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Natural radioactivity in drinking water in Finland

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The mean annual effective dose from natural radionuclides for users of drilled wells was estimated to be 0.41 mSv, for users of wells dug in the ground 0.05 mSv and for people using water from waterworks 0.02 mSv. The highest effective dose from drinking water was caused by ^{222}Rn (Fig. 2) constituting 75% and 60% of the total effective dose caused by all natural radionuclides for drilled-well users for users of wells dug in the ground, respectively. ^{210}Po and ^{210}Pb contributed the most to the effective dose caused by the long-lived radionuclides. Contribution of the isotopes of radium (^{226}Ra and ^{228}Ra) to the total effective dose from drinking water was minor.

Introduction

The activity concentrations of natural radionuclides in groundwater are connected to the activity concentrations of uranium (^{238}U and ^{235}U) and thorium (^{232}Th) and their decay products in the ground and bedrock. This is due to groundwater reacting with the ground and bedrock and releasing quantities of dissolved components that depend on the mineralogical and geochemical composition of the soil and rock, chemical composition of the water, the degree of weathering of the rock, redox conditions and the residence time of groundwater in the soil and bedrock.

Natural radionuclides in the Finnish groundwater originate mainly from the decay series of ^{238}U (Asikainen 1981a, 1981b, Salonen 1994, Vesterbacka 2005). The most harmful of these, from the point of view of radiation protection, is ^{222}Rn. Other alpha-active isotopes include ^{238}U, ^{234}U, ^{210}Po and ^{226}Ra. In addition, beta-active ^{210}Pb, ^{228}Ra and ^{40}K isotopes are also found in drinking water. The isotope ^{228}Ra originates from the decay series of ^{232}Th. In addition to the radiation dose from the ingested ^{222}Rn, the water-born ^{222}Rn is a source of indoor-radon because ^{222}Rn is released into indoor air during water usage and inhaled ^{222}Rn daughters affect lungs.

Approximately 90% of Finns use water from waterworks as their daily drinking and household water. The number of people using private wells is approximately 500 000 that corresponds to about 10% of the Finnish population (Mäkeläinen *et al.* 2001). The activity concentration of natural radionuclides depends on the water source. In the surface water, activity concentrations are typically very low. Occasionally increased concentrations are found in dug-well water, whereas in drilled-well water activity concentrations can be exceptionally high.

In Finland, the annual effective radiation dose in household water should not exceed 0.5 mSv (Finnish Centre for Radiation and Nuclear Safety 1993). The limit for ^{222}Rn in public water has been set to 300 Bq l^{-1}, and for the other radionuclides to 0.5–20 Bq l^{-1}, depending on the radionuclide. For private wells, the limit for ^{222}Rn has been set to 1000 Bq l^{-1} (Ministry of

Social Affairs and Health 2001); for the other radionuclides the limits have not yet been set.

^{238}U has a special position. In addition to radioactivity, it has a chemical toxicity that predominately affects the kidneys (Auvinen *et al*. 2002, Kurttio *et al*. 2002, 2005). In Finland, there is no national guideline for ^{238}U based on its chemical toxicity. However, according to the Radiation and Nuclear Safety Authority recommendations, concentration of ^{238}U in drinking water should not exceed 0.1 mg l^{-1}).

In Finland activity concentration of natural radionuclides in drinking water has been studied since 1960. By the year 2005, 9000 drilled wells, 5000 dug wells and over 1000 waterworks or water catchments were measured. This paper summarises results of measurements of natural radioactivity in drinking water in Finland.

Material and methods

Water sources

In this study, the results from randomly selected 288 drilled and 184 dug wells used as sources of drinking water are reported (Vesterbacka *et al*. 2005b).

The results on waterworks are based on over 1000 measurements from waterworks or water catchments (Mäkeläinen *et al*. 2001). In order to obtain representative estimates for average radionuclide concentrations in the consumed water, the proportions of surface and ground water at each individual waterwork were taken into account. The database of the Finnish Environment Institute was utilised in these calculations. The missing values for ^{226}Ra, ^{234}U, ^{238}U, ^{210}Po and ^{210}Pb were obtained with regression analysis using the values of gross alpha, gross beta and ^{222}Rn as explanatory variables. To obtain values for waterworks without any measurements, means from waterworks using the same water type (surface water, groundwater from soil or groundwater from bedrock) were used.

Water sampling

The samples for determination of ^{222}Rn were collected either into liquid scintillation vials that had been pre-filled with a liquid scintillation cocktail or into one-litre glass bottles in which the aliquot samples for ^{222}Rn measurement were taken to the laboratory. The samples for determination of other natural radionuclides (^{234}U, ^{238}U, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po) were collected in polyethylene or glass bottles. In the laboratory, the water samples were acidified with hydrochloric acid (4.5 ml concentrated HCl in one-litre water sample).

The samples from the private wells were collected mainly by municipal health inspectors but also by the owners of the wells. The samples from water catchments and waterworks were collected by municipal health inspectors and waterworks managers in accordance with the instructions given by the Radiation and Nuclear Safety Authority.

Determination of activity concentration

The ^{222}Rn concentration was determined using either the liquid scintillation spectrometry (most of the samples) or gamma spectrometry (Kahlos and Asikainen 1973, Salonen 1993a, Salonen and Hukkanen 1997).

The gross alpha activity — the total amount of ^{234}U, ^{238}U, ^{210}Po and ^{226}Ra — was determined using either a zinc sulphide counter or liquid scintillation spectrometry (Kahlos and Asikainen 1973, Salonen and Hukkanen 1997).

The ^{226}Ra was determined using liquid scintillation spectrometry via ^{222}Rn and its short-lived daughters from the gross alpha spectrum (Salonen 1993b, Salonen and Hukkanen 1997). Activity concentration of ^{226}Ra was also determined radiochemically based on the $BaSO_4$ precipitation (Kahlos and Asikainen 1973).

The activity concentration of ^{228}Ra was determined via its daughter nuclide ^{228}Ac using gamma spectrometry. The water samples (two litres) were first evaporated to a volume of 0.5 litres and thereafter they were transferred into a Marinelli beaker for gamma spectrometric counting.

The activity concentrations of ^{234}U, ^{234}U and ^{238}U were separated from other radionuclides using the ion-exchange method and alpha spectrometry (Alpha Analyst).

The activity concentrations of ^{210}Pb and ^{210}Po were determined using spontaneous deposition of ^{210}Po on a silver disk and alpha spectrometric measurement of the ^{210}Po activity (Häsänen 1977). ^{210}Pb was determined 200 days later from the same solution as the ^{210}Po. The final results were calculated from these two depositions (Vesterbacka and Ikäheimonen 2005). An alternative method for determination of ^{210}Pb was based on the extraction chromatography and liquid scintillation spectrometry (Vajda *et al*. 1997).

The effective annual dose from ingestion of radionuclides was calculated on the basis of the mean activity concentrations of the radionuclides presented in Table 1. The daily water consumption was considered to be 2.2 litres for long-lived radionuclides and 0.5 litres for ^{222}Rn. The conversion factors provided by the National Academy of Sciences were used for ^{222}Rn (National Research Council 1999); for the other radionuclides, those given in the Internal Commission on Radiological Protection were used (Internal Commission on Radiological Protection 1996).

Results

Activity concentrations

The mean activity concentrations in the drilled-well water were — depending on the radionuclide — from 2 to 20 times higher than in the dug-well water or in the waterworks (Table 1). The low activity concentrations in the water from the waterworks were due to the fact that many waterworks use surface water or groundwater as the water source.

^{222}Rn concentrations exceeded 1000 Bq l^{-1} in 10% of the drilled wells. In dug wells this limit was not exceeded. ^{222}Rn concentration of 100 Bq l^{-1} was exceeded in 59% of the drilled wells and in 11% of the wells dug in the ground. ^{238}U concentrations of 100 μg l^{-1} and 15 μg l^{-1} were exceeded in 5% and 18% of the drilled wells, respectively. In wells dug in the ground, the ^{238}U concentration was clearly lower and in only 3% it exceeded 15 μg l^{-1}. The reference concentrations of 0.2 Bq l^{-1} for ^{210}Pb and 0.1 Bq l^{-1} for ^{210}Po given by the Commission of the European Communities were exceeded in 4% and 9% of the drilled wells, respectively. The reference concentrations for ^{210}Po was exceeded in only 1% of the dug wells. High radium activity concentrations in the drilled-well water were rare: ^{226}Ra and ^{228}Ra concentrations exceeded 0.5 and 0.2 Bq l^{-1} in 2%–4% and 1%–2% of the drilled wells, respectively.

The highest activity concentrations of ^{222}Rn were found in southern Finland (Fig. 1), where the bedrock typically consists of granites. In addition, occasional high activity concentrations were found all over Finland. The spatial distributions of ^{234}U, ^{238}U, ^{210}Pb and ^{210}Po were essentially similar to that of ^{222}Rn. In contrast to other natural radionuclides, the highest ^{226}Ra activity concentrations were often found in coastal areas.

Effective doses

The mean annual effective dose from natural radionuclides for users of drilled wells was estimated to be 0.41 mSv, for users of wells dug in the ground 0.05 mSv and for those using water from waterworks 0.02 mSv (Table 2). The contribution of natural radionuclides in drinking water to the average annual effective dose of 3.7

Table 1. Mean activity concentration (Bq l^{-1}) of natural radionuclides in Finnish drinking water according to the water source (Mäkeläinen *et al.* 2001, Vesterbacka *et al.* 2005b, 2006).

Water source	^{222}Rn	Gross alpha*	^{238}U	^{238}U	^{234}U	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po
Drilled-well water	460	0.61	20.9	0.26	0.35	0.05	0.034	0.040	0.048
Dug-well water	50	0.05	1.2	0.015	0.02	0.02	–[1)]	0.013	0.007
Waterworks	27	0.04	1.2	0.015	0.02	0.003	–[1)]	0.003	0.003

* total amount of ^{238}U, ^{234}U, ^{226}Ra and ^{210}Po.
[1)] activity concentration not determined.

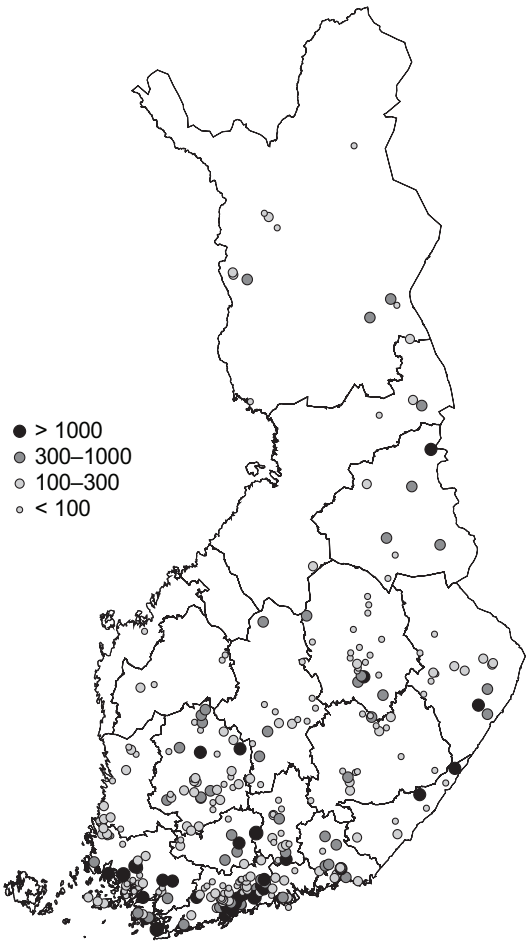

Fig. 1. The regional distribution of ^{222}Rn (Bq l^{-1}) in 288 drilled wells (Vesterbacka *et al.* 2005b).

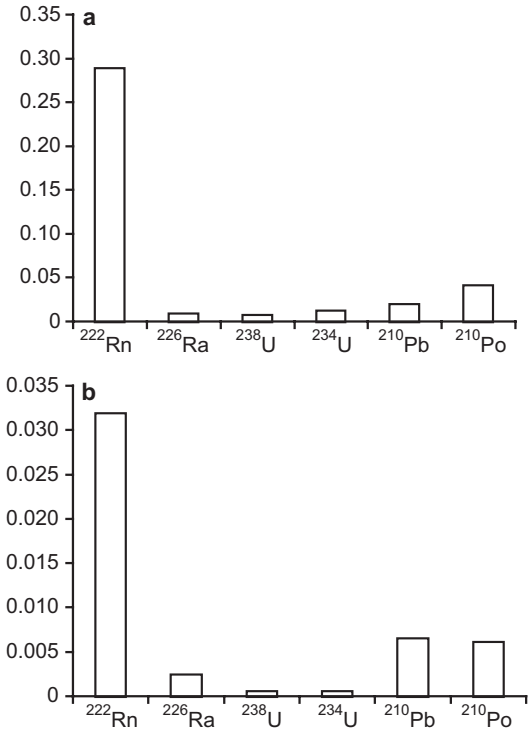

Fig. 2. Distribution of the dose (mSV) from ^{222}Rn, ^{210}Po, ^{210}Pb, ^{238}U, ^{234}U, ^{226}Ra in (**a**) drilled-well and (**b**) dug-well water (Vesterbacka *et al.* 2005b).

mSv from all sources for Finnish users of drilled wells was 11%.

The highest effective dose from drinking water was caused by ^{222}Rn (Fig. 2) constituting 75% and 60% of the total effective dose caused by all natural radionuclides for drilled-well users for users of wells dug in the ground, respectively (Vesterbacka *et al*. 2005b). ^{210}Po and ^{210}Pb contributed the most to the effective dose caused by the long-lived radionuclides. Contribution of the isotopes of radium (^{226}Ra and ^{228}Ra) to the total effective dose from drinking water was minor (Vesterbacka *et al*. 2006).

Discussion

In Finland, Sweden, Norway, Spain and Ukraine, natural radioactivity in groundwater has been found to be very similar. The overall radionuclide activity concentration is dominated by that

Table 2. Mean annual effective doses (mSv) from various radionuclides for users of drilled wells, dug wells and for people using water from waterworks (Mäkeläinen *et al.* 2001, Vesterbacka *et al.* 2005b, Vesterbacka *et al.* 2006).

Water source	Number of users	^{222}Rn	^{238}U	^{234}U	^{226}Ra	^{228}Ra	^{210}Pb	^{210}Po	Total
Drilled well water	200000	0.29	0.008	0.014	0.010	0.017	0.022	0.046	0.41
Dug well water	300000	0.032	0.001	0.001	0.003	–	0.007	0.007	0.05
Waterworks	4700000	0.02	0.0005	0.0008	0.0007	–	0.0015	0.003	0.02

of ^{222}Rn (Fernandez *et al*. 1992, Zelensky *et al*. 1993, Midtgård *et al*. 1998, Strand *et al*. 1998, Isam Salih *et al*. 2002, Vesterbacka *et al*. 2005b). A high mean activity concentration of ^{226}Ra (> 0.5 Bq l^{-1}) was found in Sweden, Ukraine and Spain (Fernandez *et al*. 1992, Zelensky *et al*. 1993, Isam Salih *et al*. 2002). In central Europe the mean ^{238}U concentration was typically low (< 10 μg l^{-1}), whereas mean concentrations larger than 20 μg l^{-1} were found in Finland, Sweden, Norway, Ukraine and Spain (Fernandez *et al*. 1992, Zelensky *et al*. 1993, Midtgård *et al*. 1998, Strand *et al*. 1998, Isam Salih *et al*. 2002, Vesterbacka *et al*. 2005b). Additionally, in France the mean ^{238}U concentration was elevated as compared with that (0.5–1.1 μg l^{-1}) in Slovenia and Germany (Saumande *et al*. 1973, Gans *et al*. 1987, Kobal *et al*. 1990). Studies including determinations of ^{210}Po and ^{210}Pb in European counties are scarce as compared with those examining ^{222}Rn, ^{238}U and ^{226}Ra.

Estimates of annual effective doses from natural radionuclides have only been reported from a few countries. Different radionuclides have been included in the dose estimates and comparison of the results was therefore difficult. However, the results indicated that large variation exists in dose estimates.

Effective doses for the users of drilled wells have been estimated in Finland, Sweden, Denmark and Ukraine (Ulbak and Klinder 1984, Zelensky *et al*. 1993, Isam Salih *et al*. 2002). In all these countries, ^{222}Rn caused the largest part of the dose. Generally, effective doses were similar to each other and varied between 0.2 and 0.5 mSv. In Switzerland, the main dose from drinking water was caused by ^{228}Ra (Deflorin *et al*. 2004). The median and maximum annual doses caused by ^{238}U, ^{226}Ra and ^{228}Ra were approximately 0.003 and 0.03 mSv, respectively.

In Spain, Galan Lopez *et al*. (2004) estimated that the annual effective dose from ingested ^{222}Rn varied between 0.0004 and 3.3 mSv. This is similar to the results obtained in Finland. Fernandez *et al*. (1992) estimated that the highest dose from long-lived radionuclides for users of drilled well water comes from radium and for users of dug wells from uranium. This differs from the results obtained in Finland, where the highest dose was caused by ^{210}Po and ^{210}Pb.

In Scotland, Al-Doorie *et al*. (1993) estimated that the dose from ingested ^{222}Rn from well water was 0.03 mSv, and at maximum 0.05 mSv. In Austria, Schönhofer (1992) estimated that a significant dose from ^{222}Rn from drinking water was caused by the inhalation of ^{222}Rn released from water to indoor air and that the additional effective dose from inhalation of ^{222}Rn was 0.4–0.7 mSv. The dose from ingestion of ^{222}Rn was an order of magnitude lower than the dose from inhalation of ^{222}Rn released from water. Ulbak and Klinder (1984) recorded the same kind of observations as Schönhofer (1992) and estimated the annual effective dose due to inhalation of ^{222}Rn released from water to indoor air to be 0.5 mSv.

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