

increasing the amount of air, either by using more air or by recirculating the water several times through ejector mouth-pieces.

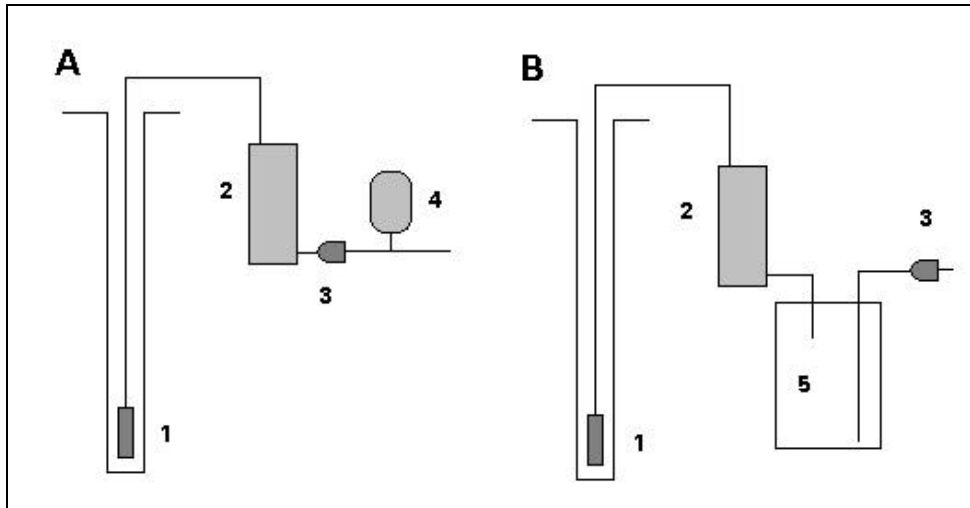
Eleven installations of equipment for radon removal by aeration supplied by four commercial companies were studied. Seven units were selected for long-term monitoring. The main criteria for the selection of the households where aerators were installed were the concentrations of radon ( $^{222}\text{Rn}$ ), iron (Fe), manganese (Mn) and organic matter (TOC, total organic carbon) (Table I).

Different types of installation were also studied (Figure 1). Basically, there are two ways to carry out the installation of an aerator. The aerated water can either be directed into a pressure tank (type A) or a storage tank (type B). The pressure tank maintains the hydraulic pressure in the household water line. When a storage tank is applied, the booster pump starts up every time water is taken from the taps. An additional pressure tank before the aerator, piping and three valves are needed to by-pass the system. The types of installation presented in the figure are largely simplified.

**Table I.** Water quality data at the selected test locations. The concentrations of  $^{222}\text{Rn}$ , Fe, Mn and TOC may have varied, the maximum values are presented. The installation types A and B are set out in Figure 1.

Test Location	Company	Model	Install. Type	Volume (L)	$^{222}\text{Rn}$ (Bq/L)	Fe (mg/L)	Mn (mg/L)	TOC (mg/L)
1A	Overcraft	Radox	B	290	510	69	3	-
1B		Radox	B	690	18 900	410	240	2.0
1C		Radox	B	300	17 000	130	59	3.4
2A	Vartiainen	modified	B	200	9 000	18	43	1.5
2B		RA 300/35	B	300	27 500	21	11	1.2
2C		RA 300/35	B	300	14 200	170	77	0.8
3A	WatMan	RF-150/KR6	A	150	22 200	65	95	0.9
3B		RF-150/KR6	A	150	15 200	390	170	7.6
3C		RF-150/3R	A	150	1 200	170	92	-
3D		Rn-A1	A/B	300	670	64	22	-
4A	Sarholms	Radonett	A	100	42 000	45	3	1.1

- not available or determined.



**Figure 1.** Two types of installation of a treatment system based on aeration. In the figure, number 1 corresponds to the (submersible) well pump, 2 to the aerator, 3 to the booster pump, 4 to the pressure tank and 5 to the storage tank. In type A, the water is stored under pressure (normally 1.5–5 bar) whilst in type B the storage tank operates under atmospheric pressure.

Radon concentration as well as the relevant water quality parameters (Fe, Mn, TOC, pH, CO<sub>2</sub>, Redox, O<sub>2</sub>, alkalinity and temperature) were monitored in both influent and effluent at approximately three month intervals. Respectively, the microbiological quality of the water was studied by determining the heterotrophic plate count (HPC in 22°C and 35°C). At an early stage in the study it was noticed that the radon removal efficiency of an individual unit is not always constant. The concentration of radon in the aerated water varied with the volume of water flowing, with the flow rate, and with the water usage prior to sampling. Furthermore, the installation (e.g. the power of the well pump and the booster pump) had an effect on the removal efficiency. One radon sample taken from the effluent did not provide sufficient information on the removal efficiency and therefore a standard test protocol was developed. This test protocol revealed malfunctions that could not be detected with conventional sampling where only one or two samples taken were from the effluent (Figure 2).

The flow rate was adjusted to a constant value (4, 8, 12 L/min). Samples were taken every 10–20 litres. If the flow rate did not remain constant (installation type A, Fig. 1) the flow rate was measured with a stop-watch and a measuring cylinder after each sample was collected. When more than

100 litres of water had flowed the sampling was stopped (time recorded) and a second sample of the influent was taken.

The radon concentrations (Bq/L) were plotted against the volume of water that had flowed. The removal efficiency for the water sample that was first taken, was calculated according to the equation

$$(1) \quad R_0 = \left(1 - \frac{C_0}{C_{i0}}\right) \cdot 100\% ,$$

where  $R_0$  is the *initial removal efficiency*,  $C_0$  is the radon concentration in the first effluent sample, and  $C_{i0}$  is the concentration in the first influent sample.

The *average removal efficiencies* for 50 and 100 litres were calculated according to the equation

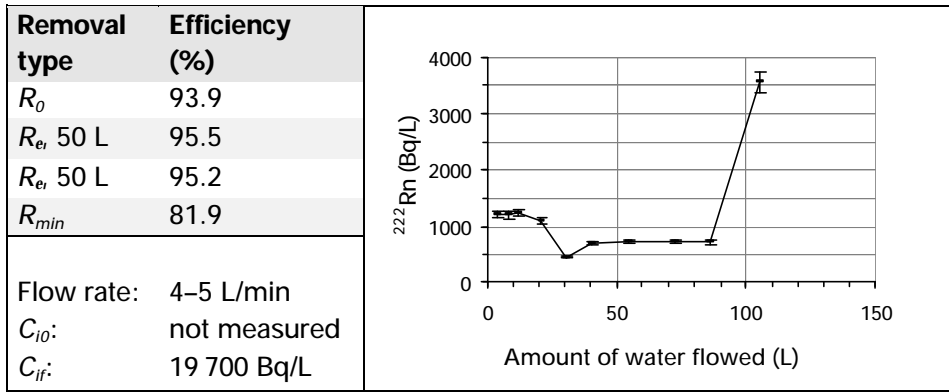
$$(2) \quad R_e = \frac{(C_0 \cdot V_0) + \sum_n \left[ \frac{1}{2} \cdot (C_n + C_{n-1}) \cdot (V_n - V_{n-1}) \right]}{V_{tot} \cdot \frac{C_{i0} + C_{if}}{2}} \cdot 100\% ,$$

where  $C_0$  is the radon concentration in the first effluent sample,  $V_0$  is the volume of water that had flowed before the first sample was taken,  $C_n$  is the radon concentration of the effluent sample  $n$ ,  $V_n$  is the volume of water that had flowed when sample  $n$  was taken,  $C_{i0}$  and  $C_{if}$  are the radon concentrations in the initial and final influent samples respectively, and  $V_{tot}$  is the volume at the last sampling, for which the efficiency was calculated (50 or 100 litres).

The *minimum removal efficiency* during the sampling run was calculated according to the equation

$$(3) \quad R_{\min} = \left(1 - \frac{2C_{\max}}{C_{i0} + C_{if}}\right) \cdot 100\% ,$$

where  $C_{\max}$  is the maximum radon concentration measured in effluent.  $C_{i0}$  and  $C_{if}$  are the radon concentrations in the initial and final influent samples, respectively.



**Figure 2.** The results obtained at test location 3A. The O-ring of a solenoid valve was broken and insufficiently aerated water was released into the waterline.

The rate of radon removal from water was also determined. A high removal efficiency may naturally be attained if the aeration time is prolonged, but by doing this less water is produced. The rate of removal will ultimately control the throughput of the system. Since radon removal by aeration is a random process, the rate of removal can be modelled by first-order kinetics. The kinetics equation can be presented in the form of

$$(4) \quad K_s = \frac{-\ln\left(\frac{C_t}{C_0}\right)}{t}, \text{ where}$$

$C_t$  is the radon concentration in the treated water after the aeration (processing) time,  $t$  has passed,  $C_0$  is the radon concentration in the raw water and  $K_s$  is the (*first-order kinetics*) *stripping constant*, which represents the rate of removal. As such, the  $K_s$  constant is not sufficient to be used when comparing the capacity of different aerators. The size of the aerator must also be considered, because various volumes of water (batches) are treated by different aerators. Since a removal efficiency of 99% is sufficient in most cases, a new parameter, the *effective flow rate*,  $Q_{eff}$  is introduced. The  $Q_{eff}$  is the flow rate by which the aerator attains the removal efficiency of 99%. It is calculated by the following equation:

$$(5) \quad Q_{eff} = 0.217 \cdot K_s \cdot V_{batch},$$

where  $K_s$  is the stripping constant of the aerator and  $V_{batch}$  the volume of the water (batch) that is aerated in one go.

The aerator “Radonett” (4A) had the highest removal rate. Radon was released from water rapidly and only short aeration times were needed to mitigate even extremely high levels of radon. The Radox aerator (1A–1C) was also very efficient and the removal rate was high. The aerator needs longer aeration times than Radonett, but larger volumes of water are aerated in one batch in the corresponding time. Radonfällan RF-150 (3A–3C) had a good removal efficiency but it required rather long aeration times, and only a small batch of water could be aerated in one go. The Orwa aerator (2A–2C) had problems with both the removal efficiency and the removal rate. Even during low consumption, radon laden water could get into the plumbing. A summary of the radon removal performance of the aerators studied is presented in Table II.

**Table II.** The removal efficiencies, rates of removal and effective flow rates calculated for different aerators.

Test location	$R_0$ (%)	$R_e$ , 50L (%)	$R_e$ , 100L (%)	$R_{min}$ (%)	$K_s$ ( $\text{min}^{-1}$ )	$Q_{eff}$ (L/min)
1A	97.0	97.0	97.0	97.0	0.7	44
1B	99.2–99.7	99.2–99.7	99.2–99.7	99.2–99.7	0.25	37
1C	94.7–99.0	94.7–99.0	94.7–99.0	94.7–99.0	0.6	39
2A	87.1–92.0	81.3–99.4	85.8–99.5	60.7–98.9	0.2	9
2B	>99.9	95.9	91.2	77.0	0.2	11
2C	62.5–95.4	62.0–95.0	61.2–93.0	59.4–76.7	0.2	11
3A	93.9–99.9	95.5–99.4	95.2–99.5	81.9–91.2	0.6	14
3B	96.3–98.5	94.4–98.4	95.1–98.4	86.1–98.1	0.5	12
4A	>99.9	>99.9	>99.9	>99.9	3.7	48

The water quality remained good. Some iron may precipitate as ferric hydroxide during aeration. The precipitates can settle down on the bottom of the aeration tank, be removed by a sediment filter installed after the aerator, or be released in the water line. Therefore, the concentrations of iron are usually different in raw and aerated water. Manganese may co-precipitate with ferric hydroxide. Iron and manganese precipitates can cause fouling of the treatment system. Water becomes virtually saturated with oxygen during aeration. This improves the taste of the drinking water, and reduces its corrosiveness. Carbon dioxide is removed, which causes a rise in pH value. Water with a higher pH value is less corrosive for the

plumbing. Other water quality parameters change only slightly. No clear trends could be observed in their results.

### **3.5 Removal of Radon by Aeration: Testing of Various Aeration Techniques for Small Waterworks**

The aim of this study was to design and test different aeration techniques for radon removal, to compare their cost-effectiveness and to write guidelines on how to build aeration systems. For that purpose one waterworks, where radon removal was based on spray and diffused bubble aeration, was designed and installed in Finland and experiments were made in Germany with a counter current packed tower column in half technical scale to evaluate its ability to remove radon and carbon dioxide.

The aim of this study was also to compare the different aeration techniques already applied for radon removal at waterworks in Finland, Germany and Sweden. The data on the radon removal efficiencies, on the descriptions of the aeration principles and on the other water treatments applied simultaneously with radon removal were gathered from several waterworks in these countries. The most important water quality parameters were determined in raw and treated water at the Finnish waterworks to see the effect of the water treatment on its quality. In most of the waterworks studied here, aeration was applied together with other water treatments. In Finland and Sweden typical treatments are the removal of iron, manganese or humus or the alkalisation of too acidic and soft waters. In Germany the typical water treatment in the areas where increased radon levels occur in groundwater, is de-acidifying, but iron and manganese removal is also necessary.

The aim of this project was also to collect data on radon removal efficiencies in those waterworks which apply aeration for removing Fe, Mn, CO<sub>2</sub> or H<sub>2</sub>S. For this purpose water samples from the raw and treated water were collected at several Finnish waterworks.

A number of radon measurements were carried out at Finnish, Swedish and German waterworks which apply different aeration techniques either for radon removal or in other water treatment processes, with the aim of surveying their radon removal capabilities.

A water treatment plant for the removal of radon ( $^{222}\text{Rn}$ ), uranium ( $^{238,234}\text{U}$ ), manganese and hardness was designed and installed at one Finnish waterworks supplying water to 350 people. Its radon removal is based on combined spray and diffused aeration. Water is sprayed into a cylindrical basin through four spray nozzles located about 30 cm above water level. Four diffusers are placed at the bottom of the basin to accomplish aeration by diffused bubble aeration having an air-to-water ratio of 11. After aeration the water is discharged into a storage basin. At this waterworks uranium, manganese and hardness are removed with separate anion and cation exchangers before aeration. Thus raw water first enters the cation exchanger, where manganese and hardness are removed and then the water flows through the anion exchanger for uranium removal. The ion exchangers are regenerated automatically every night with saturated sea salt solution.

Additionally, six other Finnish waterworks where radon is removed by various aeration methods, were located and their radon removal efficiencies were determined. Radon removal efficiencies in these seven waterworks varied between 67% and 98% and were sufficient in all waterworks to attain the limit for radon (300 Bq/L) set in the Finnish regulations. The concentration of radon in the raw water varied from 330 to 5 800 Bq/L and at some waterworks they varied greatly at different sampling times. Practically all uranium was removed by an anion exchanger.

Radon removal efficiencies were compiled from 18 small Swedish waterworks where various types of commercial aerators had been installed for radon removal. The results showed that the removal efficiencies varied between 93% and 99%. All these aerators were efficient enough to reduce the radon levels (in raw waters between 400–4 000 Bq/L) below the limit of 100 Bq/L set in Sweden and to supply water even to dozens of households.

Radon removal efficiencies were determined at nine Finnish waterworks where various types of aeration or oxidation techniques were applied in the removal processes for iron and manganese. The radon levels in the raw waters were quite low (8–110 Bq/L) but the water treatment capacities varied largely (24–5 600 m<sup>3</sup>/d). The radon reduction varied greatly (from 13% to 94%). It seems that better rates of removal are attained by applying packed tower or drip aeration (from 72% to 94%) than by using spray aeration or cascade gravitation (from 13% to 58%). Aeration removed radon and carbon dioxide at a very similar rate. The radon reduction was usually a little higher than that of carbon dioxide. Iron and manganese were removed efficiently at all of these waterworks.

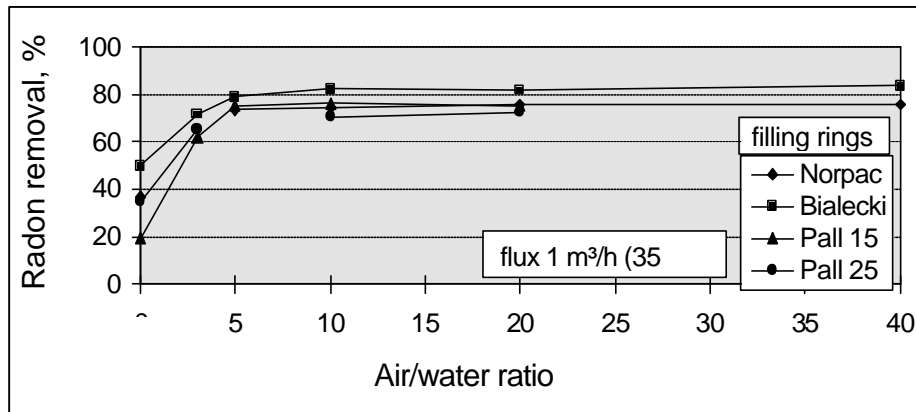
Radon removal efficiencies were studied at 11 German waterworks where conventional water treatment methods are applied mainly for de-acidification or for iron and manganese removal. Due to the geological situation elevated levels of radon in ground waters are not so common in Germany as in Scandinavia, and also the concentrations of radon are lower in German raw waters which are used for the production of drinking water (max. about 1500 Bq/L). The radon reductions at these waterworks varied from 0% to 98%.

At one of the German waterworks (water consumption 700 m<sup>3</sup>/d) a Venturi water aeration device was installed as a first treatment step to remove carbon dioxide and radon (300 Bq/L in raw water) and to add oxygen. The aeration process takes place in the container room of the waterworks at a water flux of approximately 30 m<sup>3</sup>/h. The removal efficiency of radon was always from 70% to 80%. The transfer of radon from water to air leads to a strong enrichment of radon in the indoor air. To attain a continual exchange of the indoor air with outdoor air, ventilation windows were set into the outer walls. Nevertheless, average indoor radon level of 62 000 Bq/m<sup>3</sup> was found in the container room during a four-day measurement period.

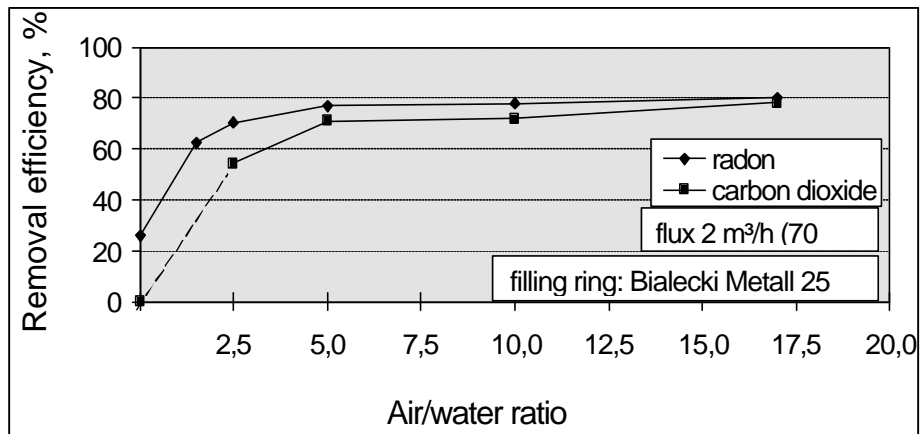
At another German waterworks (water throughput 1200 m<sup>3</sup>/d) shallow bed cross-flow aeration equipment was installed as a first treatment step to remove carbon dioxide and radon (130 Bq/L in raw water) and to add oxygen. The equipment is operated at a water throughput of approximately 50 m<sup>3</sup>/h and an air-to-water ratio of 16. Water passes through the equipment on a slightly inclined bed while air is blown through many nozzles set into the bed in a cross-flow direction. To avoid radon and carbon dioxide enrichment in the indoor air of the waterworks, the process-air is led out of the building through a pipe. The radon reduction was above 98%. Average indoor air radon levels were only 500 Bq/m<sup>3</sup> because the aerator was equipped with the ventilation pipe.

Experiments were made in Germany with a counter-current packed tower column in half-technical scale to compare the removal behaviour of radon and carbon dioxide from water during its aeration. In a packed tower column water flows downstream while air is blown upstream. The filling rings inside the equipment form a large surface where dissolved gases transfer from the liquid to the gaseous phase. The process-air can be led out of the equipment by a pipe.

Water with a radon content of 1000 Bq/L and a carbon dioxide content of 1 mmol/L was used for the experiments. The column was 150 cm high and had a diameter of 19 cm. In several experiments it was filled with four different filling rings made of metal or polypropylene. The filling height of the column was 1 m. By using throughputs of 1 to 4 m<sup>3</sup>/h (specific fluxes: 35 to 140 m<sup>3</sup>m<sup>-2</sup>h<sup>-1</sup>) and air/water ratios of up to 40, removal efficiencies up to 85 % were achieved (Figure 3 and Figure 4).



**Figure 3.** Radon removals attained by various filling rings as a function of air-to-water ratio.



**Figure 4.** Radon and carbon dioxide removals as a function of air-to-water ratio.

From a packed tower column radon or carbon dioxide-rich air can be diverted directly out of a waterworks building. The tested column in half-technical scale removed 85% of the water-dissolved radon. By using a typical filling ring heights of two to three metres in a practical application at waterworks a removal efficiency of 95% can be expected. Additionally the experiments showed that high radon removal efficiencies were attained by using air-to-water ratios as low as five and that carbon dioxide and radon showed very similar removal behaviour.

### **3.6 Removal of Radionuclides from Private Well Water with Granular Activated Carbon (GAC): Removal of Radon**

In drinking water treatment GAC filtration has been primarily used for removing taste, odour, colour and synthetic organic chemicals. Activated carbon removes effectively low or trace concentrations of impurities occurring in water by adsorption. The adsorption capacity of GAC is directly related to the extremely high internal surface area within the porous structure, which consists of macropores and micropores.

Radon ( $^{222}\text{Rn}$ ) can be effectively adsorbed by a GAC filter. Since radon is chemically inert and radon does not form bonds, the adsorption process is one of purely physical adsorption. Also the short-lived decay products of radon are retained on GAC. As a consequence the filter matrix will emit gamma radiation. The dose rate in the vicinity of the filter can approach 100  $\mu\text{Sv/h}$ . The radioactivity in the GAC masses can be a problem when they are disposed of.

The main objective of this study was to investigate radon removal by GAC filtration in everyday household use. Test locations were selected so that the water types most typically found in bedrock were covered. The classification of water types was based on the concentration of radon, iron (Fe), manganese (Mn), organic matter (TOC) and the long-lived radionuclides of the uranium series (U,  $^{226}\text{Ra}$ ,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ ). Their removal and effect on the GAC filter's performance and the effect of pre-filters were also studied. Other aspects considered in this study were: the effect of GAC filtration on water quality parameters (including microbiological quality), gamma radiation levels on the surface of the filter and in its vicinity, necessary shielding, and waste disposal of spent carbon.