the bone surface, brain, salivary glands and skin it is 0.01 (ICRP, 2007).

Considering the linkage between the cancer risk and effective dose, the International Commission on Radiological Protection has assessed the detriment-adjusted cancer risk to be $5.5 \cdot 10^{-2}$ Sv^{-1} for the whole population and $4.1 \cdot 10^{-2}$ Sv^{-1} for adult workers (ICRP, 2007). This means that if a person is exposed to an effective dose of one sievert, his or her cancer risk increases by 5.5 percentage points over a long period of time. The dose-response relationship is hence thought to be linear. The effective dose can also be understood as a quantity of projected risk.

It must be noted that dosimetric risk assessments may hold large uncertainties because the risk is extrapolated from high exposure situations and it may not therefore be representative for a low exposure range. Epidemiological data on the dose-response relationship in this range are generally lacking. However, evidence supporting a linear dose-response relationship exists for inhaled radon, although evidence is still absent for ingested radon (UNSCEAR, 2006).

1.2.1 Ingested radon

Following its discovery in 1900, radon (or radium emanation as radon was first named) was thought beneficial for human health. Israël-Köhler *et al.* (1936) published a review of the therapeutic effects of radon. They were considered to

include an increase in the leukocyte count, prevention of the coagulation of blood and activation of red bone marrow (treatment of anaemia). The acceleration of metabolism, tranquilization of nerves and relaxation of hypertonia were also reported. Belief in the beneficial health effects of radon still persists, and several "radon spas" remain in operation across the world.

Research on the adverse effects of ingested radon started in the 1950s. In those days, radiation protection was still an emerging science and the quantities when assessing doses or exposure were yet to be defined. This makes it hard to decipher old assessments of doses from ingested radon. What we now define as the radiation weighting factor was first referred to as the radiation biological effectiveness (RBE). In the 1951 recommendations of the International Commission on Radiological Protection (ICRP), a value of 20 was adopted as the RBE for alpha radiation, but in 1954, a value of 10 was given (ICRP, 1951; ICRP, 1955; Snyder and Neufeld, 1957).

In the early 1960s, confusion caused by the usage of the term RBE in both radiation biology and radiation protection was recognised and a new parameter, the quality factor (QF), was introduced for use in radiation protection (Neary, 1963; ICRP, 1964). In radiation biology, RBE continued to specify the capacity of a specific ionizing radiation to produce a specific biological effect, expressed relative to a reference radiation. At this time, the dose equivalent (DE) was also introduced and its unit was defined as the rem (roentgen equivalent man), which equals 10^{-2} J kg^{-1}. The dose equivalent corresponds to the present day equivalent dose, and it was obtained by multiplying the absorbed dose by QF, the dose distribution factor (DF) and other necessary modifying factors. The value of QF depended on the collision stopping power of the radiation (L_∞) and was defined as 10 for alpha radiation (ICRP, 1964). In the 1977 recommendations of the ICRP, the quality factor for alpha particles was fixed at 20 (ICRP, 1977).

In 1977, the tissue-weighting factor, w_T, was also introduced. In 1978, the product of dose equivalents and tissue weighting factors summed over the whole body was defined as the effective dose equivalent, which corresponds to the present day effective dose (ICRP, 1978). The value of w_T for the stomach was defined as 0.06, and it remained so until 1990 when it was fixed at 0.12 (ICRP, 1977; ICRP, 1991).

One of the earliest studies investigating the behaviour of ingested radon was by Nussbaum and Hursh (1957). They studied the solubility of radon in rat tissues in order to provide information on the fate of radon inside human bodies. This publication is still a major source of information when performing dosimetric calculations for radon, although questions about the solubility of radon in adipose tissue and bone marrow have subsequently been raised (Kursheed, 2000).

Hursh *et al.* (1965) measured radon in exhaled air, blood and urine after the administration of radon-bearing water to volunteers. They found that about 56% of radon exits the body with a biological half-life of 18 min. About 38% of radon exhibits a half-life of 33 min and only 6% resides for longer in the body, with a half-life of 4.7 hours. The equivalent dose to the stomach was estimated as $120 \cdot 10^{-9}$ and $110 \cdot 10^{-9}$ Sv Bq^{-1} for ingestion into an empty stomach and a full stomach, respectively. Equivalent doses to other tissues were very small compared to that of the stomach.

Von Döbeln and Lindell (1964) used the data set of Hursh *et al.* and assessed the average equivalent dose to the stomach as $120 \cdot 10^{-9}$ Sv Bq^{-1}. Suomela and Kahlos (1972) employed whole-body counting to determine the longitudinal distribution of ^{214}Bi (radon daughter) in the body after the ingestion of radon bearing water. The calculated equivalent doses to a full and empty stomach were $130 \cdot 10^{-9}$ and $210 \cdot 10^{-9}$ Sv Bq^{-1}, respectively.

UNSCEAR (1977) adopted a reference value of $100 \cdot 10^{-9}$ Sv Bq^{-1} for the equivalent dose to the stomach. Two years later, WHO (1979) recommended a value of $0.25 \cdot 10^{-9}$ Sv Bq^{-1} as the dose equivalent due to ingested radon. This figure most probably corresponds to the equivalent dose to the whole body and assumes a uniform distribution of radon. Kendall *et al.* (1988) criticized the 1979 estimate by WHO and proposed $100 \cdot 10^{-9}$ Sv Bq^{-1} as an equivalent dose conversion factor for the stomach. The 1988 UNSCEAR report also suggested the same value.

Crawford-Brown (1989) fitted values obtained from previous investigations into a newly developed "biokinetics model" and estimated the dose conversion factor for the stomach as $300 \cdot 10^{-9}$ Sv Bq^{-1}. Two years later, he adjusted the estimate to $110 \cdot 10^{-9}$ Sv Bq^{-1} (Crawford-Brown, 1991). Sharma *et al.* (1997) used breath analysis data from Brown and Hess (1992) and derived a dose conversion factor of $82 \cdot 10^{-9}$ Sv Bq^{-1} for the stomach. A much smaller dose conversion factor of $1.6 \cdot 10^{-9}$ Sv Bq^{-1} was suggested by Harley and Robbins (1994). This estimate was based on the assumption that the vascular structure of the mucosa prevents radon from diffusing into the stomach wall deep enough to be able to irradiate the cells sensitive to radiation (NRC, 1999).

Following a request by the U.S. EPA, the National Research Council appointed a committee to conduct a study on health risks related to the ingestion of radon in 1997 (NRC, 1999). The committee carried out a comprehensive review of available literature and formulated a new compartmental model for the behaviour of ingested radon in the body. A refined model for the diffusion of radon through the stomach wall was developed based on the finding that mucine molecules and water constantly secreted by the stomach wall mucosa significantly slow down radon diffusion into the stomach wall. By taking this into consideration, the dose conversion factor for the stomach was assessed as $24 \cdot 10^{-9}$ Sv Bq^{-1} for

adults. The report by the committee also estimated doses to several organs and presented age-dependent equivalent dose conversion factors. The effective dose conversion factor for adults was estimated as $3.5 \cdot 10^{-9}$ Sv Bq^{-1} based on the tissue weighting factors from ICRP (1991). The new tissue weighting factors from ICRP (2007) will not affect this value because most of the effective dose is due to exposure to the stomach, and the w_T for the stomach has remained unchanged.

Kursheed (2000) developed a dynamic model for the retention of radon in the body and estimated the stomach dose conversion factor as $84 \cdot 10^{-9}$ Sv Bq^{-1}. The effective dose conversion factor was estimated to be about $10 \cdot 10^{-9}$ Sv Bq^{-1}. It was, however, pointed out that the model presents a robust upper limit for the dose, as the rate of diffusion to the stomach wall still remained unresolved. Kendall and Smith (2002) continued the work by Kursheed and separately examined doses from radon and its short-lived daughters. Radon daughters (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) were assessed to contribute only 3% to the effective dose. Later, Kendall and Smith (2005) agreed with the NRC's stomach model and supplemented the estimates with calculations for radon daughters. This, however, had little effect on the effective dose.

At present, there seems to be a general consensus among scientists to adhere to the model and dose conversion factors reported by NRC (1999). In this model, waterborne radon is mostly absorbed by the stomach and small intestine walls, from where it enters the blood flowing through the portal vein into the liver. Only a small part is transported into the large intestines and excreta. From the liver, radon enters the large veins and through the right heart into the lungs, where it is released via exhalation.

The association between ingested radon and stomach cancer has been difficult to establish by epidemiology. Collman *et al.* (1988) carried out an ecological study in North Carolina in which inhabitants were divided into an unexposed group (radon in drinking water was below 8.4 Bq L^{-1}) and an exposed group (radon concentration 8.4–403 Bq L^{-1}). The respective number of people in the unexposed and exposed groups was 3.2 million and 0.78 million. The analysis revealed no association between the radon concentration and mortality from gastrointestinal tract cancer. A similar study carried out among children, however, provided evidence that exposure to waterborne radon increases the risk of mortality from childhood cancer (Collman *et al.*, 1991). The ecological study carried out by Kjellberg and Wiseman (1995) also found a positive correlation between radon levels and gastric mortality in Pennsylvania. The groups, however, were selected based on the projected radon concentration in indoor air. It must be noted that ecological studies have an inherent tendency to produce biased results, as there are no data on individual exposures or other factors such as exposure to other contaminants.

Table 1. Review of the literature reporting dose conversion factors for the stomach from the ingestion of radon in adults.

Author	Equivalent dose, stomach ($Sv \cdot Bq^{-1}$)	Effective dose, ($Sv \cdot Bq^{-1}$)
von Döbeln & Lindell, 1964	$120 \cdot 10^{-9}$ †	$14 \cdot 10^{-9}$ ‡
Hursh *et al.*, 1965	$110 \cdot 10^{-9}$ (full) $120 \cdot 10^{-9}$ (empty)	$13 \cdot 10^{-9}$ ‡ $14 \cdot 10^{-9}$ ‡
Suomela & Kahlos, 1972	$130 \cdot 10^{-9}$ (full) † $210 \cdot 10^{-9}$ (empty) †	$16 \cdot 10^{-9}$ ‡ $25 \cdot 10^{-9}$ ‡
UNSCEAR, 1977	$100 \cdot 10^{-9}$	$12 \cdot 10^{-9}$ ‡
WHO, 1979	$0.25 \cdot 10^{-9}$ (whole body)	
Kendall *et al.*, 1988	$100 \cdot 10^{-9}$	$10 \cdot 10^{-9}$
UNSCEAR, 1988	$100 \cdot 10^{-9}$	$12 \cdot 10^{-9}$ ‡
Crawford-Brown, 1989	$300 \cdot 10^{-9}$	$42 \cdot 10^{-9}$ ‡
Crawford-Brown, 1991	$110 \cdot 10^{-9}$	$24 \cdot 10^{-9}$ ‡
Brown & Hess, 1992	$280 \cdot 10^{-9}$	$33 \cdot 10^{-9}$ †
Harley & Robbins, 1994	$1.6 \cdot 10^{-9}$	$0.19 \cdot 10^{-9}$
Sharma *et al.*, 1997	$82 \cdot 10^{-9}$	$10 \cdot 10^{-9}$ ‡
NRC, 1999	$24 \cdot 10^{-9}$	$3.5 \cdot 10^{-9}$
Kursheed, 2000	$84 \cdot 10^{-9}$	$10 \cdot 10^{-9}$
Kendall & Smith, 2005	$24 \cdot 10^{-9}$	$3.5 \cdot 10^{-9}$

† The radiation-weighting factor was first presented as the relative biological effectiveness and later as the quality factor. In publications before 1977, a weighting factor of 10 was applied, and this has been corrected here to a value of 20.

‡ The effective dose was calculated by the author using a tissue weighting factor of 0.12 for the stomach and assuming that the majority of the effective dose is due to stomach.

Auvinen *et al*. (2002) carried out a case-cohort study investigating the association between naturally occurring radionuclides in water and leukaemia, and found no correlation between exposure and risk. Another case-cohort study by Auvinen *et al*. (2005) investigated the association between stomach cancer and exposure to waterborne radon, but did not find an increased risk at the exposure levels studied. Kurttio *et al*. (2006) examined the association between cancers in urinary organs and naturally occurring radionuclides in well water in a case-cohort study. They concluded that radionuclides found in well waters are not associated with a substantially increased risk of bladder or kidney cancers.

The few epidemiological studies that have been carried out so far provide little evidence of an association between stomach cancer, or any other cancer, and exposure to waterborne radon. Doses to the stomach from ingestion of radon remain generally low and only a small number of people are exposed. The projected cancer risk is hence small and not easily observable epidemiologically.

Dosimetric calculations, therefore, presently remain the only tool for assessing risks related to radon in drinking water.

1.2.2 Inhaled radon

Radon gas in air accounts for the largest portion of the annual effective dose to the public both globally and in Finland. The majority of the radon found in indoor air originates from the soil underneath buildings. Certain building materials may also release radon into indoor air. Radon dissolved in household water readily escapes from the water during usage, and hence may transfer into indoor air and incur exposure through inhalation as well as ingestion (Nazaroff. 1992). Therefore, a brief description of the health effects of inhaled radon is also presented here. These effects have been extensively investigated. Detailed reviews provided by ICRP (1994a, 2010), the Committee on Health Risks of Exposure to Radon (1999) and UNSCEAR (2006) should therefore be referred to for further information.

As briefly discussed, radon is a monatomic and inert gas. The majority of radon gas that enters the lungs via inhalation therefore exits the lungs via exhalation, and only a very small part of it enters the blood via gas exchange. However, radon constantly produces short-lived daughters (Figure 1). These daughters (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) are solid and due to ionization during their decay, very reactive. The first daughter is ^{218}Po, which forms molecule clusters whose size is typically below 0.01 µm in less than a second. This portion is termed the unattached fraction. The subsequent daughters, ^{214}Pb and ^{214}Bi, have longer half-lives and thus more time to react with particles in the air. Hence, they form larger particles (0.1–2.5 µm) that are referred to as the attached fraction of radon daughters.

Due to their small aerodynamic diameter, the radon daughters in the unattached fraction most efficiently deposit in the thoracic (mostly alveolar) region of the respiratory tract. As the aerodynamic diameter increases, more particles are deposited in the extrathoracic region (the anterior nose, nasal passages, larynx, pharynx and the mouth). The thoracic region is more sensitive to radiation (with a tissue weighting factor of 0.12) than the extrathoracic region, which is considered as one of the remainder tissues (ICRP, 1994b).

As alpha particles have the highest mass and energy, they release their energy in a very short distance (tens of micrometres), and hence incur more local damage to tissues than beta particles or gamma photons. The health detriment from radon is therefore largely due to isotopes of polonium, which are alpha emitters. These radon daughters have short half-lives, and due to an insufficient time for transport the exposure is mostly confined to the lungs. Presently,

an increased lung cancer risk is thought to be the only relevant health effect of inhaled radon.

Abundant congruent epidemiological data on the radon-induced cancer risk is presently available. These studies form the basis for assessing risks to the public. Dosimetric quantities are hence used only when comparing the risk from radon to risks from other radionuclides or when assessing, for instance, the efficiency of protective countermeasures among exposed groups. UNSCEAR (2006) recommends an effective dose coefficient for inhaled radon of $9 \cdot 10^{-9}$ Sv (Bq h $m^{-3})^{-1}$, while the ICRP (1994a) recommends a dose coefficient of $6 \cdot 10^{-9}$ Sv (Bq h $m^{-3})^{-1}$. The dose coefficients of the ICRP, however, are being revised and the new values should be published in the near future (ICRP, 2010).

Until the 2000s, epidemiological studies in residential dwellings generally lacked the statistical power to verify the increased cancer risk associated with low-dose exposure to radon. The dose-response relationship of radon in indoor air was assessed from epidemiological studies among miners or other groups, who had been exposed to high radiation doses. Miners are, however, exposed to other airborne contaminants at work such as exhaust gases and stone dust as well as smoking, which is common among them. These factors added to the uncertainty in extrapolating the results to radon exposure in homes.

In the 2000s, direct evidence of a link between an increased lung cancer risk and exposure to radon in residential indoor air was obtained when data from separate epidemiological studies were combined. In the first study, data from two Chinese case-control studies were pooled and it was found that the excess relative risk (ERR) of lung cancer was 13.3% (with a 95% confidence interval of 1–36%) per 100 Bq m^3 of radon in indoor air for whole life exposure (Lubin, 2004).

Darby *et al.* (2005) combined the data from 13 European case-control studies. The ERR per 100 Bq m^3 of radon in indoor air for whole life exposure was assessed as 16% (5–31%). Pooled analysis of seven North-American residential radon studies showed an ERR of 11% (0–28%) per 100 Bq m^3 of radon in indoor air for whole life exposure (Krewski *et al.*, 2005). All these studies thus indicate an association between the lung cancer risk and exposure to indoor air radon. It is worth noting that the previously used ERR extrapolated from miner radon studies was 12% (2–25%), which is in the same range. This suggests that the linear hypothesis of the dose-response for radiation exposure is not ungrounded.

1.3 Radon in water sources

Lindell (1968) provided a review of the first measurements of radon in groundwater. Measurements carried out in Sweden as early as in 1915 showed

that typical radon concentrations in groundwater in sedimentary rocks ranged between 30 and 40 Bq L^{-1}, while concentrations in igneous rocks ranged between 74 and 740 Bq L^{-1}. In the late 1950s, German tap waters were surveyed. Radon concentrations in groundwater resources were 4 – 90 Bq L^{-1} and in surface waters less than 2 Bq L^{-1}. In the early 1960s, an average of 40 Bq L^{-1} was recorded in Cornish groundwaters. In 1966, high concentrations of waterborne radon, up to 7 400 Bq L^{-1}, were found in Maine and New Hampshire. The maximum value was similar to that found in Sweden in the early 1960s.

Measurements of waterborne radon in Finland were initiated in 1965. In a four-year period, dozens of drilled wells were examined and the concentrations were high, with a mean of 1 600 Bq L^{-1} and maximum of 10 000 Bq L^{-1} (Kahlos, 1969; Kahlos and Asikainen, 1973). This prompted the surveying of waterborne radon throughout Finland. By the end of 1978, a total of 2 300 samples from waterworks, dug wells, springs and drilled wells had been analysed. The mean radon concentrations in wells dug in soil and wells drilled in bedrock were 60 and 630 Bq L^{-1}, respectively (Asikainen and Kahlos, 1980).

A survey encompassing more than 1 000 samples from drilled wells in southern Finland was carried out in 1982 – 1986. Median radon concentrations varied between rock types. The highest median value, 349 Bq/L, was found in wells drilled in granite. The median value of the total data set was 210 Bq/L (Juntunen, 1991).

By 1994, the well database of the Radiation and Nuclear Safety Authority (STUK) held data on radon concentrations from over 4 000 drilled wells and nearly 3000 dug wells. Salonen (1994) estimated that the mean concentration in drilled wells was 930 Bq L^{-1} and in dug wells 76 Bq L^{-1}. About 160 000 people were believed to be using water in which the naturally occurring radioactivity caused an effective dose exceeding 0.5 mSv per year.

These estimates were downsized in 2001 when data from regional laboratories were incorporated into STUK's dataset. The regional laboratories had carried out small-scale surveys in their areas of operation, and water samples had only been sent to STUK for further analysis if the measured radon concentration exceeded 300 Bq L^{-1}. The new mean values for the radon concentration in drilled wells and dug wells were 590 and 60 Bq L^{-1}, respectively. About 20 000 people were estimated to use water in which the radon concentration exceeded 1 000 Bq L^{-1}, which is the present-day recommended maximum for private water supplies (Mäkeläinen *et al*., 2001; Ministry of Social Affairs and Health, Decree 401/2001).

The most recent representative survey of radon in private wells was carried out in 2005. In this survey, the mean radon concentrations in drilled wells and dug wells were 460 and 50 Bq L^{-1}, respectively. This survey also

covered other radionuclides of the uranium series, namely ^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb and ^{210}Po. On average, 75% of the dose arising from well water consumption among users of drilled wells was caused by radon. This proportion was 60% among users of dug wells (Vesterbacka *et al.*, 2005).

At present, it is evident that radon is the most significant radionuclide in private water supplies. The lack of a national well registry, however, has made it difficult to estimate the number of wells that are employed by homes and by holiday houses. The portion of drilled wells among all wells has increased. In the 1950s, one well out of 1000 was a drilled well, but by the early 1990s the portion had increased to 23% (Korkka-Niemi, 2001). According to STUK's radon survey in 2006, the proportion of drilled wells was already 38% (Mäkeläinen *et al*, 2009). Another more recent survey carried out at STUK suggested even a higher proportion of 43% (Muikku *et al.*, 2009). It must be kept in mind, however, that when conducting questionnaire surveys, people who suspect a high radon concentration in their well water are more inclined to reply diligently.

Based on the 91% of households connected to water services, we can estimate that there are almost 500 000 people who rely on wells to supply their water (Isomäki *et al*, 2007). The best estimate of the number of users of drilled wells is therefore presently about 200 000 people.

1.4 Legislation controlling radon in drinking water

In 1993, STUK issued Radiation Safety Guide ST-12.3, in which the maximum effective dose arising from the ingestion of drinking water was set as 0.5 mSv per year. The guide obliged waterworks and professional water distributors to comply with the limit and also have their water sources investigated. The maximum concentration of radon in water was set as 300 Bq L^{-1} provided that no other radionuclides were present in the water (STUK, 1994).

The EU Drinking Water Directive (Council Directive 98/83/EC) was finalised in December 1998. Radioactivity (other than tritium, potassium-40, radon and radon daughters) was limited to 0.1 mSv per year, which meant that the most relevant radionuclide was excluded from the directive. The monitoring frequencies, monitoring methods and the most relevant locations for monitoring points were to be set later.

Further information on the monitoring was never given, but instead a revision of the directive was planned. In February 2011, however, the Environment Directorate-General stated that there would be no revision of the Drinking Water Directive. Instead, a new directive laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption is now being proposed

by the Commission (2012/0074 (NLE)). The parametric value for radon in the proposal is 100 Bq L^{-1}. Legislation and recommendations by public authorities are discussed in more detail in Chapter 4.

1.5 Aim of this research

According to Salonen's (1994) estimate, the 0.5 mSv limit was exceeded in water among 160 000 users of private wells. At that time, only one company was selling an aeration unit for removing radon and only few had been installed in homes. Therefore, reliable household water treatment units for removing natural radionuclides (radon, uranium, radium, lead and polonium) from drinking water were urgently needed.

Only little data on ^{228}Ra were available in the database of STUK, since the determination method of this isotope required laborious radiochemical separation using barium sulphate (Salonen, 1994). The survey conducted in the early 2000s also dismissed this isotope (Vesterbacka *et al.*, 2005). It was felt that representative information on the occurrence of this isotope was, however, needed.

In view of the afore-noted challenges, the main aims of this work were:

1. To investigate the suitability of an aeration technique for removing radon from drinking water in homes by observing changes in water quality, by assessing the durability and reliability of the units, and by recording problems encountered by the residents.
2. To study the applicability of activated carbon filtration in removing radon in homes by observing changes in water quality, by investigating if any water constituent would limit its applicability, and by studying external radiation and waste generated by the units.
3. To evaluate the impact of having radon removal units on the market by assessing the magnitude of exposure to waterborne radon averted among the members of public.
4. To develop a screening method for ^{228}Ra in drinking water and to assess the occurrence of both isotopes of radium (^{226}Ra and ^{228}Ra) in drilled wells.

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2 Radon removal by aeration: observations on testing, installation and maintenance of domestic treatment units

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Abstract
Radon is one of the contaminants that sometimes impair the water quality of wells, especially those drilled in bedrock. Domestic radon removal units based on aeration have been commercially available for more than ten years. In order to determine how effectively these units remove radon a new test protocol applying frequent sampling while letting 100 litres of water flow, was developed. This way, removal efficiencies can be more accurately calculated and possible malfunctions detected. Seven models of domestic aerators designed for removing radon from household water were tested. The aerators were based on diffused bubble aeration, spray aeration or jet aeration. The average removal efficiencies for 100 litres with a medium flow rate were 86–100% except for a unit that circulated the aerated water back to the well that had removal efficiency of 80% at the maximum. By conducting a questionnaire study usual problems related to the aeration units were localized and recommendations on maintenance and installation are given accordingly.

Keywords
aeration, domestic water treatment, radon, sampling

Introduction
One of the potential contaminants of domestic wells, especially of those drilled in bedrock, is radon. Radon is a radioactive gas that originates from naturally occurring uranium that has been present in the Earth's crust since its formation. Elevated uranium concentrations in bedrock lead generally to high concentration of radon in bedrock wells (Lahermo & Juntunen 1991). Uraniferous rocks are found in many parts of Europe. The highest radon concentrations that have

been found in wells drilled in crystalline bedrock of the Fennoscandian shield in Norway, Sweden and Finland have been 31,900, 57,000 and 130,000 Bq/L, respectively (Banks *et al*. 1998; Mjönes & Åkerblom 1998; STUK 2008). In Central Iberian zone in Spain the highest concentration has been 31,000 Bq/L (Soto *et al*. 1995) and in Bohemian Massif in Austria and in Czech Republic 758 and 4,000 Bq/L, respectively (Katzlberger *et al*. 2001; Hanslík 2008). The highest radon concentration in well water reported in the US was in New Hampshire, 96,000 Bq/L (Lamarre 1989). Residents using radon bearing water are exposed to radon through ingestion and through inhalation because radon is partly released into indoor air during water usage. This exposure increases mainly the risk of stomach and lung cancer (NRC 1999).

According to the recommendation by European Commission, radon concentration in water from commercial or public water supply plants should not exceed 100 Bq/L but "a level higher than 100 Bq/L may be adopted if national surveys show that this is necessary for implementing a practical radon programme". Concentration higher than 1,000 Bq/L is not considered acceptable and this concentration is also recommended as the maximum concentration for private water supplies (European Commission 2001).

The first choice for a household that has an elevated radon concentration in their well water is to connect to a water distribution network. These networks, however, are sometimes unavailable owing to long distances to water mains from the estate or due to projected low end-point consumption. The only viable alternative in these cases is to remove radon with a domestic water treatment unit.

Research on removing radon from drinking water was started in Czechoslovakia in the 1970s (Hanslík *et al*. 1978). Aeration was found a suitable method for stripping radon from water. Removal efficiency of 99% was achieved using aeration porous discs with an air-to-water ratio of 8 : 1 and 8 minutes aeration time. Various aeration techniques and activated carbon adsorption were tested in Sweden in the early 1980s. Aeration under atmospheric pressure was reported to be the best method with removal efficiency up to 75% (Hedberg *et al*. 1982). Removing radon from potable water was also studied in the US at the same time (Lowry 1983). Three methods were tested and found effective; granular activated carbon (GAC) adsorption, diffused aeration and spray aeration. At first, GAC adsorption was regarded as the most promising method because it was found very effective and it had low investment and maintenance costs. The usability of GAC adsorption was re-evaluated later when it was found that external gamma radiation caused by the units may exceed residential guideline values (Rydell *et al*. 1989).

Several aeration techniques for removing radon have been introduced including packed tower aeration, diffused bubble aeration, spray aeration, tray

aeration, jet aeration, shallow-tray aeration, cascade aeration and pressure aeration in hydrophor (NRC 1999). Domestic aerators designed for removing radon are available from several manufacturers and they conventionally combine jet aeration and spray aeration (Lindén 1997; Mjönes 2000; Vesterbacka *et al*. 2008). In these applications, water is pumped from the well through a spray nozzle into an aeration tank. A level sensor is used to control the filling of the tank. The water is then circulated by a pump through an ejector that aspirates air into the water until the pre-set aeration time is reached. The water is then directed into a hydrophor applying solenoid valves. The excess air in the tank is ventilated outdoors (Figure 1). Aerators are always installed to treat all household water i.e. point-of-entry because radon is partly released into indoor air during water usage and thus causes exposure through respiration as well.

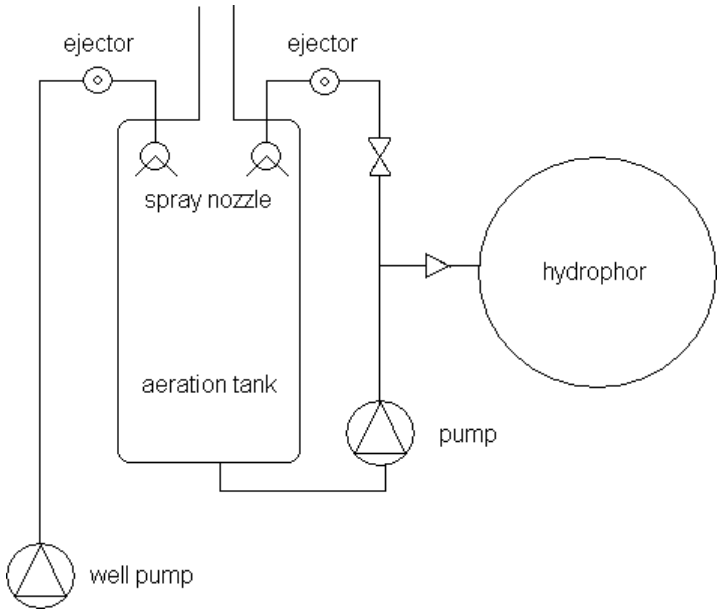

Figure 1. A simplified schematic of the conventional type domestic aerator.

The objective of this study was firstly to develop a reliable method for evaluating radon removal efficiencies of domestic style aerators. Secondly, the most common problems related to the aeration units were localized by sending a questionnaire to the residents who have radon removal units at home. Ultimately, recommendations on maintenance and installation of the units could be given.

Experimental

Seven types of aerators from six manufacturers were tested (Table 1). Radonett from Sarholms Plåtdetaljer Ab is a conventional domestic aerator of hybrid spray/jet aeration type. In this application, the pump that circulates water through the ejector is placed inside the aeration tank, which muffles the noise from the pump. Also, an UV-unit and an air filter are used to prevent microbiological fouling of the aerator. Radon-X100 from HOH Vattenteknik Ab and RF-150 from Oy WatMan Ab are rather similar in structure and match the conventional type of aerator. RnAI-500 from Oy WatMan Ab has a large plastic aeration tank where the pump is also placed. The Vesivahti aerator was originally developed to prevent freezing of the household water pipes and to remove odour and improve taste. It circulates water 5.5 minutes at a time from well to hydrophor and back to the well through two ejectors. The frequency of the 5.5 minute circulation can be adjusted. Radox-aerator from Overcraft Oy is based on diffused bubble aeration where bubbles are created by a hollow, perforated cylinder that is rotated by an electric motor. In accordance with Bernoulli's theorem, a pressure difference is created between the inner and outer surface of the cylinder and air is aspirated into the water as the cylinder rotates. RA 300 from Ins. tst Vartiainen Oy has a separate air compressor and aeration discs. The last two models, however, are no longer available in the market as of March 2009.

Table 1. Domestic aerators that were tested.

Manufacturer	Brand/model	Aeration type	Remarks
Sarholms Plåtdetaljer Ab	Radonett B2	Spray/jet	Tank 80 l
HOH Vattenteknik Ab	Radon X100	Spray/jet	Tank 210 l
Oy WatMan Ab	RF-150	Spray/jet	Tank 150 l
Oy WatMan Ab	RnAI-500	Spray/jet	Tank 500 l
Ins. tst. Vartiainen Oy	RA 300*	Spray/diffused bubble	Tank 300 l
Overcraft Oy	Radox*	Diffused bubble	Tank 300 l
Sednove Oy	Vesivahti	Jet	Well aeration

*No longer available.

First, a sample of raw water was taken from a separate raw water line. In most cases this was possible because a separate line was installed for usage of water which does not require radon removal e.g. watering the garden or washing the car. In this sampling, about 10 mL of water was let flow in a thin stream directly into a tared liquid scintillation vial pre-filled with 12 mL of liquid scintillation cocktail (Ultima

Gold XR from Perkin-Elmer). If sampling of raw water was not possible, radon concentration determined before the installation of the aeration unit was used.

Then, a 25-cm hose was connected to the kitchen tap and the flow rate was adjusted to 4–8 L/min with a stopwatch and a measuring cylinder and the time the flowing started was recorded. Water was let flow into the bottom of a 2 litre flask through the hose and the air bubbles were removed by letting water overflow from the flask. Every two or three minutes a 10 ml water sample for radon measurement was taken with a pipette that was filled by exerting positive water pressure in the bottom of the flask (Kitto 1994). The water sample was then injected into a liquid scintillation vial that was pre-filled with the cocktail. The time of each sampling was recorded. When more than 100 litres of water had flown the sampling was stopped, time recorded and a second sample from the raw water was taken. The radon concentration was determined by counting the samples with Guardian™ 1414 from Wallac (Salonen & Hukkanen 1997).

The measured radon concentrations were plotted as a function of the volume of water that had flowed. The removal efficiency for the water sample that was first taken was calculated according to equation:

$$R_0 = (1 - \frac{c_{a0}}{c_{r0}}) \cdot 100\% , \quad (1)$$

where R_0 is the initial removal efficiency, c_{a0} is the radon concentration in the first aerated water sample, and c_{r0} is the concentration in first raw water sample.

The average radon removal efficiency for 100 litres was calculated according to equation:

$$R_{100} = \frac{\sum_n \left[\left(c_n + c_{n+1} \right) \cdot \left(V_n - V_{n-1} \right) \right]}{\left(c_{r0} + c_{rf} \right) \cdot V_{tot}} , \quad (2)$$

where V_n is the volume of water flowed when sample n was taken and c_n is the radon concentration of this sample. c_{rf} is the radon concentrations in the final raw water sample and V_{tot} is the volume at the last sampling i.e. 100 litres. The minimum removal efficiency during the sampling run was calculated according to:

$$R_{\min} = 1 - \frac{2 \cdot c_{a,\max}}{c_{r0} + c_{rf}} \cdot 100\% , \quad (3)$$

where $c_{a,max}$ is the highest radon concentration measured in the treated water.

In order to gather experiences of the end-users, a questionnaire was sent to 192 households who had purchased equipment to remove natural radionuclides. The questions concerning aeration units were: specifications of the unit, commissioning date, means of storage of aerated water, possibility to

by-pass the unit, location of the unit, data on the room where the unit is placed (floor drain, window, water-proof materials, room temperature and ventilation cannel assembly), subjective experience on the noise from the unit, maintenance that has been carried out and malfunctions that may have occurred.

Results

Figure 2 presents the radon concentration in treated water attained by the tested aerators. Normally, radon concentration in the treated water does not remain constant but can vary greatly depending on how much water is used. In many cases aerators are designed to be able to produce water also during high consumption and hence insufficiently aerated water may enter the household water lines. Among the tested aerators, only Radox produced water that had constant radon concentration. This is due to the large storage tank where aerated water was directed after aeration.

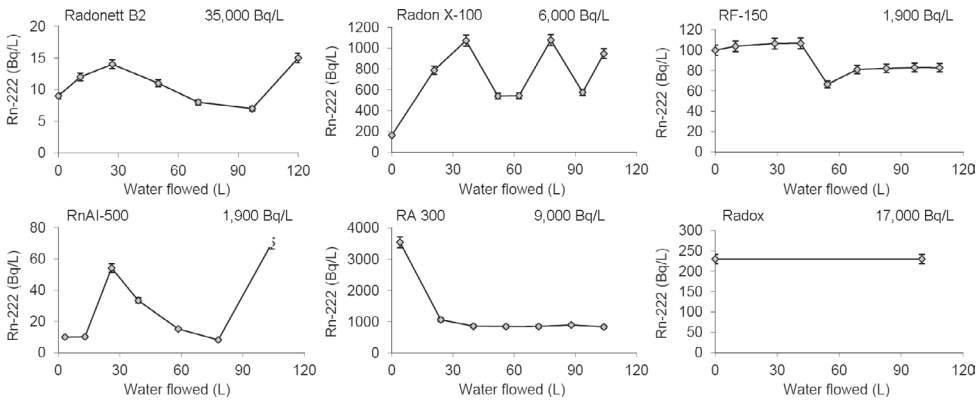

Figure 2. Radon concentration in treated water attained by six aerator models. The mean radon concentration in the raw water is presented on the upper-right corner of the respective graph.

The best removal efficiencies (Table 2) were attained by Radonett B2, RnAI-500 and Radox all of which were able to remove nearly 100% of radon from 100 litres of water. Radon-X100 and RA 300 showed slightly lower removal efficiency and concentrations exceeding 1,000 Bq/L were recorded for both. Before testing RA 300, the residents had been using a lot of water and therefore, there was insufficiently aerated water in the plumbing and the first sample had an unacceptably high radon concentration.

Table 2. Radon removal efficiencies attained by six aerator models.

Model	Mean flow rate (L/min)	Aeration time (min)	R_0	R_ε	R_{min}
Radonett B2	5.5	5.5	100%	100%	100%
Radon-X100	5.2	9	97%	90%	82%
RF-150	7.5	7	94%	95%	94%
RnAl-500	6.5	Cont.	100%	99%	96%
RA 300	4.0	Cont.	61%	86%	61%
Radox	–	7	99%	99%	99%

Radon-X and Radonett have also been evaluated by Lindén (1997). According to the measurements where only two samples of aerated water were taken, the removal efficiency of Radon-X and Radonett were 95–98% and 99–99.9%, respectively. For Radonett, the results between the two studies are similar but for Radon-X removal efficiency presented here is smaller. The model tested by Lindén was Radon-X and in this study Radon-X100, which is probably an updated model.

Similar test runs were carried out at additional locations and repeated at the first test sites. This sort of sampling was able to reveal malfunctioning better than taking a single water sample. In Figure 3, the test run for RF-150 carried out with a flow rate 4.5 L/min and 10 minutes aeration time showed 95% removal efficiency for 100 litres and 82% for minimum removal efficiency. A previous test run with 10 L/min and 10 minutes aeration time had shown 99% removal efficiency for 100 litres at this site. As can be seen on the figure, the radon concentration increases rapidly after 90 litres has flown, which indicated that during the fill-up of the aeration tank, untreated water enters the plumbing. The reason was a broken gasket in the solenoid valve. In the same figure, a test run performed on a revamped RA 300 is shown. Since the removal efficiency of the unit was not satisfactory, a second aeration disk was placed whereby the removal efficiency for 100 litres improved to 99%.

Vesivahti aerator was studied with well water that had shown rather stable radon concentration at three sampling carried out over four months (10,200–11,600 Bq/L). The well was 120 meters deep with a diameter of 125 mm and the pump was located 100 meters deep. First, the 5.5 minutes circulation was adjusted to take place every 45 minutes. After a few days of operation radon samples were taken. Then, the circulation frequency was adjusted to 22.5 minutes and new sampling was carried out a few days later. Radon concentrations at the first and the second sampling were 4,000 Bq/L and 2,500 Bq/L, respectively (Figure 4). Since aeration follows the first-order rate law, we can estimate that

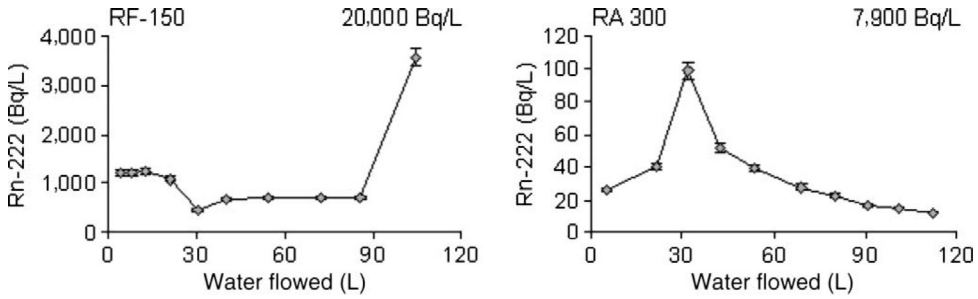

Figure 3. Radon concentration in treated water. The RF-150 had malfunction due to a broken gasket of a solenoid valve. The RA 300 shows the results from a revamped model with two aeration discs.

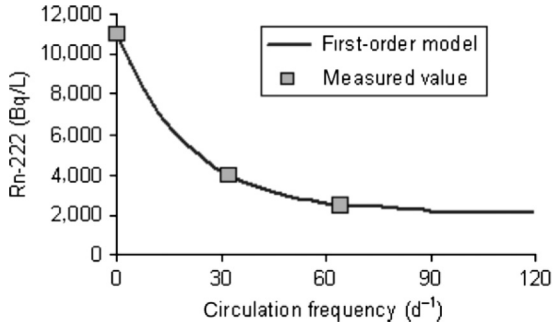

Figure 4. Results from the Vesivahti aerator. Assuming first-order rate law for the aeration process the best removal efficiency that can be obtained using this aerator was about 80% in this well.

in this well, the radon concentration cannot be reduced to smaller than about 2,000 Bq/L with this unit. Radon is constantly transported to the well by ground water flow-trough and from radium-226 that decays into radon. The removal efficiency of this technique depends, thus, greatly on the ground water dynamics of an individual well and therefore sufficient removal efficiencies cannot be assured based on these tests.

The questionnaire was answered by 70 households of which 41 removed radon by granular activated carbon filtration and 17 by aeration. Ten of the respondents reported malfunctions or troubles in their aeration units. Together with those encountered during our tests the most common troubles emerge from the solenoid valves – they had seized up by particles in water, once a gasket was broken and once the coil was burnt. In some cases radon removal efficiency was not sufficient due to a malfunction or increased radon concentration in

raw water. At one location, a level sensor had failed which caused overflow and water damage in the room. The piping joints has also leaked and damaged the floor materials. Sometimes the aerator was very inconveniently placed so that cleaning was very difficult and the taps for by-passing the unit could not be conveniently reached. The circulation pumps had caused obtrusive noise, especially if the aeration started in the middle of the night after e.g. someone flushing the toilet. In some cases, water production rate of the aerator was not sufficient and breaks occurred during high water consumption e.g. showering. The cold climate in Finland had also caused problems, in one case a water pipe had frozen and in another case the exhaust air vent had collected ice and finally gotten blocked. This was caused by the condensed and frozen water vapour in the exhaust vent. Every third of the respondents did not have a possibility to by-pass the aeration unit.

Discussion

This new test protocol gives a much better overview on the functioning and efficiency of the aerator than merely taking a single sample from treated water. Household water usage is not evenly distributed during the day; on the contrary, water usage peaks in the mornings and evenings which now can be better emulated and removal efficiencies calculated accordingly. Newly designed testing has also helped to identify malfunction of the units e.g. the jamming of the solenoid valves.

The type and model of aerator should be selected according to the required water production rate and radon concentration in the raw water. Factors affecting the aerators water production rate are the aeration time and the volumes of the aeration tank and the hydrophor(s). Longer aeration times are needed to reduce higher concentrations of radon and the aeration time should be adjusted so that a sufficient reduction is achieved and no breaks in water supply occur. However, it should be noted that the radon concentration of the raw water may vary significantly (at one of our test locations radon concentration in raw water varied from 3,900 to 15,000 Bq/L) and hence aeration time should be adjusted to achieve sufficient reduction even when the radon concentration in raw water is at its maximum level. The effective volume of the pressure tank should be large enough to enable sufficient water feed into the plumbing during aeration. If the aeration time is 7 minutes and the flow rate from the taps is 18 L/min, there should be a 126-litre effective volume in the tank during the aeration which corresponds to a 500-litre tank with 1 bar pre-pressure.

A possibility to by-pass the aerator should always be considered. Large volumes of water that doesn't have to be treated is often needed e.g. for washing

the car or watering the garden. It is also convenient to have a possibility to by-pass the system if there is a failure in the treatment unit since service is not always available the same day.

The most common malfunction of the aerators is the clogging of the solenoid valves. This normally occurs because of particles in the water, but may also take place because of build-up of calcium scale, ferric hydroxide or manganese oxide. These compounds can be formed during the aeration due to the loss of carbon dioxide, rise in the pH value and oxidising conditions. Later they can transfer to the valves. If iron and manganese removal or water softening is needed, the units should be installed before the aerator to protect the valves from clogging. Particles originating from the drilling can be found in well water several months after commissioning the well and again, after the well has been washed. A filter before the aerator will prevent the particles from entering the unit and causing jamming of the valves.

There are also special requirements for the room where the aerator is installed. Some aerators are rather noisy and therefore the aerator should be located far from the bed rooms. Low pitch sounds are difficult to reduce but sometimes a rubber mat under the pump and the aerator helps. The room temperature should be kept low to prevent the water temperature from rising during aeration, and enhancing microbiological growth. The aerator should not be placed under direct sun light because this can enhance growth of biofilms in systems that are not light-proof. The incoming air should be clean of impurities such as dust and pollen. This is especially important for aerators that are not equipped with separate air filters. The room should also be regularly cleaned to keep the air cleaner. The ventilation channel, which leads the radon-bearing air released from the water into outdoor air, should be wide enough and directed to the roof. The air is saturated with water vapour; hence, the outlet has to be designed properly to avoid build-up of ice during winter. Furthermore, there should always be a floor drain to prevent water damage in case joints leak or solenoid valves clog.

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3 Radon removal from different types of groundwater applying granular activated carbon filtration

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Abstract

Granular activated carbon (GAC) filters were installed in 12 private homes or vacation homes for removing unacceptably high concentrations of radon from household water. Radon removal efficiency was nearly 100% in most locations, although different water types were encountered. Other radionuclides such as uranium, radium, lead and polonium were removed less efficiently. Treated water quality remained good and no significant external radiation dose was caused to the residents.

Introduction

Radon (^{222}Rn) removal from household water is required in tens of thousands of homes in Finland and Sweden. These households are located mainly in the sparsely-populated countryside, where public water supplies are usually far away. The residents, therefore, obtain their household water from private wells. Radon must be removed from all the household water because radon is readily released into indoor air when water is used (e.g. washing machine, shower). High radon concentrations occur mostly in drilled wells, whose number has increased during recent years. In about 40% of the roughly 100,000 drilled wells in Finland, ^{222}Rn concentration exceeds 300 Bq/l which is the limit set for public water supplies.[1,2] In about 20% of the Finnish drilled wells, the radon concentration is higher than 1,000 Bq/l. This is the recommended action level for private households in Sweden.[3]

Two basic techniques are used for removing radon from water: aeration and granular activated carbon (GAC) filtration. Advantages and disadvantages have been reported for both techniques.[4] Information about experiences of radon removal by GAC filtration is mainly from the USA, where several studies

have been conducted.[5–7] The US Environmental Protection Agency (EPA) has proposed aeration as the best available radon removal technology because of serious concerns regarding safe daily operation of GAC treatment and proper disposal of the spent carbon.

The objective of this study was to investigate radon removal by GAC filtration in real household use. Bedrock waters in Finland are typically soft, carbon dioxide or carbonate rich and slightly alkaline. Test locations were selected so that the water types most typically found in Finnish bedrock were covered. Iron, and manganese as well as uranium series' long-lived nuclides (natural uranium, ^{226}Ra, ^{210}Po and ^{210}Pb) may occur simultaneously with radon. Their removal and effect on the GAC filter's performance were also studied. Other aspects considered in this study were: changes in water quality due to filtration, hygiene quality of treated water, external dose rate in the vicinity of the filter and necessary shielding.

Experimental

Installation

Two commercial companies installed GAC-filters at several test locations (July 97 – June 98), which were all private homes or vacation homes (Table 1). Two GAC filters were equipped with a pre-filter and backwash system, one with a pre-filter and the rest without any other filters. In addition, two test locations had a 21-liter anion exchange unit for removing uranium installed before the GAC-filter. Flow meters and sampling taps for influent water were installed in most locations. Whenever an anion exchange filter was installed before the GAC filter, a sampling tap was installed between the two filters. Test locations were selected such that different concentrations of iron and manganese and of total amount of organic carbon (TOC) would occur. Any high concentrations of U, ^{226}Ra, ^{210}Po and ^{210}Pb could not be included because knowledge on their removal is limited. In Table 1, test location C (b) is actually the same household as test location C (a) but with a new filter combination and the GAC batch changed.

A GAC filter operates under normal plumbing pressure, 2–6 bar. It is installed after the pressure tank, so that all household water will be treated (point-of-entry). The filter vessel is made of fiberglass-strengthened plastic and operates in the down-flow mode. Vessels equipped with a backwash system need free volume for the backwashing that is about one third of the total vessel volume. Capacity of 39- and 63-liter GAC filters is at least 12.3 l/min (Fig. 1).

The carbon type used during this study was selected based on the results of a study carried out in Finland.[8] The GAC bed sizes were calculated by applying the first-order kinetics model presented by Lowry.[9]

Table 1. Installation data and some water quality parameters at the test locations.

Test location	GAC vol., l	Residence type	Location	Flow meter	Back-wash	Pre-filter	Anion exch.	Fe, mg/l	Mn, mg/l	TOC, mg/l	U, mg/l
A	39	Permanent	Cupboard	–	–	–	–	0.021	0.018	–	0.052
B	40	Permanent	Tech. room	x	x	x	–	0.67	0.13	1.1	0.013
C (a)	39	Permanent	Cellar	x	–	–		0.015	0.066	2.3	0.20
C (b)	39	Permanent	Cellar	x	–	x	x	0.015	0.066	2.3	0.20
D	39	Permanent	Cupboard	x	–	–	–	0.14	0.26	3.4	0.043
E	40	Vacation	Cellar	x	x	x	–	0.033	0.014	1.2	0.022
F	63	Vacation	Cellar	–	–	–	–	0.016	0.008	2.4	0.12
G	63	Permanent	Shed	x	–	–	–	0.017	0.027	1.8	0.067
H	39	Permanent	Cellar	x	–	–	x	0.130	0.004	1.9	0.26
I	63	Permanent		x	–	–	–	–	–	–	–
J	63	Permanent		x	–	–	–	–	–	–	–
K	63	Permanent		x	–	–	–	0.70	–	–	–
L	63	Vacation	Cellar	x	–	x	–	0.16	0.066	2.1	0.007

x Installed.
– Not installed or not measured.

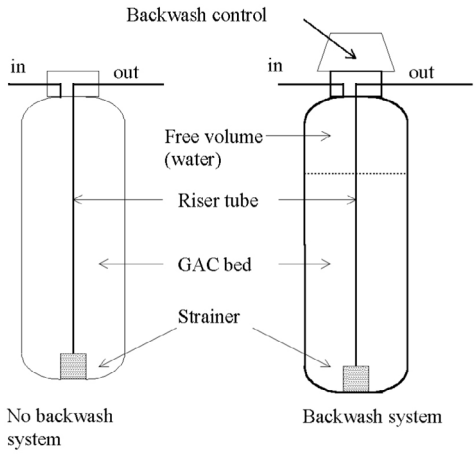

Figure 1. A schematic picture of the two GAC vessel types used in the study. The vessel on the right is equipped with a backwash system.

Sampling and analyses

The radon sampling protocol was as follows: At least 100 l of water was used, with a flow rate of 10 l/min or more. Water flowed into the bottom of a 2 l flask through a hose connected to the tap. The air bubbles were removed by allowing water overflow from the flask. A 10 ml water sample for radon measurement

was taken with a pipette that was filled by exerting positive water pressure in the flask.[10] The sample was injected into a liquid scintillation vial pre-filled with scintillation cocktail. The sample was analyzed in the laboratory using an EG&G Wallac Guardian 1414 liquid scintillation counter (LSC).[11]

^{226}Ra was determined applying gross-alpha determination with an LSC spectrometer, Quantulus™.[11] Uranium was determined using radiochemical separation and alpha spectrometry.[12] The ^{210}Po was determined using spontaneous deposition on a silver disk and alpha spectrometry.[13] The ^{210}Pb was determined after a 200-day in-growth period by measuring its daughter ^{210}Po.[13]

Water quality samples were taken according to the instructions provided by the accredited laboratory that performed the analyses (City of Helsinki Environment Centre). Several parameters were determined but only some of them are discussed here.

External gamma dose rate measurements were made by using a DGM-Turva radiation counter from KATA-Electronics OY. Dose rate was measured at different heights on the filter surface to monitor the operation of the filter. Horizontal dose rate measurements at different distances from the filter were performed to evaluate the radiation exposure to the residents.

Results and discussion

Radon removal

Radon was removed very effectively by most filter combinations at test locations (Table 2). The removal efficiency remained practically constant throughout the whole period of operation in which the filters were studied. The only exceptions were test locations B and C (a). The results in test location B can be explained by insufficient packing of the carbon (channeling) during the first sampling, when only 64 bed volumes were filtered (Fig. 2). High levels of Mn, Fe and TOC (mainly humus) did not decrease radon removal efficiency.

Test location C (a) had the lowest reduction of all the test locations from the beginning of the test period (Fig. 2). The reason for this cannot be explained definitively. When the water quality at this site (Fe, Mn, TOC, SiO_2, turbidity, bacteria, CO_2, total hardness, etc.) was compared with that of the other test locations no explanation could be found. When the vessel was emptied and filled with a new carbon batch no visible malfunction, such as a lumpy GAC or a loose riser tube, were noticed. One explanation for the poor retention could be the high concentration of uranium (0.2 mg/l) in the influent water. To investigate the effect of uranium on radon removal, the unit was loaded with a new batch of GAC. An additional pre-filter (5 µm) and a 21-liter anion exchange filter

(uranium removal) were installed before the GAC filter. This new installation is referred as test location C (b). Following this installation, the filter removed more than 99.8 % of the radon.

Table 2. Range of influent and effluent water radon concentration at different times during the period of operation of the filters and radon reduction in the most recent sampling and corresponding treated water volume (in bed volumes).

Test location	Radon influent water	Effluent water, Bq/l	Time in usage, months	Water treated BV	Radon reduction, %
A	1900–2000	1.2–2.8	8	~2100	99.9
B	3500–4200	12–190	11	5350	98.9
C (a)	2600–2700	160–340	13	4050	92.9
C (b)	3100	<0.4	1	230	99.8
D	3700–4100	<0.4–15	13	1570	>99.9
E	910–1100	<0.4–3.1	8	150	>99.9
F	1700–3000	0.9–2.2	13	~400	>99.9
G	5100–7400	<0.4–2.2	10	1586	>99.9
H	1600–2200	1–16	13	3210	99.2
I	1300–2200	1–2.3	10	880	99.9
J	4100–4600	1.2	13	3430	>99.9
K	1300–1600	<0.4	13	3180	>99.9
L	5600–6000	69–76	4	180	98.7

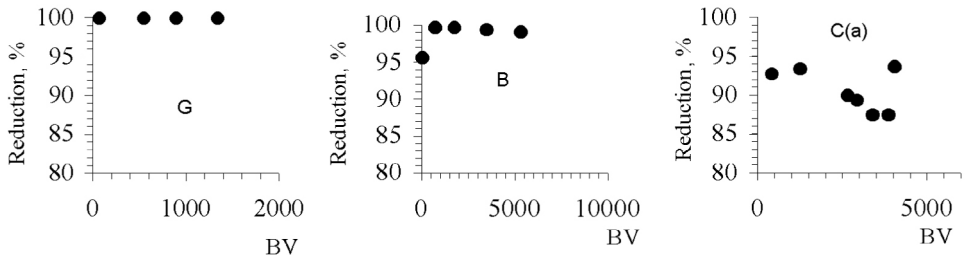

Figure 2. Radon reduction percentages as a function of treated water (in bed volumes) at three test locations. The chart for test location G shows typical radon reduction; test location B and C (a) have abnormal reduction charts.

Removal of U, ^{226}Ra, ^{210}Po and ^{210}Pb

In Finland the long-lived uranium series' radionuclides (^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb and ^{210}Po) quite often occur simultaneously with radon in bedrock water. It would be desirable to be able to remove all the nuclides simultaneously with the same filter. Because our knowledge about removal of these nuclides by GAC was limited, test locations with only low concentrations of these nuclides in the water were selected.

All these nuclides have shown different retention rates in the GAC filters studied (Table 3, Fig. 3). The results for the same nuclide may also vary among filters. It is obvious that GAC filtration is not capable of removing all these nuclides simultaneously. The same results were obtained at the test locations where only gross alpha measurement was used to screen removal of the long-lived radionuclides (Table 4). The only exception was test location L where, according to previous radiochemical determinations, gross alpha comprises mainly ^{210}Po and ^{226}Ra.

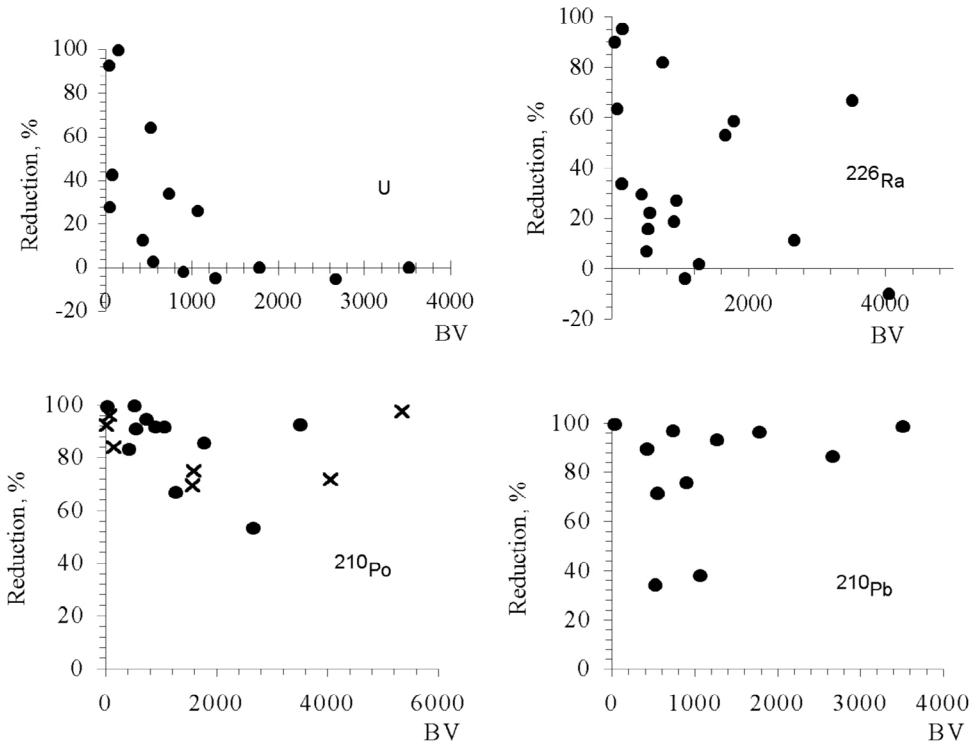

Figure 3. Uranium, ^{226}Ra, ^{210}Po and ^{210}Pb reduction as a function of treated water (in bed volumes). A cross indicates a preliminary result.

Table 3. Radionuclide concentrations in influent and effluent water at test locations where regular sampling was carried out.

Test location	Treated BV	U influent	Effluent, µg/l	^{226}Ra influent	Effluent, Bq/l	^{210}Pb influent	Effluent, Bq/l	^{210}Po influent	Effluent, Bq/l
B	741	12.9	8.5	0.198	0.036	0.283	0.009	0.122	0.007
	1786	14.8	14.8	0.169	0.070	0.371	0.014	0.102	0.015
	3518	13.0	13.7	0.198	0.066	0.328	0.005	0.154	0.012
	5348	–	–	0.198	0.095	–	–	(0.477)	(0.012)
C	431	207	181	0.211	0.149	0.375	0.040	0.266	0.045
	1274	202	212	0.228	0.224	0.640	0.045	0.126	0.042
	2670	200	211	0.247	0.219	0.228	0.031	0.109	0.051
	4051	–	–	0.240	0.264	–	–	(0.106)	(0.030)
D	202	–	–	–	0.401	–	0.117	–	0.027
	528	35.9	12.8	0.280	0.236	0.354	0.234	0.169	<0.002
	1069	42.9	31.8	0.260	0.270	0.313	0.195	0.520	0.045
	1565	–	–	0.542	0.429	–	–	(0.141)	(0.043)
E	41	18.9	1.4	0.052	<0.01	0.461	0.003	0.416	0.003
	151	21.7	0.2	0.050	<0.01	–	–	(0.031)	(0.005)
G	75	70	40	0.303	0.108	–	–	(0.455)	(0.018)
	555	67	65	0.319	0.248	0.517	0.149	0.461	0.043
	903	67	68	0.320	0.263	0.616	0.151	0.360	0.031
	1586	–	–	0.289	0.263	–	–	(0.203)	(0.051)

– Not determined.
( ) Preliminary result.

Table 4. Gross alpha and ^{226}Ra concentrations in influent and effluent water at test locations where regular sampling was not performed.

Test location	BV	Gross-α, Bq/l		^{226}Ra, Bq/l	
		Influent	Effluent	Influent	Effluent
I	142	6.85	5.90	0.288	0.191
	509	6.09	5.51	0.203	0.189
J	943	1.64	1.01	0.085	0.062
K	184	0.58	0.23	0.08	0.07
	1656	0.45	0.33	0.149	0.070
L	23	2.32	0.13	0.185	0.011

High rates of uranium reduction were obtained only when less than 200 BV's (bed volumes) of influent water were filtered, though most filters showed low reduction even then. There seemed to be no particular breakthrough volume for uranium: rather, the retention decreased gradually. When large amounts of water were treated, the uranium concentration in the effluent water was the same or slightly higher than in the influent water.

Radium-226 reduction varied considerably. Two of the filters removed radium fairly well: 67% at test location B after 3520 BV's and 53% at test location K after 1660 BV's. The water at both test locations contained a large amount of iron (> 0.4 mg/l). At test location B iron was analyzed three times; its reduction rate was 99%. Iron removal may increase radium retention. Very low ^{226}Ra concentrations (< 0.05 Bq/l) are omitted from the chart in Fig. 3.

There are only eleven final results for ^{210}Po and ^{210}Pb because the analysis requires a 200-day in-growth period. However, several preliminary results on ^{210}Po are available, with errors of up to ±20%. Until now, polonium, and lead reduction has been fairly good.

No clear tendency regarding the removal of uranium, radium, polonium, and lead could be discerned. It is obvious that the chemical forms of these radionuclides, vary greatly in the waters studied. This depends on the physico-chemical properties of the groundwater, e.g. redox potential, pH-value, different complexing agents, colloids, bacteria, and salinity.

Physico-chemical quality of treated water

Several water quality parameters that were selected using the results of a comparative study of different carbons, were monitored.[8] The selected parameters were those that had been affected by GAC filtration. The most important parameters (Fe, Mn, TOC, temperature and pH) were monitored regularly (Table 5) as was the hygienic quality. Other parameters were monitored less frequently (Table 6).

The iron concentration decreased 50 – 99 % during GAC filtration (Fig. 4). The best iron retention was obtained at test location B, where the raw water iron concentration was the highest (0.4 – 0.7 mg/l). Very low iron concentrations (< 0.05 mg/l) are omitted from this chart due to the uncertainty of the results.

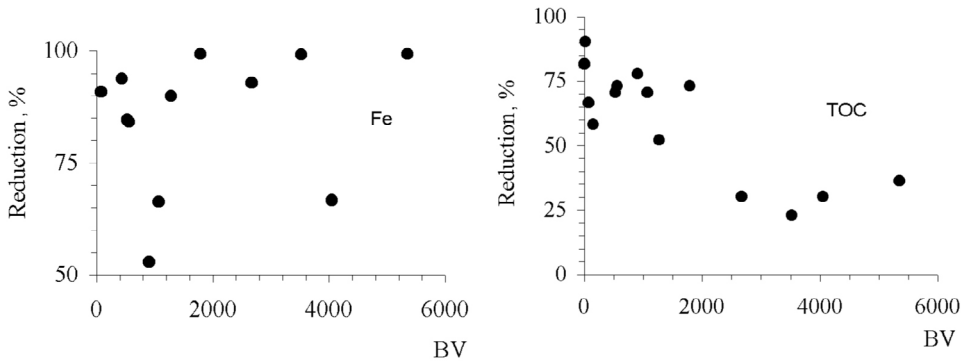

Figure 4. Iron and TOC (total organic carbon) reduction as a function of treated water (in bed volumes).

TOC (total organic carbon) indicates the amount of organic matter present in the water. TOC was also removed fairly well, with a reduction rate of 55–90% when less than 1000 BV's were filtered. The reduction rate decreased gradually, so that after 2000 BV's had been filtered it was 25–35%.

GAC filtration did not directly increase the water temperature. The main reason for increased effluent water temperatures were the pressure tanks. If the tanks are in a warm place and the water is stored for some time prior to use, the water temperature rises. An ideal place for the filter is in a cellar, where there is no concern about the filter freezing or the water warming up. The risk of bacterial growth is also the lowest.

Table 5. Fe, Mn, TOC, temperature and pH in influent and effluent waters at different bed volumes at test locations where regular sampling was carried out.

Test location	BV	Fe, mg/l		Mn, mg/l		TOC, mg/l		Temperature, °C		pH	
		Infl.	Effl.	Infl.	Effl.	Infl.	Effl.	Infl.	Effl.	Infl.	Effl.
B	1786	0.410	0.003	0.120	0.110	1.5	0.4	8.7	9.8	7.7	7.7
	3518	0.390	0.003	0.110	0.100	1.3	1.0	8.8	9.8–11.3	7.73	7.70
	5348	0.670	0.005	0.130	0.086	1.1	0.7	13.2	13.9–17.6	7.8	7.7
C	431	0.016	<0.002	0.064	0.070	–	–	9.0	9.0	7.74	7.71
	1274	0.020	0.002	0.086	0.068	2.3	1.1	8.0	7.9	7.6	7.6
	2679	0.014	<0.002	0.074	0.078	2.3	1.6	5.3	5.3–7.9	7.44	7.38
	4051	0.015	0.005	0.066	0.057	2.3	1.6	12.3	12.3–13.7	7.6	7.6
D	528	0.150	0.023	0.250	0.250	3.4	1.0	6.5	7.0	7.1	7.1
	1069	0.140	0.047	0.260	0.230	3.4	1.0	6.9	7.1	7.0	7.1
E	41	0.020	0.004	0.009	0.021	1.1	0.2	8.3	8.3–13.0	8.49	8.44
	151	0.033	0.058	0.009	0.014	1.1	0.2	8.5	12.5–14	7.2	7.2
F	~100	0.019	<0.002	0.024	0.029	–	–	–	–	8.29	8.24
	~400	0.016	0.004	0.008	0.009	2.4	0.8	9.5	9.5	8.1	8.3
G	75	0.033	0.003	0.027	0.022	1.5	0.5	5.9	5.9	8.1	8.4
	555	0.019	0.003	0.024	0.023	1.5	0.4	6.1	4.7–5.1	8.1	8.1
	903	0.017	0.008	0.027	0.026	1.8	0.5	–	–	8.1	8.2
H	2270	0.005	0.004	0.005	0.004	1.1	0.6	13.1	13.1	7.1	7.2
L	23	0.160	0.015	0.066	0.005	2.1	0.2	4.5	8–9	7.3	8.9

Microbiological quality of treated water

After a GAC filter has been installed bacterial populations start to develop on the GAC. The development of bacterial populations is affected by the length of time a filter is in service, water temperature, flow rate, the material of which it is made, and the quality of the influent water.[14] The hygiene quality of the water should be investigated at different times of day (e.g. morning, afternoon and evening) and the samples should also be collected at frequent intervals to determine how the hygiene quality of effluent water changes as the water is used.

Table 6. Turbidity, electrical conductivity, alkalinity, total hardness, and SiO_2 in influent and effluent waters at different bed volumes at test locations where regular sampling was carried out.

Test location	BV	Turbidity, FTU		Electical conductivity, mS/m		Alkalinity, mmol/l		Total hardness, mmol/l		SiO_2, mg/l	
		Infl.	Effl.	Infl.	Effl.	Infl.	Effl.	Infl.	Effl.	Infl.	Effl.
B	0	2.3	–	55.0	–	1.44	–	1.45	–	–	–
	64	–	<0.05	–	53.2	–	1.49	–	1.36	–	–
	741	–	<0.05	–	53.7	–	1.54	–	1.41	–	–
	1786	–	–	53.2	53.1	1.56	1.59	1.40	1.39	–	–
	3518	2.5	0.30	55.4	55.4	–	–	–	–	–	–
	5348	–	–	51.8	51.4	–	–	–	–	16.9	16.7
C	0	0.14	–	44.5	–	2.3	–	1.51	–	–	–
	431	<0.05	<0.05	–	–	–	–	–	–	–	–
	1274	0.05	<0.05	–	–	–	–	–	–	–	–
	2670	0.06	0.07	–	–	–	–	–	–	14.5	14.5
	4051	0.09	0.01	–	–	–	–	–	–	14.7	14.7
D	0	0.17	–	57.8	–	2.47	–	0.68	–	–	–
	202	–	–	–	72.7	–	–	–	–	–	–
	528	–	–	50.9	44.1	2.43	2.41	0.61	0.61	–	–
	1069	–	–	51.4	53.0	–	–	0.63	0.60	–	–
E	0	0.72	–	24.5	–	1.81	–	0.72	–	–	–
	41	–	–	24.2	24.0	–	–	0.71	0.72	–	–
	151	0.42	0.23	23.6	23.5	–	–	–	–	14.5	14.5
F	~100	0.24	<0.05	43.2	45.7	3.01	3.01	0.67	0.59	–	–
	~400	0.19	0.1	44.0	44.1	–	–	–	–	11.9	11.3
G	75	<0.05	0.05	25.4	25.4	1.94	1.95	0.82	0.81	–	–
	555	<0.05	<0.05	24.7	24.7	1.92	1.94	0.80	0.80	–	–
	903	–	–	24.7	24.7	–	–	–	–	–	–
H	2273	0.23	0.23	–	–	1.25	–	–	–	–	–
L	23	–	–	29.5	30.3	–	–	–	–	–	–

The results of the heterotrophic plate counts (HPC) presented here were collected after the applied radon sampling protocol (100 liters of usage) and are therefore not characteristic of general effluent water quality (Table 7). Two samplings were carried out to analyze water quality after the water had been standing in the plumbing for several hours (Table 8).

The Ministry of Social Affairs and Health in Finland has set target values for HPC. For public water supplies HPC should not exceed 100 cfu/ml (colony forming units per milliliter) and 10 cfu/ml at 22 °C and 35 °C, respectively. At 22 °C, effluent water HPC exceeded 100 cfu/ml in three cases. Only in one of those cases was the influent water HPC less than this. HPC in effluent water at 35 °C exceeded 10 cfu/ml in six cases. In three of those cases influent HPC

Table 7. Heterotrophic plate counts at 22 °C and 35 °C in colony-forming units (cfu) per milliliter at test locations where regular sampling was carried out. Sampling after letting 100 liters of water flow.

Test location	BV	22 °C influent	Effluent, cfu/ml	35 °C influent	Effluent, cfu/ml
B	1786	2	13	0	8
	3518	90	4	4	2
	5348	23	210	1	230
C (a)	1274	3	6	0	2
	4051	5	16	1	1
D	528	>3000	140	9	5
	1069	180	8	1	0

Table 8. Heterotrophic plate counts at 22 °C and 35 °C in colony-forming units (cfu) per milliliter at test locations C (a) and E after letting 5 and 200 liters of water flow.

Test location	BV	22 °C, cfu/ml			35 °C, cfu/ml		
		Influent	Effluent (5 l)	Effluent (200 l)	Influent	Effluent (5 l)	Effluent (200 l)
C (a)	431	13	76	–	10	43	–
E	36	190	180	36	220	190	33

was 10 cfu/ml or less. In many cases influent HPC was higher than effluent HPC. According to these results HPC is higher in water that has stood in the plumbing for several hours. Similar sampling for studying aerators has been carried out and indicated much higher HPC. Often the HPC at 22 °C exceeded 3000 cfu/ml in aerated waters.[15]

Dose rate and shielding

After radon has been adsorbed on a GAC, it decays into its short-lived progeny, ^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po. These daughters are also retained on the GAC and they reach equilibrium with radon in a few hours.[16] The ^{214}Pb and ^{214}Bi also emit gamma radiation and hence the GAC filter becomes a source of gamma radiation. Within about a month after a new GAC batch has been used, the adsorption and decay rates of radon will be equal. During this steady state the gamma radiation reaches its maximum. In vacation homes, where water usage is not regular, the steady state is not attained and radiation levels vary significantly.

The external gamma dose rate depends on the radon concentration in the influent water, on daily water usage, and on the GAC vessel dimensions.

Due to these factors gamma dose rates varied at different test locations (Table 9). At test locations A and D, the GAC filter was placed in a cupboard, inside the house. Therefore, radiation shields were installed. The filter was encased in three 1 mm-thick sheets of lead. Lead attenuated the gamma radiation very efficiently: a 3 mm thick layer reduced radiation on the filter surface about 40% at test location D. On a desk 150 cm away from the filter, the radiation field was 0.16 µSv/h after installation of the shield. Other places in the house where the residents spend time are not near the filter. At test location A a separate shed for the pressure tank and the GAC unit is under construction. At the other test locations GAC filters were installed either in a cellar, in a technical room (where e.g. boiler and fuse panel are located), or in a separate shed where the filter would not cause significant exposure to the residents.

Table 9. GAC bed size, daily usage, effluent water ^{222}Rn concentration, and maximum dose rate on the GAC filter surface at different test locations.

Test location	GAC size, l	Daily usage, l/day	Effluent ^{222}Rn, Bq/l	Maximum dose rate on filter surface, µSv/h
A	39	–	1900 – 2000	47
B	40	627	3500 – 4200	103
C (a)	39	510	2600 – 2700	37
D	39	175	3700 – 4100	18.5
G	63	327	5100 – 7400	111
H	39	340	1600 – 2200	26.8
Vacation homes, no steady-state				
E	40	–	900 – 1100	3.9
F	63	–	1700 – 2800	12.6
L	63	–	5800 – 6000	35.6
Lead shield 3 mm				
A	39	–	1900 – 2000	14
D	39	175	3700 – 4100	11.1

Gamma emission profiles on GAC vessel surfaces are usually similar: the highest gamma dose rate is measured in the top few centimeters of the bed. Test location C (a), which had the lowest radon reduction rate, also had a different emission profile (Fig. 5). When the removal efficiency of the carbon is not good, maximum gamma activity moves towards the center of the GAC bed.

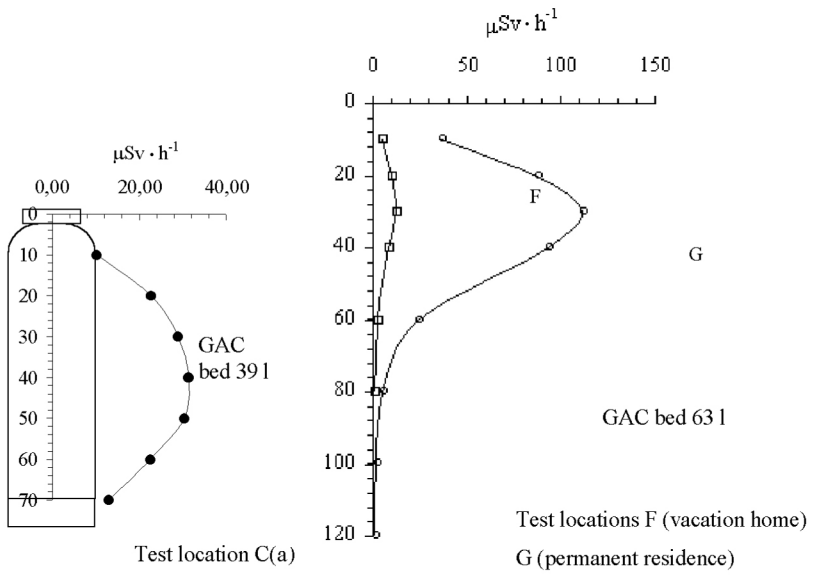

Figure 5. Gamma emission profiles at different test locations. Vertical distance measured from the top of the filter cap. Test location C (a) has a profile indicating poor radon reduction. Test locations F and G have typical gamma profiles.

Spent GAC as radioactive waste

At present there are no regulations in Finland specifically applicable to the disposal of GACs containing natural radionuclides. GACs accumulate mainly radon decay products but also long-lived radionuclides like uranium and radium. Previous studies have shown that nearly 100% of the ^{210}Pb produced from the decay of radon retains on the GAC.[17] Therefore, the major part of the accumulated activity will be due to ^{210}Pb and its progeny (^{210}Bi and ^{210}Po). A spent GAC batch should be allowed to stand at least three weeks to allow for the decay of radon and its short-lived daughters.

A GAC batch (14 liters, 8.2 kg dry weight) was studied in order to investigate the accumulated activity of ^{210}Pb and other long-lived nuclides. The GAC filter had operated without a sediment pre-filter for three months when the various GAC types were tested. Radon concentration in the effluent water had varied significantly (840 – 9300 Bq/l). Therefore, the calculated maximum ^{210}Pb concentration varied between 42 and 88 kBq.

The GAC batch was divided into four parts. Samples 1, 2 and 3 were the first three 0.5-liter fractions from the top of the bed. Sample 4 is a subsample from the rest of the bed, which was homogenized. Samples were dried and measured in 0.5-liter Marinell containers with an n-type HPGe detector (Table 10).

Table 10. Mass and activity measurements for different 0.5 liter fractions of a used GAC batch.

Sample	Mass, kg	Density, kg/l	^{210}Pb, kBq/kg	^{226}Ra Bq/kg
1	0.367	0.73	32	260
2	0.366	0.73	27	346
3	0.272	0.54	29	685
4	0.313	0.63	6.4	341
Total	8.2	0.58	9.1	342

Calibration error was ± 10% because of the low gamma energy (46.5 keV) of ^{210}Pb. The results show that most of the ^{210}Pb produced concentrates in the top layers of the GAC bed whilst ^{226}Ra is dispersed to the larger bed volume. The first two fractions showed about a 14% weight-gain, which is likely due to retention of iron and organic substances on the GAC.

The accumulated activity of ^{210}Pb was thus determined to be 75 kBq, which is in accordance with the value calculated based on the radon concentration and flow rate. The activity mass-concentration of the used GAC batch is 9 Bq/g. When GAC filters are in operation for longer periods, they accumulate much more ^{210}Pb. The total activity of ^{226}Ra on the GAC was 2.8 kBq.

Conclusions

During this study, experiences with radon removal using GAC filtration were good. Radon removal efficiency was very high at most test locations. Removal efficiency did not degrade because of high concentrations of iron, manganese, or humus. The results suggest that uranium can cause decreased radon retention. However, this problem can be solved by installing an anion exchange unit before the GAC. Retention of other uranium series radionuclides varies.

Water quality was not impaired. Some iron and organic substances could be removed, which improved the water quality. According to the results, the hygienic quality of treated water was satisfactory; no enhanced bacteria levels could be discovered.

The filters installed were technically reliable. No clogging or malfunctioning was reported. The filters were installed such, that the residents were not exposed to gamma radiation in excess of the annual average radiation dose.

Radiation and Nuclear Safety Authority (Finland) will prepare guidelines for the safe installation and daily use of GAC filters as well as proper disposal of spent carbon. Generally, the effective dose caused by the direct radiation

originating from a GAC filter should not exceed 0.1 mSv per year. The effective dose rate at one meter distance should be less than 1 µSv/h.

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4 Prevention measures against radiation exposure to radon in well waters: analysis of the present situation in Finland

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Abstract

Naturally occurring radioactive elements are found in all groundwaters, especially in bedrock waters. Exposure to these radioactive elements increases the risk of cancer. The most significant of these elements is radon which, as a gas, is mobile and dissolves in groundwater. In Finland, water supply plants are obliged to carry out statutory monitoring of the water quality, including radon. Monitoring of private wells, however, is often neglected. In this paper, we outline the problem by reviewing the outcomes of the studies conducted in Finland since the 1960s. We also summarise the development of legislation, regulations and political decisions made so far that have affected the amount of public exposure to radon in drinking water. A review of the studies on radon removal techniques is provided, together with newly obtained results. New data on the transfer of radon from water into indoor air are presented. The new assessments also take into account the expanding use of domestic radionuclide removal units by Finnish households.

Keywords

groundwater, radioactivity, radon, public communication, water policy, water treatment

Abbreviations and notations

Bq	Becquerel. Unit of radioactivity, equal to one nuclear transformation per second.
Bq/l	Becquerel per litre.
GAC	Granular activated carbon.
Sv	Sievert. Unit of equivalent dose or effective dose equal to Jkg^{-1}.
man Sv	man Sievert. Unit of collective effective dose, equal to the sum of effective doses of individuals among a group.

Introduction

In a modern society, access to high-quality household water and appropriate sewerage and waste water treatment is a necessity that affects the everyday life of its citizens and their means of livelihood, thus making it a service of general interest. A variety of contaminants, such as micro-organisms, organic and inorganic compounds, and radionuclides can occur in household water and thus lead to health risks unless water reserves, water treatment methods and the materials of water mains and pipes are selected and maintained appropriately. Surveillance of water quality and proper waste water treatment are especially challenging in sparsely populated rural areas where water distribution networks have not been built and people have to pump their household water from private wells.

Naturally occurring uranium has been present in terrestrial matter since the formation of the Earth. Uranium is widely distributed in the Earth's crust and can be found at low levels in all rocks, soils and water. As uranium decays it forms a successive chain of decaying radioactive elements known as the uranium series (Figure 1). The bedrock of Finland is largely composed of igneous and metamorphic rocks, more specifically granite as well as granitoid, migmatite and gneiss complexes (Korkka-Niemi 2001). Rapakivi granite and granite, which make up large areas of bedrock, especially in southern Finland, contain high concentrations of uranium. Therefore, bedrock water in these areas may also contain high concentrations of other radionuclides found in the uranium series (Lahermo & Juntunen 1991). In terms of radiation dose, the most significant of these radionuclides is radon-222, which accounts for 71% of the effective dose received by users of drilled wells and 64% of that received by users of dug wells (Table 1). Apart from polonium-210, which accounts for 11% of the effective dose among users of drilled well water, doses from the rest of the nuclides are generally low (Vesterbacka *et al.* 2005, 2006).

In this paper we focus on radon, firstly, because in terms of dose it is the most significant of the naturally occurring radionuclides in drinking water. Secondly, new data are now available on the number of users of private wells and on the transfer of radon from water to indoor air in an average Finnish house. With these new data we are able to assess doses and cancer risks associated with use of well water in more detail. An average detriment from the inhalation of radon released from water was found about equal to that from ingestion. Here, we also summarise the development of legislation, regulations and political decisions made so far that have affected the amount of public exposure to radon in drinking water. A review of the outcomes of removal studies is provided, together with newly obtained results. The new dose assessments also take into account the expanding use of domestic radon removal units by Finnish households.

Uranium-238 → Thorium-234 → Protactinium-234 m → Uranium-234 → Thorium-230 → Radium-226 → Radon-222 → Polonium-218 → Lead-214 → Bismuth-214 → Polonium-214 → Lead-210 → Bismuth-210 → Polonium-210 → Lead-206 (stabile)

Figure 1. The uranium series.

Table 1. Mean concentrations of naturally occurring radionuclides in water from Finnish drilled wells and dug wells and assessed mean annual effective doses, accordingly (Vesterbacka *et al.* 2005, 2006).

Radionuclide	Drilled wells		Dug wells	
	Mean concentration (Bq/l)	Mean effective dose (mSv/a)	Mean concentration (Bq/l)	Mean effective dose (mSv/a)
Rn-222	460	0.29	50	0.032
U-238	0.26	0.008	0.015	0.0005
U-234	0.35	0.014	0.020	0.0008
Ra-228 *	0.030	0.017		
Ra-226	0.050	0.010	0.016	0.003
Pb-210	0.040	0.022	0.013	0.007
Po-210	0.048	0.046	0.007	0.007

* From Vesterbacka *et al.* 2006.

Surveying radon in drinking water

Systematic surveys of the occurrence of radon in Finnish groundwaters began at the end of the 1960s. In the first phase, 66 bedrock water samples from drilled wells in the Helsinki area were surveyed (Kahlos & Asikainen 1973). Radon concentrations were generally high, with a mean of 1,600 Bq/l, which prompted the surveying of waterborne radon throughout Finland. By the end of 1978, in total 735 samples of tap water from public water supply plants using both surface and groundwaters, 690 samples from dug wells and springs and 878 samples from drilled wells had been measured. The mean radon concentrations in groundwater wells and bedrock water wells were 60 and 630 Bq/l, respectively (Asikainen & Kahlos 1980).

After the end of the 1970s, the surveys were mainly directed at private wells, although all new groundwater sources were assessed before they were connected to the water distribution networks. Bedrock wells were particularly studied in the risk areas where concentrations of radon, uranium or other natural radionuclides had been found high (Salonen 1988). Municipal health inspectors had an important role in surveying radon in private wells: water

samples were measured at regional laboratories and sent to the Radiation and Nuclear Safety Authority (STUK) for more detailed analysis if the measured radon concentration exceeded 300 Bq/l. Private wells were also investigated in co-operation with Geological Survey of Finland and the Finnish Environment Institute, which performed nationwide studies on the physico-chemical and microbiological quality of well waters (Juntunen 1991).

In a more recent population-based random study, in which 184 water samples from dug wells and 288 from drilled wells were analysed, the observed mean concentrations of radon were 50 and 460 Bq/l, respectively (Vesterbacka *et al.* 2005). In dug wells, all measured radon concentrations were below 1,000 Bq/l, but 10.8% of drilled wells exceeded this level. The availability of water services has steadily increased during the past three decades, especially in the more densely populated southern Finland, where the highest radon concentrations are also generally found. Today, over 93% of people in southern Finland have access to water distribution services, which could explain the lower mean obtained in the most recent study.

At present, STUK's database contains information on more than 7,600 drilled wells and 3,800 dug wells. Over 50,000 results from the analysis of radon, uranium, radium-226, radium-228, lead-210 and polonium-210 have been recorded. The basic data on the wells includes parameters such as well type, commissioning year, location, depth, purpose of use, annual duration of use, number of users and installed water treatment units. The database is very useful in planning new research projects dealing with topics such as epidemiology, radiation hygiene or water treatment methods.

Finland presently has a population of 5.3 million, about 4.8 million of whom have access to water distribution services (Isomäki *et al.* 2007). All water distributors that service water to more than 50 people are recorded in a national database and all water supply plants and beverage companies are required to comply with statutory monitoring of water quality (Lapinlampi & Raassina 2002). About 500,000 people use private wells or have formed small local co-operatives for abstracting groundwater. These people typically live in one-family houses in sparsely populated rural areas where municipal water networks are still unavailable, or they are often impracticable to build owing to the long distances to water mains and low end-point consumption. The number of drilled wells has steadily increased. In the 1950s, one well out of 1,000 was a drilled well, but by the early 1990s the portion had increased to 23% (Korkka-Niemi 2001). According to a survey conducted in 2007, the portion of drilled wells was already 32.5% (Vienonen 2007). At the end of 2006, 996,000 households with a total of 2.65 million residents were living in one-family houses (Statistic Finland 2007*a*). The average number of persons living in one-family houses is thus 2.66.

From these data, we can estimate that about 160,000 people permanently use water from drilled wells and that the number of drilled wells in permanent use is about 61,000. The number of summer cottages is presently 475,000, and in nearly half of them water is derived from a private well (Statistics Finland 2007*b*). These dwellings, however, are used only during the vacations and weekends, and the usage of well water is thus typically limited to less than two months per year.

Legislation

The preparation of legislation concerning water resources and steering of its implementation are divided between three ministries in Finland: the Ministry of Social Affairs and Health is responsible for the health aspects of drinking water, the Ministry of Environment for waste water discharges and the Ministry of Agriculture for the management of water resources. According to the Radiation Act (592/1991), STUK issues general instructions, known as Radiation Safety Guides (ST Guides), that regulate the use of radiation and operations involving radiation.

Regulations concerning radon in drinking water

In order to reduce the harmful effects emerging from radioactivity in drinking water, legislation covering both water supply plants and private wells has been passed on national and European Union levels. In Finland, the Radiation Safety Guide ST 12.3, Radioactivity of Household Water, was passed in 1993 by STUK. It concerns water supply plants and manufacturers of bottled beverages using their own water supply. The objective of Guide ST 12.3 is to limit the effective dose from radionuclides in drinking water to less than 0.5 mSv per year, and its implementation is assessed by an activity index that is calculated from activity concentration measurements of radon, gross alpha and gross beta activity. If the index exceeds a reference value, a more detailed analysis of radionuclides is required. The EU Drinking Water Directive (Council Directive 98/83/EC) introduced another screening parameter, the total indicative dose (TID), by which the effective dose from drinking water was limited to 0.1 mSv per year. The TID corresponds to the annual dose received from all the natural and artificial radionuclides in water except for tritium, potassium-40 and radon, along with its decay products. Its monitoring frequencies were to be set later. However, this work is not finished as of November 2009, and the Commission is currently preparing a revision of the directive. According to recommendation 2001/928/Euratom by the European Commission, the radon concentration in water from water supply plants shall not exceed 100 Bq/l but “a level higher

than 100 Bq/l may be adopted if national surveys show that this is necessary for implementing a practical radon programme. For concentrations in excess of 1,000 Bq/l, remedial action is deemed to be justified." Moreover, an action level of 1,000 Bq/l shall be used for private wells (European Commission 2001).

Decree 401/2001 by the Ministry of Social Affairs and Health recommends water supply plants distributing drinking water in a lesser amount than 10 m^3 per day or to less than 50 persons to limit radon in drinking water below 300 Bq/l. For private wells the recommended maximum concentration is 1,000 Bq/l. The municipal health authority can require an investigation of a private well in the case of a suspected health risk. The municipal health authority is obliged to ensure that households and companies relying on their own water supply receive enough information on the quality and the associated health risks of the local drinking water and on the possibilities of mitigating those risks.

Radon is, thus, included in regulations that concern drinking water from water supply plants and beverage manufacturers, but also water from private wells. Private well owners are under the supervision of municipal health authorities and, as will be shown in the following sections, this supervision still requires elaboration.

Legislation concerning organizing water services

The Act on Water Services 2001/119 aims to ensure water services for Finnish citizens by obliging municipalities to draw up sufficient development plans for their territories in co-operation with the water supply plants. When required due to health considerations, a municipality must ensure that appropriate measures are taken to establish a water supply plant to meet the needs, to expand the area of operation or to otherwise secure the availability of sufficient water services. In addition, properties in operational areas of water supply plants must be connected to the existing water distribution network and sewer system. This has expedited the expansion of water distribution networks and the use of their water. Starting from 2005, municipalities and home-owners are entitled to apply for government subsidies to organise water services on the basis of Government Act 686/2004. The amount of the subsidy is up to 50% if the work is carried out by a private or municipal constructor and up to 75% if the government commissions the work. Subsidies can be paid to home-owners for joining water distribution networks, but they may also be given for acquiring water service equipment intended for removing radionuclides from drinking water.

Government Decree 542/2003 on Treating Domestic Wastewater in Areas outside Sewer Networks sets strict requirements for domestic waste water systems that should be met before 2014. According to the Government

Programme of Prime Minister Matti Vanhanen's second Cabinet (Prime Minister's Office 2007), the government will channel additional funds to finance water projects in rural areas in an attempt to connect sparsely populated areas to water distribution networks and sewer systems, and involve them in integrated waste management projects in order to comply with Government Decree 542/2003. The recent political decision-making thus promotes the joining of water distribution networks, which will most likely reduce the number of households dependent on private wells in the future.

Finnish housing

More than half of Finns (56%) have aspirations towards living in a privately owned one-family house. The majority of Finns prefer housing in sparsely built-up areas and only a quarter desire to live in cities, towns or municipal centres. Families with children, in particular, appreciate peaceful neighbourhoods; about 40% of them wish to live in the countryside (Statistics Finland 2007*c*). During the past two decades, the proportion of Finns living in one-family houses has remained at 50%. Due to the declining number of persons per household, the absolute number of one-family houses has increased from 820,000 to nearly one million. During the first decade of the twenty-first century, the number of new planning permissions for one-family houses per year has steadily increased. New one-family houses are being built especially in rural areas, since the real estate prices in cities are high, and in the greater Helsinki area over 100% higher than in the rest of the country (Statistics Finland 2008*a*). Not all sites with planning permission have water mains in the area.

Most one-family houses are owned by the residents. The inclination of Finns towards owning their homes can be explained by the generally acknowledged fact that in Finland, owner-occupied housing has the lowest housing costs in the long term. This is partly due to Finnish fiscal legislation, which allows 28–30% tax deductions on mortgage interest. People aged 18–39 years buying their first home are exempted from the 1.6–4% transfer tax (Ministry of Finance 2005). It is also possible to apply for collateral security from the state up to 20% of the loan. In 2006, more than 70% of Finns lived in owner-occupied houses.

Radioactivity measurement services for drinking water

There are about 40 regional laboratories committed to measuring radioactivity in foodstuffs in the case of a fallout situation according to STUK's instructions. STUK provides these laboratories with similar NaI-type gamma spectrometers and organises training and validation for the measurements. Most of these

laboratories also use the spectrometers for measuring radon in drinking water. If the measured radon concentration exceeds 1,000 Bq/l, the regional laboratories advise the customers to send a water sample to STUK for more comprehensive analysis. Well-owners or municipal health officers can also send water samples directly to STUK for analysis.

If the second measurement confirms the radon concentration to be higher than 1,000 Bq/l, the customer is advised to consider remedial action. The principle alternative is to join a water distribution network. This, however, is not always possible, and purchasing a radon removal unit may therefore remain the only viable option. Before buying a treatment unit, the occurrence of other natural radionuclides is screened by gross alpha and gross beta activity measurement and analysis of certain water quality parameters is recommended. Sometimes, water contains primary (health) contaminants, e.g. uranium, fluoride or arsenic, or secondary (technical-aesthetic) contaminants such as iron, manganese or humic substances that should be removed together with radon.

Health effects of radon

Both ingestion and inhalation of radon increases the cancer risk. For individual tissues or organs the dosimetric quantity used is the equivalent dose, and for the whole body the effective dose, which is the sum of the weighted equivalent doses for all organs and tissues. The unit of both is the sievert (Sv). The collective effective dose describes the sum of effective doses among a group of individuals and its unit is the man sievert (man Sv). The cancer risk for a long-term exposure of one sievert effective dose has been estimated at 5.5% for the whole population and 4.1% for adult workers (ICRP 2007). Stochastically, this would mean that if a person is exposed to an effective dose of one sievert during a long period of time, the individual risk of cancer is increased by 5.5 percentage points. If the same dose is distributed among a hundred people, the risk that one of them gets cancer would still be increased by 5.5 percentage points. For an individual, the increase would then be only 0.055 percentage points. There are, however, limitations when using these radiation-related cancer risk coefficients, which are based on epidemiological data on medium and high doses. Large uncertainties may be related to their use in the low dose range, and the calculation of radiation-related cancer risk from a collective dose that is comprised of small individual doses or doses ranging over several orders of magnitude is not justified. For collective effective doses that are smaller than the reciprocal of the risk coefficient (i.e. 18 man Sv), the number of excess cancer cases is most probably zero (NCRP 1995).

Ingested radon

The National Research Council has evaluated risks from ingested radon and has assessed equivalent doses for different organs and tissues (NRC 1999). The highest equivalent dose is caused to the stomach walls, and about 80% of the cancers related to ingested radon are estimated to be stomach cancers. The effective dose per unit intake of ingested radon is estimated to be $3.5 \cdot 10^{-9}$ Sv/Bq for adults. The dose per unit intake increases with decreasing age: for five-year-old children the factor is estimated to be $1.0 \cdot 10^{-8}$ Sv/Bq and for infants $4.0 \cdot 10^{-8}$ Sv/Bq.

In order to assess the doses from the intake of well water we must first know the amount of well water consumed. Water used for preparing food or hot beverages contains no radon, since radon is released from water when boiled (Abulfaraj & Mamoon 1995). Drinking water consumption has been investigated among the adult Finnish population in the age groups 25–34, 35–44, 45–54, 55–64 and 65–74 years. The average daily intakes among female and male populations representing these age groups were 0.667–0.929 and 0.441–0.676 litres, respectively (Paturi *et al.* 2008). No data are available on water intake among children. The closest country from which these data are available is Germany. There, water intake (mineral water and tap water) among age groups 2–3 years and 4–8 years was 0.175 and 0.215 litres per day, respectively (Sichert-Hellert *et al.* 2001). In the age group 9–13 years, boys consumed 0.344 and girls 0.298 litres per day. Packaged waters normally have long storage times and, owing to the relatively short half-life of radon, contain little radon. Until now, the consumption of packaged water has been very low in Finland: in 2004 the per capita consumption was only 14.1 litres per year (Gleick 2006).

Naturally, not all drinking water is consumed at home: water is also consumed at day-care centres, schools, work places, cafeterias and other places. The proportion of children attending day care is higher in older age groups, being 36.6% on average among 0- to 6-year-olds. The average number of days in day care is 175 per year (STAKES 2008). All children aged between 7 and 15 attend school and the mean number of school days per year is 188. The average number of days at work per year is 220 for those who are employed. The proportion of pensioned or unemployed people increases with age (Statistics Finland 2008*b*). Among the elderly, days spent in nursing homes or hospitals increases with age: 5 day per year among the age group 65–74 years, 26 among 75- to 84-year-olds and 97 among people older than 85 years (STAKES 2008). People also spend time travelling and at summer houses. People from rural municipalities travel about 19% less than the national average and families with children make about 20% more domestic trips than the average population.

Travelling is most common among the age group 25–34 years, being 29 nights per year spent outside the home (Statistics Finland 2007*d*).

We assumed that two-thirds of water is consumed at home on day-care/school/work days, and no well water is consumed on days spent travelling and at nursing homes or hospitals. In addition, data on the missing age groups were interpolated or extrapolated when assessing the water intake among different age groups (Tables 2 and 3). The age-weighted mean effective doses received from the ingestion of drilled well water by females and males were calculated as 0.35 and 0.26 mSv/year, respectively. Respective effective doses received by users of dug well water are 0.04 and 0.03 mSv/year, respectively. Among users of water where radon exceeds 1,000 Bq/l, the mean radon concentration is 2,700 Bq/l and the effective doses from ingestion for female and male users are 2.0 and 1.5 mSv/a.

Next, we assess the detriments related to ingested radon for different users of drilled wells. Life expectancy at birth of female and male Finns is 75.8 and 82.8 years, respectively. If a person uses drilled well water that contains 2,700 Bq/l of radon throughout life, the risk of having cancer is 0.9 and 0.6% among female and male users, respectively. It is, however, rare that a person lives in the same house for his or her entire life. According to Strandell (2005), people from the age group 20–29 years live in one-family houses substantially less often than those from other age groups. This is easily explained by young adults moving away from their childhood home and studying in towns or cities. As mentioned above, families with children generally value living in the countryside and buy their house when the parents are around 30 years old. The mean age of women at first birth was 28.0 in 2006 (STAKES 2007). Therefore, we also present estimates of detriments among age groups 0–18 and 30–82/75 years (Table 4).

We can estimate that the collective effective dose is 31 man Sv annually among users of drilled well water in which radon exceeds 1,000 Bq/l. This translates into approximately two cancers attributable to ingested radon. For the users of drilled well water in which the radon concentration is lower that 1,000 Bq/l, the collective effective dose is roughly 19 man Sv per year and among users of dug wells 11 man Sv, so radiation-related health effects among these groups should theoretically be even less.

In the latest survey, only 30 water samples exceeded the guideline value of 1,000 Bq/l. Extreme concentrations of radon in well water are, however, occasionally found. In summer 2007, two well waters containing 62,000 and 130,000 Bq/l of radon were measured. The latter family had been using the water regularly for a period of 10 years. The effective dose from ingested radon to the five family members was about 1 Sv each, so it can be estimated that the

Table 2. Assessed water intake among the Finnish female population.

Age (years)	No.$_{female}$ ($\times 10^3$)	Water intake (l/d)	Day-care/school/work (d/y)	Hospital/nursing home/travelling (d/y)	Intake of well water (l/y)
0	29	0.025	13	20	9
1	28	0.120	54	20	39
2–3	56	0.175	87	20	55
4–8	139	0.215	154	20	63
9–13	155	0.298	188	20	84
14–17	130	0.639	203	22	174
18–24	224	0.929	206	23	254
25–34	315	0.929	202	24	255
35–44	350	0.850	198	21	236
45–54	382	0.709	190	20	200
55–64	365	0.755	126	19	230
65–74	252	0.667	6	20	229
75–84	195	0.667	0	37	219
85–	71	0.667	0	103	175

Table 3. Assessed water intake among the Finnish male population.

Age (years)	No.$_{male}$ ($\times 10^3$)	Water intake (l/d)	Day-care/school/work (d/y)	Hospital/nursing home/travelling (d/y)	Intake of well water (l/y)
0	30	0.025	13	20	9
1	29	0.120	54	20	39
2–3	59	0.175	87	20	55
4–8	146	0.215	154	20	63
9–13	161	0.344	188	20	97
14–17	136	0.518	203	22	150
18–24	235	0.633	206	23	185
25–34	331	0.633	202	24	185
35–44	362	0.536	198	21	164
45–54	385	0.448	190	20	160
55–64	358	0.429	126	19	142
65–74	213	0.429	6	20	151
75–84	115	0.429	0	37	145
85–	23	0.429	0	103	115

risk of one of the family members getting cancer attributable to ingested radon is about 30%. Presently, 129 wells with water exceeding 10,000 Bq/l have been found in Finland.

Table 4. Mean detriments related to ingested radon among different well water users groups.

Group		Detriment, whole life	Detriment, age 0–18	Detriment, age 30–*
Female	Drilled wells	0.16%	0.03%	0.11%
	Drilled wells (Rn >1000 Bq/l)	0.92%	0.17%	0.61%
	Dug wells	0.02%	0.003%	0.01%
Male	Drilled wells	0.11%	0.03%	0.07%
	Drilled wells (Rn >1000 Bq/l)	0.64%	0.16%	0.37%
	Dug wells	0.01%	0.003%	0.007%

* 30– refers to females aged 30–82 years and males aged 30–75 years.

Radon released into indoor air

Normally, the most significant source of radon in indoor air is the soil beneath the house. In Finnish one-family houses, the average radon concentration in indoor air is 145 Bq/m^3 (Arvela *et al.* 1993). Other sources include building materials and household water. Because radon is a gas and has a solubility in water similar to carbon dioxide, it is partly released into indoor air during water usage. The proportion of radon released from water during certain types of water usage (e.g. shower, toilet) is expressed as a transfer coefficient. By measuring indoor-air volume of the house V; the air-exchange rate of indoor air λ; the use-weighted transfer coefficient e; and the water consumption rate W, we can calculate the transfer factor $f = We/V\lambda$, by which the amount of waterborne radon released to indoor-air can be assessed (Nazaroff *et al*. 1987). Water usage by Finnish households has been well investigated and the average water usage in one-family houses has been defined as 125 litres per person per day. The largest share of the water is used for showers (39%), drinking and cleaning (27%) and the toilet (14%) (Etelämäki 1999). From transfer coefficients reported by Nazaroff *et al*. (1987), we can estimate that the use-weighted mean transfer efficiency is 0.57. The average living area of Finnish one-family houses is 134 m^2, and assuming a room height of 2.5 m we can estimate the average volume to be 340 m^3 (Statistic Finland 2007*d*). The mean air exchange rate in Finnish one-family houses is 0.64 (Ruotsalainen *et al*. 1992). Using these data we come to a value $0.36 \cdot 10^{-4}$, i.e. 1,000 Bq/l of radon in water increases the indoor air radon concentration by 36 Bq/m^3. It must be noted that large variations in the transfer factor occur because the house dimensions and water consumption rates are highly case-specific.

To validate this value, 268 houses where radon in both drilled well water and indoor air had been measured were randomly selected from STUK's indoor air database. The regression analysis performed on these data indicated that the baseline value for indoor air was 210 Bq/m^3 and the transfer factor was $0.36 \cdot 10^{-4}$

(R^2 = 0.35). By setting the baseline value to the average radon concentration of indoor air in one-family houses (145 Bq/m^3), we obtain a value $0.39 \cdot 10^{-4}$ (R^2 = 0.33) for the transfer factor.

By applying the transfer factor of $0.39 \cdot 10^{-4}$ we can estimate that the additional radon concentration in indoor air is 2 Bq/m^3 in houses with dug wells and 18 Bq/m^3 in houses with drilled wells, which are small values compared to the average indoor air concentrations in Finland. The average additional indoor air radon concentration in houses where the recommended maximum value for radon is exceeded is 100 Bq/m^3.

Breathing radon and its progeny causes an equivalent dose almost exclusively to the lungs, and the prominent detriment is thus lung cancer. The dose is mostly comprised of short-lived radon progeny consisting of metal ions or atoms, which can cluster and attach to indoor air particles. The aerodynamic diameter of these products determines where in the respiratory system they are deposited. In addition to particle size distribution, the ratio of radon progeny to radon (equilibrium factor) also varies. Assessed effective dose conversion factors for radon in the indoor air of homes range from 6×10^{-9} to 15×10^{-9} Sv/(Bq h m^{-3}) (UNSCEAR, 2000). STUK has chosen to apply a value of 6×10^{-9} Sv/(Bq h m^{-3}) with an equilibrium factor of 0.4, as is recommended in ICRP65 (ICRP 1994). Hence, we can estimate that the average effective dose from radon released from water is 0.03 mSv/year among users of dug wells and 0.30 mSv/year among users of drilled wells. Among the users of drilled wells where radon exceeds 1,000 Bq/l the mean effective dose can be estimated as 1.8 mSv/year.

According to a collaborative analysis of data from 13 European case-control studies, the risk of lung cancer increases by 16% per 100 Bq/m^3 of radon in indoor air of homes (Darby *et al.* 2005). The lung cancer incidence in Finland is about 4.2 per 10,000 citizens per year (Finnish Cancer Registry 2007). From these data we can assess that among users of dug wells, lung cancer cases attributable to water usage are likely to be less than one case per year. Among users of drilled wells, about two lung cancers are annually attributable to radon in water.

Summing up the different exposure pathways, we can estimate that radon in private wells causes only a few cancer cases per year in Finland. The main target group for radiation protection is those whose water contains radon in excess of 1,000 Bq/l, which is the recommended maximum concentration for private wells. Among this group, the theoretical number of radon-related cancers is only three per year. Epidemiological studies also support the assessment made above (Auvinen *et al.* 2005; Kurttio *et al.* 2006). In these studies, the risks of stomach, kidney and bladder cancers and leukaemia from the radionuclides ingested with drinking water were assessed using a case-cohort method.

The results did not indicate increased risks of any of these four cancers at the exposure levels of the epidemiological study.

Methods for removing radon from drinking water

Research on radon removal from water supplies was initiated in the 1970s in Czechoslovakia (Hanslik *et al.* 1978). Aeration was found to be a suitable method for stripping radon gas out of water. A removal efficiency of 99% was recorded for 8 minutes aeration time and an air-to-water ratio of 1 : 8. In the early 1980s, different aeration techniques and activated carbon adsorption were tested in Sweden. Aeration under atmospheric pressure was reported to be a viable method, with a removal efficiency of up to 75% (Hedberg *et al*. 1982). In the USA, studies were also begun in the early 1980s (Lowry 1983). Three methods were tested and found effective: granular activated carbon (GAC) adsorption, diffused aeration and spray aeration. Initially, GAC adsorption was considered the most auspicious method due to its effectiveness and low investment and maintenance costs. After a few years the viability of GAC adsorption was re-evaluated due to external gamma radiation caused by the units and waste problems emerging from spent GAC batches (Rydell *et al*. 1989).

Removal studies in Finland began in the early 1990s by first reviewing the literature on the viable treatment methods (Jokela 1993). Experimental studies were initiated a couple of years later in collaboration between STUK, the Finnish Environment Institute and Helsinki University of Technology. From the first project onwards, the main objective has been to bring suitable and reliable treatment units to the market so that home-owners can easily purchase them. Therefore, companies specializing in water treatment have been invited to participate in the research projects (Myllymäki 1996; Myllymäki *et al.* 1999).

In 1997, a new research project was initiated within the 4th Framework Programme financed by the European Commission. Seven institutions from four member countries participated in a project in which removal of the most commonly occurring natural radionuclides (radon, uranium, radium-226, lead-210 and polonium-210) from groundwaters was investigated (Annanmäki & Turtiainen 2000). Water works were also included in the research (Salonen *et al.* 2002). After the project, dozens of removal units were in use in Finnish homes. In order to obtain long-term experiences of their operation, a follow-up project was initiated and the units were monitored until the end of 2002 (Vesterbacka *et al.* 2003). At this time, a few hundred units had been installed and problems associated in their use had been recognized. The companies involved in the projects had gained expertise and most of the consumer guidance can now be channelled there.

Granular activated carbon adsorption

As a non-polar monatomic gas, radon is effectively adsorbed on activated carbon. Granular activated carbon filters sold in Finland are typically pressure vessels with carbon bed volumes of 39–105 litres. An automatic backwash system is employed in cases where the influent contains large amounts of iron, manganese or humic substances. These filters are always installed to treat all household water so that exposure to radon through inhalation is also prevented. The units operate passively under normal plumbing pressure (2–6 bar) and therefore do not require additional pumps. This keeps the acquisition price of the unit low compared to the aeration technique (Turtiainen *et al.* 2000*a*).

The amount of radon accumulated in the unit depends on the water usage and the radon concentration in raw water. The short-lived progeny of radon that emit gamma radiation are also retained on the carbon bed, which thus becomes a source of external gamma radiation. Therefore, GAC filters cannot be installed inside residential buildings and their use should be limited to radon concentrations below 5,000 Bq/l (Annanmäki *et al.* 2000).

The radon removal efficiency of GAC filters was followed at ten households for a period of 3 to 9 years. The carbon bed was replaced with a fresh batch in three locations during the follow-up. In the selection of the households, different types of water were covered, including iron- and manganese-rich water and water with a high content of organic carbon. Radon concentrations in raw water were 1,500–7,400 Bq/l (Vesterbacka *et al.* 2003). All GAC filters removed more that 90% of radon, and most of them nearly 100%. Some units showed a decline in removal efficiency over time, and hence to ensure an adequate removal efficiency it was recommended that the carbon batches be replaced every three years. Water quality, both microbiological and chemical, remained good. Iron and organic substances were partly removed by GAC filtration (Turtiainen *et al.* 2000*b*).

Some equipment designed for iron and manganese removal can be partly filled with activated carbon and thus simultaneously removes iron, manganese and radon (Vesterbacka *et al.* 2003). Iron and manganese removal units that are based on aeration-oxidation normally do not produce a sufficient amount of air to effectively strip radon and hence cannot be recommended for radon removal (Vesterbacka & Salonen 2008).

Aeration

As a dissolved gas, radon can be removed from water by aeration. Three alternative principles are customarily employed in aeration: a large number of air bubbles are produced in the water (diffused aeration); water is sprayed into air as small droplets (spray aeration); or a large surface is created between

water and air on an inert material (tower aeration) (Annanmäki & Turtiainen 2000). Domestic aeration units typically employ a combination of the first two principles and the aeration time needed is 4–10 minutes. Most commercial systems work under atmospheric pressure and hence a water pump and a pressure tank are needed after the aeration unit to give the water enough pressure to deliver it throughout the plumbing (Mjönes 2000). Water flow from a well though an aeration unit into a pressure tank is electronically controlled. The cost of acquiring aeration units is two to three times as high as that of a GAC filter. This difference, however, will be partly compensated by the higher operating costs due to changing the carbon batch every three years (Mäkeläinen & Turtiainen 2003). Low-cost alternatives where aeration takes place in the bore hole have also been introduced (Vesterbacka *et al.* 2003).

In total, aeration units from eight manufacturers have been tested. The lowest radon removal has been recorded for aeration in the bore hole, where efficiencies varied from 3% to 77% (average efficiency during 100 litres flow). More sophisticated aeration units, however, were all able to remove more than 90% of radon. Presently, four brands are available on the Finnish market, all with adequate radon removal efficiency.

Customer insights on radon removal units

A survey among customers who have bought radon removal units was conducted with a questionnaire (Vesterbacka *et al.* 2003). Activated carbon adsorption was the most prevalent technique of radon removal, with a 70% share. Only 7% reported defects or inconveniences, the most common being difficulty in replacing the carbon batch. By contrast, more than half of the users of aerator units reported defects or inconveniences. Malfunction in solenoid valves was the most common complaint.

Another survey was conducted in 2008 by a telephone interview. All well-owners who had learned that the radon concentration in their well water exceeded 1,000 Bq/l (53 families) were asked what measures they had taken to reduce the concentration, if any. Already, 32 families had reacted: 19 of them had purchased activated carbon filters, ten families had joined or were joining public distribution networks and three families had acquired aerators. Twelve families were still contemplating the options, while nine families did not intend to react. The latter had radon concentrations ranging from 1,000–4,000 Bq/l. This survey implies that most people are ready to take measures against radon in well water when they receive information.

This is also shown when considering the number of removal units sold. Companies were interviewed and asked how many radon removal units that they had sold. As of in March 2007, this number was about 1,000. According to

the latest survey, the number of wells where radon exceeds 1,000 Bq/l is about 6,600, which means that measures against radon have been taken for about 15% of the wells with elevated radon concentrations.

Effect of radon removal units on the collective effective dose

Assuming an average removal efficiency of 95% for the removal units, the effective dose through both ingestion and inhalation of radon has been reduced by 3.4 mSv per year on average among those who have purchased them. Expressed as collective effective dose, the reduction is about 9 man Sv per year. Two uncertainties relating to the assessment reversely affect the amplitude of averted doses: the number of households that have connected to water networks is not known and thus the amplitude of averted doses may be higher. However, a proportion of the removal equipment is used at summer houses, which would suggest that averted doses estimated above may be smaller. If the remaining households that still use water with an elevated radon concentration would take measures to reduce radon, the averted collective effective dose could be about 50 man Sv annually.

Public communication

As discussed earlier, decree 401/2001 by the Ministry of Social Affairs and Health obliges municipal health authorities to ensure that households relying on their own water supply receive enough information on the quality and the associated health risks of the local drinking water and on the possibilities of mitigating those risks. Far too often, municipal health authorities have failed to do this, as was the case with two high radon concentrations found in summer 2007. In both these cases the residents had not been informed by the municipality about the associated health risk, but had taken the initiative themselves to have their water measured after receiving the information elsewhere. As was shown, the collective effective doses and the associated number of fatal cancers are small, but regarding the ALARA principle of limiting radiation exposure to "as low as reasonably achievable", counter-measures must be taken in cases where the radon concentration of an individual's drinking water is high.

During the past decade, STUK has organised courses for municipal health authorities and the personnel of water supply plants on the health risks of radon and removal methods for drinking waters. These have usually been held two or three times per year, and there have generally been a few dozen attendees at the lectures. Considering that there are 415 municipalities in Finland with occasionally changing personnel, the training has therefore been insufficient.

For this reason, STUK sent letters in May 2008 to 67 municipalities where more than 20% of investigated drilled wells contain radon above 1,000 Bq/l. The letters, containing information on the occurrence, measurement and removal of radon, were addressed to health inspectors. Since Finland has two official languages, all materials and information were provided in both Finnish and Swedish. A few days later a press release was published and the list of the municipalities along with statistical information on wells was provided on STUK's Internet site. This press release was reported in several regional newspapers and on radio stations, which resulted in increased measurement activity in these radon-prone areas.

Statistically, the number of cancer cases related to the consumption of water from wells is only a few per year. However, we feel that Finnish citizens have the right to be informed about this risk, and this right is also implemented in national legislation. Radiation is a sensitive issue for many families, especially those with children. Therefore, it is important that the work to locate the remaining 5,000 to 6,000 wells with elevated radon concentrations continues.

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5 Activity concentrations of ^{226}Ra and ^{228}Ra in drilled well water in Finland

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Abstract

The activity concentrations of ^{226}Ra and ^{228}Ra in drinking water were determined in water samples from 176 drilled wells. ^{226}Ra activity concentrations were in the range of < 0.01 to 1.0 Bq l^{-1} and ^{228}Ra activity concentrations in the range of < 0.03 to 0.3 Bq l^{-1}. The mean activity concentration of ^{226}Ra and ^{228}Ra were 0.041 and 0.034 Bq l^{-1}, respectively. High radium activity concentrations in drinking water were rare. Only 2–4% of the drilled wells exceeded a ^{226}Ra concentration of 0.5 Bq l^{-1} and 1–2% of the wells exceeded a ^{228}Ra concentration of 0.2 Bq l^{-1}. These are the activity concentrations that cause a 0.1 mSv annual effective dose for users of drinking water. The maximum annual effective doses from ^{226}Ra and ^{228}Ra for users of drilled wells were 0.21 mSv and 0.16 mSv, respectively. The elevated activity concentrations of ^{226}Ra and ^{228}Ra did not occur simultaneously in the same groundwaters and the correlation between ^{226}Ra and ^{228}Ra was small.

Introduction

Natural radionuclides in Finnish groundwater mainly originate from the decay series of ^{238}U [1–3]. The most harmful of these from the point of view of radiation protection is ^{222}Rn. Other alpha-active isotopes include ^{238}U, ^{234}U, ^{210}Po and ^{226}Ra. In addition, beta-active ^{210}Pb and ^{228}Ra isotopes are also found in drinking water. The isotope ^{228}Ra originates from the decay series of ^{232}Th.

Groundwater is affected by the composition of bedrock and soil. In areas with high concentrations of radionuclides in groundwater, the bedrock typically consists of granites[2–4]. Finnish groundwater has high activity concentrations of ^{238}U and ^{234}U compared to ^{226}Ra, differing significantly from those in most other European countries[3, 5–7]. The activity concentration of ^{228}Ra in Finnish groundwater has not extensively been examined. Earlier data on ^{228}Ra activities in Finnish drilled well water have been based on 130 measurements in the south of Finland.

The activity concentration of radium in groundwater is controlled by chemical and physical processes such as adsorption-desorption, complexation and precipitation-dissolution. These processes are related to the chemical composition of the groundwater. The activity concentration of ^{226}Ra in groundwater is unrelated to dissolved uranium and that of ^{228}Ra exceeds its dissolved parent ^{232}Th by several orders of magnitude[8–9]. Significant associations have been observed in groundwater between ^{226}Ra, chloride, calcium, potassium and bromine, the latter four of which are typical elements in saline groundwater[8].

The importance of ^{228}Ra and ^{226}Ra has increased due to the EU drinking water directive (DWD), which lays down for waterworks a reference dose of 0.1 mSv per year[10]. The reference dose excludes ^{3}H, ^{40}K, ^{222}Rn and ^{222}Rn progenies and thus, in practice, it includes isotopes of uranium and radium. A reference dose of 0.1 mSv per year corresponds to an activity concentration of ^{226}Ra and ^{228}Ra of 0.5 Bq l^{-1} and 0.2 Bq l^{-1}, respectively.

The goal of this study was to obtain an estimate of the activity concentration of ^{228}Ra in drilled well water and whether the two radium isotopes, ^{226}Ra and ^{228}Ra, occur in high activity concentrations in same waters. Furthermore, the radiation dose from drinking water for people living outside the public water supply in Finland was estimated. The survey was also necessary for the implementation of the DWD and the development of monitoring practices in Finland. The results will be utilised in further studies investigating the occurrence of ^{228}Ra, ^{226}Ra and other ^{238}U-series radionuclides.

Materials and methods

Selection of wells

The selected wells were the same as in a previous representative random sampling study by Vesterbacka *et al*.[3]. In that study, 2000 persons not using a public water supply as their source of drinking water in 2000 were selected for the survey from the Population Register Centre of Finland. Of the 779 private well users who consented to participate, 288 drilled wells were selected for investigation. Of these drilled wells, 167 yielded an activity concentration of ^{226}Ra higher than 0.01 Bq l^{-1} and 134 of the wells were included in the present study. From the 121 drilled wells whose activity concentration of ^{226}Ra was lower than 0.01 Bq l^{-1}, 42 were randomly selected. Thus, the final number of wells examined here was 176.

Water sampling

Samples for determination of ^{226}Ra and ^{228}Ra were collected in two 1 l polyethylene bottles from taps without abundant water flow. In the laboratory, water samples

were acidified with hydrochloric acid (4.5 ml concentrated HCl in 1 l water sample). All water samples were taken by the owner of the private well and were collected between July and August 2004.

Determination of activity concentration of radionuclides

The determination method used at STUK for analysing ^{228}Ra is accredited according to the European Standard EN ISO/IEC 17025:2000[11]. The method is validated and its analytical quality assurance includes calibration, internal quality control measurements, measurement of reference material and participation in proficiency testing or interlaboratory testing. ^{226}Ra determination is not included in the accreditation, but analytical quality has been controlled by the same methods used in the ^{228}Ra determination. The minimum detection limit (MDA) was calculated using the EPA definition[12]. Brief descriptions of ^{226}Ra and ^{228}Ra determination are given below.

The activity concentration of ^{226}Ra was determined with a 1220 Quantulus™ (PerkinElmer) low-background liquid scintillation spectrometer. The sample was prepared by evaporating off the water until totally dry with a freeze-dryer in a teflon-coated polyethylene vial (Zinsser). The residue was dissolved in a small amount of acid and then a scintillation cocktail was added. The sample was counted 1 month after preparation. During this time ^{226}Ra attains equilibrium with ^{222}Rn and its short-lived daughters. The ^{226}Ra activity concentration was calculated from the alpha spectrum on the basis of the counts measured in a window set in the area of the ^{214}Po peak[13,14]. This provides accurate results for ^{226}Ra, since no other natural radionuclides have alpha emissions in the same energy range. The counting efficiency of ^{214}Po (and thus of ^{226}Ra) in the selected window is 86 ± 3%.

The activity concentration of ^{228}Ra was determined via its daughter nuclide ^{228}Ac using gamma spectrometry. Water samples (2 l) were evaporated to a volume of 0.5 l, after which they were transferred to a Marinelli beaker for gamma spectrometric counting.

The measured concentrations of ^{226}Ra or ^{228}Ra can be below the lower limit of detection values. Due to background subtraction from low gross count rates, also negative values can be obtained. In order to avoid data distortion, all measured concentrations were accepted and all values were employed as such in data analyses.

The effective annual dose from ingestion of ^{226}Ra and ^{228}Ra was calculated on the basis of the mean, median and maximum concentration of the radionuclide presented in Table 1. The daily water consumption for adults was assumed to be 2 l and for children in the age group of 1–7 years 0.8 l. The conversion factors used are those given in ICRP publication 72[15].

Table 1. The mean activity concentrations of ^{226}Ra and ^{228}Ra in the present study and the mean activity concentration of ^{226}Ra from the population-based random study.

	Number of samples	Mean
All samples		
^{228}Ra (Bq l^{-1})	176	0.034
Weighted		
^{226}Ra (Bq l^{-1}) ^{228}Ra (Bq l^{-1})	176 176	0.041 0.030
The population based random study[3]		
^{226}Ra (Bq l^{-1})	288	0.050

The weighted mean activity concentrations of ^{226}Ra and ^{228}Ra were calculated using the weighting factors derived from the earlier population-based random study. The randomly selected 42 wells in which the activity concentration of ^{226}Ra was <0.01 Bq l^{-1} represent 42% of the original 288 wells in the earlier study. The rest 134 wells in which the activity concentration of ^{226}Ra was >0.01 Bq l^{-1} represent 58% of the wells.

Results

Activity concentration of ^{226}Ra and ^{228}Ra in drilled wells and annual effective dose

^{226}Ra activity concentrations were in the range of <0.01 to 1.0 Bq l^{-1} and ^{228}Ra activity concentrations in the range of $<0.03-0.32$ Bq l^{-1}. The levels 0.01 and 0.03 represent the detection limits for ^{226}Ra and ^{228}Ra, respectively. The mean activity concentrations of ^{228}Ra for the measured samples were 0.034 Bq l^{-1} (Table 1). The weighted mean activity concentrations (the weighting is described above) of ^{226}Ra and ^{228}Ra were 0.041 and 0.030 Bq l^{-1}, respectively.

The activity concentrations of ^{226}Ra and ^{228}Ra in drilled well water differed from each other (Table 2). Differences were found in mean activity concentrations of ^{226}Ra and ^{228}Ra and in maximum activity concentration, which was clearly higher for ^{226}Ra than for ^{228}Ra. In the activity concentration group where the ^{226}Ra activity was lower than 0.01 Bq l^{-1} (in the earlier population-based random study), the mean ^{228}Ra activity was two times higher than that of ^{226}Ra. In the activity concentration group where the ^{226}Ra activity concentrations were higher than 0.01 Bq l^{-1}, the mean ^{228}Ra activity was, on the contrary, only half the activity concentration of ^{226}Ra.

High radium activity concentrations in drinking water were rare. The ^{226}Ra concentration of 0.5 Bq l^{-1} was exceeded in 4% of the analyzed samples

Table 2. Activity concentrations of ^{226}Ra and ^{228}Ra (Bq l^{-1}) in water from drilled wells at two activity concentrations groups of ^{226}Ra (the total number of samples is 176).

Radio-nuclide	Number of samples	Mean	Quartile 1	Median	Quartile 3	95th Percentile	Maximum
^{226}Ra < 0.01 Bq l^{-1}*							
^{226}Ra (Bq l^{-1})	42	0.009	0.005	0.010	0.012	0.019	0.023
^{228}Ra (Bq l^{-1})	42	0.019	0.006	0.017	0.030	0.052	0.092
^{226}Ra > 0.01 Bq l^{-1}*							
^{226}Ra (Bq l^{-1})	134	0.063	0.011	0.018	0.045	0.238	1.037
^{228}Ra (Bq l^{-1})	134	0.038	0.012	0.031	0.052	0.108	0.317

* Classified according to the ^{226}Ra results from the earlier population-based random study.

of the present study. From the earlier population-based random study, the proportion of wells exceeding the concentration of 0.5 Bq l^{-1} represents 2% of the wells. A reference concentration, 0.5 Bq l^{-1}, for waterworks was calculated from a reference dose of 0.1 mSv a^{-1} according to the Drinking Water Directive. A ^{228}Ra concentration of 0.2 Bq l^{-1}, which results in the same annual effective dose, was exceeded in 1% of the samples analyzed. Taking into account the uncertainties in the representativeness of the samples in the range of ^{226}Ra < 0.01, the proportion of the wells exceeding ^{228}Ra concentration of 0.2 Bq l^{-1} may be 1–2%.

The regional distribution of ^{228}Ra and ^{226}Ra of the data is presented in Figure 1. The regional occurrences of ^{228}Ra and ^{226}Ra differed from each other. The reasons affecting the different occurrence of ^{226}Ra and ^{228}Ra was not within the scope of this study since the sampling was based on population-based random study, not on geological survey. It is probable that the difference is due to different concentrations of uranium- and thorium series' radionuclides in the host rock. The distribution of ^{226}Ra and ^{228}Ra activity concentrations levels in water samples from drilled wells in the present study are presented in Figure 2.

Since ^{226}Ra and ^{228}Ra have similar chemical properties and thus similar solubility properties, we examined the correlation between the occurrences of these isotopes. The Spearman's correlation was small between ^{226}Ra and ^{228}Ra ($\rho = 0.27$). The activity concentration of ^{228}Ra as a function of the activity concentration of ^{226}Ra is presented in Figure 3a. The figure indicates that there is large scattering between activity concentration of ^{226}Ra and ^{228}Ra. The isotopic ratio of ^{226}Ra/^{228}Ra as a function of activity concentration of ^{226}Ra is presented in the Figure 3b. In the calculation of the ^{226}Ra/^{228}Ra ratio, only values that exceeded the limit of detection were taken into account ($n = 53$). The mean, median and maximum ^{226}Ra/^{228}Ra ratios were 2.3, 0.6 and 27, respectively. The activity concentration of ^{226}Ra is dominating the isotopic ratio of ^{226}Ra/^{228}Ra since the range of ^{226}Ra activities is larger than ^{228}Ra activities.

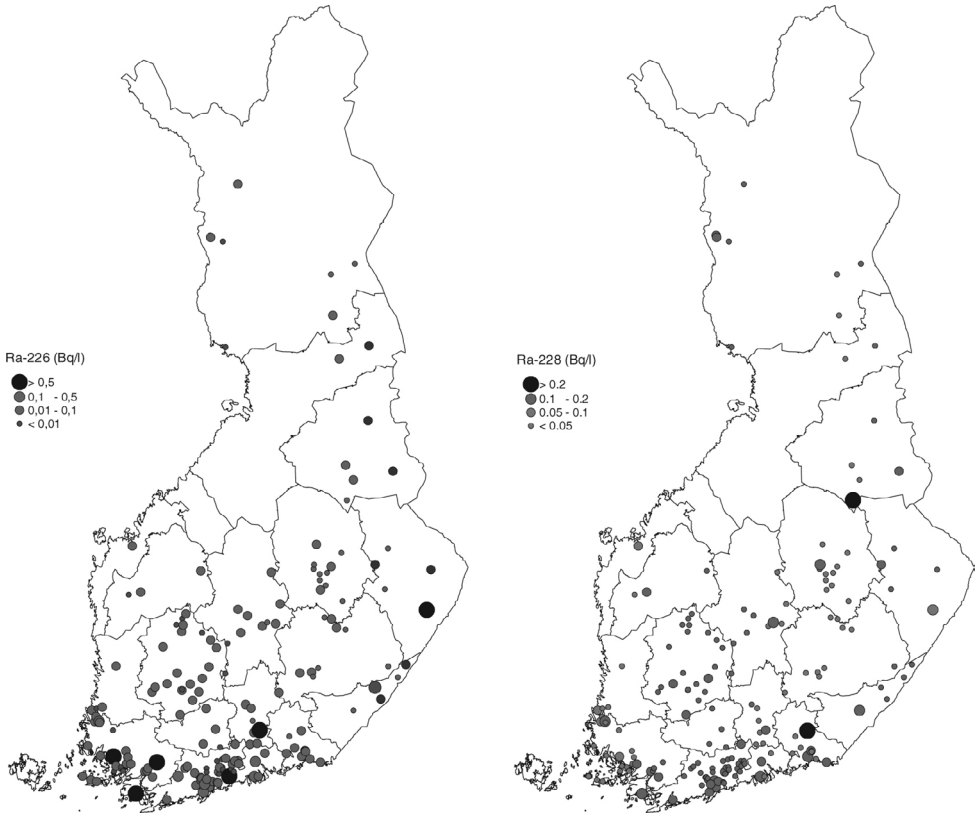

Figure 1. Regional occurrence of ^{226}Ra (on the left) and ^{228}Ra (on the right) activity concentrations in water samples from drilled wells.

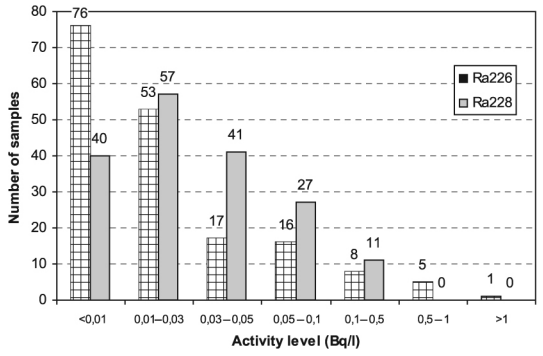

Figure 2. The distribution of ^{226}Ra and ^{228}Ra activity concentrations levels in water samples from drilled wells based on the data from the present study. The number of samples in the range of ^{226}Ra < 0.01 Bq l^{-1} was lower than that of samples in the range of ^{226}Ra > 0.01 Bq l^{-1}.

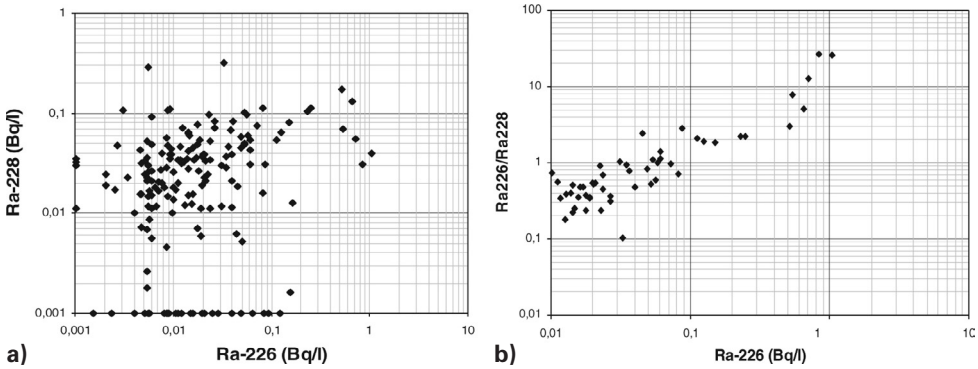

Figure 3. (a) Activity concentration of ^{228}Ra and (b) the isotopic ratio of ^{226}Ra/^{228}Ra as a function of the activity concentration of ^{226}Ra. In the figure (a) the negative ^{228}Ra values are set to level of 0.001 Bq l^{-1}.

Table 3. Mean, median and maximum annual effective doses (mSv) arising from ^{226}Ra and ^{228}Ra for an adult user of drilled wells at two activity concentrations groups of ^{226}Ra.

Radionuclide	Effective dose (mSv)		
	Mean	Median	Maximum
^{226}Ra < 0.01 Bq l^{-1}*			
^{226}Ra	0.002	0.002	0.005
^{228}Ra	0.010	0.009	0.046
^{226}Ra > 0.01 Bq l^{-1}*			
^{226}Ra	0.013	0.004	0.212
^{228}Ra	0.019	0.016	0.160

* Classified according to the ^{226}Ra results from the earlier population-based random study.

The annual effective dose from ^{226}Ra and ^{228}Ra for an adult user of drilled wells varied from 0.001 mSv to 0.21 mSv (Table 3). Depending on the age of the child, the corresponding dose from ^{226}Ra is 0.9–1.4 times higher and from ^{228}Ra 2.0–3.3 times higher than for adults. At low ^{226}Ra concentrations, the dose arising from ^{228}Ra was multifold compared to that from ^{226}Ra. The maximum effective dose from radium, however, was caused by the isotope ^{226}Ra. The effective dose for an adult from the mean activity concentrations of ^{228}Ra (0.034 Bq l^{-1}) and ^{226}Ra (0.041 Bq l^{-1}) were 0.017 and 0.008 mSv a^{-1}, respectively.

Discussion

In a previous study in Finland carried out in the early 1980s, the highest measured activity concentration of ^{228}Ra in water sampled from 125 private wells was 0.6 Bq l^{-1}, which corresponds to an annual effective dose of 0.3 mSv. In the present study such a high concentration could not be found[1,16,17].

^{226}Ra activity concentrations were previously examined in the population-based random study[3]. The weighted mean activity concentration of ^{226}Ra, 0.041 Bq l^{-1}, in the present study was close to the mean activity concentration obtained in the population-based random study, 0.050 Bq l^{-1}. In case of ^{228}Ra there may be more uncertainty in the representativeness of the results due to the selection of the data.

The activity concentrations of ^{226}Ra and ^{228}Ra in groundwater samples were very similar to those reported in Germany, China, Switzerland and Brazil (Table 4). However, the results reported from Germany, China and Brazil were from both drilled wells and dug wells and are therefore difficult to compare with this study. Since the studies were not based on representative sampling, the values presented in Table 3 can only be compared on a general level.

The mean ^{226}Ra concentrations in Sweden and Spain were high compared with the results obtained from other countries, which was mainly due to the restricted study areas consisting of granites, with a high uranium and therefore also a high ^{226}Ra concentration in the bedrock[18,19]. The mean ^{228}Ra concentration was high in Brazil and differed significantly from the results reported from other countries. High ^{228}Ra concentrations have been observed in many Brazilian environmental samples, indicating that ^{232}Th has enriched relative to ^{238}U in Brazilian soils[20].

In low salinity groundwater, ^{226}Ra/^{228}Ra ratios have varied over a very wide range. Values from 0.07 to 41 have been reported in the literature[9]. In most studies the ^{226}Ra/^{228}Ra ratio has been higher at greater activity concentrations of radium[9]. The results of this study indicated the same trend.

In Finnish groundwater, the activity concentration of ^{226}Ra has been studied much more extensively than that of ^{228}Ra. It has been observed that the activity concentration of ^{226}Ra is unrelated to that of dissolved uranium[2,4]. Similar observations have also been reported in a separate study on California groundwater[21]. The activity concentration of radium exhibits a closer relationship with the groundwater chemistry than with the radium content of the host rock. Associations have been detected between ^{226}Ra and chlorine, bromide, iodide, sulphate, calcium, inactive strontium, sodium and lithium[8]. These stable elements are typically found in saline water.

The earlier population-based random study gave the mean annual effective dose of 0.39 mSv for the users of drilled wells from the radionuclides ^{222}Rn, ^{234}U,

^{238}U, ^{226}Ra, ^{210}Pb and ^{210}Po[3]. Taking into account the dose, 0.017 mSv, arising from the mean activity concentration of ^{228}Ra, 0.034 Bq l^{-1}, the total annual effective dose amounts to 0.41 mSv. The dose from ^{222}Rn released from water to indoor air is not included in the dose estimate. The average annual effective dose for Finns from all sources of ionising radiation is 3.7 mSv[22]. Thus, the effective dose from the consumption of drilled well water is on the average 11% of this.

Table 4. Studies of natural radionuclides in drinking water from Finland, Sweden, Germany, Spain, China, Switzerland and Brazil.

Nuclide	Country	Water source	Number of samples	Mean	Maximum	Reference
^{226}Ra (Bq l^{-1})	Finland	D G	288 184	0.05 0.02	1.3 0.03	3 3
	Sweden	D	328	0.26[b]	4.9	18
	Germany	G + D	192	0.15	6.29	6
	Spain	D G	345[a] 345[a]	0.84 0.03	9.26 0.07	19 19
	China	G + D	428	0.03	0.94	24
	Brazil	G + D	428	0.02[c]	0.49	20
^{228}Ra (Bq l^{-1})	Finland	D	176	0.03	0.32	This study
	Sweden	D	328	0.09[b]	1.6	18
	Germany	G + D	12	0.02	0.04	6
	China	G + D	137	0.05	0.49	24
	Brazil	G + D	88	0.26[c]	1.5	20
	Switzerland	DW M	360 42	– 0.03[d]	0.06[d] 0.4[d]	23 23

D, drilled well; G, groundwater (dug wells and springs); DW, drinking water; M, mineral water.

[a] Number of drilled well and dug well samples all together.

[b] Geometric mean.

[c] Median.

[d] Median and maximum values are estimated from cumulative frequency diagrams.

Conclusions

^{226}Ra activity concentrations were in the range of < 0.01 to 1.0 Bq l^{-1} and ^{228}Ra activity concentrations in the range of < 0.03 to 0.32 Bq l^{-1} in water samples from 176 drilled wells. A ^{226}Ra concentration of 0.5 Bq l^{-1} was exceeded in 2–4% of wells, while 1–2% of the wells exceeded a ^{228}Ra concentration of 0.2 Bq l^{-1}. The activity concentration of radium in Finnish drilled well water was generally low and high activity concentrations were rare.

The maximum annual effective doses from ^{226}Ra and ^{228}Ra for users of drilled wells were 0.21 mSv and 0.16 mSv, respectively. The effective doses from ^{228}Ra and ^{226}Ra isotopes for users of drilled wells were estimated to be 0.017 mSv a^{-1} and 0.008 mSv a^{-1}, respectively. This means that only ~6% of the total effective dose due to natural radionuclides in drinking water was caused by radium isotopes.

Activity concentration of ^{226}Ra is not related to the activity concentration of ^{228}Ra and the correlation between them was small. Therefore, it is important to analyse these nuclides independently of each other.

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6 General Discussion

6.1 Aeration technique

Research on removing radon from drinking water was started in Czechoslovakia in the 1970s (Hanslík *et al.*, 1978). Aeration was found a suitable method for stripping radon from water. A removal efficiency of 99% was achieved using porous aeration discs with an air-to-water ratio of 8:1 and an aeration time of 8 minutes. In Finland, Castrén (1980) reported an aerator installed at a school that reduced the radon concentration in water from 45 000 Bq/L to 190–550 Bq/L, depending on the consumption rate. Various aeration techniques were also tested in Sweden in the early 1980s. Aeration under atmospheric pressure was reported to be the best method, with a removal efficiency of up to 75% (Hedberg *et al.*, 1982). In the US, diffused aeration and spray aeration were also found effective methods for removing radon from drinking water (Lowry, 1983). Based on these pioneering studies, the manufacture of aerators for household use was started.

Lindén (1997) reported removal efficiencies for nine commercially available aerators that were able to remove over 90% of radon in raw water, in most cases between 96 and 99%. Several technical problems, however, were reported. Clogging and malfunctioning of the solenoid valves was the most common problem. In several cases, malfunction of the mechanical floats used for regulating the water level inside the aerators had also occurred, which could lead to severe damage to the dwelling.

Therefore, the main aim of our studies concerning aeration techniques was to concentrate on improving the usability, durability and reliability of the available units together with the distributors and manufacturers, and to carefully monitor water quality in order to ensure high quality drinking water for the residents (Myllymäki, 1996; Myllymäki *et al.*, 1999; Turtiainen *et al.*, 1999; Annanmäki and Turtiainen, 2000; Vesterbacka *et al.*, 2003; Chapter 2). Initially, two Finnish companies designed new aerators and started to manufacture them. The demand on the market, however, was much less than anticipated and these units are therefore no longer available. Two new models designed by another company, however, proved to be very robust, and due to their competitive pricing they are still on the market (Vesterbacka *et* al., 2003; Chapter 2).

The present work indicated that the removal efficiency of the aerators does not necessarily remain constant when the water consumption rate is high, and unacceptably high concentrations of radon may be found in drinking water (Turtiainen *et al.*, 1999). Therefore, a new test protocol was developed that applies frequent sampling while allowing 100 litres of water flow (Chapter 2).

It also was observed that iron and manganese may precipitate during aeration, which was also reported earlier by Lindén (1997). During aeration, water becomes virtually saturated with oxygen, which improves the taste of the drinking water. Carbon dioxide is removed during aeration, which causes a slight rise in pH and reduces the corrosiveness of the water. No clear trend in other water quality parameters, including bacterial densities, could be observed and the water quality criteria set for drinking water were complied with (Turtiainen *et al*., 1999). Guidance on the installation of aeration units was also given (Chapter 2).

6.2 Activated carbon filtration

Activated carbon filtration was first investigated for removing waterborne radon in Sweden in the early 1980s. Hedberg *et al.* (1982) tested a 0.6-litre granular activated carbon (GAC) unit that was mounted in a household water line before the pressure tank. Initially, the unit could remove only about 30% of the radon. The removal efficiency showed a decreasing trend, and after treating 2.5 m^3 of water more radon was found in treated water than raw water. This was explained by ^{226}Ra, which accumulated in ferric precipitates formed inside the carbon unit and acted as a radon source. In addition, measurements performed next to the GAC unit showed external gamma dose rate levels that were about 300 times higher than before the test due to radon daughters ^{214}Pb and ^{214}Bi. The authors stated that gamma radiation emerging from GAC units cannot be neglected from a radiation protection viewpoint, and the spent carbon may be classified as radioactive waste due to the accumulated radionuclides. Therefore, activated carbon filtration was not further investigated and aeration was regarded as the best viable alternative for removing radon from drinking water.

Lowry (1983) investigated much larger GAC units (28–70 litres) in the US and found them effective in radon removal and more cost effective than spray or diffused aeration techniques. However, it became apparent that both the external gamma radiation emerging from the GAC units and the ^{210}Pb accumulated in the carbon were problematic, as the national guidelines in the US could be exceeded (Rydell *et al.,* 1989). Lowry *et al.* (1989) also reported that approximately 5% of GAC units had exhibited lower than expected removal efficiencies or premature fouling, which was assumed to be caused by raw water characteristics, most probably iron.

Regardless of the disadvantages reported by these earlier studies, testing of granular activated carbon filtration was included in our research projects aiming at bringing reliable radon removal units to the market (Myllymäki, 1996; Myllymäki *et al.*, 1999; Turtiainen *et al.*, 2000; Chapter 3, Vesterbacka

et al., 2003). Based on the results presented by Lowry (1983), the bed volumes of the units employed were 39–63 litres. In our investigations, several water quality parameters were monitored in order to determine possible limiting factors in using this technique and to guarantee high quality drinking water for the residents. Therefore, wells exhibiting different water characteristics were selected for testing (Chapter 3). In addition, external gamma radiation and waste issues were considered and recommendations given accordingly (Annanmäki *et al.*, 2000).

Our investigations revealed that activated carbon units removed radon efficiently from all types of well water. Iron and organic carbon concentrations in raw water were not found to correlate with the removal efficiencies (Vesterbacka *et al.*, 2003). The sample, however, was limited and no definite conclusion based on the data could be drawn. Ten units were further followed for a period of 3 to 9 years in total, during which no breakthrough of radon occurred (Chapter 4). Premature fouling reported by Lowry *et al.* (1989) was hence not observed in our study.

One 39-litre unit exhibiting lower than average removal efficiencies was redesigned, and an anion exchange unit was installed before a fresh batch of GAC. This was carried out in order to remove the unacceptably high uranium concentration (200 µg/L). Consequently, the radon removal efficiency increased from 87–95% to >98%, which suggests that anionic compounds may impair the efficiency of GAC filtration (Turtiainen *et al*, 2000). Lowry *et al.* (1988) had also observed a decrease in the radon removal efficiency simultaneously with the breakthrough of uranium, which mainly occurs as an anionic complex in ground waters.

GAC units efficiently removed iron from raw water, generally more than 70%, and the removal efficiency remained good for up to 12 000 bed volumes. The total organic carbon concentration decreased during GAC filtration, initially by 60–90%, but the efficiency decreased over time. The microbiological quality was monitored with a heterotrophic plate count for up to 3.8 years and no significant increase was observed. Little influence on other water quality parameters was detected (Turtiainen *et al.*, 2000; Chapter 3; Vesterbacka *et al.*, 2003).

GAC units were capable of initially removing uranium, but the total capacity was approximated as 50 mg per litre of GAC. The removal of ^{226}Ra varied between units, and a higher concentration was occasionally recorded in treated water than in raw water. The removal of ^{210}Pb and ^{210}Po was generally >60%, but also varied between units. Therefore, GAC filtration can only be reliably applied for removing radon (Turtiainen *et al.,* 2000).

The external radiation emerging from GAC units was found manageable with shields attenuating gamma radiation if the daily radon flow through the

units was below 4 MBq (i.e. radon concentration × daily water consumption). Shielding of the units, however, was believed to sometimes fail in practice, and it was therefore recommended that the units not be installed inside homes but in an outhouse shed or inside a service well (Markkanen, 2005). After considering the activity of ^{210}Pb in spent GAC and the projected number of GAC units finding their way to municipal dumps, it was recommended that spent GAC be discharged as ordinary solid household waste (Annanmäki *et al.*, 2000).

6.3 Mitigation of exposure to radon by household water treatment

As of in March 2007, the number of radon removal units installed in homes was already about 1 000. Based on a survey conducted by Vesterbacka *et al*. (2005), we estimated the number of wells where radon exceeds 1 000 Bq/l to be about 6 600. This means that measures against radon had been taken for about 15% of the wells with elevated radon concentrations. A few projected cancer deaths had hence been averted by the use of these units (Chapter 4).

Nevertheless, we know that there are still thousands of wells in permanent use that have a higher than recommended level of radon. The only way to reach the house owners who are still unaware of the problem is through campaigns. Local media have been the best channel of communication in reaching these people (Chapter 4).

6.4 Occurrence of radium isotopes in well water

In the present work, it was demonstrated that waterborne isotopes of radium (^{226}Ra and ^{228}Ra) rarely cause significant exposure to users of drilled wells. The maximum annual effective doses from ^{226}Ra and ^{228}Ra found in the survey were 0.21 mSv and 0.16 mSv, respectively. No correlation was observed between the concentrations of these two isotopes (Chapter 5). In the rare cases where radium removal is required, this can be accomplished, for instance, by cation exchange (Annanmäki and Turtiainen, 2000).

6.5 Conclusions

Finland is a large, sparsely populated country. More than half of Finns wish to live in a privately owned one-family house, and the majority of them prefer housing in sparsely built-up areas. This brings great challenges in organising water services, as more private wells will be constructed along with new buildings in dispersed settlements. It is also interesting to note that even though

the number of private wells in permanent use has greatly declined, the number of drilled wells has in fact increased. Greater awareness of the occurrence of radon in drilled wells is needed.

Decree 401/2001 by the Ministry of Social Affairs and Health obliges municipal health authorities to ensure that households relying on their own water supply receive enough information on the quality and the associated health risks of the local drinking water and on the possibilities for mitigating these risks. This has, however, proven to be very difficult to bring into practice, as no registry of wells presently exists in Finland. There are still thousands of homes in which water with elevated radon levels is used. We should emphasize that if there is a possibility to connect a home to a public water supply, this should be considered as the primary mitigation option. When the property is sold, there is a risk that information on the maintenance of the radon removal unit will not be passed on to the new owner. This is especially problematic with activated carbon units that emit gamma radiation when they are in operation. However, a public water supply is not always available.

From today's perspective, our agenda of involving companies in our research projects was a successful one. Companies specialising in water treatment now have a high level of expertise in designing water treatment alternatives that meet the needs of house owners. These companies are nowadays the primary contact for house owners seeking additional information on the units.

The occasional problems of elevated radon levels in wells located in sparsely populated areas can nowadays be easily solved. Consumers know that scientific research has been carried out when bringing these units to the market, and water quality together with the radon concentration has been followed in a number of radon removal units. Since the investment costs of these units are often high, this fact may be reassuring when contemplating the purchase of a treatment unit. Information on treatment alternatives is also available on the Internet, which home owners can familiarize themselves with.

In 2007, more than a thousand radon removal units were in use and the collective radon exposure from drinking water had been reduced by approximately 15% among users of private wells in which the health-based guideline value set for radon is not met. This is a great achievement considering that the first removal units were installed only ten years earlier.

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