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TURTIAINEN Tuukka, KOKKONEN Pauliina, SALONEN Laina. Removal of Radon and Other Natural Radionuclides from Household Water with Domestic Style Granular Activated Carbon Filters. Helsinki 2000, 82 pp. + Appendices 18 pp.

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ABSTRACT

Removal of radon and other natural radionuclides from household water was studied applying granular activated carbon filtration. The study was part of the TENAWA project (Treatment Techniques for Removing Natural Radionuclides from Drinking Water), which was carried out on a cost-shared basis with the CEC under contract No. FI4PCT960054 and under the supervision of the Directorate-General XII Radiation Protection Research Unit. The overall objective of the TENAWA project was to accumulate information and to test methods and equipment currently available for removing natural radionuclides from drinking water. The project was divided into 13 work packages. In this report, the field research carried out within work packages 3.1 and 3.2 will be described.

Reliable methods of removing radon from ground water supplies are needed in thousands of homes in Finland. Twelve granular activated carbon (GAC) filters were installed for radon removal in homes using water from drilled wells. The activity concentration of ^{222}Rn in different wells varied from 900 Bq/L to 7,400 Bq/L. In addition, different types of groundwater were covered. The filters were studied for 9–27 months. Radon removal efficiency and the adsorption rate of radon on GAC were calculated from samples taken every 1–3 months. Several water quality parameters and the long-lived radionuclides of the uranium series were also determined. Dose equivalent rates in the vicinity of the filters were measured, and the accumulation of different radionuclides in GAC was determined by gammaspectrometrical methods.

In most cases, the radon removal efficiency was over 99%. Only high concentrations of uranium, and possibly organic matter, in the raw water were observed to lower the adsorption rate of radon. GAC filtration also removed variable amounts of iron, organic matter and the long-lived radionuclides of the uranium series. The microbiological quality of water remained good.

The dose equivalent rate on the surface of the GAC units was 1–110 µSv/h and it depended on the daily water usage and radon concentration. With an appropriate placement of the unit and adequate shielding the dose rates in the residential rooms were less than 0.3 µSv/h. Materials such as lead, concrete, bricks and water jacket were found to be suitable for shielding. The long-lived radon daughter, ^{210}Pb ($t_{½}$=22.6 a) originating from the radon retained in the filter, was totally retained on GAC. Therefore, high specific activity of ^{210}Pb may be found in spent GAC. This brings up a problem in the disposal of spent GAC. The filters were technically reliable and no problems were reported during the study.

TURTIAINEN Tuukka, KOKKONEN Pauliina, SALONEN Laina. Radonin ja muiden luonnon radionuklidien poistaminen talousvedestä kotitalouskäyttöön suunnitelluilla aktiivihiilisuodattimilla. Helsinki 2000, 82 s. + liitteet 18 s.

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Avainsanat radon, radonin poisto, aktiivihiili, vedenkäsittely, pohjavesi, säteilysuojus

TIIVISTELMÄ

Radonin ja muiden luonnon radionuklidien poistoa talousvedestä tutkittiin aktiivihiilisuodatuksella. Tutkimus oli osa TENAWA-projektia (Treatment Techniques for Removing Natural Radionuclides from Drinking Water), joka toteutettiin yhteisrahoitteisesti Euroopan Komission kanssa. Valvojana toimi pääosasto XII:n Radiation Protection Research Unit. TENAWA-projektin päätavoite oli tutkia olemassa olevia menetelmiä ja laitteita, jotka soveltuvat radionuklidien poistoon juomavedestä. Projekti koostui kolmestatoista työpaketista. Tässä raportissa selostetaan työpaketteihin 3.1 ja 3.2 liittyvien kenttäkokeiden järjestelyt sekä tarkastellaan saatuja tuloksia. Raporttiin kuuluu lisäksi kirjallisuuskatsaus aiemmin tehdyistä aktiivihiilitutkimuksista.

Luotettavia menetelmiä radonin poistamiseksi pohjavesikaivojen vesistä tarvitaan tuhansissa suomalaisissa kodeissa. Kaksitoista aktiivihiilisuodatinta asennettiin talouksiin, jotka ottivat talousvetensä porakaivoista. Vesien radonpitoisuudet olivat välillä 900 Bq/l ja 7 400 Bq/l. Lisäksi vedet olivat laadultaan erityyppisiä. Suodattimia tutkittiin 9–27 kuukautta. Radoninpoistotehokkuus ja adsorptionopeus määritettiin näytteistä, joita otettiin 1–3 kuukauden välein. Lisäksi määritettiin useita veden laatuparametrejä sekä uraanisarjan pitkäikäisten radionuklidien pitoisuudet. Annosekvivalenttinopeudet mitattiin suodattimien läheisyydestä ja radionuklidien kerääntyminen hiileen määritettiin gammaspektrometrisesti.

Useimmissa tapauksissa radonin poistotehokkuus oli yli 99 %. Ainoastaan raakaveden korkeilla uraani- ja mahdollisesti humuspitoisuuksilla havaittiin radonin adsorptionopeuden hidastumista. Aktiivihiilisuodatus poisti lisäksi vaihtelevia määriä rautaa, orgaanisia aineita ja uraanisarjan pitkäikäisiä radionuklideja. Veden mikrobiologinen laatu säilyi hyvänä.

Annosekvivalenttinopeudet suodattimien ulkopinnoilla vaihtelivat veden radonpitoisuudesta ja veden kulutuksesta riippuen välillä 1–110 μSv/h. Suodattimien asennuspaikan sopivalla valinnalla ja toteutetuilla riittävillä säteilysuojuksilla annosnopeudet asuintiloissa jäivät alle 0,3 μSv/h. Materiaalit, kuten lyijy, betoni, tiilet ja vesivaippa, havaittiin sopiviksi säteilysuojusmateriaaleiksi. Radonin pitkäikäinen tytärnuklidi lyijy 210 ($t_{½}$=22.6 a), jota radon jatkuvasti tuottaa hajotessaan suodattimen sisällä, pidättyy kvantitatiivisesti aktiivihiilimassaan. Sen vuoksi korkeita lyijy 210:n ominaisaktiivisuuksia voi esiintyä käytetyssä aktiivihiilessä, mikä voi tuoda esiin ongelmia käytetyn hiilen hävittämisessä. Suodattimet olivat teknisesti luotettavia eikä tekniseen toimivuuteen liittyviä ongelmia esiintynyt tutkimuksen aikana.

CONTENTS

PREFACE

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

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

The TENAWA project was divided into 13 work packages:

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

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

In this report, the field research carried out within work packages 3.1 and 3.2 will be described. Four test locations that are discussed in this report were shared between the TENAWA project and the joint research project carried out by the Finnish Environment Institute, STUK and three commercial companies (Myllymäki et al. 1999). All the data collected at these test locations are also presented in this report. Many discussions have taken place among our colleagues in and beyond the TENAWA project and we gratefully acknowledge these.

1 INTRODUCTION

Radon (^{222}Rn) in household water may result in excessive radiation doses from ingestion of drinking water and from inhalation of radon gas that is released in the air during water use. High radon concentrations occur mostly in drilled wells, the number which has increased during recent years. In about 30% of the roughly 70,000 drilled wells in Finland, ^{222}Rn concentration exceeds 300 Bq/L, which is the maximum limit set for public water supplies (Mäkeläinen et al. 1999, ST-Guide 12.3 1993). In about 10% of Finnish drilled wells, the radon concentration is higher than 1,000 Bq/L. In Sweden, the radon levels exceed 100 Bq/L in about 50% of the drilled wells and in about 4% levels higher than 1,000 Bq/L are found (Kulich et al. 1988). 1,000 Bq/L is the recommended action level for private households in Sweden (SSI 1998). 500 Bq/L is exceeded in about 13% of the roughly 130,000 drilled wells in Norway (Banks et al. 1998).

Consequently, techniques for removing radon from household water are required in thousands of homes in the Nordic countries. These households are located mainly in the sparsely populated countryside, where the residents obtain their household water from private wells. When this study was started in 1996, there were only a few treatment units installed for radon removal in domestic use in Finland. The need for suitable equipment, however, was great.

Radon follows radium (^{226}Ra) in the uranium series. The highest concentrations of radon are found in granitic and granodiorite areas, where the uranium content of the bedrock is the highest (Juntunen 1991). The occurrence of radon in bedrock water is affected by the uranium content of the mineral matter of the bedrock and the dynamics of the groundwater. The migration rate of radon is high in the shattered zones of crystalline bedrock. The chemical composition of groundwater does not control the occurrence of radon (Juntunen 1991).

Two basic techniques can be applied to remove radon from water: aeration and granular activated carbon (GAC) filtration. Information on experiences of radon removal by GAC filtration has mainly come from the USA, where several studies have been conducted since 1981. In Finland, comprehensive research into radon removal from household water was commenced in 1995. Only a few small-scale experiments had been carried out earlier. The research was initiated as a joint research project carried out by the Finnish Environment Institute, the Helsinki University of Technology and the Finnish Centre for Radiation and

Nuclear Safety (STUK). After the basic study, the research was continued as a joint research project carried out by the Finnish Environment Institute, STUK and three commercial companies. The performance of several GAC types was tested and two new aeration units were designed (Myllymäki 1996). The study was continued as full-scale field research with the best GAC type and the aerators (Myllymäki et al. 1999).

Based on literature, GAC filtration offers an inexpensive, maintenance-free and silent method of removing radon and it does not require much free space. Advantages and disadvantages, however, have been reported for both techniques (Kinner et al. 1990, Hiltebrand et al. 1988). The most significant drawback in GAC filtration is the gamma radiation that is emitted from the filters when they are in service. The US Environmental Protection Agency (USEPA) has proposed aeration as the best available radon removal technology because of serious concerns regarding the safe daily operation of GAC treatment and the proper disposal of the spent carbon (Paris 1993).

Lowry et al. (1990) reported that approximately 6 per cent of the 121 GAC filters studied had experienced premature failure. The failure was believed to be water quality related. Bedrock waters in Finland are typically soft, bicarbonate rich and slightly alkaline. Owing to the alkalinity, carbon dioxide is present in low concentrations, if it is present at all (Juntunen 1991). Therefore, the main objective of this study was to investigate how GAC filters remove radon from different types of groundwater in everyday household use and in vacation residences. Test locations were selected such that the water types most typically found in Finnish bedrock were covered. Several water quality parameters were determined. Iron and manganese, as well as the long-lived nuclides of the uranium series (^{238}U, ^{234}U, ^{226}Ra, ^{210}Po and ^{210}Pb) may occur simultaneously with radon. Their removal and their effect on the performance of the GAC filter were also studied. Other aspects considered in this study were: changes in water quality due to filtration, the microbiological quality of treated water, the accumulation of radionuclides in GAC, the dose equivalent rate in the vicinity of the filters and the necessary shielding.

2 SUMMARY OF THE STUDIES CONDUCTED 1985–1997

The first GAC unit installed for the removal of radon from a water supply was in Maine (USA) in February 1981 (Lowry and Brandow 1985). Lowry discussed radon removal by GAC filtration in question-and-answer format to provide information for persons considering a method of removing radon from their household groundwater supply (Lowry 1983). The information was based upon the laboratory research and the field tests of 11 GAC units. Radon removal applying GAC filtration was found to be extremely effective. Several GAC filters were installed in private households, and more comprehensive laboratory and field research was initiated.

In this chapter, the main characteristics of GAC filtration in radon removal will be discussed. The data presented here are based on the articles and study reports from different research groups as well as on the information gained from commercial companies that specialise in water treatment technology in Finland.

2.1 Occurrence of radon in ground water

Radon (^{222}Rn) is a naturally occurring radioactive gas, which follows radium (^{226}Ra) in the uranium decay series. Uranium is widely distributed in nature and is a minor constituent of all rock and soil, as is radon. The highest concentrations of radon in groundwater occur close to uranium ore deposits or in rocks with elevated uranium contents. The most typical geological settings are crystalline complexes and granitoids. The occurrence of radon in bedrock water is affected by the uranium content of the mineral matter of the bedrock and the dynamics of the groundwater. The migration rate of radon is high in the shattered zones of crystalline bedrock. The chemical composition of groundwater does not control the occurrence of radon (Juntunen 1991).

2.2 Physico-chemical properties of radon

Radon (^{222}Rn) is an alpha-emitting radionuclide. The half-life of radon is 3.82 days, and the decay constant is $0.181\ d^{-1}$. Because of the short half-life, the ratio between mass and activity is small. One thousand becquerels of radon correspond to only $0.18 \cdot 10^{-12}$ g. The short-lived radon progeny (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) attain equilibrium with radon within approx. four hours, which is why these daughter products occur simultaneously with radon in water. The progeny, however, may be adsorbed on the walls of the drill hole, or even on the water pipes. Therefore, a full secular equilibrium is seldom attained (Swedjemark and Lindén 1998).

The most important properties of radon in terms of the removal process are:

- Radon is a noble gas and has the electronic configuration $6s^2 6p^6$. Radon does not readily form compounds and it is virtually chemically inert (can be oxidised with BrF_3 or O_2SBF_6). Therefore, radon removal based on chemical reaction is not possible.
- Radon is soluble in water. Henry's law constant for radon at 10°C is 3,300 mg/(L·atm). Consequently, radon can be removed by aeration.
- Radon has a relatively short half-life. Therefore, the activity of radon decreases rapidly and is only approx. two per cent of its original value after three weeks. Radon, therefore, is not regarded as long-lived radioactive waste (Kinner et al. 1990).

2.3 Preparation and properties of activated carbon

Activated carbon is a term used to describe a wide range of materials that exhibit a high degree of porosity and an extremely large surface area (Scarpitta and Harley 1990). Activated carbon can exist in e.g. pulverised or granular form, the latter of which is more commonly applied in water treatment technology. Activated carbon can be made of several different raw materials. These materials can be e.g. peat, coconut, hard coal, and brown coal. Prior to activation, the raw material is carbonised to drive off non-carbon volatiles, thereby forming a fixed carbon mass containing a rudimentary pore structure. In the activation process, or heating in the absence of oxygen, both macropores and micropores are created in the solid matrix of the initial material (Scarpitta and Harley 1990). Due to the porosity, one gram of activated carbon contains about 400–1,300 m^2 of active surface area. The density of GAC is usually 0.4–0.5 kg/L; thus an ordinary 40-litre GAC filter may have an active surface area equal to 20 square kilometres.

2.4 Adsorption of radon on GAC

The ability of activated charcoal to adsorb radon was discovered by Rutherford in 1906. Since radon is chemically inert and does not form bonds, the adsorption process is one of purely physical adsorption. Physical adsorption is not chemically specific and is readily reversible. The forces of attraction between the adsorbate and adsorbent are weak and similar to van der Waals' forces.

Jonas (1978) represented the removal of gas by impregnated activated carbon as a series of seven consecutive steps (Figure 1). Activated carbon can adsorb various non-polar gases, for which the reaction steps were described. In the case of radon adsorption, the first five steps are valid. Since radon is chemically inert, the sixth and the seventh steps do not pertain. Radon, however, undergoes radioactive decay, which can be regarded as the sixth and final step.

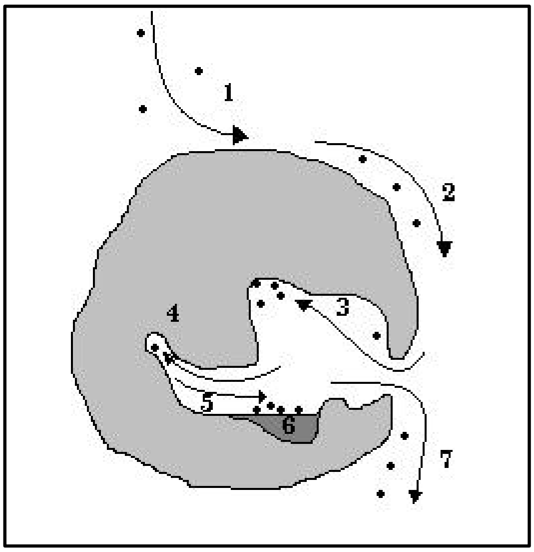

Figure 1. *The seven reaction steps in gas sorption by an impregnated carbon (Jonas 1978). There are only six reaction steps in the adsorption of radon, the last of which is radioactive decay.*

1. *Mass Transfer, wherein the gas molecules are transported from the vicinity of neighbouring fluid molecules to the vicinity of the adsorbent granule outer surface.*
2. *Surface Diffusion, wherein gas molecules are transported over a two-dimensional granule surface, to the mouth or entrance of the adsorbent micropore.*
3. *Intragranular Diffusion, wherein the transport of gas molecules within the small pores causes the gas to collide with or come within the force field of an active site.*
4. *Physical adsorption, wherein the gas molecule loses translational freedom and is held by and occupies the active site.*
5. *Gas desorption, wherein gas molecules gain translational freedom and desorb from the active sites in the micropore region, traversing the transition and macropore regions of the adsorbent on which the chemical impregnants were deposited.*
6. *Chemical Reaction, wherein the desorbing gas molecules react with the chemical impregnants on the surfaces of the larger adsorbent pores.*
7. *Surface Renewal, wherein the products of the chemical reaction are both highly volatile and poorly adsorbed by the adsorbent, thus leaving the macropore region and entering the fluid stream surrounding the carbon granule where they are swept by mass transport away from the carbon granule bed.*

The first step (mass transfer) is affected by transport phenomenon such as mass flow if a pressure gradient is present or mass diffusion if a concentration gradient is present. The second step (surface diffusion) is rapid compared to the third step (intragranular diffusion), which is dependent on the pore radius, temperature, and molecular weight of the gas molecule. For the fourth step (physical adsorption), pseudo first order kinetics should pertain. The kinetic equation can be shown in the form of the equation

(1) $$t_b = \frac{W_e}{C_0 Q}\left[W - \frac{r_b Q}{k_u}\ln(\frac{C_0}{C_x})\right]$$, where

t_b	is the gas breakthrough time in minutes at which the concentration
C_x	appears in the exit stream,
C_0	the inlet concentration in g/cm^3,
Q	the volumetric flow rate in cm^3/min.
r_b	the bulk density of the packed bed in g/cm^3,
k_u	the pseudo first order adsorption rate constant in min^{-1},
W	the adsorbent weight in g, and
W_e	the kinetic saturation capacity in g/g at the arbitrarily chosen ratio of C_x/C_0.

Values of t_b plotted as a function of W yield a straight-line from whose slope and x-axis intercept the properties W_e and k_u can be calculated. The x-axis intercept $(r_b Q/k_u)\ln(C_0/C_x)$ is considered to be the critical bed weight W_c, or the weight of carbon just sufficient to reduce the influent concentration to the chosen effluent concentration.

In the early studies conducted by Lowry and others, different activated carbons were compared by performing batch experiments, and the adsorptive properties were reported as Freundlich adsorption isotherms (Lowry and Brandow 1985, Lowry 1983, Lowry et al. 1987). The column experiments, however, revealed that no break-through could be observed as the filtration continued. The curves drawn showed that after the filter had been in service for three weeks, the effluent-influent ratio reached a constant value. The phenomenon could be explained by the radioactive decay of radon. As radon is adsorbed on GAC, it decays and the active sites are freed. After three weeks' operation with a constant flow rate, the adsorption rate of radon equals the rate of its decay. This state was referred to as the *adsorption-decay steady-state*. The net result of the dependence of these two opposing processes upon the amount of adsorbed radon is that for any given installation there is a constant amount of radon adsorbed on the carbon after the establishment of steady-state operation.

Later Lowry and Lowry (1987) adopted a different approach for modelling radon removal by GAC. First order kinetics was applied, the equation of which was presented as

(2) $C_i = C_0 \cdot e^{-K_{ss} \cdot t}$, where

(3) $t = \frac{V_b}{Q}$, and

C_t	is the effluent concentration in Bq/L,
C_0	the influent concentration in Bq/L,
K_{ss}	the adsorption-decay steady-state constant in h^{-1},
V_b	the volume of the GAC bed in L, and
Q	is the average volumetric flow rate in L/h.

Parameter t is called the empty bed detention time (EBDT) that represents the average time water would stay in an empty vessel the volume of which is equivalent to the volume of the wet carbon bed. The K_{ss} constant indicates the rate of adsorption; the higher the constant the faster the adsorption, and the less carbon is needed to obtain the same removal efficiency.

The adsorption-decay steady-state constants were determined for different carbons using an experimental GAC vessel, equipped with several outlets at different bed heights (volumes) and at a constant flow rate. After the steady-state was obtained, radon samples were collected from the inlet and from the outlets exhibiting different bed volumes. From these data, the bed volumes (V_b) were plotted against $\ln(C_t/C_o)$ and a straight-line curve was obtained. K_{ss} could be calculated from the slope and the known flow rate according to Equation 4.

(4) $V_b = -\frac{Q}{K_{ss}} \ln\left(\frac{C_t}{C_0}\right)$

Lowry suggested that a K_{ss} constant can be calculated for any installation from the volume of the GAC bed, the influent and effluent concentration and the water usage over a period of two to three weeks prior to the sampling (Lowry and Lowry 1987). He described a GAC filter as a kind of decay-storage device that retains radon for a period of two to three weeks. During that time most of the radon has decayed and no desorption of “old radon” occurs. Therefore, a GAC unit needs the same period to respond to any changes in water usage.

Different brands of GAC have been investigated in several studies (Lowry and Lowry 1987, Myllymäki 1996, Haberer et al. 1997, Myllymäki et al. 1999). Mostly only the adsorption isotherms, however, have been determined. Table I summarises the results of the experiments where K_{ss} constants were determined.

Table I. *Summary of the* K_{ss} *constants determined by Lowry and Lowry (1987), Kinner et al. (1990), Myllymäki (1996) and Myllymäki et al. (1999).*

Carbon	Size (mesh)	K_{ss} (h^{-1})	Reference
HydroDarco 4000	12x40	2.09	Lowry and Lowry, 1987
Calgon F-400	12x40	1.53	Lowry and Lowry, 1987
Norit Peat	8x20	1.35	Lowry and Lowry, 1987
Barneby Cheney 299 or 1002	–	3.02	Lowry and Lowry, 1987
Barneby Cheney 1002	–	2.98–3.29	Kinner et al. 1990
Calgon F-400	12x40	2.4–3.4	Myllymäki, 1996
Aqua Sorb	12x40	4.4	Myllymäki, 1996
Norit PK	8x30	2.9	Myllymäki, 1996
Silcarbon K825	16x60	2.8	Myllymäki, 1996
F.A.W. Jacobi Ab*	–	3.6–3.8	Myllymäki et al. 1999
Bang & Bonsomer Oy*	–	3.7–3.8	Myllymäki et al. 1999
Sutcliffe Speakman Carbons Ltd*	–	3.3–3.4	Myllymäki et al. 1999
B. Jacobssons Embellage & Kemikalie Ab*	–	3.4–3.5	Myllymäki et al. 1999

* Supplier, the carbon type is not given.

Two brands of GAC were investigated in two studies. The K_{ss} constant of Barneby Cheney 1002 determined by Kinner et al. (1990) compared favourably to that determined by Lowry and Lowry (1987). The K_{ss}

constants of Calgon F-400 determined by Myllymäki (1996) and Lowry and Lowry, however, differed significantly and therefore the results of these two studies are not comparable. In the 1996 study conducted by Myllymäki, the experimental GAC filter did not operate under normal plumbing pressure (1.5–6 bar), which was the main reason for the difference in the results. In the 1999 study, Myllymäki et al. determined the K_{ss} constants under ordinary household conditions. Nevertheless, the K_{ss} constants determined in both studies reveal the respective order of the GAC types.

2.5 Setting up a GAC filter

As a gas, radon readily transfers into the air as water is used (Nazaroff et al. 1987). Release of radon from water is the most significant when a dishwasher or washing machine and a shower are being used (Table II). Consequently, radon results in radiation doses from both ingestion and inhalation and must, therefore, be removed from all the household water. The GAC filter is installed in the incoming water line directly after the pressure tank. This type of installation is called point-of-entry (Figure 2). A sediment filter can be mounted at the inlet of the filter to protect the bed against premature fouling by particles. If removal of other nuclides (i.e. uranium) needs to be carried out with another filter, this filter is placed between the sediment filter and the GAC filter (Kinner et al. 1990).

Table II. *Transfer coefficients for the release of radon from water to the air, by use (Nazaroff et al. 1987).*

Type of use	Per cent of radon liberated
Dishwasher	90–98
Laundry	90–95
Shower	63–71
Bath	30–50
Drinking and cleaning	10–50
Toilet	29–30

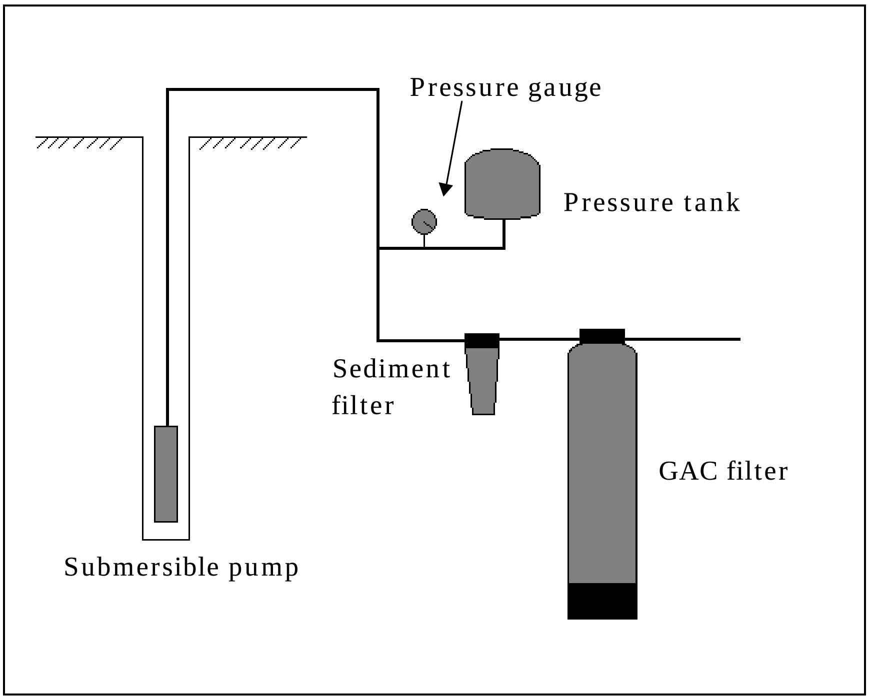

Figure 2. *A schematic representation of the installation of a POE (point-of-entry) GAC filter. The pressure tank maintains pressure in the plumbing. The water pressure is controlled by the gauge, which regulates the operation of the submersible pump. A sediment filter is mounted in the water line before the GAC filter.*

The first order kinetics model presented by Lowry and Lowry (1987) can be utilised when dimensioning adequate bed volumes for GAC filters. Table III describes the conditions of an arbitrary household that needs to remove radon from the water. The dimensioning of the filter is presented accordingly.

Table III. *An example of an arbitrary household where GAC filtration is to be applied. The parameters affecting the filter size are the daily water usage (3 weeks' average), the radon concentration in raw water and the removal efficiency desired.*

Parameter	Value	
Water usage	600	L/d
^{222}Rn concentration in water	5,000	Bq/L
Desired effluent concentration	10	Bq/L
Removal efficiency	99.8	%
K_{ss} of the GAC	2.5	h^{-1}

Applying Equation 4, the result is

$V_b = -\frac{Q}{K_{ss}} \ln(\frac{C_t}{C_0})$, where

Q is 600 L/d = 25 L/h,
K_{ss} is 2.5 h^{-1},
C_t is 10 Bq/L, and
C_o is 5,000 Bq/L. Hence,

$$V_b = -\frac{25 \text{ L h}^{-1}}{2.5 \text{ h}^{-1}} \cdot \ln(\frac{10 \text{ Bq/L}}{5000 \text{ Bq/L}}) = 62 \text{ L} .$$

The filter housing that is used for GAC filtration is sometimes equipped with automatic backwashing. Filter housings without this feature, however, is cheaper and more common. The filter vessels are usually made of fibreglass or fibreglass-strengthened plastic. They generally operate in a down-flow mode, which means that the water enters the GAC bed from above and flows down through the bed. A riser tube on the bottom of the vessel allows the water to flow up again into the plumbing. Vessels equipped with a backwash system need free volume, that is about one third of the total vessel volume. During backwashing, a three-way valve directs water from the well through the riser tube onto the bottom of the tank. The water rises through the carbon bed and rinses off particulates and metal precipitates. The hydraulic capacity of the filter depends on the grain size of GAC and the diameter of the filter housing. Typically, the capacity of 40–60-litre GAC filters is approximately

12 L/min minimum. The two types of GAC filters are presented in Figure 3.

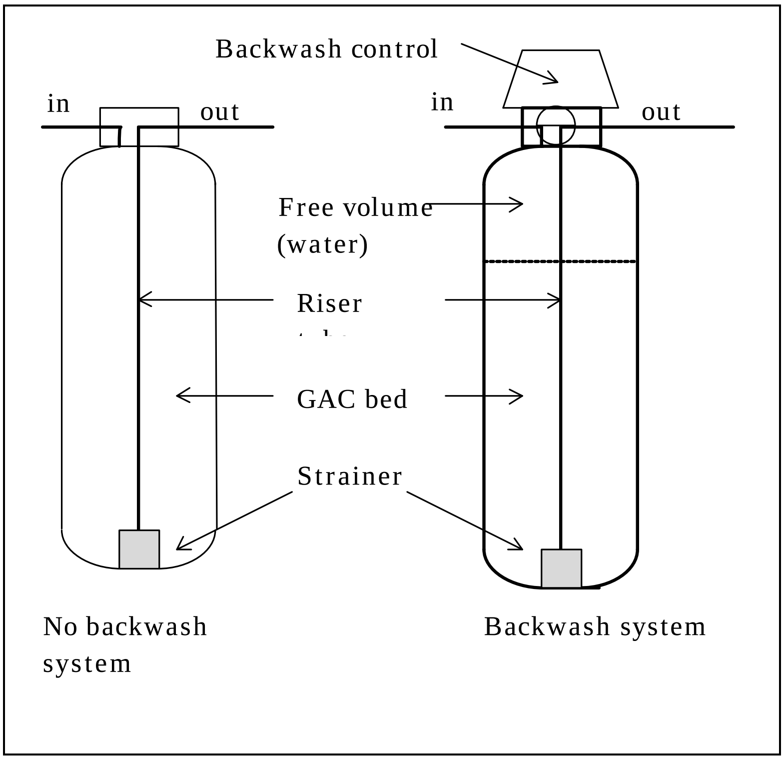

Figure 3. *A schematic representation of the two types of GAC filters.*

Before the filter vessel is commissioned, the carbon must be rinsed thoroughly in order to remove carbon dust and particles that are not retained by the strainer in the vessel. Soaking the carbon is important to avoid channelling of the bed. Generally, a few days' soaking and rinsing is sufficient.

GAC filters with automatic backwashing are often employed when the household water contains a high concentration of iron. During backwashing iron precipitates and other particulates are removed from the filter bed and drained into the sewer. Backwashing, however, should not be performed too frequently. During backwashing, the bed undergoes a very high radon loading. This causes a loss in subsequent removal efficiency due to desorption of radon from the bottom of the bed. Field data indicate that backwashing annually is sufficient in most cases (Kinner et al. 1990).

2.6 Effect of GAC filtration on water quality

Kinner et al. (1990), Myllymäki (1996) and Myllymäki et al. (1999) monitored changes in water quality when GAC filtration was applied to remove radon from water supplies. In Table IV, the main results of the influence of GAC filtration on water quality are summarised. The water quality parameters are explained and the target values or maximum contaminant levels set by the Ministry of Social Affairs and Health in Finland are given in APPENDIX 1 (Sosiaali- ja Terveysministeriö et al. 1994).

Reasoner et al. (1987) studied bacteria in water after water treatment with point-of-use activated carbon filters impregnated with copper and silver. After a new GAC filter has been installed, bacterial populations begin to develop on the GAC. The development of bacterial populations is affected by the length of time a filter is in service, the water temperature, the flow rate, the material of which the filter media are made, and the quality of the influent. According to Reasoner et al. (1987), the microbiological quality of the water should be investigated at different times of day (e.g. morning, afternoon and evening) and the samples should be collected at frequent intervals to determine how the microbiological quality of water changes as the water is used.

By removing organic matter, a GAC filter provides a substratum for microbiological growth (Kinner et al. 1990, Reasoner et al. 1987). Furthermore, iron is removed very efficiently during filtration, which may cause fouling in long-term use. This, however, has not been reported.

Table IV. *Summary of the effects of GAC filtration on water quality.*

pH	Immediately after commissioning the filter, the pH value may increase. Later the pH does not change significantly as water passes through GAC units.
Alkalinity	Alkalinity can initially increase but will return to near influent value after a few weeks' operation. If the pH value of the influent decreases, the alkalinity may increase temporarily.
Calcium	Calcium is initially retained on GAC but will soon break through and the concentration in influent and effluent become the same.
Turbidity	Turbidity decreases during GAC filtration. The opposite effect, however, has also been occasionally observed.
Iron	GAC filtration removes iron very efficiently, nearly 100% reduction has been documented. Coring experiments have shown that most of the iron is retained as ferric hydroxide in the top of the GAC bed.
Manganese	Concentration of manganese does not significantly change during GAC filtration
Organic Matter	Organic matter can be removed effectively with GAC filtration. The removal efficiency decreases over time and after the breakthrough there is a potential to have greater concentrations of organic matter in the effluent than in the influent.
Phosphate	It has been noticed that some phosphate is dissolved from certain brands of GAC during the first few weeks in service.
Potassium	It has been noticed that some potassium is dissolved from certain brands of GAC during the first few weeks in service.
Electric Conductivity	Initially electric conductivity increases, but after a few weeks in service conductivity in influent and effluent shows no significant difference. This is caused by dissolution of ions such as PO_4^{3-} and K^+.
Magnesium	No significant changes have been observed.
Sodium	No significant changes have been observed.
Total hardness	Total hardness may decrease during the first few weeks in service.
Fluoride	No significant changes have been observed.
Chloride	No significant changes have been observed.
Microbial numbers	It has been documented that GAC filters are capable of supporting microbial populations. Coring studies have shown that spent GAC batches contain large microbial numbers, which suggests that micro-organisms could potentially be released into the water passing through the unit.

2.7 GAC unit as a source of gamma radiation

Radon and its daughters build up in the GAC unit. The short-lived progeny of radon (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po) come into secular equilibrium with radon in about four hours. The isotopes of lead and bismuth, which are beta emitters, also emit gamma radiation. The daughter products formed are quantitatively retained on GAC. Thus, the bed becomes a source of gamma radiation (Lowry et al. 1989). For example, under the conditions set out in Table III the activity of radon accumulated (and the short-lived progeny) in steady-state is about 16 MBq. The formation of steady-state is presented in Figure 4. The activity of radon can be calculated from the equation

(5) $X_{ss} = \frac{Q(C_0 - C_{ss})}{l}$, where

X_{ss} is the activity of radon accumulated in steady-state operation in Bq,
Q the volumetric flow rate in L/d,
C_0 the influent activity in Bq/L,
C_{ss} the effluent activity in Bq/L in steady-state, and
l the decay constant of radon, 0.1813 d^{-1} (Lowry and Brandow 1985).

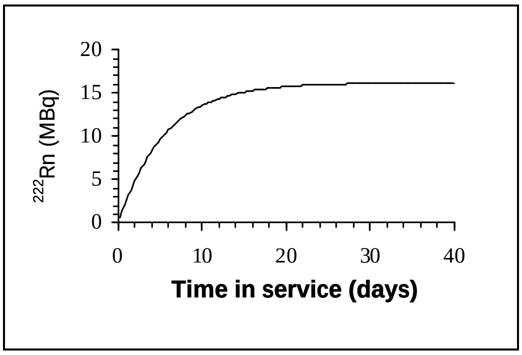

Figure 4. *The accumulation of radon and formation of steady-state in a GAC filter as the water usage is 600 litres per day, influent activity 5,000 Bq/L and effluent activity 10 Bq/L.*

In continuous use, the steady-state is always obtained in about three weeks. As can be seen in Equation 5, the amount of radon accumulated in the filter, and hence the intensity of gamma radiation in steady-state, is dependent on water consumption and the concentration of radon in influent. Most of the radon and the short-lived progeny are retained in the top few centimetres of the GAC bed. Therefore, the highest intensity of gamma radiation is emitted there. Figure 5 illustrates typical profiles of the dose equivalent rate on GAC filters (Turtiainen 1999).

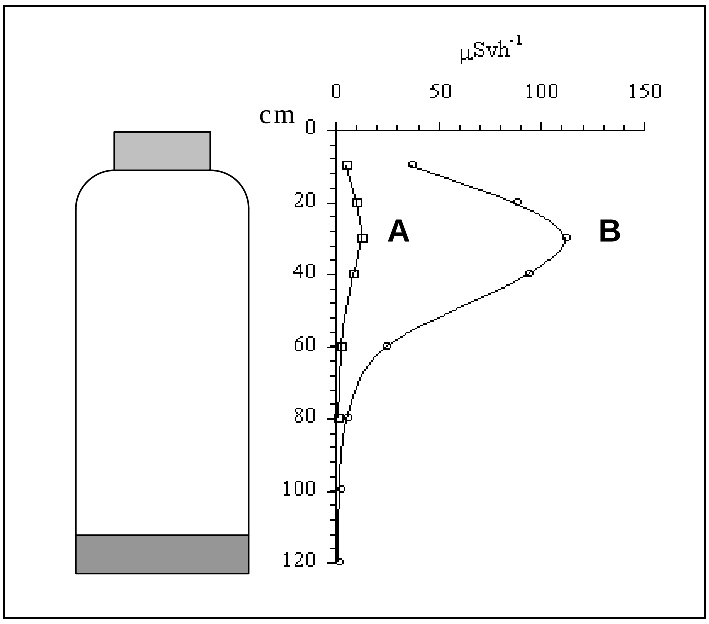

Figure 5. *Profiles of the dose equivalent rate on the surfaces of two GAC filters, A and B. Filter A is used in a secondary residence, and B is in permanent use. Due to the greater water usage, the dose rate on Filter B is much higher (Turtiainen 1999).*

The intensity of radiation originating from such daughter products that are retained on GAC directly from the influent, is negligible. Assuming full secular equilibrium and 100% adsorption for radon progeny in Table III, and applying Equation 3, an activity of 80 kBq is obtained for ^{214}Pb in steady-state. This is only 0.5% of the activity of ^{214}Pb resulting from radioactive decay of radon and the subsequent formation of secular equilibrium (16 MBq).

In 1987, a computer program called *Carbdose* that models radon removal by a domestic style GAC filter was written by Rydell et al. (1989). The program calculates several parameters including estimated health risks, the probable dose rate at specific distances greater than 91 centimetres, the distance at which the probable dose rate is less than the NCRP (the US National Council on Radiation Protection) guideline, and the in-growth rate of ^{210}Pb on the filter.

The “probable dose” is based on the configuration of a domestic style fibreglass or plastic GAC tank, 137 cm tall with an inside diameter of 25 cm and containing 57 litres of activated carbon. The “probable dose” considers the self-absorption of gamma radiation by the carbon and water contained in the GAC filter, the geometry of the filter and the resultant dose build-up as well as the air absorption. It is based on a conservative calculation wherein all the activity is contained in a cylindrical layer of carbon 25 cm in diameter by 12 cm high located on the top of the carbon column. Distances are measured from the tank wall.

It is difficult to estimate the maximum dose rate on the vessel surface because factors such as vessel geometry and steepness of the dose rate profile (affected by the rate of adsorption and water usage) have an effect on the result. The radiation is not emitted by a point-source; therefore reliable estimates of the dose rate can only be obtained at a distance of approximately one metre. Figure 6 illustrates the effect of water usage and the radon concentration in influent on the most probable dose rate at a distance of one metre, assuming the radon removal efficiency is 99.5%.

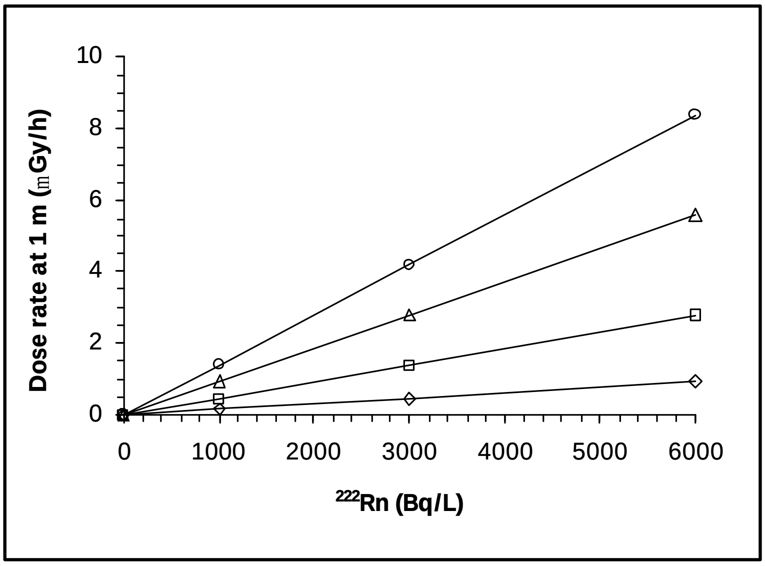

Figure 6. *The probable dose rates at a distance of one meter from a GAC filter with different daily water usage and the radon concentration in influent and assuming 99.5% removal efficiency (Carbdose v4.0).*

GAC filtration has also been applied on a small water works scale (Kinner et al. 1989). Exposure received by the workers at the treatment plant was monitored by film dosimeter badges. The data indicated that exposure problems were potentially significant; while changing GAC the workers were exposed to 0.4–1 mSv in a period of 1.5 hours.

Different radiation shields have been considered. Lead attenuates gamma radiation most efficiently. Lead, however, is expensive and, as a heavy metal, harmful to health. Bricks and concrete can be used, but the thickness of the shield must be considerably greater than for lead. A case made of concrete or bricks must be disassembled when the spent GAC batch is changed. A water jacket can be built by placing the GAC vessel in a tank with an outlet. Water attenuates gamma rays, and is cheap (Kinner et al. 1990, USEPA 1987). Nevertheless, it is not always sufficient to eliminate the problems associated with residential radiation exposure, especially when the influent radon activity is high. Therefore, according to Rydell et al. (1989), radon removal applying GAC filtration often remains a viable treatment only at the low end of the radon concentration range.

2.8 Accumulation of radionuclides on GAC

Studies have shown that nearly 100% of the ^{210}Pb produced in the decay of radon is retained on GAC (Watson and Crawford-Brown 1991). The accumulation of ^{210}Pb can be calculated based on the constant activity of radon in the steady-state (Equation 5) and the in-growth rate of ^{210}Pb:

(7) $A_{Pb} = \frac{Q \cdot (C_0 - C_t)}{l_{Rn}} \cdot (1 - e^{-l_{Pb} \cdot t})$, where

A_{Pb}	is the activity of ^{210}Pb accumulated in the filter (Bq),
Q	is the volumetric flow rate (L/d),
C_0	is the average steady-state activity of radon in influent (Bq/L),
C_t	is the average steady-state activity of radon in effluent (Bq/L),
l_{Rn}	is the decay constant of radon (0.181 d^{-1}),
l_{Pb}	is the decay constant of ^{210}Pb ($8.52 \cdot 10^{-5}$ d^{-1}) and
t	is the time the filter was in service (d).

If the radon concentration in influent and effluent or the water use vary greatly, the more accurate method of calculating the activity of ^{210}Pb in spent GAC is to determine the total amount of radon adsorbed on GAC. This activity is converted into ^{210}Pb activity by decay constants and corrected by radioactive decay:

(8) $A_{Pb} = \frac{l_{Pb}}{l_{Rn}} \cdot e^{-\frac{l_{Pb} \cdot t_s}{2}} \cdot \sum_i Q_i \cdot (C_{0i} - C_{ti}) \cdot t_i$, where

A_{Pb}	is the activity of ^{210}Pb accumulated in the filter (Bq),
l_{Rn}	is the decay constant of radon (0.181 d^{-1}),
l_{Pb}	is the decay constant of ^{210}Pb ($8.52 \cdot 10^{-5}$ d^{-1}),
t_s	is the total time the filter was in service (d),
Q_i	is the flow rate during sampling interval i (L/d),
C_{0i}	is the radon concentration in influent during sampling interval i (Bq/L),
C_{ti}	is the radon concentration in effluent during sampling interval i (Bq/L), and
t_i	is the duration of sampling interval i (d).

The dry weight of the carbon in a typical 40-litre GAC filter is about 20 kg. If daily water usage is 500 L/d and the radon concentration in influent is 3,000 Bq/L, the specific activity of ^{210}Pb in the carbon is approx. 40 Bq/g after three years in service. The ^{210}Pb progeny (^{210}Bi, ^{210}Po) are also present; thus the total specific activity is about 110 Bq/g.

At present, there are no regulations in Finland specifically applicable to the disposal of spent GAC batches containing natural radionuclides. In the USA, the U.S. Nuclear Regulatory Commission (NRC) and the U.S. Department of Transportation (DOT) consider materials whose specific activity exceeds 74 Bq/g as low-level radioactive waste which must be disposed of appropriately.

2.9 Longevity of a GAC bed

Lowry et al. (1990) have reported GAC filters that have been in service for several years without any sign of decreased efficiency in radon removal. This is a consequence of the steady-state performance of the GAC unit: as radon decays, more active sites are freed for adsorption. In a way, the GAC bed is constantly regenerated by radioactive decay.

It has been predicted that other factors, unrelated to ^{222}Rn, will ultimately control the life of the GAC bed. These factors could be fouling by bacteria or ferric hydroxide precipitates, channelling of the bed, loss of carbon due to backwashing, or clogging of the filter by particles (Kinner et al. 1990, Lowry and Brandow 1985).

Some GAC units have failed prematurely (Lowry et al. 1990). Reasons for this, however, have not been found among the factors mentioned above. Lowry et al. (1988) reported that high gross alpha content of influent can impair the radon removal efficiency. The results suggested that the GAC filter's removal efficiency for radon gradually becomes lower and finally attains a constant value (steady-state) after the breakthrough of alpha-emitters (mainly uranium) has occurred.

The effective service time of a GAC batch is most likely dictated by the amount of ^{210}Pb and its progeny, ^{210}Bi and ^{210}Po accumulated in the filter. This is subject to the national regulations concerning radioactive wastes.

3 MATERIALS AND METHODS

3.1 Test locations

Two commercial companies installed GAC filters at several test locations (July 97–June 98), which were all private homes or vacation homes (Table V). Four of the test locations were also monitored in the study conducted by Myllymäki et al. (1999). Most GAC filters were installed as such, without having any pre-treatment units prior to GAC filtration. Two GAC filters were equipped with both a sediment filter and a backwash system and one GAC filter was equipped with a sediment filter. In addition, two test locations had a 21-litre anion exchange unit for removing uranium installed before the GAC filter. Flow meters and sampling taps for influent were installed in most test locations. Whenever an anion exchanger was installed before the GAC filter, a sampling tap was installed between the two filters. In Table V test location C(b) is the same household as test location C(a) but with a new filter combination and the GAC batch changed.

GAC filters operated under normal plumbing pressure, 2–6 bar. They were installed after the pressure tank; thus all household water became treated (point-of-entry). The filter vessels were made of fibreglass-strengthened plastic and operated in the down-flow mode. The hydraulic capacity of 39- and 63-litre GAC filters was 12.3 L/min minimum.

The carbon type (from Bang & Bonsomer Oy) used during this study was selected based on the results of a study carried out by Myllymäki et al. (1999). At test locations C(a) and F the GAC beds had a small proportion of another carbon (from F.A.W. Jacobi Ab) that had the same K_{ss} constant. The bed sizes were calculated applying the first-order kinetics model presented by Lowry and Lowry (1987). The available sizes of the filter vessels, however, determined the final volumes of the carbon beds.

Table V. *Installation data at the test locations.*

Test location	GAC volume (L)	Residence type	Placement	Flow meter	Back-wash	Sediment filter	Anion exch.
A	39	permanent	cupboard	–	–	–	–
B	40	permanent	tech. room	x	x	x	–
C(a)	39	permanent	cellar	x	–	–	–
C(b)	39	permanent	cellar	x	–	x	x
D	39	permanent	cupboard	x	–	–	–
E	40	vacation	cellar	x	x	x	–
F	63	vacation	cellar	–	–	–	–
G	63	permanent	shed	x	–	–	–
H	39	permanent	cellar	x	–	–	x
I	63	permanent	tech. room	x	–	–	–
J	63	permanent	garage	x	–	–	–
K	63	permanent	tech. room	x	–	–	–
L	63	vacation	cellar	x	–	x	–

installed (**x**), not installed (–).

It has been documented that iron and organic substances are removed during GAC filtration (Kinner et al. 1990). Bacteria occur in all groundwater and they can digest biodegradable organic carbon retained in the filter (Reasoner et al. 1987). Manganese and iron can form precipitates in water treatment units (Kinner et al. 1990). High gross alpha levels have been observed to lower the adsorption rate of radon on GAC (Lowry et al. 1988). When GAC filtration is applied to mitigate high levels of radon, the potential hazard of elevated effective dose to the residents increases (Rydell et al. 1989). Therefore, the main criteria for selecting the test locations were the concentration of radon, iron, manganese, uranium and organic matter in the raw water. The desired groundwater types are set out in Table VI (Weingartner and Staubmann 1997). Type 1 represents good water and Type 4 the worst case in terms of water treatment. Any high concentrations of U, ^{226}Ra, ^{210}Po and ^{210}Pb could not be included because knowledge concerning their removal was limited. The selected test locations and their main water quality parameters are listed in Table VII.

Table VI. *Classification of water types in respect of the concentration of radon, iron, manganese and organic matter according to Weingartner and Staubmann (1997). Type 1 represents good water and Type 4 the worst case in terms of water treatment. The SOC (the spectral absorption coefficient at 254 nm, in m*$^{-1}$*) suggested for organic matter has been converted to TOC (mg/L) by ratio 1* **:** *2.5.*

		Type 1	**Type 2**	**Type 3**	**Type 4**
^{222}Rn	Bq/L	> 5,000	1,000–5,000	1,000–5,000	> 5,000
Fe	mg/L	< 0.2	> 1.0	< 0.2	> 1.0
Mn	mg/L	< 0.05	> 0.5	< 0.05	> 0.5
TOC	mg/L	< 1.6	< 1.6	> 4	> 4

Table VII. *The main water quality parameters of raw water at the test locations during the first sampling.*

Test location	**^{222}Rn (Bq/L)**	**Fe (mg/L)**	**Mn (mg/L)**	**TOC (mg/L)**	**U (mg/L)**
A	2,000	0.021	0.018	–	0.052
B	4,000	0.410	0.120	1.5	0.013
C(a)	3,000	0.016	0.064	2.3	0.210
C(b)	3,000	0.016	0.064	2.3	0.210
D	3,700	0.089	0.270	3.4	0.035
E	910	0.032	0.011	1.1	0.017
F	3,000	0.019	0.024	2.4	0.046
G	5,100	0.033	0.027	1.5	0.070
H	1,800	0.034	0.002	2.5	0.260
I	2,000	–	–	–	–
J	4,600	–	–	–	–
K	1,300	0.70	–	–	–
L	5,800	0.16	0.066	2.1	0.007

(–) not determined

As can be seen in Table VII, the desired water types could not be covered. Waters with very high concentrations of iron, manganese and TOC could not be found among the locations willing to participate in the study. Test locations B and K represent water with fairly high iron content, test location D water with high TOC and Mn. Water at test locations B and D was slightly saline. The chloride and sodium concentrations were 115 mg/L and 50 mg/L at test location B and 73 mg/L and 94 mg/L at test location D, respectively.

3.2 Sampling and analysis methods

The radon sampling protocol was as follows: Generally, about 100 litres of water was made to flow with a flow rate that was recorded. 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 (Kitto 1994). The sample was injected into a liquid scintillation vial pre-filled with scintillation cocktail. The sample was analysed in the laboratory using an EG&G Wallac Guardian 1414 liquid scintillation counter (Salonen and Hukkanen 1997).

Gross alpha and radium were determined with a low-background liquid scintillation spectrometer 1220 Quantulus™ from Wallac (Salonen and Hukkanen 1997). The spectrometer is equipped with an anti-coincidence guard counter and a pulse shape analyser (PSA). The sample was prepared by evaporating (with a freeze-dryer) 38 mL of water in a Teflon-coated polyethylene vial until it was dry. The residue was dissolved in 1 mL of 0.5 M HCl, to which 21 mL of scintillation cocktail (OptiPhase HiSafe 2, Wallac) was then added. The sample was counted (for 6 hours) one month after the sample had been prepared.

During the one month period after the sample preparation ^{226}Ra attains equilibrium with its daughter product ^{222}Rn. ^{214}Po, which is also in equilibrium, has a high α-energy (7.7 MeV) and can be detected as a separate peak in the α-spectrum. ^{226}Ra activity was calculated from the ^{214}Po peak with an efficiency of 86%. Gross alpha (uranium, radium and polonium) activity was calculated from the α-spectrum, from which the counts caused by the short-lived alpha emitting daughter products (^{222}Rn, ^{218}Po, ^{214}Po) of ^{226}Ra were subtracted. The lower limits of detection at 95% confidence level were 0.02 Bq/L for gross alpha and 0.01 Bq/L for radium.

Uranium was determined using radiochemical separation and alpha spectrometry (Sill and Williams 1981). The water sample was concentrated applying iron scavenging. The precipitate was dissolved in concentrated HCl and purified by ion exchange separation (Dowex 1x4, 50/100 mesh). The sample was prepared by co-precipitating uranium as fluoride with CeF_3. The sample was counted with an AlfaAnalyst (Canberra) alphaspectrometer. ^{232}U was used as a yield tracer.

Both lead and polonium were determined using spontaneous deposition on a silver disk, and alpha spectrometry (Häsänen 1977). The water sample was pre-concentrated with evaporation. The initial ^{210}Po was deposited on a silver disk and counted with the AlfaAnalyst. The residual solution was stored for 200 days to allow the in-growth of ^{210}Po that is a daughter product of ^{210}Pb. The deposition was carried out again and the ^{210}Po formed during the in-growth period was counted. The activities were then calculated.

Several water quality parameters that were selected using the results of the previous studies (Kinner et al. 1990, Myllymäki 1996) were monitored. The selected parameters were mostly those that had been affected by GAC filtration (Chapter 2.6). The most important parameters (Fe, Mn, TOC, pH and heterotrophic plate count) were monitored regularly, as was the temperature. Other radionuclides of the uranium series (^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb and ^{210}Po) were also determined regularly. In addition, SiO_2, redox potential, O_2, CO_2, colour, $KMnO_4$, acidity, PO_4^{3-}, NO_3^- and NH_4^+ were occasionally analysed. Water quality samples were taken according to the instructions provided by the accredited laboratory that performed the analyses (City of Helsinki Environment Centre).

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

3.3 Gammaspectrometric measurements for spent GAC

Two batches of spent GAC were investigated in order to determine the activity of radionuclides accumulated in the carbon. The first batch (14 litres, 8.2 kg dry-weight) had operated without a sediment filter for three months when various brands of GAC were investigated (Myllymäki et al. 1999). Radon concentration in the influent had varied significantly (840–9,300 Bq/L). Therefore, the theoretical ^{210}Pb activity varied between 42 and 88 kBq (Equation 8).

The GAC batch was divided into four parts. Three 0.5-litre samples were cored from the top of the bed. The rest of the bed was homogenised, from which the fourth sample was taken. Samples were dried under an infra-red lamp and measured in Marinelli geometry (500 mL) with an n-type HPGe detector. The calibration error was ±10% because of the low gamma energy (46.5 keV) of ^{210}Pb.

The second batch of activated carbon (39 L) had been used for 407 days at test location C(a). Water usage, radon concentration and ^{210}Pb concentration in both influent and effluent were monitored at 1 to 3-month intervals (Table VIII). The theoretical ^{210}Pb activity was 240 kBq (Equation 8).

Table VIII*. Sampling dates, flow meter readings and the radon and lead concentration in influent and effluent at test location C(a).*

Date (d.m.y)	Time in service (d)	Water treated (m^3)	^{222}Rn concentration (Bq/L)		^{210}Pb concentration (Bq/L)	
			influent	effluent	influent	effluent
1.7.1997	0	0	–	–	–	–
9.10.1997	100	17	2,596	190	0.375	0.040
17.12.1997	169	50	2,746	180	0.640	0.045
25.3.1998	267	104	2,628	285	0.228	0.031
20.4.1998	293	115	–	290	–	–
25.5.1998	328	133	–	340	–	–
14.7.1998	378	156	2,660	170	0.132	0.015
12.8.1998	407	168	–	–	–	–

(–) not determined

The spent batch of GAC was homogenised and part of it was dried under an infra-red lamp. Four sub-samples were taken for gammaspectrometric determination. Two samples were counted in Marinelli geometry (500 mL) and two in cylindrical containers (diameter 43 mm, height 25 mm) with an n-type HPGe detector.

4 RESULTS AND DISCUSSION

4.1 Radon removal

Radon was removed very efficiently by most filter combinations (Table IX). Six units out of thirteen were capable of removing more than 99.9% of radon, which proves that the filters were properly dimensioned. Figures 7 and 8 set out the radon removal efficiencies as a function of the volume of treated water at the test locations where the residents live permanently. The volume of treated water is presented as bed volumes. One bed volume (BV) is equivalent to the volume of wet carbon inside the vessel.

Table IX. *The ranges of radon concentration in influent and effluent during the operating period of the filters, the radon reduction at the most recent sampling and the corresponding treated water volume (in bed volumes).*

Test location	^{222}Rn Influent (Bq/L)	Effluent (Bq/L)	Time in service (months)	Water treated (BV)	Radon reduction (%)
A	1,900–2,850	1.2 – 41	27	~8,000	98.5
B	3,500–4,200	12 – 190	11	5,350	98.9
C(a)	2,600–2,700	150 – 340	13	4,050	92.9
C(b)	1,850–3,100	<0.4 – 31	11	4,460	98.6
D	2,750–4,100	<0.4 – 15	23	3,030	>99.9
E	910–1,100	<0.4 – 3.1	8	150	>99.9
F	1,700–3,000	0.9 – 2.2	13	~400	>99.9
G	5,100–7,400	<0.4 – 2.2	23	3,790	>99.9
H	1,600–2,200	1 – 37	21	4,860	97.8
I	1,300–2,200	1 – 5.3	15	1,501	99.7
J	4,100–4,600	1.2 – 2.6	23	6,650	>99.9
K	1,300–1,600	<0.4 – 0.5	23	5,550	>99.9
L	3,040–6,380	45 – 99	9	389	98.5

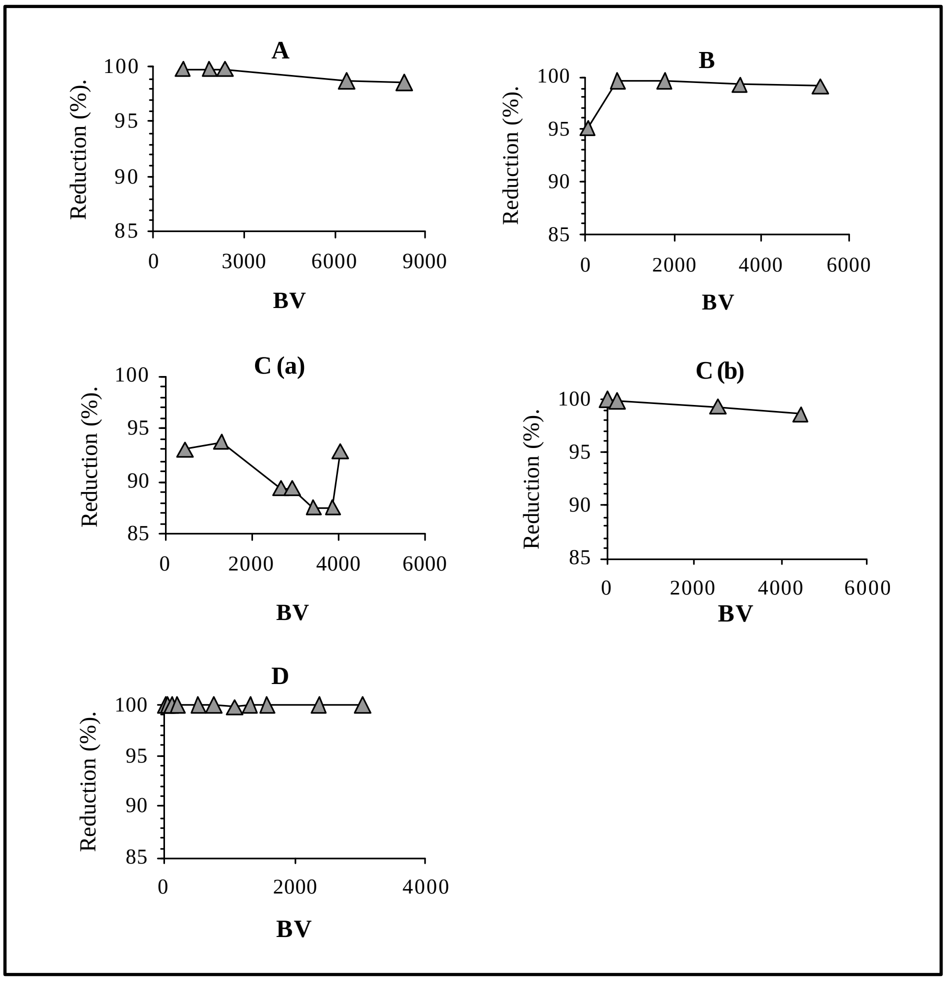

Figure 7. Radon reduction at test locations A, B, C(a), C(b), D, and G when different amounts of water had been treated. The volume of treated water is expressed as bed volumes. One bed volume is the volume of wet GAC in the vessel (Table IV). The BV's for test location A were estimated based on 400 L/d consumption.

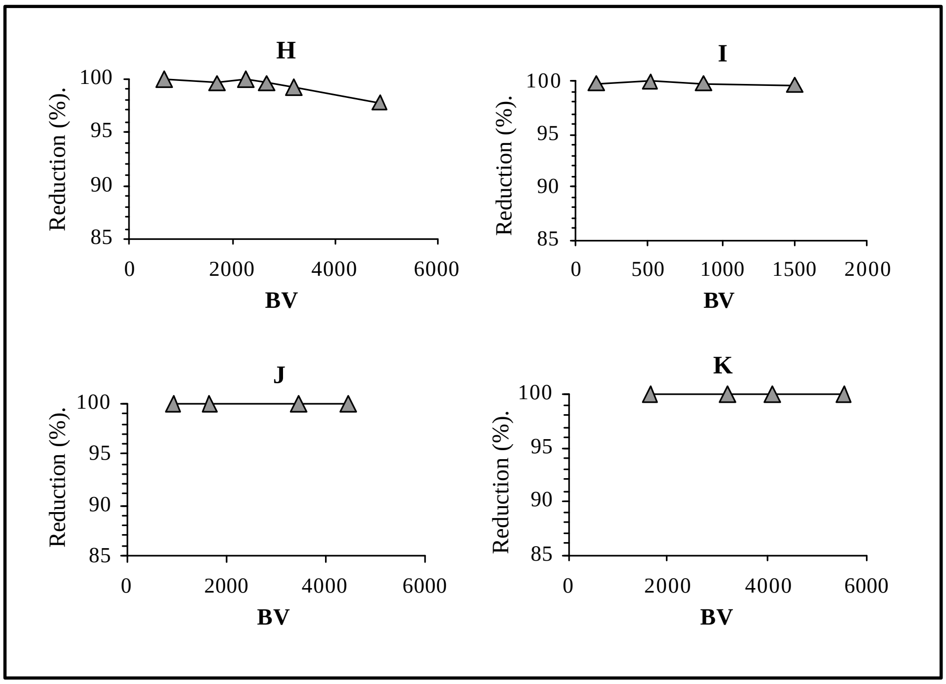

Figure 8. Radon reduction at test locations H, I, J and K when different amounts of water had been treated.

Effluent activity was very low in most cases. The only exceptions were test locations B (12–190 Bq/L), C(a) (160–340 Bq/L), and L (45–99 Bq/L). The removal efficiencies remained close to constant throughout the study except at test locations B and C(a). The first result obtained from test location B (Figure 7), when only 64 bed volumes were filtered, may be explained by insufficient packing of the carbon (channelling). It was not documented if the GAC filter had been backwashed during the installation. If this was the case, desorption of radon from the bottom of the bed might have impaired the removal efficiency at the first sampling. At the subsequent samplings the removal efficiency (99%) was close to the value expected from the previously determined K_{ss} constant.

Test location C(a) had the lowest radon reduction of all the test locations from the beginning of the test period (Figure 7). The reason for this could not be explained with certainty. When the filter vessel was emptied and filled with a new carbon batch, no visible malfunction such as a lumpy

GAC or a loose riser tube was noted. The only water quality parameter that was clearly different from other test locations was uranium (0.2 mg/L). The effect of water quality on radon removal is discussed later in this chapter.

The GAC filters at the two vacation homes did not show impaired removal during the second summer season. In fact, at test location E the radon levels were lower than the detection limit during the second sampling.

The 39-litre GAC filters did not attain constant removal efficiency for radon during peak consumption (Figure 9). When the flow rate was 6 L/min, radon concentration in effluent increased by 0.5 Bq/L for the first 100 litres. For flow rates of 10 and 16 L/min the concentration increased by 4 and 11 Bq/L, respectively. The results, however, proved that the filters were properly dimensioned and that no radon-laden water was released into the water line even during high consumption. For the 63-litre GAC filters the increase of radon concentration during flowing was smaller.

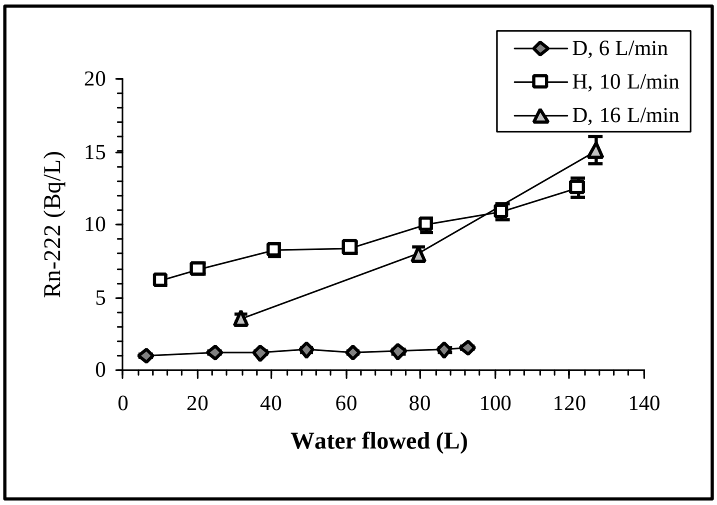

Figure 9. *Radon concentration as a function of the volume of water flowing applying three different flow rates. Both test locations (D and H) had a 39-litre GAC filter.*

The longevity of the GAC bed in terms of radon removal could not be estimated reliably. A straight line was fitted to the data set out in Figures 7 and 8 applying the “least squares” method. The fitting was successful (r^2>0.95) only for the data collected at test locations A, B and C (b). The fitting at test location A is discarded since the flow rate was based on an estimate. The first data point at location B is also discarded due to the divergent result. Provided the linearity in the charts continues, the data suggest that a 5% breakthrough of radon occurs at 16,000 bed volumes (650 m^3) at test location C(b) and 35,000 bed volumes (1,400 m^3) at test location B. Considering the daily water usage, the 5% breakthrough would occur at test location C(b) 3.4 years after the commissioning and at test location B after 6.1 years.

During this study, no true breakthrough of radon was noted. Theoretically, a GAC filter is constantly regenerated in respect of radon due to the radioactive decay, and can therefore continue in service for several years. When competitive substances occupy the active sites of the carbon, radon removal efficiency may decrease. This, however, depends on the other water quality parameters.

4.1.1 Adsorption rate of radon on GAC

As described in Chapter 2.3, the rate of adsorption (the removal rate of radon) can be described with the adsorption-decay steady-state constant, K_{ss}. The higher the K_{ss} constant, the more rapid the adsorption. K_{ss} constants were calculated according to Equation 3 for all the GAC filters that were equipped with a flow meter and were in permanent use. Table X sets out the average daily water use and the empty bed detention time at different test locations on different sampling dates. The removal efficiencies and K_{ss} constants in Table X were calculated for the effluent sample that was collected after approximately 50 litres of water had flowed.

Table X. *The daily water usage, empty bed detention time (EBDT), removal efficiency and* K_{ss} *constants at the test locations on different sampling dates. 100% removal of radon stands for over 99.95% removal.*

Test Location	Date	Water usage (L/d)	EBDT (h)	Removal (%)	K_{ss} (h^{-1})
B	23.9.97	644	1.49	99.7	3.82
	3.12.97	589	1.63	99.6	3.39
	25.3.98	618	1.55	99.3	3.22
	14.7.98	659	1.46	99.1	3.21
C(a)	17.12.97	476	1.97	93.8	1.41
	25.3.98	556	1.68	89.4	1.33
	20.4.98	418	2.24	89.5	1.00
	25.5.98	514	1.82	87.6	1.15
	1.7.98	486	1.92	87.6	1.09
	14.7.98	537	1.74	92.9	1.51
C(b)	9.9.98	431	2.17	99.8	2.87
	2.3.99	525	1.78	99.3	2.78
	14.7.99	551	1.69	98.6	2.50
D	27.11.97	169	5.53	100	1.51
	5.2.98	181	5.16	99.9	1.43
	11.6.98	168	5.58	99.9	1.26
	24.9.98	184	5.09	100	1.55
	11.4.99	157	5.97	100	1.28
	14.9.99	166	5.64	99.9	1.26
G	10.3.98	340	4.45	100	1.94
	18.5.98	318	4.75	100	1.72
	13.8.98	321	4.71	100	1.81
	23.9.98	368	4.11	100	2.11
	15.3.99	334	4.53	100	1.78
	4.10.99	399	3.79	100	2.15
H	29.9.98	282	3.32	99.6	1.64
	8.6.99	337	2.78	97.8	1.38
I	25.6.98	254	5.95	99.9	1.21
	29.9.98	242	6.25	99.9	1.09
	17.3.99	232	6.51	99.7	0.89
J	11.6.98	556	2.72	99.9	2.74
	15.12.98	601	2.51	99.9	2.97
	13.4.99	532	2.84	100	2.87
	10.11.99	660	2.29	99.9	3.27
K	2.6.98	523	2.89	100	2.78
	2.12.98	526	2.88	100	2.78
	23.3.99	513	2.95	100	2.70
	14.9.99	525	2.88	100	2.86

The results in Table X show that the calculated K_{ss} constants differed significantly between different test locations, but remained quite constant at an individual test location. The best adsorption rate was obtained at test location B (3.4 h^{-1} on average) while test location I had the poorest adsorption rate (1.1 h^{-1} on average). Because the volume of the GAC bed and the daily water usage were distinct for each GAC filter, low K_{ss} constants did not necessarily mean poor radon removal efficiency (Figure 10A). For example, at test location I the removal efficiency was always greater than 99.7%. Figure 10 shows how the adsorption rate (K_{ss} constant), empty bed detention time (EBDT) and radon removal efficiency were correlated with each other.

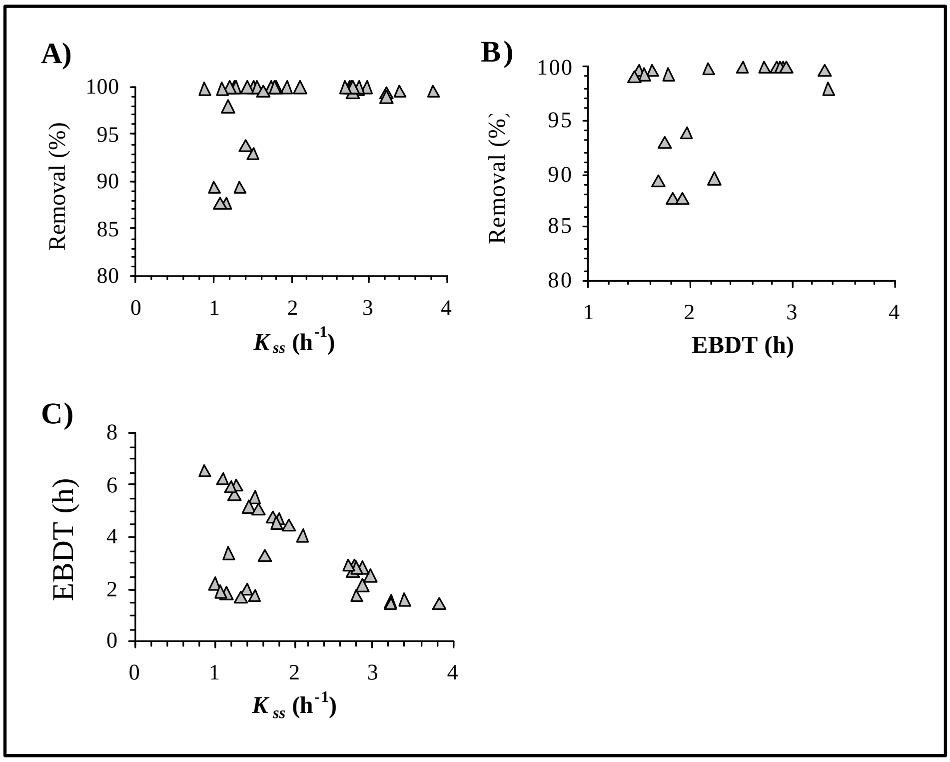

Figure 10. A) *Radon removal efficiency plotted against K_{ss} constant,* ***B)*** *Radon removal efficiency plotted against empty bed detention time (EBDT) and* ***C)*** *EBDT plotted against K_{ss} constant. The data points are from Table X.*

It can be seen in Figure 10C that in many cases an extended EBDT resulted in a low K_{ss} constant. This suggests that the adsorption rate of radon slows down as the radon concentration becomes very low. For low concentration of radon, the reaction that determines the rate of removal process may be not only the adsorption, but also mass transfer, which is affected by the concentration gradient in the vicinity of macropores (see Chapter 2.3). Furthermore, the radon concentration in effluent was in most cases very close to the detection limit, which is why the calculated K_{ss} constants may hold large overall errors. A vessel with several outlets at different bed heights should have been used in order to determine the adsorption rate of an individual GAC unit more accurately.

4.1.2 Effect of water quality on adsorption rate

An ideal situation to determine the effect of water quality on radon removal would have been a set of filters of the same size, with the same radon concentration and the same daily water use but with different influent composition. Naturally, this is not possible in field research. Therefore, the K_{ss} constants set out in Table X are used here to evaluate the effect of different water quality parameters on the adsorption rate of radon and thereby on the removal process.

The effect of the gross-α and uranium concentration on the K_{ss} constant is set out in Figure 11. Gross-α mainly consists of uranium; thus in practice the charts represent the effect of uranium on the adsorption rate. It is apparent that a high concentration of uranium in influent contributes a negative effect on the removal rate of radon.

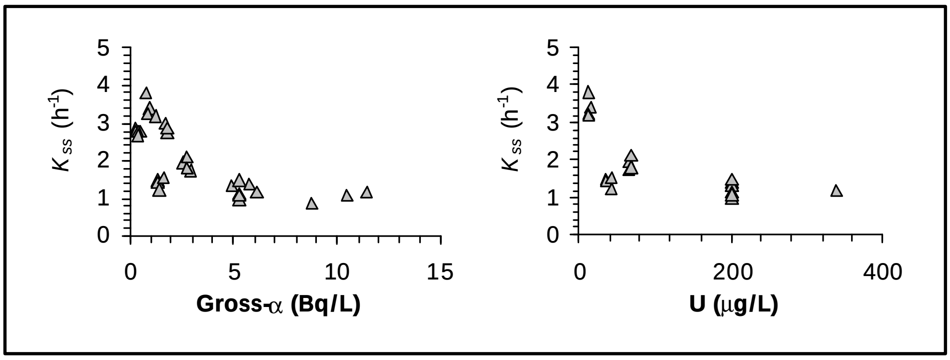

Figure 11. *The gross-a and uranium concentration plotted against the respective K_{ss} constants (all test locations). The results suggest that a high concentration of uranium impairs the adsorption rate of radon on GAC.*

Figure 12 presents the effect of iron, manganese and organic matter (TOC and $KMnO_4$ titration) on the K_{ss} constant. The results suggest that high TOC concentrations may have a negative effect on radon removal. The data, however, are more limited than for the gross-α. The data obtained from $KMnO_4$ titration did not confirm the negative effect of organic matter on radon removal. The $KMnO_4$ titration, however, is not as reliable a parameter for organic matter as is TOC. Organic matter as well as uranium is partly retained during GAC filtration which implies that they have the potential to compete with radon and thereby impair the adsorption of radon (see Chapter 4.3).

The results suggest that the concentrations of iron and manganese have no marked effect on radon removal although iron may be removed very efficiently by GAC filtration (see Chapter 4.3). Ferric hydroxide precipitates can be formed inside the filter where they occur as large flakes and therefore do not potentially clog the GAC micropores. According to the results of this study, manganese is not removed by GAC filtration.

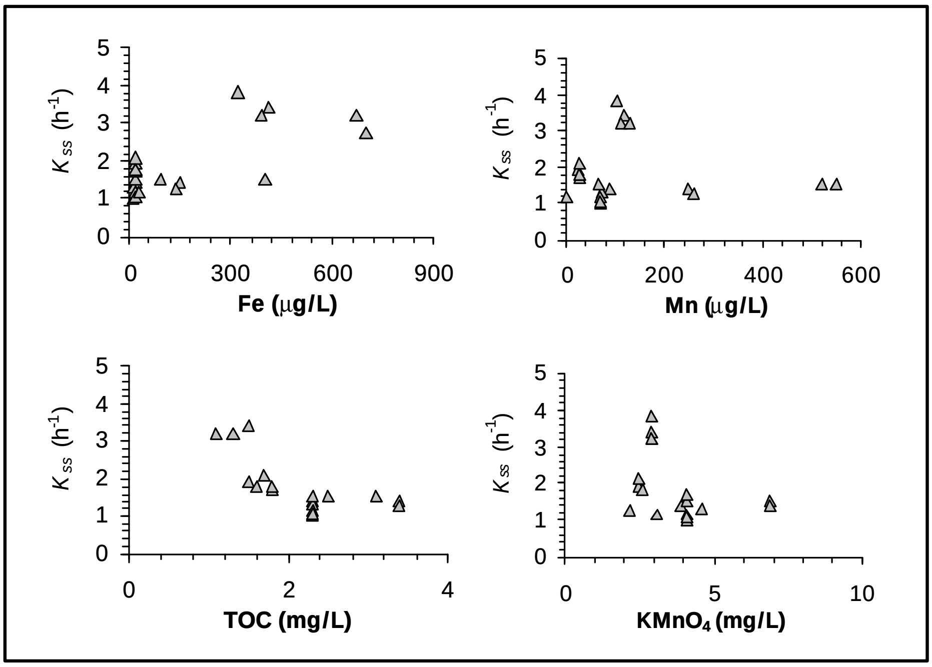

***Figure 12**. Concentration of iron, manganese and organic carbon (TOC and $KMnO_4$ titration) plotted against the respective K_{ss} constants.*

Lowry et al. (1988) reported that high gross alpha levels in influent may decrease the removal efficiency of radon. To investigate the effect of uranium on radon removal in more detail, the GAC filter at test location C(a) was loaded with a fresh batch of GAC. An additional sediment filter (5 microns) and a 21-litre anion exchange unit were installed before the GAC filter. This new installation is referred to as test location C(b). Following this installation, the filter removed more than 99 % of the radon, and the K_{ss} constant changed from value 1.2 h^{-1} to 2.8 h^{-1}. It is evident that the interfering substance in the water was some anionic species, most probably uranium or humus.

It has been reported that GAC filters are capable of supporting large microbial populations (Kinner et al. 1990). The microbiological quality of water was investigated with heterotrophic plate count (HPC) determination. The effect of the waterborne bacteria on the K_{ss} constant is presented in Figure 13. The plate counts are expressed as colony forming units per millilitre (cfu/mL). According to the results, the microbiological quality of the influent has little effect on the removal rate of radon. The microbial density of the influent, however, does not represent the microbial populations inside the filter. In order to investigate the effect of microbes on the adsorption of radon, the biofilms on different carbons should have been studied.

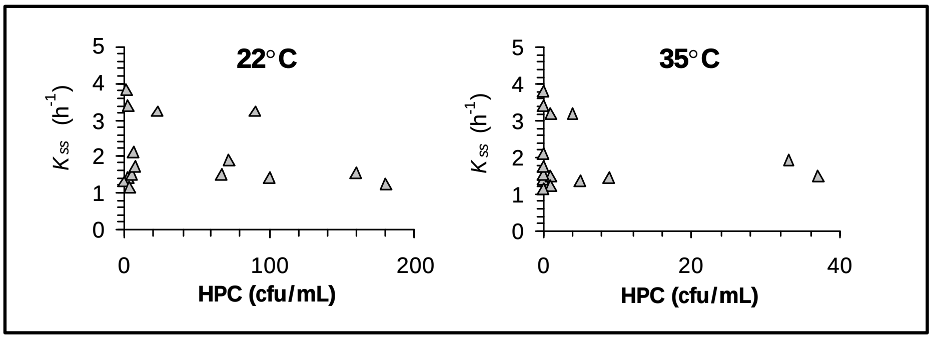

Figure 13. *Heterotrophic plate counts at 22°C and 35°C plotted against the respective K_{ss} constants.*

More data on the anions in influent are needed in order to reach more definite conclusions about their interference in radon removal. Nevertheless, based on the results iron and manganese do not impair the removal rate of radon. Therefore, GAC filtration can be regarded as a viable technique for removing radon too from iron and manganese-rich waters.

High concentrations of uranium and organic matter may lower the adsorption rate of radon. In such cases, the empty bed detention time must be prolonged in order to attain satisfactory removal efficiencies. Based on the results, a K_{ss} constant of 1 h^{-1} should cover all water types when dimensioning the filters. For example, if the radon concentration is 3,000 Bq/L, the water usage 500 L/d and the desired removal efficiency 99%, the volume of the GAC bed must be a minimum of 96 litres.

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

The results revealed that GAC filtration is not a viable technique for removing the long-lived radionuclides of the uranium series along with radon (Table XI). The best reduction was obtained for polonium and the poorest for uranium. The results for the same radionuclide varied between different test locations. Results obtained at the test locations where only gross alpha measurement was used to screen removal of the long-lived radionuclides are set out in Table XII. The results indicated fair or poor reduction for gross-alpha. The only exception was test location L where, according to previous radiochemical determinations, gross alpha mainly comprises ^{210}Po and ^{226}Ra.

Table XI. *Concentration of U, ^{226}Ra, ^{210}Pb and ^{210}Po in influent and effluent at the test locations where regular sampling was carried out.*

Test Location	BV	U (mg/L)		^{226}Ra (Bq/L)		^{210}Pb (Bq/L)		^{210}Po (Bq/L)	
		inf.	eff.	inf.	eff.	inf.	eff.	inf.	eff.
B	741	12.9	8.5	0.20	0.04	0.283	0.009	0.122	0.007
	1,786	14.8	14.8	0.17	0.07	0.371	0.014	0.102	0.015
	3,518	13.0	13.7	0.20	0.07	0.328	0.005	0.154	0.012
	5,348	–	–	0.20	0.10	0.660	0.005	0.454	0.013
C(a)	431	207	181	0.21	0.15	0.375	0.040	0.266	0.045
	1,274	202	212	0.23	0.22	0.640	0.045	0.126	0.042
	2,670	200	211	0.25	0.22	0.228	0.031	0.109	0.051
	4,051	–	–	0.24	0.26	0.132	0.015	0.104	0.033
D	202	–	15.9	–	0.40	–	0.117	–	0.027
	528	35.9	12.8	0.28	0.24	0.354	0.234	0.169	<0.002
	1,069	42.9	31.8	0.26	0.27	0.313	0.195	0.520	0.045
	1,565	–	–	0.54	0.43	0.477	0.162	0.134	0.039
E	41	18.9	1.4	0.05	<0,01	0.461	0.003	0.416	0.003
	151	21.7	0.2	0.05	<0,01	0.014	0.001	0.033	0.006
F	~150	47	34	0.22	0.07	–	–	–	–
	~400	115	112	0.13	0.11	0.075	0.017	0.137	0.047
G	75	70	40	0.30	0.11	–	–	0.455	0.018
	555	67	65	0.32	0.25	0.517	0.149	0.461	0.043
	903	67	68	0.32	0.26	0.616	0.151	0.360	0.031
	1,586	–	–	0.29	0.26	0.451	0.200	0.206	0.047
L	23	–	–	0.19	0.01	0.417	0.085	1.92	0.133
	181	6.5	0.3	0.14	0.14	–	–	–	–

– not determined

Table XII. *Gross alpha and ^{226}Ra concentrations in influent and effluent at the 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	0.29	0.19
	509	6.09	5.51	0.19	0.20
	878	10.6	8.12	0.23	0.25
	1,501	8.78	9.81	0.19	0.17
J	943	1.64	1.01	0.09	0.06
	1,649	1.81	1.54	0.08	0.07
	3,434	1.73	1.45	0.08	0.07
	4,438	1.80	1.03	0.09	0.07
K	184	0.58	0.23	0.08	0.07
	1,656	0.45	0.33	0.15	0.07
	3,180	0.38	0.20	0.14	0.11
	4,087	0.40	0.25	0.11	0.11
	5,545	0.38	0.39	0.13	0.09
L	23	2.32	0.13	0.19	0.01
	181	2.63	0.18	0.14	0.14
	389	1.67	0.46	0.29	0.26

Efficient reduction of uranium was obtained only when less than 200 BV's of water had been filtered, though most filters showed low reduction even then (Figure 14). There seemed to be no particular breakthrough volume for uranium: rather, the retention decreased gradually. When a large amount of water had been treated, the uranium concentration in the effluent was the same or slightly higher than in the influent.

Gammaspectrometric measurements performed on a 39-litre batch of spent GAC (see Chapter 4.7) indicated that a 39-litre batch of GAC is capable of retaining only approx. 2 grams of uranium. The reduction of uranium observed is not likely due to adsorption of uranium that occurs in particulate species because firstly, uranium occurs mostly as soluble anionic carbonate complexes in ground water and secondly, the breakthrough of uranium occurs much sooner than for e.g. turbidity and TOC, which are parameters describing the particles and colloids in water. The retention, therefore, is probably due to precipitation of uranium on GAC or an ion exchange reaction.

The adsorption efficiency for ^{226}Ra varied considerably (Figure 14). Two of the filters removed radium fairly well: 67% at test location B after 3,520 BV’s and 53% at test location K after 1,660 BV’s. Radium mostly occurs as a hydrated cation in ground water. Part of the retention observed, however, may be due to the complexes which radium can form with humus- and fulvic acids because no breakthrough could be observed. Some retention may also occur by adsorption onto ferric hydroxide precipitates that are formed in the filter vessels during filtration. Ion exchange reaction may also be possible. The best adsorption was observed at the test location where the concentration of iron was the highest.

^{210}Pb was removed quite efficiently at test locations B and C(a) but only fairly at test locations D and G. Lead, as well as ^{210}Po, is readily adsorbed on particles, surfaces and colloids. Therefore, it is to be expected that the mechanism by which these radionuclides are removed is particle filtration. The ratio between particulate species and dissolved species may vary in natural waters, which explains the different removal efficiencies obtained.

No clear tendency regarding the removal of uranium, radium, polonium, and lead could be discerned (Figure 14). It is obvious that the chemical forms (speciations) of these radionuclides vary greatly in the waters that were studied. The speciations depend on the physico-chemical properties of the groundwater, e.g. redox potential, pH value, different complexing agents, colloids, bacteria, and salinity.

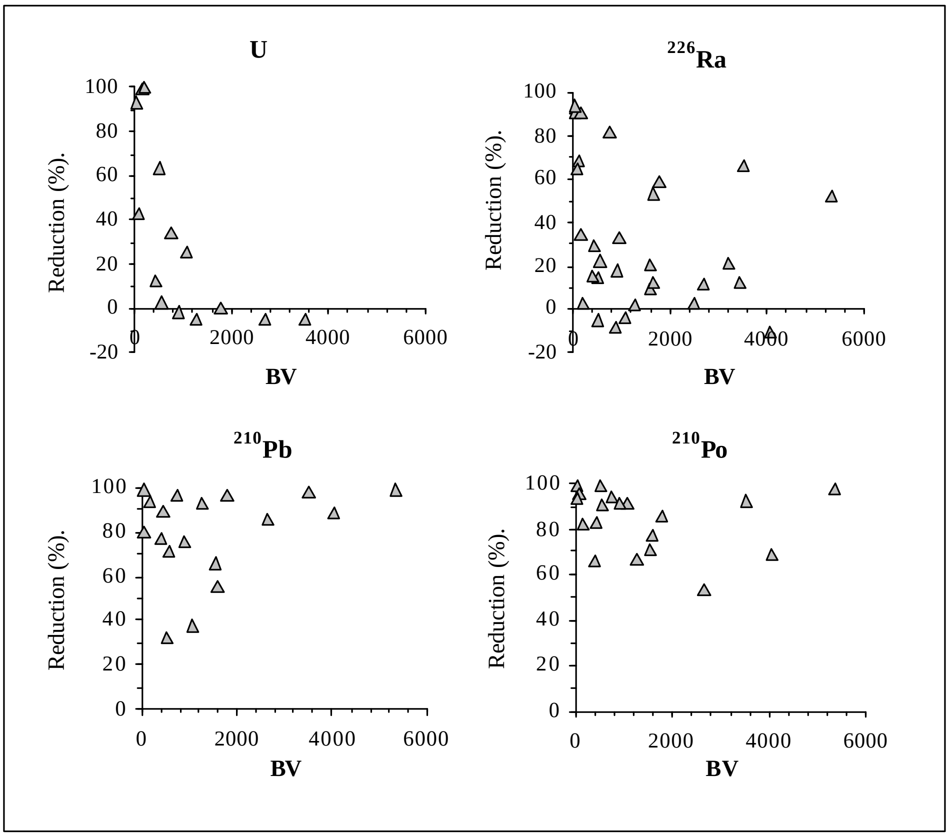

Figure 14. *Uranium, ^{226}Ra, ^{210}Po and ^{210}Pb reduction as a function of treated water (in bed volumes). The charts are a compilation of the results obtained from all the test locations.*

4.3 The physico-chemical quality of treated water

Several water quality parameters were monitored. The most important parameters (Fe, Mn, TOC and pH) were monitored regularly (Table XIII), as was the temperature. Turbidity, electric conductivity, alkalinity and total hardness were determined sporadically (Table XIV). In addition, SiO_2, redox potential, O_2, CO_2, colour, $KMnO_4$, acidity, PO_4^{3-}, NO_3^- and NH_4^+ were occasionally monitored. All the data collected are set out in Appendix 2.

Table XIII. *Fe, Mn, TOC and pH in influent and effluent 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)		pH	
		inf.	eff.	inf.	eff.	inf.	eff.	inf.	eff.
B	1,786	0.410	0.003	0.120	0.110	1.5	0.4	7.7	7.7
	3,518	0.390	0.003	0.110	0.100	1.3	1.0	7.7	7.7
	5,348	0.670	0.003	0.130	0.086	1.1	0.7	7.8	7.7
C(a)	431	0.016	<0.002	0.064	0.070	–	–	7.7	7.7
	1,274	0.020	0.002	0.086	0.068	2.3	1.1	7.6	7.6
	2,679	0.014	<0.002	0.074	0.078	2.3	1.6	7.4	7.4
	4,051	0.015	0.005	0.066	0.057	2.3	1.6	7.6	7.6
D	528	0.150	0.023	0.250	0.250	3.4	1.0	7.1	7.1
	1,069	0.140	0.047	0.260	0.230	3.4	1.0	7.0	7.1
	1,565	0.400	0.036	0.550	0.550	3.1	1.3	6.8	6.9
E	41	0.020	0.004	0.009	0.021	1.1	0.2	8.5	8.4
	151	0.033	0.058	0.009	0.014	1.1	0.2	7.2	7.2
F	~100	0.019	<0.002	0.024	0.029	–	–	8.3	8.2
	~400	0.016	0.004	0.008	0.009	2.4	0.8	8.1	8.3
G	75	0.033	0.003	0.027	0.022	1.5	0.5	8.1	8.4
	555	0.019	0.003	0.024	0.023	1.5	0.4	8.1	8.1
	903	0.017	0.008	0.027	0.026	1.8	0.5	8.1	8.2
	1,586	0.016	0.005	0.025	0.026	1.6	0.5	7.9	7.9
H	1,714	0.008	0.003	0.0026	0.0024	1.4	0.5	6.9	7.1
	2,273	0.005	0.004	0.005	0.004	1.1	0.6	7.1	7.2
	2,683	0.005	0.003	0.0021	0.0013	1.2	0.4	6.6	7.5
L	23	0.160	0.015	0.066	0.005	2.1	0.2	7.3	8.9

(–) not determined

Table XIV. *Turbidity, electric conductivity, alkalinity and total hardness in influent and effluent at different bed volumes at test locations where regular sampling was carried out.*

Test Location	BV	Turbidity (FTU)		El. conductivity (mS/m)		Alkalinity (mmol/L)		Tot. hardness (mmol/L)	
		inf.	eff.	inf.	eff.	inf.	eff.	inf.	eff.
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
	1,786	–	–	53.2	53.1	1.56	1.59	1.40	1.39
	3,518	2.5	0.30	55.4	55.4	–	–	–	–
	5,348	–	–	51.8	51.4	–	–	–	–
C(a)	0	0.14	–	44.5	–	2.3	–	1.51	–
	431	<0.05	<0.05	–	–	–	–	–	–
	1,274	0.05	<0.05	–	–	–	–	–	–
	2,670	0.06	0.07	–	–	–	–	–	–
	4,051	0.09	0.09	–	–	–	–	–	–
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
	1,069	–	–	51.4	53.0	–	–	0.63	0.60
	1,565	–	–	76.8	76.0	–	–	–	–
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	–	–	–	–
F	~100	0.24	<0.05	43.2	45.7	3.01	3.01	0.67	0.59
	~400	0.19	0.10	44.0	44.1	–	–	–	–
G	75	<0.05	0.05	25.4	25.4	1.95	1.92	0.82	0.81
	555	<0.05	<0.05	24.7	24.7	1.94	1.94	0.80	0.80
	903	–	–	24.7	24.7	–	–	–	–
	1,586	–	–	25.4	25.1	–	–	–	–
H	1,714	0.18	–	–	–	1.43	–	–	–
	2,273	0.23	0.23	–	–	1.25	–	–	–
	2,683	0.13	0.08	–	–	1.37	–	–	–
L	23	–	–	29.5	30.3	–	–	–	–

(–) not determined

4.3.1 Iron, manganese, organic matter, pH and temperature

The iron concentration usually decreased 50%–99% during the treatment (Figure 15). In some cases, the removal may have partly occurred in the sediment filter. The best iron reduction was obtained at test location B, where the iron concentration in the influent was high (0.4–0.7 mg/L). At test location E (secondary residence), the iron concentration increased during the second sampling. The higher concentration of iron in the effluent may have been caused by the release of some ferric precipitate in the water line.

The concentration of manganese did not significantly change during GAC filtration (Figure 16). During the first few weeks in service, the concentration of manganese might slightly decrease. After 500 BV's had been treated, an average reduction of 11% was obtained.

TOC indicated fair removal of organic substances with a reduction efficiency of 67%–90% when less than 1,000 BV's were filtered (Figure 17). The reduction efficiency decreased gradually, so that after 2,000 BV's had been filtered it was 23%–67%. $KMnO_4$ titration also indicated good reduction (average 62%).

The pH value changed only slightly during GAC filtration (Figure 18). At test location H, where the influent had the lowest pH values, the increase was the greatest.

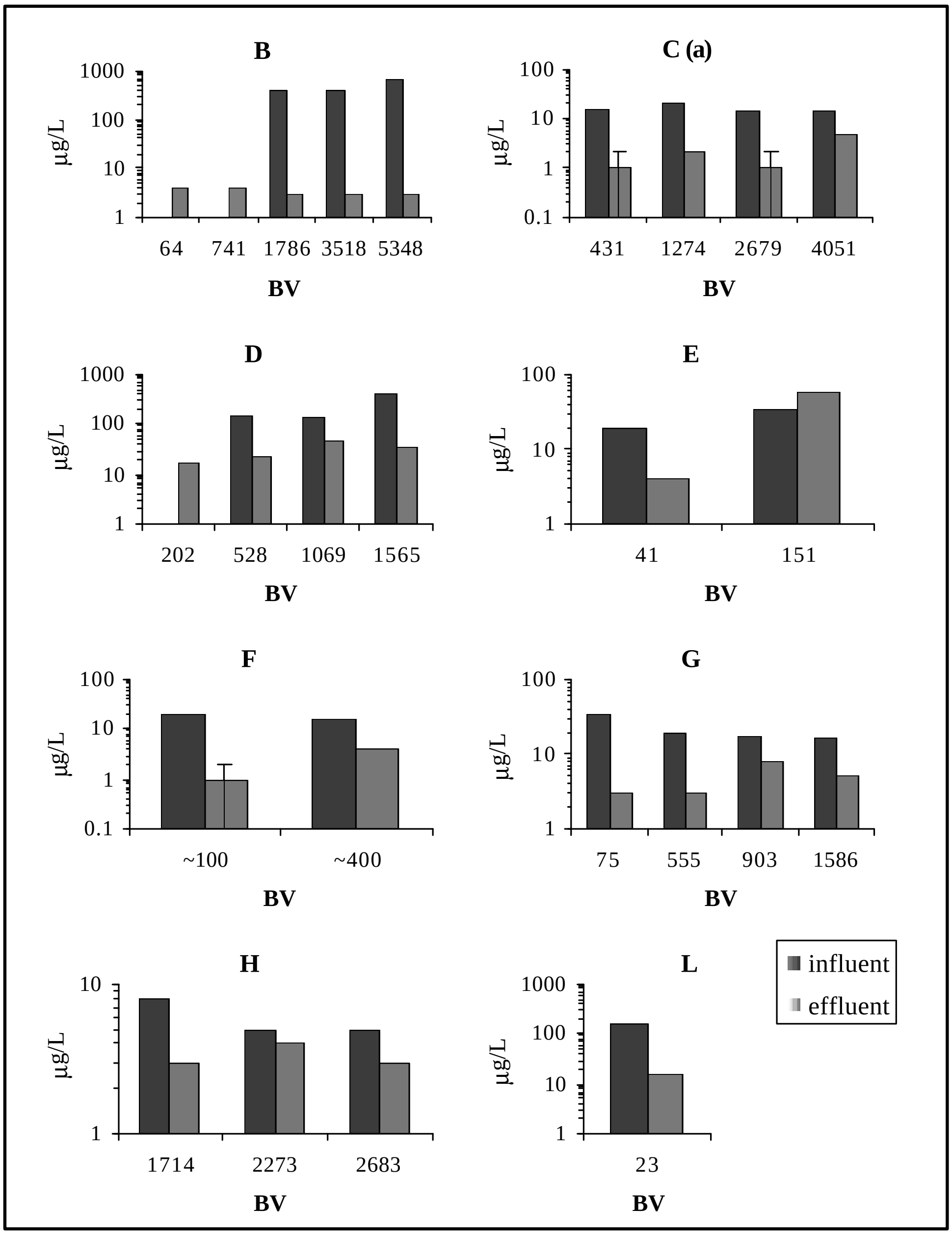

Figure 15. *Concentration of iron in influent and effluent at test locations B, C, D, E, F, G, H and L. The charts are presented on a logarithmic scale. The best reduction was observed at test location B, where the reduction was partly due to the sediment filter installed before the GAC filter. The lowest limit of detection (LLD) was 2 mg/L. Results less than the LLD are presented as 1 ±1 mg/L.*

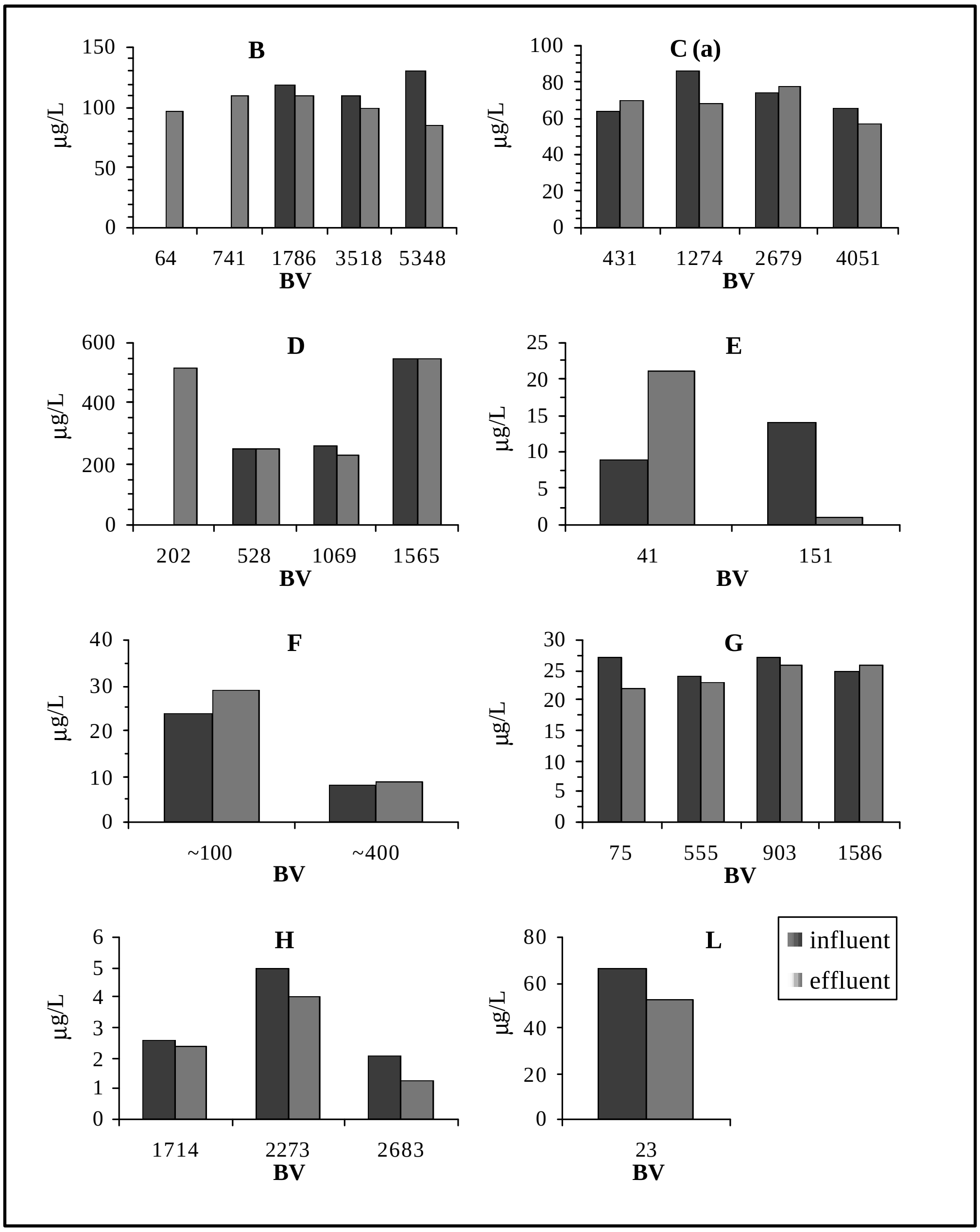

Figure 16. *Concentration of manganese in influent and effluent at test locations B, C, D, E, F, G, H and L. The charts are presented on a linear scale. Only minor reduction could be observed.*

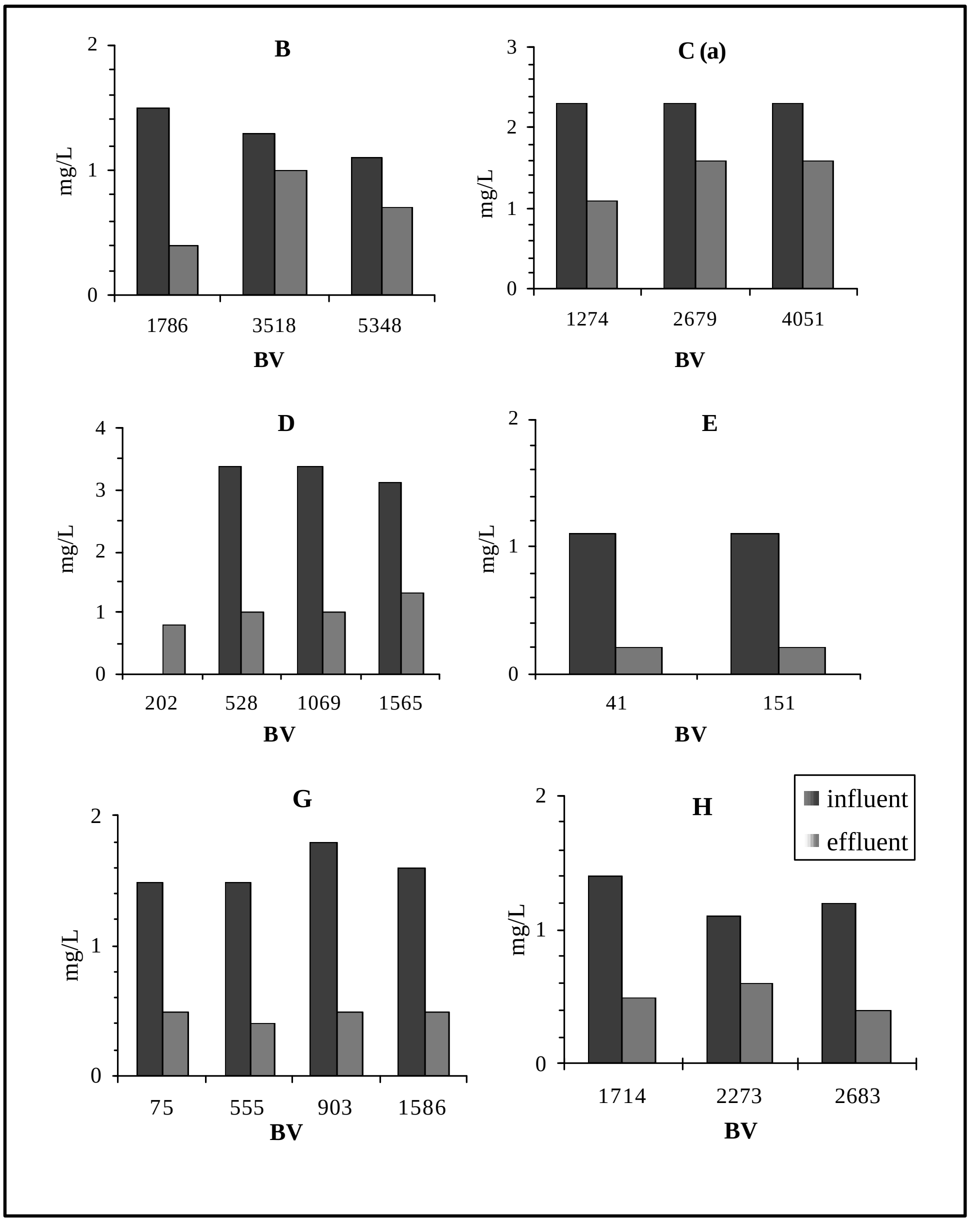

Figure 17. *Concentration of total organic carbon (TOC) in influent and effluent at test locations B, C, D, E, G and H. The charts are presented on a linear scale. Reduction decreased gradually as larger volumes of water were treated.*

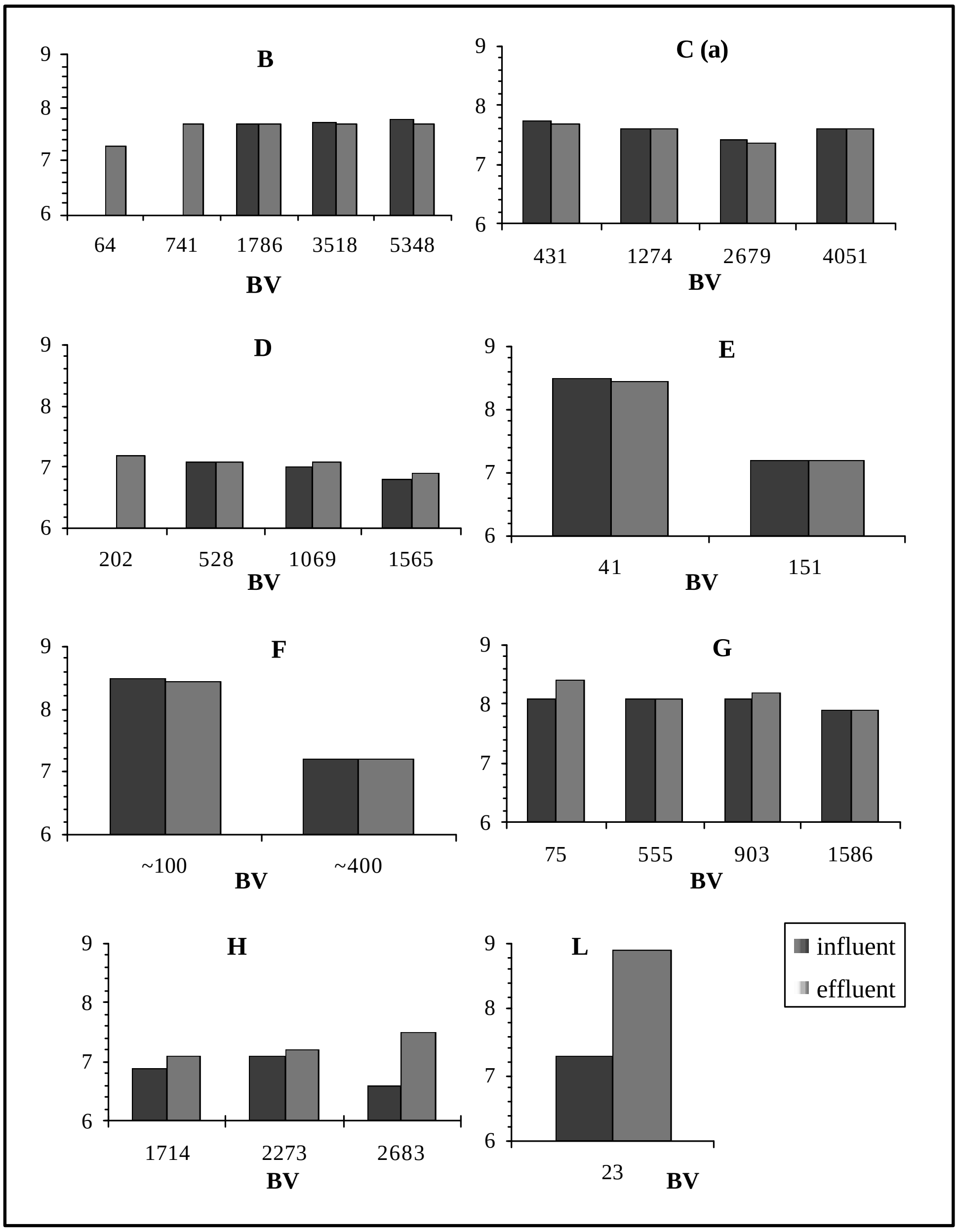

Figure 18. *The pH values in influent and effluent at test locations B, C, D, E, G and H. No clear trend discerning the change of the pH value could be observed.*

GAC filtration did not directly increase the water temperature. The pressure tanks were the main reason for the increased effluent temperature. If the tanks are installed 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 and the pressure tank is in a cellar, where there is no concern about the filter's freezing or the water's warming up. Then the risk of bacterial growth is also the lowest. A refrigerated pressure tank can be used if the pressure tank has to be placed in a warm room. Temperatures in influent and effluent at different test locations are set out in Table XV.

Table XV. *Temperature in influent and effluent at the test locations on different dates.*

Test Location	Date	Temperature (°C) influent	Temperature (°C) effluent
B	3.12.97	8.7	9.8
	25.3.98	8.8–9.3	9.8–11.3
	14.7.98	13.2	13.9–17.6
C(a)	9.10.97	9.0	9.0
	17.12.97	8.0	7.9
	25.3.98	6.3–6.6	5.3–7.9
	14.7.98	12.3	12.2–13.7
D	5.2.98	6.5	7.0
	11.6.98	7.5	7.7–11.2
	24.9.98	8.6	8.6–14.3
E	30.11.97	8.3	8.0–13.0
	15.7.98	–	12.5–16.0
F	15.7.98	9.5	10.0
G	11.12.97	5.9	5.9
	10.3.98	6.1	4.7–5.1
	18.5.98	7.2	7.0–8.0
	23.9.98	9.6	11.3–12.4
H	18.5.98	7.6	7.7
	21.7.98	13.1	13.3
	29.9.98	8.5	7.5–11.0

The temperature in both influent and effluent was lower during the winter. The highest effluent temperature was measured at test location B, where a 500-litre pressure tank was placed in the same room with the furnace of the central heating, which made the water warmer. When water flowed from the tap, the well pump started feeding fresh water into the pressure tank and the influent became colder. Figure 19 illustrates how water temperature in effluent changes as water flows from the taps at test locations B and C(a).

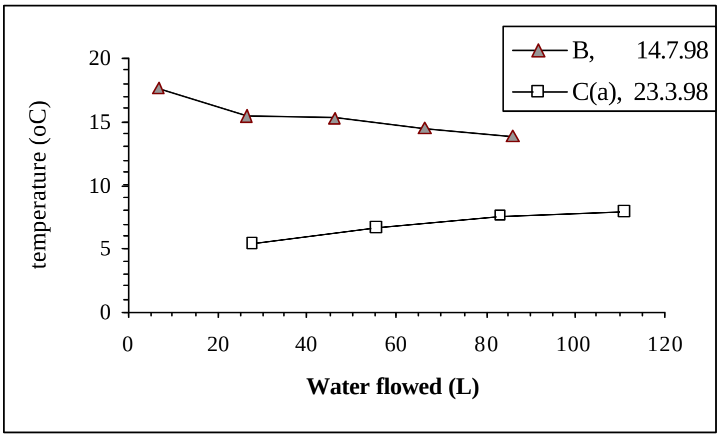

Figure 19. *Temperature decreases at test location B when water flows from a tap. A furnace increases the temperature in the room where the pressure tank and the GAC filter are placed. At test location C(a) the GAC filter is placed in a cellar and the pressure tank in a shed. In wintertime, the temperature of the ground water is higher than the tap water.*

Figure 20 summarises all the data on Fe, Mn, TOC and pH collected at different test locations. Table XVI summarises the statistical data on reduction of iron manganese and TOC, and the change in pH value. The standard deviation was calculated using the equation

$$s = \sqrt{\frac{n\Sigma x^2 - (\Sigma x)^2}{n(n-1)}}\,. \tag{8}$$

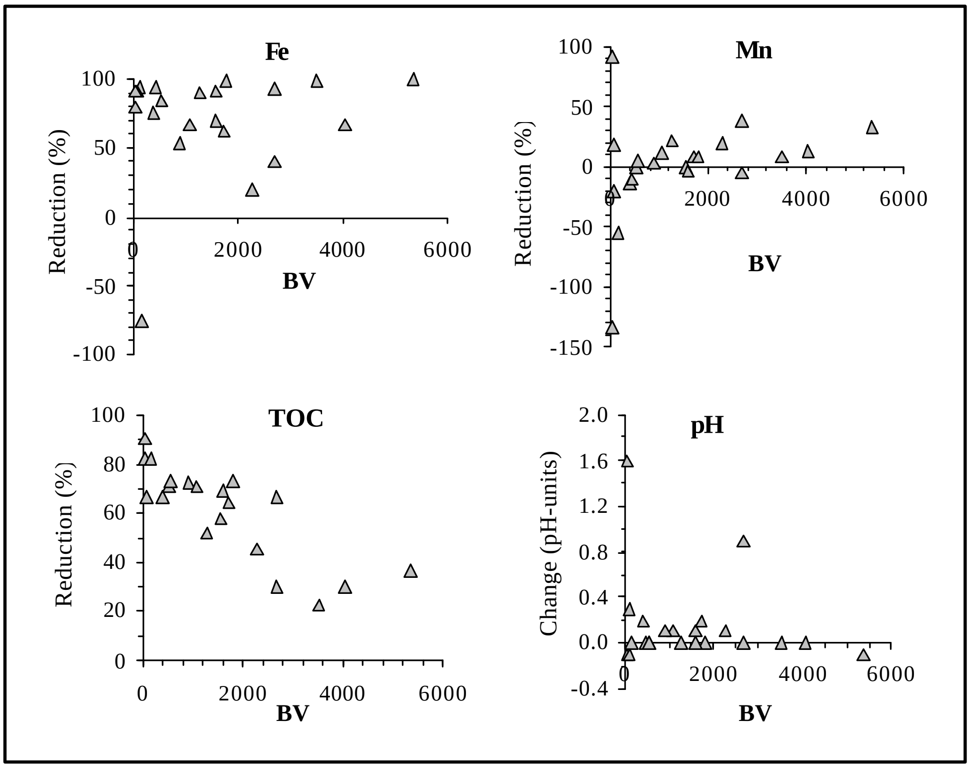

Figure 20. *Reduction of iron, manganese and organic matter (TOC), and change in pH value as a function of the volume of water treated (in BV's). The data were collected from all the test locations.*

Table XVI. *Statistical data on the reduction percentage of Fe, Mn and TOC, and the change in pH value induced by GAC filtration. The data were collected from all the test locations.*

Parameter	unit	No. of data	Average	Median	Min.	Max.	Std. deviation
Fe	%	22	71.2	84.4	–75.8	99.6	39
Mn	%	22	1.9	5.9	–133	92.4	40
TOC	%	20	61.2	66.7	23.1	90.5	19
pH	pH unit	22	0.2	0.0	–0.1	1.6	0.4

4.3.2 Other physico-chemical parameters

Statistical data on the reduction of other water quality parameters are set out in Table XVII. The best reduction of turbidity was obtained at test location B, where the turbidity was the highest due to the high concentration of iron in the influent. Since iron was reduced very efficiently, the turbidity also decreased. At many test locations, the FTU values of turbidity were lower than the lowest limit of detection.

When a new GAC filter has been commissioned, the electric conductivity increases for a few days. After this, no significant changes in this parameter are observed. A chart was drawn for test location D, where four samples were collected during the first few weeks after the filter had been commissioned (Figure 21).

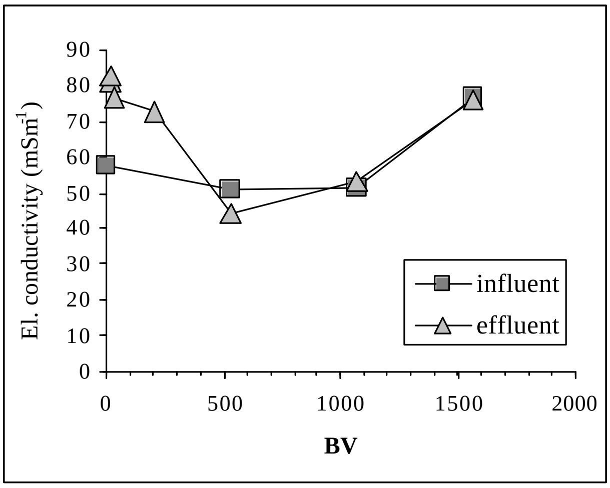

Figure 21. *Electric conductivity in influent and effluent at test location D. The chart shows how GAC filtration increases the electric conductivity during its first few days in service.*

GAC filtration did not alter the alkalinity or total hardness of the waters studied.

Ammonium (NH_4^+) and nitrate (NO_3^-) were analysed three times in both influent and effluent. Mostly, the concentration of NO_3^- was below the detection limit. No significant changes in their concentration occurred.

Phosphate (PO_4^{3-}) was analysed four times in both influent and effluent. The concentration of phosphate increased significantly when the filter had been in service for less than 400 BV's. The detection limit for phosphate was mostly too high for the selected waters.

Table XVII. *Statistical data on the reduction percentage of* O_2, *KMnO*$_4$, *electrical conductivity,* CO_2, *total harness, alkalinity,* PO_4^{3-}, *turbidity,* NO_3^-, NH_4^+ SiO_2 *and acidity, and the change in redox potential (in mV). The data were collected from all the test locations.*

Parameter	Unit	No. of data	Average	Min.	Max.	Median	Std. deviation
O_2	%	17	16	–230	88	33	93
$KMnO_4$	%	15	62	14	88	67	22
El. cond.	%	15	0.4	–5.8	13.4	0	4.1
Redox	mV	13	28	–351	548	0	200
CO_2	%	11	15	–44	93	15	42
Tot. hardness	%	5	3.3	–1.4	11.9	1.2	5.3
Alkalinity	%	5	0.1	–1.9	1.5	0	1.3
PO_4	%	4	–640	–2200	0	–	–
Turbidity[1]	%	4	68	45	90	–	–
NO_3	%	3	0.8	–18	21	–	–
NH_4	%	3	14	0	25	–	–
SiO_2	%	3	2.6	0	5	–	–
Acidity	%	2	–5.0	–10	0	–	–

– data too limited.

[1] Results below 0.2 FTU and test locations with pre-treatment units not included.

No clear trend in the results of CO_2, O_2 and redox potential could be observed. The concentrations of CO_2 and O_2 may be affected by microbial respiration. Most of the results indicated that the concentration of oxygen decreased during filtration. An increase of oxygen, however, was observed in two water samples, which may have been due to an error in sampling. CO_2 seemed to be retained on GAC when small amounts of water had been treated. When larger volumes (>2,000 BV's) were treated the concentration of CO_2 mostly increased.

Redox potential is proportional to the equivalent free energy change per mole of electrons associated with a given reduction. The activity of hydroxyl ions (OH^-) in water influences the activity of hydrogen ions. Redox potential was not significantly affected by GAC filtration.

4.3.3 Summary of the effect of GAC filtration on the physico-chemical quality of water.

Table XVIII. *Summary of the effect of GAC filtration on water quality.*

Iron	Iron was usually removed efficiently by GAC filtration (>50%). The highest reductions were observed with waters that had the highest iron content.
Manganese	The concentration of manganese did not significantly change during filtration.
Organic Matter	Organic matter was removed well when less than 1,000 BV's had been treated. The reduction efficiency decreased gradually and after 2,000 BV's reduction efficiency was less than 70%.
pH	The pH value may increase for a few weeks after a new filter has been commissioned.
Temperature	The changes in water temperature depended on the placement of the GAC filter and the pressure tank, the seasonal variations in outdoor and groundwater temperature, and the size of the pressure tank. Water use also affects the temperature of the effluent.
Turbidity	Turbidity decreased. Correlation between iron reduction and decrease in turbidity was observed.
Electrical Conductivity	Electrical conductivity increased for a couple of weeks after a new filter had been commissioned. No significant changes were observed later.
Alkalinity	No significant changes were observed.
Total Hardness	No significant changes were observed.
Phosphate	The concentration of PO_4^{3-} may increase significantly when less than 400 BV's had been filtered.
Nitrate	No significant changes were observed.
Ammonium	No significant changes were observed.
Silica	No significant changes were observed.
Oxygen	In most cases the concentration of oxygen decreased during the filtration.
Carbon dioxide	After a new filter has been commissioned, the CO_2 concentration may decrease for a while. When 2,000 BV's have been filtered the concentration of CO_2 may increase.
Redox-potential	No significant changes were observed.
Uranium	Uranium was retained fairly well on GAC when less than 1,000 BV's of water had been treated.
Radium	Radium retention on GAC was apparently dependent on water quality; reduction varied greatly.
Lead	Lead was efficiently retained on GAC in most cases.
Polonium	Polonium was well retained on GAC.

4.4 Microbiological quality of treated water

The results of the heterotrophic plate counts (HPC) set out here were collected after the applied radon sampling protocol (100 litres of flowing) and are therefore not characteristic of general effluent water quality (Table XIX). Two samplings were carried out to analyse water quality after the water had been standing in the plumbing for several hours (Table XX).

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

Test Location	BV	22°C (cfu/mL)		35°C (cfu/mL)	
		influent	effluent	influent	effluent
B	64	–	110	–	–
	741	–	1	–	0
	1,786	2	13	0	8
	3,518	90	4	4	2
	5,348	23	210	1	230
C(a)	431	13	76	10	43
	1,274	3	6	0	2
	2,670	0	1	5	1
	4,051	5	16	1	1
D	528	>3000	140	9	5
	1,069	180	8	1	0
	1,565	160	16	0	10
E	41	190	36	220	33
	151	190	17	200	23
F	~100	4	25	1	2
	~400	5	22	2	1
G	555	72	8	33	0
	1,586	6	7	0	1
H	1,714	100	330	24	9
	2,273	290	220	32	25
	2,683	12	95	0.1	14
L	23	0	48	0	42

– not determined

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

Test Location	BV	22°C (cfu/mL) influent	22°C (cfu/mL) effluent (5 L)	22°C (cfu/mL) effluent (200 L)	35°C (cfu/mL) influent	35°C (cfu/mL) effluent (5 L)	35°C (cfu/mL) effluent (200 L)
C(a)	431	13	76	–	10	43	–
E	36	190	180	36	220	190	33

– not determined

The Ministry of Social Affairs and Health in Finland has set target values for HPC. In public water supplies, the HPC should not exceed 100 cfu/mL (colony forming units per millilitre) and 10 cfu/mL at 22°C and 35°C, respectively. At 22°C, HPC in effluent exceeded 100 cfu/mL in three cases. Only in one of these cases was the influent HPC less than this. HPC in effluent at 35°C exceeded 10 cfu/mL in six cases. In three of these cases, HPC in influent was 10 cfu/mL or less. In many cases, HPC in influent was higher than in effluent. Similar sampling for studying aerators has been carried out and indicated much higher HPC. Often the HPC at 22°C exceeded 3 000 cfu/mL in aerated waters (Turtiainen 1999).

According to the results set out in Table XX, HPC is slightly higher in water that has stood in the plumbing for several hours.

Generally, the heterotrophic plate counts did not increase during GAC filtration. Although bacterial populations develop on GAC, no significant release of microbes into the effluent was observed under the conditions studied.

4.5 Technical reliability of the filters

All the GAC filters and filter combinations operated very reliably and no problems were documented. The hydraulic pressure in the household water line remained sufficient at all the test locations studied. No fouling of GAC units could be observed. No difference between the performance of the units equipped with sediment filters and those without could be observed. Therefore, sediment filters are needed only if the well is newly drilled and there is a lot of sand, for instance, in the water.

Backwashing was not carried out at test locations where this system had been installed because no fouling could be observed and the plumbing pressure remained sufficient throughout the whole period that the GAC units were in service.

Test location B had the highest concentration of iron in influent. The iron was removed very efficiently, partly by the sediment filter but mostly by the GAC unit. When 5,300 BV's had been filtered about 100 grams of iron had been retained on GAC and on the sediment filter. The hydraulic pressure, however, remained sufficient. Therefore, no backwashing was carried out.

4.6 Dose rate and shielding

Dose equivalent rates varied at different test locations because radon concentration, radon removal efficiency, daily water use and the geometry of the GAC vessel affect the intensity of gamma radiation (Table XXI). 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 1mm-thick sheets of lead. The shields attenuated the gamma radiation efficiently: a 3mm-thick layer reduced radiation on the filter surface about 35% at test location A and 40% at test location D. At test location A, the dose equivalent rate was 0.16 μSv/h on a desk 150 cm away from the filter 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 has been built. At the other test locations the 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 does not cause significant exposure to gamma radiation for the residents.

Table XXI. *GAC bed size, daily usage, ^{222}Rn concentration in influent, and maximum dose rate on the GAC filter surface at different test locations.*

Test Location	GAC size (L)	Daily usage (L/d)	^{222}Rn in influent (Bq/L)	Max. dose rate on filter surface (μSv/h)
A	39	–	1,900–2,000	47
B	40	590–660	3,500–4,200	84–103
C (a)	39	420–560	2,600–2,700	30–37
D	39	170	3,800	21
G	63	320–370	5,100–7,400	65–111
H	39	340	1,600–2,200	18–27
Vacation homes, no steady-state				
E	40	–	900–1,100	3.9
F	63	–	1,700–2,800	12.6
L	63	–	5,800–6,000	35.6
Lead shield 3 mm				
A	39	–	1,900–2,000	14
D	39	170–180	3,700–4,100	9–12

(–) not available.

Gamma emission profiles on GAC vessel surfaces were usually similar: the highest dose equivalent rate occurred in the top few centimetres of the bed. The only exception was test location C(a), which had the lowest radon reduction (Figure 22). When the rate of adsorption is not sufficient in respect of water use, the maximum gamma activity moves towards the centre of the GAC bed.

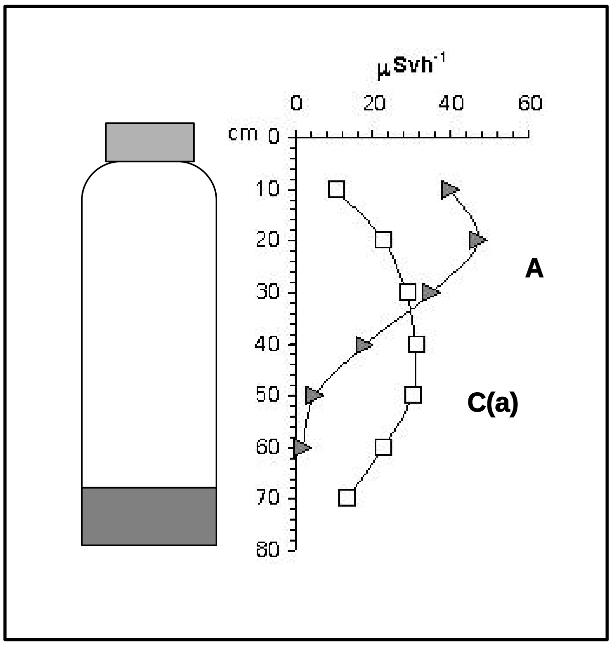

Figure 22. *Gamma emission profiles on filter surfaces at test locations A and C(a). The filters were of the same size and had no pre-treatment units installed. Vertical distance was measured from the top of the filter cap.*

Different shielding materials were studied at test location G (a 63-litre filter). The measurements were carried out at a distance of one metre from the GAC unit. The height of the measuring point was 1 metre from floor level (the height on which the maximum dose equivalent rate occurred). The results of different materials are set out in Figure 23. The attenuation is presented as the percentage of dose rate absorbed by different shields. The background dose rate was 022 µSv/h and the net dose rate without shielding was 1.78 µSv/h. The error values were estimated using ±0.1 µSv/h for the measurements with and without the shield.

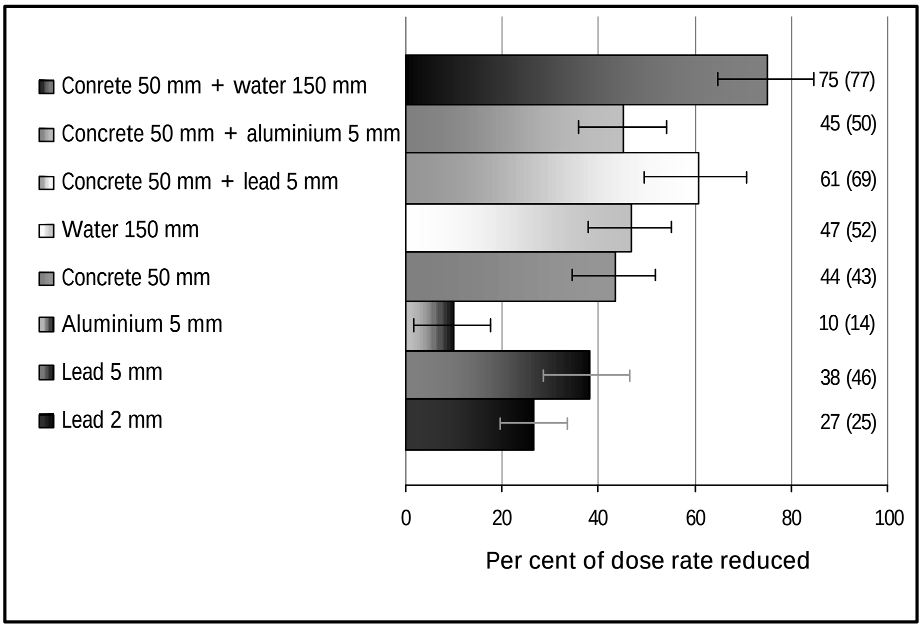

Figure 23. *The measured attenuation of dose rate with different types of shields. The results are presented as the percentage of dose rate reduced by different types of shields. The calculated values are given in parentheses.*

Lead, as the densest shield material selected, attenuated the gamma rays the most efficiently relative to the shield thickness. Five millimetres of lead attenuated the gamma rays about 40% while 15 centimetres of water reduced the radiation about 50%. Lead is, however, a heavy metal and therefore cannot be recommended for households where some other type of shield can be installed. The best available materials are concrete, bricks and a water jacket. All these materials can easily be installed and are inexpensive. A concrete well ring can be split lengthwise and the two halves can be erected around the GAC unit. A water jacket can be built of a container (e.g. a tun). The filter is placed in the container, after which water is poured in. There must be a floor drain if a water jacket is applied. Bricks can be used to build a case around the GAC filter.

STUK will produce a guide that provides information about the proper placement and shielding of a GAC unit. Generally speaking, the effective dose rate should not exceed 1 μSv/h at a distance of one metre from the

GAC unit. The annual additional dose received by the residents from the direct gamma rays from the filter should be less than 0.1 mSv.

4.7 Spent GAC as radioactive waste

Two spent batches of GAC were investigated with respect to the radioactivity accumulated in the carbon. The results of the gammaspectrometric measurement carried out for the first batch of GAC (14 litres) show that most of the ^{210}Pb produced is concentrated in the top layer of the GAC bed whilst ^{226}Ra is dispersed to the larger bed volume (Table XXII). Samples 1–3 were the first three 0.5-litre fractions on the top of the bed. Sample 4 was a subsample of the rest of the (homogenised) bed. The first two fractions showed about a 14% weight-gain, which is likely due to retention of iron and organic matter on the GAC.

Table XXII. *Weight and activity of different 0.5-litre fractions of a spent batch of GAC. Total represents the whole batch of GAC.*

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

The activity of ^{210}Pb accumulated in the first batch of spent GAC was 75 kBq, which is in good agreement with the value calculated based on the radon concentration and flow rate (42–88 kBq). The specific activity of the spent batch of GAC was 9 Bq/g. The total activity of ^{226}Ra in the batch was 2.8 kBq.

The radionuclides identified in the second batch of spent GAC and the activities determined are set out in Table XXIII.

Table XXIII. *The nuclides identified and the respective activities of four sub-samples collected from the second (homogenised) batch of spent GAC. Two types of measurement geometry were applied.*

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

The results indicated that the carbon was homogenised well enough. The average specific activity of ^{210}Pb calculated from the four sub-samples was 11.2 Bq/g. The density of the sub-samples was 0.6 kg/L. When the carbon was dried, its volume was compressed approximately 10%. Hence, the activity of the batch on the day of determination was 236 kBq. When returned back to the date when the GAC filter was taken out of service the activity is 240 kBq.

^{238}U had attained secular equilibrium with ^{234}Th that was detected from the gamma spectrum. The total amount of uranium retained on GAC was two grams. The activity of ^{235}U was low.

The measured activity of ^{210}Pb (240 kBq) compared favourably to the calculated value (240 kBq). Although the adsorption rate of radon was rather low (K_{ss}=1.2 h^{-1}), the retention of radon progeny was 100%. This experiment gave further evidence of the quantitative retention of radon daughters on GAC. Additionally, ^{210}Pb was monitored in both influent and effluent during the experiment. An 89% average efficiency for direct adsorption of ^{210}Pb from influent was recorded. The low concentration of ^{210}Pb in the effluent was another argument for the 100% retention of radon daughters. If some of the ^{210}Pb were released from the GAC matrix the ^{210}Pb activity in effluent would increase.

A spent batch of GAC is a source of radiation and therefore should not be transported before the radiation has decreased to an acceptable level. According to present regulations in Finland the dose rate on the surface of a postal packet may not exceed 5 µSv/h. This limit can be achieved when the activity concentration of wet carbon is approximately less than 70 Bq/g (the sum of radon and four daughter products). The time that is needed to allow the retained radon to decay and the dose rate to decrease to the acceptable level is set out in Table XXIV.

Table XXIV. *The times that are needed to allow the adsorbed radon to decay and the filter to achieve the acceptable radiation level. Radon loading is equal to the daily water use (L/d) multiplied by the radon concentration in influent (Bq/L).*

Radon loading (MBq/d)	Time to allow decay (d)
< 0.5	8
0.5–1	11
1–2	15
2–3	17
3–4	19

At present, there are no regulations in Finland specifically applicable to the disposal of GAC containing natural radionuclides. Spent GAC should not be regenerated because both lead and polonium may vaporise at the regeneration temperatures (up to 700–900°C). This would cause contamination of the furnaces and a potential health risk to the workers. Therefore, it is recommended that spent batches of GAC should be disposed of properly and the units filled with fresh material. Granular activated carbon is relatively inexpensive. A batch of 40 litres costs approx. 150 euros. If the carbon is changed every two years or every 250 m^3, the cost of water treatment is 0.02 € per day or 1.7 €/m^3, respectively.

5 CONCLUSIONS

The results of this study were in good agreement with the studies conducted previously. Radon removal efficiency was very high (>99.9%) at most test locations. It seems that GAC filtration can be applied to mitigate unacceptably high concentrations of radon in ground waters that are most typically found in Finnish bedrock. The water types studied were (1) water that is soft and low in mineral content, (2) iron- and manganese-rich water, (3) humus-rich water and (4) slightly saline water.

It is possible that high concentrations of uranium, and possibly organic matter, lower the adsorption rate of radon. Uranium, as a toxic heavy metal, is harmful to human health and therefore high concentrations (>0.1 mg/L) should always be removed from drinking water. The two filter combinations, which had a 21-litre anion exchanger installed before a 39-litre GAC filter, were capable of removing both uranium and radon very efficiently. Neither high concentration of iron and manganese in influent nor sediment filters prior to GAC units had any detectable effect on the performance of the GAC filters.

The longevity of a GAC batch was not found out, because most filters exhibited constant removal efficiency for radon throughout the study. Therefore, no recommendations regarding the effective lifetime of a GAC batch can be given. STUK continues sampling at the test locations in order to find out the longevity of a GAC bed.

The intensity of gamma radiation originating from GAC units in permanent use was high. Considering the radiation safety aspects, radon levels higher than 5,000 Bq/L should generally be removed with aeration. Shielding is often necessary even for radon levels from 1,000 to 5,000 Bq/L, With the proper shielding, instructions and placement of the unit, elevated doses to the residents can be avoided. STUK is preparing a guide intended for the companies that supply GAC units. The guide will give instructions for designing the necessary shielding and the safe transport and disposal of spent GAC.

The water quality at the test locations remained good. As a result of this study, it can be expected that the number of domestic GAC filters used in radon removal will increase. GAC filtration can be considered an inexpensive and easy way of mitigating unacceptably high concentrations of radon in household water.

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QUALITY REQUIREMENTS OF HOUSEHOLD WATER

APPENDIX 1

Iron commonly occurs in Finnish groundwaters. It can also be released into household water from components of the water line (cast iron, galvanised steel). Under certain conditions, microbial populations that bind iron from water, may develop. Therefore even a small concentration of iron may form precipitates that are released due to pulsation in water pressure. Iron is not harmful to human health but may cause discoloration of sinks and form precipitates in the water pipes. The technical-aesthetic limit set for iron is 0.2 mg/L.

Manganese often occurs simultaneously with iron in groundwaters. High concentration of manganese impairs the taste of water. It also causes discoloration and precipitates. The technical-aesthetic limit for manganese is 0.05 mg/L.

Humic substances form most of the organic matter in groundwaters. These substances can be divided into humic and fulvic acids, and humin. High-molecular-weight humic materials exhibit a colloidal structure that provides a very large surface for the adsorption of both inorganic and organic materials. Radionuclides such as ^{210}Pb, ^{210}Po, and ^{226}Ra may be adsorbed on these colloids. The microbial growth in the water line depends on the amount of biodegradable organic carbon in water. High concentration of organic matter in water causes odour and flavour.

TOC analysis indicates the total amount of organic carbon that occurs in water. TOC values are thus comparable, because in groundwater there are no components that would interfere with the analysis. The target value for TOC is <2 mg/L. $KMnO_4$ titration, however, is interfered with by Cl^-, SO_3^{2-}, S^{2-}, and NO_2^-, which all can be oxidised by $KMnO_4$. Furthermore, not all the organic carbon present in water can be oxidised with this method. The technical-aesthetic limit set for $KMnO_4$ is 12 mg/L and the target value <8 mg/L.

The pH value of Finnish groundwaters is typically slightly acidic, in the range pH 6–7. The bedrock waters are, however, often slightly alkaline. The pH value of water affects its corrosiveness and therefore the target value is 7.0–8.8. The pH value should also stay constant. The technical-aesthetic limit set for pH is 6.5–9.5.

APPENDIX 1 QUALITY REQUIREMENTS OF HOUSEHOLD WATER

Turbidity is generally caused by clay, iron and colloids that occur in water. No determinental health effects are associated with this parameter. Heavy metals and bacteria, however, have a tendency to be adsorbed on the particles and colloids. The technical aesthetic limit for turbidity is 4 FTU.

Electrical conductivity represents the amount of minerals dissolved in the water. The target value of electrical conductivity is <40 mS/m in 25 °C, which was set on a technical-aesthetic basis.

Alkalinity within the pH range of drinking water is caused mostly by bicarbonate ions. The lower the alkalinity, the more easily the pH value of water can change. If alkalinity is low (<0.06 mmol/L), corrosion in the water line may occur (depending on the concentrations of chloride and sulphate). If alkalinity is high, calcareous fur may develop in the water line. The alkalinity of household water should generally be higher than 0.6 mmol/L.

Total hardness is mostly caused by calcium and magnesium dissolved in the water. If water is too soft, it may induce corrosion in the water pipes. Therefore the guideline value set for total hardness is >1.5 mmol/L Ca. Hard water, however, enhances the formation of calcareous fur in the water line.

QUALITY REQUIREMENTS OF HOUSEHOLD WATER

APPENDIX 1

Table I. *Quality requirements of household water based on health aspects (parameters concerning microbiological and toxic substances and substances which can be harmful when consumed in excess).*

Microbiological	**Maximum plate count**
Coliformic bacteria	<1/100 mL
Escherichia Coli	<1/100 mL
Faecal streptococci	<1/100 mL
Toxic/harmfull	**Maximum concentration, mg/L**
As	0.01
Cd	0.005
Cr	0.05
Pb	0.01
F^-	1.5
NO_3^-	25
NO_3-N	6.0
NO_2^-	0.1
NO_2–N	0.03
$CHCl_3$	0.2
$CHBrCl_2$	0.06
Chlorophenole	0.01

Table II. *Quality requirements on a technical-aesthetic basis.*

Parameter	Maximum concentration, mg/L
Al	0.2
NH_4^+	0.5
NH_4-N	0.4
Ca	100
Cl^-	100
$KMnO_4$	12
COD_{Mn}	3.0
Cu	1.0
Mn	0.05
Fe	0.2
Zn	3.0
SO_4^-	150
Parameter	**Quality requirement**
pH	6.5–9.5
Turbidity (FTU)	<4
Colour	<15
Temperature (°C)	<25

Table I. *Test location A.*

Parameter	**Date**	**19.3. 96**	**17.3. 97**	**20.10.97**	**15.1.98**	**10.3.98**	**7.4.99**	**11.10.99**	
	Unit	**influent**	**influent**	**effluent**	**effluent**	**effluent**	**effluent**	**influent**	**effluent**
Rn-222	BqL^{-1}	2000	1930	1.2–2.7	1.8	2.8	36	2820	40.5
U-238	BqL^{-1}			0.647					
U-234	BqL^{-}1			1.55					
uranium	mgL^{-1}			0.052					
Ra-226	BqL^{-1}	0.02		0.03					
gross-α	BqL^{-1}	1.7		1.5					
temperature	°C		10.2						
O_2	mgL^{-1}		2.4						
O_2	sat. %		21						
pH			8.3	8.74					
tot.hardness	mmolL^{-1}		0.43						
alkalinity	mmolL^{-1}		0.91	1.88					
$KMnO_4$	mgL^{-1}		2.0	1.3					
COD_{Mn}	mgL^{-1}		0.5						
turbidity	FTU		0.35						
colour	Pt mgL^{-1}		5						
el. cond.	mSm^{-1}		23.3	23.9					
Fe	mgL^{-1}		0.021	0.006					
Mn	mgL^{-1}		0.018	0.007					
Na	mgL^{-1}		37						
K	mgL^{-1}		1.4						
Ca	mgL^{-1}		11.4						
Mg	mgL^{-1}		3.6						
F	mgL^{-1}		1.60						
Cl	mgL^{-1}		4.5						
SO_4	mgL^{-1}		17.3						
NO_3+NO_2-N	mgL^{-1}		0.125						
NO_3	mgL^{-1}			<0.5					
NH_4	mgL^{-1}		0.003						
PO_4-P	mgL^{-1}		0.025						
HPC 22°C	cfu/mL			75					
HPC 35°C	cfu/mL			25					
max. D rate	µSvh^{-1}			47					

Table II. *Test location B.*

Parameter	**Date** **Unit**	**31.10.95**	**14.4.97**	**12.8.97**	**23.9.97**	
		influent	**influent**	**effluent**	**influent**	**effluent**
Rn-222	BqL^{-1}	3400	3890	160-190		12-18
U-238	BqL^{-1}				0.159	0.105
U-234	BqL^{-1}				0.451	0.285
uranium	mgL^{-1}				0.013	0.009
Ra-226	BqL^{-1}	0.22			0.12	0.04
Pb-210	BqL^{-1}				0.283	0.009
Po-210	BqL^{-1}				0.122	0.007
gross-α	BqL^{-1}	0.88			0.76	0.36
O_2	mgL^{-1}		0.8	1.0		
pH			7.5	7.3		7.7
tot.hardness	mmolL^{-1}		1.45	1.36		1.41
alkalinity	mmolL^{-1}		1.44	1.49		1.54
$KMnO_4$	mgL^{-1}			1.4		2.0
COD_{Mn}	mgL^{-1}		< 0.5			
turbidity	FTU		2.3	< 0.05		< 0.05
colour	Pt mgL^{-1}		7.5			
el. cond.	mSm^{-1}		55.0	53.2		53.7
Fe	mgL^{-1}		0.320	0.004		0.004
Mn	mgL^{-1}		0.105	0.096		0.110
Na	mgL^{-1}		49	51		
K	mgL^{-1}		2.6	1.60		
Ca	mgL^{-1}		46			
Mg	mgL^{-1}		7.3			
PO_4-P	mgL^{-1}		0.024	0.012		
F	mgL^{-1}		0.78			
Cl	mgL^{-1}		115	112		
SO_4	mgL^{-1}		8.6	11.0		
NO_3+NO_2-N	mgL^{-1}		0.004			
NO_3	mgL^{-1}			< 0.5		
NH_4	mgL^{-1}		0.071	0.07		
colif. 35°C	cfu/100mL		0			
colif. 44°C	cfu/100ml.		0			
faec. strept.	cfu/100mL		0			
HPC 22°C	cfu/mL			110		1
HPC 35°C	cfu/mL					0
max. D rate	µSvh^{-1}					84
consumed	m^3			2.58	29.6	
	BV			64	741	

Table III. Test location B (cont.).

Parameter	**Date** **Unit**	**3.12.97** **influent**	 **effluent**	**25.3.1998** **influent**	 **effluent**	**14.7.1998** **influent**	 **effluent**
Rn-222	BqL^{-1}	4 000	13-21	4 200	24-32	3 520	31–37
U-238	BqL^{-1}	0.182	0.182	0.160	0.169		
U-234	BqL^{-1}	0.479	0.509	0.447	0.458		
uranium	mgL^{-1}	0.015	0.015	0.013	0.014		
Ra-226	BqL^{-1}	0.17	0.07	0.20	0.07	0.20	0.10
Pb-210	BqL^{-1}	0.371	0.014	0.328	0.005	0.660	0.005
Po-210	BqL^{-1}	0.102	0.015	0.154	0.012	0.454	0.013
gross-α	BqL^{-1}	0.95	0.63	0.87	0.66	1.2	0.46
temperature	°C	8.7	9.8	8.8-9.3	11.3-9.8	13.2	13.9-17.6
O_2	mgL^{-1}				0.2	0.5	0.1
CO_2	mgL^{-1}					2.6	3.1
redox	mV			374	387	264	812
pH		7.7	7.7	7.73	7.70	7.8	7.7
tot.hardness	mmolL^{-1}	1.40	1.39				
alkalinity	mmolL^{-1}	1.56	1.59				
$KMnO_4$	mgL^{-1}	2.9	2.5	2.9	1.8		
TOC	mgL^{-1}	1.5	0.4	1.3	1.0	1.1	0.7
turbidity	FTU			2.5	0.30		
el. cond.	mSm^{-1}	53.2	53.1	55.4	55.4	51.8	51.4
Fe	mgL^{-1}	0.410	0.003	0.390	0.003	0.67	0.005
Mn	mgL^{-1}	0.120	0.110	0.110	0.100	0.13	0.086
NO_3	mgL^{-1}	< 0.5	< 0.5				
SiO_2	mgL^{-1}					16.9	16.7
colif. 35°C	cfu/100mL	0	0				
HPC 22°C	cfu/mL	2	13	90	4	23	210
HPC 35°C	cfu/mL	0	8	4	2	1	230
max. D rate	µSvh^{-1}		96		103		87
consumed	m^3	71.4		141		214	
	BV	1 786		3 518		5 348	

Table IV. Test location C(a).

Parameter	Date	14.4.97	9.10.1997		17.12.1997		25.3.1998	
	Unit	influent	influent	effluent	influent	effluent	influent	effluent
Rn-222	BqL^{-1}	2 970	2 600	160–220	2 750	150–210	2 750	240–330
U-238	BqL^{-1}	2.98	2.56	2.24	2.50	2.62	2.47	2.60
U-234	BqL^{-1}	4.86	4.11	3.60	4.00	4.07	4.04	4.11
uranium	mgL^{-1}	0.24	0.207	0.181	0.202	0.212	0.200	0.211
Ra-226	BqL^{-1}	0.23	0.21	0.15	0.23	0.22	0.25	0.22
Pb-210	BqL^{-1}	0.171	0.375	0.040	0.640	0.045	0.228	0.031
Po-210	BqL^{-1}	0.121	0.266	0.045	0.126	0.042	0.109	0.051
gross-α	BqL^{-1}	6.6	5.9	5.4	5.7	5.2	4.9	4.7
temperature	$^{\circ}$C	8.0	9.0	9.0	8.0	7.9	6.3–6.6	5.3–7.9
O$_2$	mgL^{-1}	0.5					0.4	0.3
O$_2$-sat	%	4						
pH		7.4	7.74	7.71	7.6	7.60	7.44	7.38
redox	mV		252	242	282	312	460	600
alkalinity	mmolL^{-1}	2.3						
acidity	mmolL^{-1}		0.11	0.10	0.12	0.12		
KMnO$_4$	mgL^{-1}	5.6	3.9		4.6	2.2	4.1	2.7
TOC	mgL^{-1}				2.3	1.1	2.3	1.6
turbidity	FTU	0.14	$<$0.05	$<$0.05	0.05	$<$0.05	0.06	0.07
el. cond.	mSm^{-1}	44.5	0.016	$<$0.002				
Fe	mgL^{-1}	0.025	0.064	0.070	0.020	0.002	0.014	$<$0.002
Mn	mgL^{-1}	0.210			0.086	0.068	0.074	0.078
Na	mgL^{-1}	36						
K	mgL^{-1}	2.4						
Ca	mgL^{-1}	53						
Mg	mgL^{-1}	4.6						
F	mgL^{-1}	0.51						
Cl	mgL^{-1}	26.5						
SO$_4$	mgL^{-1}	64.6						
NO$_3$+NO$_2$-N	mgL^{-1}	0.155						
NO$_3$	mgL^{-1}		1.1	1.3	1.3	1.3		
NH$_4$	mgL^{-1}	0.010			0.04	0.03		
PO$_4$-P	mgL^{-1}	0.011						
SiO$_2$	mgL^{-1}						14.5	14.5
colif. 35°C	cfu/100mL	0			3	3	0	0
colif. 44°C	cfu/100mL	0						
prel. *E.Coli*	cfu/100mL				0	0		
faec.strept.	cfu/100mL	0						
HPC 22°C	cfu/mL		13	76	3	6	0	1
HPC 35°C	cfu/mL		10	43	0	2	5	1
max. D rate	μSvh^{-1}		37		30		34	
consumed	m^3		16.8		49.7		104.1	
	BV		431		1 274		2 670	

Table V. *Test location C(a) (contin.).*

Parameter	Date Unit	20.4.98 effluent	25.5.98 effluent	1.7.98 effluent	9.10.1997 influent	 effluent
Rn-222	BqL^{-1}	290	340	340	2 660	150–190
Ra-226	BqL^{-1}				0.24	0.26
Pb-210	BqL^{-1}				0.132	0.015
Po-210	BqL^{-1}				0.104	0.033
gross-α	BqL^{-1}				5.24	5.53
temperature	°C				12.3	12.2–13.7
O_2	mgL^{-1}				0.1	0.3
CO_2	mgL^{-1}				4.8	5.3
redox	mV				470	337
pH					7.6	7.6
$KMnO_4$	mgL^{-1}				1.5	<1
TOC	mgL^{-1}				2.3	1.6
turbidity	FTU				0.09	0.09
Fe	mgL^{-1}				0.15	0.005
Mn	mgL^{-1}				0.066	0.057
SiO_2	mgL^{-1}				14.7	14.7
HPC 22°C	cfu/mL				5	16
HPC 35°C	cfu/mL				1	1
max. D rate	µSvh^{-1}				31	
consumed	m^3	115	133	151	158	
	BV	2 949	3 410	3 870	4 051	

Table VI. *Test location C(b),* **"raw"** *stands for raw water,* **"AE"** *for anion exchanged water and* **"GAC"** *for first anion exchanged and then GAC filtered water, which is the finished water.*

Parameter	Date Unit	21.8.1998 raw	 AE	 GAC	9.9.1998 raw	 GAC	2.3.1999 raw	 GAC	14.7.1999 raw	 GAC
Rn-222	BqL^{-1}	3080		<0.4	1850	5.4	2980	21	2180	31
Ra-226	BqL^{-1}	0.21	0.20	0.13			0.22	0.18	0.25	0.14
Pb-210	BqL^{-1}	0.110	0.033	0.003						
Po-210	BqL^{-1}	0.080	0.029	0.006						
gross-α	BqL^{-1}	5.1	0.26	0.11			6.8	0.47	6.0	0.31
max. D rate	µSvh^{-1}		16							
consumed	m^3		0.80		8.85		100		174	
	BV		20		227		2570		4 464	

Table VII. *Test location D.*

Parameter	**Date** **Unit**	**13.9.96** **influent**	**4.2.97** **influent**	**19.8.97** **influent**	**19.10.97** **effluent**	**26.10.97** **effluent**	**9.11.97** **effluent**	**27.11.97** **effluent**
Rn-222	BqL^{-1}	2800		3720	<0.4	<1	<0.4	0.41-1.0
U-238	BqL^{-1}			0.424				0.197
U-234	BqL^{-1}			0.541				0.251
uranium	mgL^{-1}			0.035				0.016
Ra-226	BqL^{-1}	0.20		0.20				0.40
Pb-210	BqL^{-1}	0.614						0.117
Po-210	BqL^{-1}	0.104						0.027
gross-α	BqL^{-1}	0.83						0.78
O_2	mgL^{-1}			1.0				
pH			7.05	7.22				7.18
tot.hardness	mmolL^{-1}		0.57	0.68				
alkalinity	mmolL^{-1}			2.47				
TOC	mgL^{-1}			2.5				0.8
turbidity	FTU			0.17				
colour	mgL^{-1}			<5				
el. cond.	mSm^{-1}		49	57.8	81.0			72.7
Fe	mgL^{-1}		0.1	0.089				0.016
Mn	mgL^{-1}			0.270				0.520
Na	mgL^{-1}			94.0				
K	mgL^{-1}			2.50				
Ca	mgL^{-1}			17.0				
Mg	mgL^{-1}			6.50				
F	mgL^{-1}			0.48				
Cl	mgL^{-1}			72.5				
SO_4	mgL^{-1}			46.1				
NO_2	mgL^{-1}			<0.01				
NO_3	mgL^{-1}			<0.5				
NH_4	mgL^{-1}			0.04				
PO_4-P	mgL^{-1}			0.018				
colif. 35°C	cfu/100mL							2
prel. E.Coli	cfu/100mL							0
HPC 22°C	cfu/mL			67				10
HPC 35°C	cfu/mL			37				17
max. D rate	µSvh^{-1}							21/11*
consumed	m^3				1.3	2.4	5.0	7.90
	BV				33	62	130	202

* - measured with a lead shield (3 mm).

***Table VIII.** Test location D (cont.).*

Parameter	Date	5.2.98		4.4.98	11.6.98		4.8.98	24.9.98	
	Unit	influent	effluent	effluent	influent	effluent	effluent	influent	effluent
Rn-222	BqL^{-1}	3900	<3	1	4080	3.6–15.1	1.3	3000-3450	1.0–1.6
U-238	BqL^{-1}	0.444	0.159		0.393	0.291			
U-234	BqL^{-1}	0.586	0.209		0.530	0.393			
uranium	mgL^{-1}	0.036	0.013		0.043	0.032			
Ra-226	BqL^{-1}	0.28	0.24		0.26	0.27		0.54	0.43
Pb-210	BqL^{-1}	0.354	0.234		0.313	0.195		0.477	0.162
Po-210	BqL^{-1}	0.169	<0.002		0.520	0.045		0.134	0.039
gross-α	BqL^{-1}	1.3	0.59		1.4	0.98		1.7	1.2
temperature	°C	6.5	7.0		7.5	7.7-11.		8.6	8.6-14.3
O_2	mgL^{-1}				0.3	0.3		0.3	0.2
CO_2	mgL^{-1}				12	9.7		14	12
redox	mV							178	340
pH		7.1	7.1		7.0	7.1		6.8	6.9
tot. hardness	mmolL^{-1}	0.61	0.61		0.63	0.60			
alkalinity	mmolL^{-1}	2.43	2.41						
$KMnO_4$	mgL^{-1}	6.9	2.2		2.2	<1			
TOC	mgL^{-1}	3.4	1.0		3.4	1.0		3.1	1.3
el. cond.	mSm^{-1}	50.9	44.1		51.4	53.0		76.8	76.0
Fe	mgL^{-1}	0.150	0.023		0.140	0.047		0.400	0.036
Mn	mgL^{-1}	0.250	0.250		0.260	0.230		0.550	0.55
SiO_2	mgL^{-1}							14.8	14.4
colif. 35°C	cfu/100mL	0	0						
prel. E.Coli	cfu/100mL	0	0						
HPC 22°C	cfu/mL	>3000	140		180	8		160	16
HPC 35°C	cfu/mL	9	5		1	0		0	10
max. D rate	µSvh^{-1}	9.4*			12*			11*	
consumed	m^3	20.6		30	41.7		51.8	61.0	
	BV	528		770	1069		1328	1565	

* - measured with a lead shield (3 mm).

***Table IX.** Test location D (cont.).*

Parameter	Date	11.4.99		14.9.99	
	Unit	influent	effluent	influent	effluent
Rn-222	BqL^{-1}	3540	1.7	2750	2.2
Ra-226	BqL^{-1}	0.29	0.32	0.55	
gross-α	BqL^{-1}	1.32	0.88	1.24	
consumed	m^3	92.2		118	
	BV	2364		3028	

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Table X. *Test location E.*

Parameter	Date	7.7.97	30.11.97			15.7.98		5.8.99	
	Unit	influent	influent	effluents* #1	effluents* #2	influent	effluent	influent	effluent
Rn-222	BqL^{-1}	910	1090	3	1.4-2.2	1140	<0.4		<0.4
U-238	BqL^{-1}	0.207	0.233		0.0169	0.175	0.0014		
U-234	BqL^{-1}	0.308	0.338		0.0243	0.268	0.0022		
uranium	mgL^{-1}	0.017	0.0189		0.0014	0.0217	0.0002		
Ra-226	BqL^{-1}	0.04	0.05		<0.01	0.05	<0.01	0.04	0.01
Pb-210	BqL^{-1}	0.123	0.461		0.003	0.034	0.002		
Po-210	BqL^{-1}	0.181	0.416		0.003	0.033	0.006		
gross-α	BqL^{-1}	0.70	1.0		0.04	0.47	0.04	0.48	0.08
temperature	^{o}C		8.3	13.0	8.0		12.5–14		
O_2	mgL^{-1}	1.6	0.7	0.5	0.3	1.1	0.2		
CO_2	mgL^{-1}					8.8	7.5		
redox	mV					291	281		
pH		7.13	7.47	8.49	8.44	7.2	7.2		
tot.hardness	$mmolL^{-1}$	0.72	0.71	1.07	0.72				
alkalinity	$mmolL^{-1}$	1.81							
$KMnO_4$	mgL^{-1}	2.2	2.5	<1	1.5				
TOC	mgL^{-1}		1.1	0.3	0.2	1.2	0.5		
turbidity	FTU	0.72				0.42	0.23		
el. cond.	mSm^{-1}	24.5	24.2	30.2	24.0	23.6	23.5		
Fe	mgL^{-1}	0.032	0.020	<0.002	0.004	0.033	0.058		
Mn	mgL^{-1}	0.011	0.009	0.024	0.021	0.014	0.001		
F	mgL^{-1}	0.14							
Cl	mgL^{-1}	8.6							
SO_4	mgL^{-1}	16							
NO_3	mgL^{-1}	0.97							
NH_4	mgL^{-1}		0.6	<0.5	<0.5				
PO_4-P	mgL^{-1}	<0.005							
SiO_2	mgL^{-1}					14.5	14.3		
colif. 35 ^{o}C	cfu/100mL					0	0		
HPC 22^{o}C	cfu/mL	300	190	180	36	190	17		
HPC 35^{o}C	cfu/mL	290	220	190	33	200	23		
max. D rate	μSvh^{-1}		0.25			3.9			
consumed	m^3		1.65			6.03		9.71	
	BV		41			151		243	

* On 31.11.1997, two samples from the effluent were collected. Effluent #1 was collected 5 litres after the flowing started and effluent #2 after 200 litres.

Table XI. *Test location F.*

Parameter	**Date**	**12.7.1995**	**21.8.1997**		**15.7.1998**	
	Unit	**influent**	**influent**	**effluent**	**influent**	**effluent**
Rn-222	BqL^{-1}	3000	1660	2.2	2790	0.9
U-238	BqL^{-1}	0.564	0.576	0.423	0.544	0.547
U-234	BqL^{-1}	1.38	1.425	1.09	1.419	1.383
uranium	mgL^{-1}	0.046	0.047	0.034	0.115	0.112
Ra-226	BqL^{-1}	0.21	0.22	0.07	0.13	0.11
Pb-210	BqL^{-1}	0.071			0.075	0.017
Po-210	BqL^{-1}	0.134			0.137	0.047
gross-α	BqL^{-1}	2.1	2.2	1.6	1.8	1.7
temperature	^{o}C				10	9.5
O_2	mgL^{-1}		3.4	1.2	1.7	0.2
CO_2	mgL^{-1}				2.2	<1.0
redox	mV				231	220
pH			8.29	8.24	8.1	8.3
tot. hardness	mmolL^{-1}		0.67	0.59		
alkalinity	mmolL^{-1}		3.01	3.01		
$KMnO_4$	mgL^{-1}		4.1	<1	4.8	1.6
TOC	mgL^{-1}				2.4	0.8
turbidity	FTU		0.24	<0.05	0.19	0.1
el. cond.	mSm^{-1}		43.2	45.7	44	44.1
Fe	mgL^{-1}		0.019	<0.002	0.016	0.004
Mn	mgL^{-1}		0.024	0.029	0.008	0.009
Na	mgL^{-1}		71.0	75.0		
K	mgL^{-1}		3.70	3.20		
F	mgL^{-1}		1.43	1.46		
Cl	mgL^{-1}		23.2	26.1		
SO_4	mgL^{-1}		24.1	26.2		
NO_3	mgL^{-1}		0.34	0.27		
NH_4	mgL^{-1}		<0.01	<0.01		
PO_4-P	mgL^{-1}		0.012	0.038	0.011	0.011
SiO_2	mgL^{-1}				11.9	11.3
HPC 22^{o}C	cfu/mL		4	25	5	2
HPC 35^{o}C	cfu/mL		1	22	2	1
max. D rate	μSvh^{-1}		6.5		12.6	

Table XII. *Test location G. This test location has a POU anion exchanger (AE) installed in the kitchen water line.*

Parameter	Date	24.10.96	11.12.97		10.3.98		18.5.98		
	Unit	influent	influent	effluent	influent	effluent	influent	effluent	AE
Rn-222	BqL^{-1}	3700	5100	<1	5600	<1	7350	1.8-2.2	
U-238	BqL^{-1}	0.72	0.867	0.498	0.826	0.803	0.828	0.846	0.008
U-234	BqL^{-1}	1.22	1.32	0.753	1.23	1.23	1.22	1.27	0.011
uranium	mgL^{-1}	0.058	0.070	0.040	0.067	0.065	0.067	0.068	0.001
Ra-226	BqL^{-1}	0.29	0.30	0.11	0.32	0.25	0.32	0.26	0.23
Pb-210	BqL^{-1}	0.435			0.517	0.149	0.616	0.151	0.098
Po-210	BqL^{-1}	0.191	(0.455)	(0.018)	0.461	0.043	0.360	0.031	0.014
gross-α	BqL^{-1}	2.4	2.6	1.1	2.6	2.0	2.9	2.4	0.32
temp.	°C		5.9	5.9	6.1	4.7-5.1			
O_2	mgL^{-1}		0.2	0.2	0.3	0.2	0.1	0.1	
CO_2	mgL^{-1}						1.3	1	
pH			8.1	8.4	8.1	8.1	8.1	8.2	
tot.hardn.	mmolL^{-1}		0.82	0.81	0.80	0.80			
alkalinity	mmolL^{-1}		1.95	1.94	1.92	1.94			
$KMnO_4$	mgL^{-1}		2.5	<1	4.1	<1	2.6	<1	
TOC	mgL^{-1}		1.5	0.5	1.5	0.4	1.8	0.5	
turbidity	FTU		<0.05	0.05	<0.05	<0.05			
el. cond.	mSm^{-1}		25.4	25.4	24.7	24.7	24.7	24.7	
Fe	mgL^{-1}		0.033	0.003	0.019	0.003	0.017	0.008	
Mn	mgL^{-1}		0.027	0.022	0.024	0.023	0.027	0.026	
NO_3	mgL^{-1}		<0.5	<0.5					
NH_4	mgL^{-1}		0.01	0.01					
PO_4-P	mgL^{-1}		<0.005	0.058	<0.005	0.006			
colif. 35°C	cfu/100mL				0	0			
prel. E.Coli	cfu/100mL				0	0			
HPC 22°C	cfu/mL				72	8	7		770
HPC 35°C	cfu/mL				33	0	0		880
max. D rate	µSvh^{-1}		69.5		65.2			111	
consumed	m^3		4.71		35.0			56.9	
	BV		75		555			903	

Table XIII. *Test location G (cont.).*

Parameter	Date	13.8.98		23.9.98			15.3.99	
	Unit	**influent**	**effluent**	**influent**	**effluent**	**AE**	**influent**	**effluent**
Rn-222	BqL^{-1}	5440	1.1	5580-6000	0.13-1.1		6550	2.0
U-238	BqL^{-1}							
U-234	BqL^{-1}							
uranium	mgL^{-1}							
Ra-226	BqL^{-1}			0.29	0.26	0.26	0.29	0.29
Pb-210	BqL^{-1}			0.451	0.200	0.082		
Po-210	BqL^{-1}			0.206	0.047	0.013		
gross-α	BqL^{-1}			2.7	2.5	0.32	3.4	2.9
temp.	°C			9.6	11.3-12.4			
O_2	mgL^{-1}			0.3	1.0			
CO_2	mgL^{-1}			1.8	2.6			
redox	mV			257	265			
pH				7.9	7.9			
$KMnO_4$	mgL^{-1}			2.5	<1			
TOC	mgL^{-1}			1.6	0.5			
el. cond.	mSm^{-1}			25.4	25.1			
Fe	mgL^{-1}			0.016	0.005			
Mn	mgL^{-1}			0.025	0.026			
HPC 22°C	cfu/mL			6	7			
HPC 35°C	cfu/mL			0	1			
max. D rate	µSvh^{-1}	96.3		87				
consumed	m^3	84.9		99.9			158	
	BV	1347		1586			2504	

Table XIV. *Test location G (cont.).*

Parameter	Date	13.8.99	
	Unit	**influent**	**effluent**
Rn-222	BqL^{-1}	6250	1.8
Ra-226	BqL^{-1}	0.29	0.23
gross-α	BqL^{-1}	3.4	2.9
consumed	m^3	239	
	BV	3789	

***Table XV**. Test location H,* **”raw”** *stands for raw water,* **”AE”** *for anion exchanged water and* **”GAC”** *for first anion exchanged and then GAC filtered water, which is the finished water.*

Parameter	Date	18.5.1998			21.7.1998			29.9.1998		
	Unit	**raw**	**AE**	**GAC**	**raw**	**AE**	**GAC**	**Raw**	**AE**	**GAC**
Rn-222	BqL-1	2240		10	1610	1650	1	1920		6.2-15.6
U-238	BqL-1	1.77	0.0020	0.0028						
U-234	BqL-1	2.65	0.0028	0.0030						
uranium	mgL-1	0.143	0.0002	0.0002						
Ra-226	BqL-1	0.04	< 0.01	< 0.01	0.08	< 0.01	0.02	0.04	0.02	
Pb-210	BqL-1	0.116	0.071	0.010	0.190	0.052				
Po-210	BqL-1	0.074	0.015	0.010	0.062	0.018				
gross-α	BqL-1	4.4	0.08	0.06	4.5	0.09	0.09	6.4	0.12	
temp.	^{o}C				12.1	13.1	13.3	8.5		7.5-11.0
O_2	mgL^{-1}					4.0	0.7		5.1	4.3
CO_2	mgL^{-1}					7.5	9.7		13	10
redox	mV					320	320		357	340
pH		6.7	6.9	7.1	6.9	7.1	7.2	6.7	6.6	7.5
alkalinity	mmolL^{-1}	1.00	1.43		1.26	1.25			1.35	1.37
TOC	mgL^{-1}	2.5	1.4	0.5	1.9	1.1	0.6	2.0	1.2	0.4
turbidity	FTU	1.00	0.18		1.02	0.23	0.23	0.51	0.13	0.08
Fe	mgL^{-1}	0.160	0.008	0.003	0.130	0.005	0.004	0.044	0.005	0.003
Mn	mgL^{-1}	0.004	0.003	0.002	0.004	0.005	0.004	0.003	0.002	0.001
F	mgL^{-1}				0.28	0.42		0.41	0.38	
Cl	mgL^{-1}	14	17		12	15		14	16	
SO_4	mgL^{-1}				26	22		24	24	
NO_3	mgL^{-1}	14.1	6.0		9.7	9.6		8.0	6.7	
HPC 22^{o}C	cfu/mL	200	100	330	930	290	220	75	12	95
HPC 35^{o}C	cfu/mL	37	24	9	110	32	25	1	0	14
max. D-rate	µSvh^{-1}								18	
consumed	m^3		66.9			88.7			104	
	BV		1714			2273			2683	

Table XVI. *Test location H (cont.).*

Parameter	**Date**	**8.6.1999**	
	Unit	**raw**	**GAC**
Rn-222	BqL^{-1}	1700	37
Ra-226	BqL^{-1}	0.03	<0.01
gross-α	BqL^{-1}	4.1	0.07
consumed	m^3	189.5	
	BV	4860	

Table XVII. *Test location I.*

Date	**consumed**		**Rn-222 (BqL^{-1})**		**Ra-226 (BqL^{-1})**		**gross-α (BqL^{-1})**	
	m^3	**BV**	**influent**	**effluent**	**influent**	**effluent**	**influent**	**effluent**
26.3.98	8.96	142	1 990	2.1	0.29	0.19	6.9	5.9
25.6.98	32.1	509	1 300	1.0	0.19	0.20	6.1	5.5
29.9.98	55.3	878	2 160	2.3	0.23	0.25	10.6	8.1
17.3.99	94.5	1 500	1 740	5.3	0.19	017	8.8	9.8

Table XVIII. *Test location J.*

Date	**consumed**		**Rn-222 (BqL^{-1})**		**Ra-226 (BqL^{-1})**		**gross-α (BqL^{-1})**	
	m^3	**BV**	**influent**	**effluent**	**influent**	**effluent**	**influent**	**effluent**
23.3.98	59.4	943	4 580	1.2	0.09	0.06	1.6	1.0
8.6.98	103.9	1 650	4 500	2.6	0.08	0.07	1.8	1.5
15.12.98	216.3	3 430	4 200	2.4	0.08	0.07	1.7	1.5
13.4.99	279.6	4 440	4 560	1.3	0.09	0.07	1.8	1.0
10.11.99	418.9	6 650	4 480	2.5				

Table XIX. *Test location K.*

Date	**consumed**		**Rn-222 (BqL^{-1})**		**Ra-226 (BqL^{-1})**		**gross-α (BqL^{-1})**	
	m^3	**BV**	**influent**	**effluent**	**influent**	**effluent**	**influent**	**effluent**
24.11.98	11.6	184	1 300	0.5	0.08	0.07	0.58	0.23
2.6.98	104.3	1654	1 550	0.5	0.15	0.07	0.45	0.33
2.12.98	200.5	3183	1 500	0.5	0.14	0.11	0.38	0.20
23.3.99	257.5	4087	1 430	0.5	0.11	0.11	0.40	0.25
14.9.99	349.3	5545	1 480	0.4	0.13	0.09	0.38	0.39

Table XX. *Test location L.*

Parameter	**Date**	**19.7.95**	**2.7.98**		**19.10.98**		**2.5.99**	
	Unit	**influent**	**influent**	**effluent**	**influent**	**effluent**	**influent**	**effluent**
Rn-222	BqL^{-1}	6000	5780	69-76	6380	99	3040	45
U-238	BqL^{-1}	0.087			0.080	0.003		
U-234	BqL^{-1}	0.25			0.236	0.009		
uranium	mgL^{-1}	0.0070			0.0065	0.0003		
Ra-226	BqL^{-1}	1.1	0.19	0.01	0.139	0.135	0.29	0.26
Pb-210	BqL^{-1}	0.046	0.417	0.085				
Po-210	BqL^{-1}	2.26	1.92	0.133				
gross-α	BqL^{-1}	2.8	2.3	0.13	2.6	0.18	1.7	0.28
temp.	^{o}C		4.5	8-9				
O_2	mgL^{-1}		0.6	0.2				
CO_2	mgL^{-1}		7.0	<1				
redox	mV		565	214				
pH			7.3	8.9				
TOC	mgL^{-1}		2.1	0.2				
el. cond.	mSm^{-1}		29.5	30.3				
Fe	mgL^{-1}		0.16	0.015				
Mn	mgL^{-1}		0.066	0.0053				
HPC 22^{o}C	cfu/mL		0	48				
HPC 35^{o}C	cfu/mL		0	42				
max. D rate	μSvh^{-1}		35.6					
consumed	m^{3}		1.45		11.4		24.5	
	BV		23		181		389	