

Occurrence of Natural Radioactivity in Public Water Supplies in Germany ^{238}U , ^{234}U , ^{235}U , ^{228}Ra , ^{226}Ra , ^{222}Rn , ^{210}Pb , ^{210}Po and Gross- α -Activity Concentration

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INTRODUCTION

The German Federal Office for Radiation Protection (BfS) performed a representative survey on the radiological quality of drinking water in Germany. The aim of the study was to ascertain the variation of natural radionuclide concentrations and to estimate effective doses via ingestion for members of the public. The study includes the determination of the natural radionuclides ^{238}U , ^{234}U , ^{235}U , ^{228}Ra , ^{226}Ra , ^{222}Rn , ^{210}Pb , ^{210}Po and of gross- α -activity concentration in drinking water from 564 public water supplies. This represents 3 % of all water supplies providing drinking water for 37 Mio. out of 82 Mio. inhabitants in Germany.

Investigations concerning natural radioactivity in drinking water in Germany were carried out already in the past and it is known that, unlike artificial radionuclides, the radionuclide concentrations originating from natural sources in some regions of eastern and southern Germany, i.e. the Erzgebirge, Vogtland, Fichtelgebirge, Schwarzwald, Franken (Bavaria) may cause an appreciable contribution to the radiation exposure (e.g. Gans *et al.*, 1987; Gans, 1992; Bünger, Rühle, 1997).

Recently, detailed knowledge on the radiological quality of drinking water is of particular interest for the implementation of the radioactivity parameters given in the European Drinking Water Directive 98/83/EC (EU, 1998) and the Radon Recommendation 2001/928/Euratom (EU, 2001) into national regulations. The results of this study will be the basis of the monitoring strategy concerning total indicative dose to be set by the German Drinking Water Ordinance (Ministry of Health, 2001). The monitoring strategy has to take into account the radionuclides, analytical methods, detection limits and monitoring frequencies for areas with increased natural radioactivity.

MATERIALS AND METHODS

The water supplies were selected with regard to size, location and service area, population density, including regions with normal and those with enhanced natural radioactivity caused by geology and the type of water processing. The sampling was carried out in 564 water supplies between September 2003 and August 2007, supported by the local health and environment authorities and the water suppliers. Usually, 20 litre samples of raw water as well as treated water (clear water, drinking water) and unconditioned drinking water, which are fed into the distribution system, were collected and carried to the radioanalytical laboratory of the BfS in Berlin, a total of 1052 samples. The raw water samples were analysed for comparison and to obtain values concerning the reduction of the levels of natural radionuclides through the treatment of the water. Water samples for the determination of

^{222}Rn were taken separately in radon tight beverage flasks and mailed immediately to the laboratory.

Table 1: Methods used for drinking water analysis

radionuclide	analytical method	volume	detection
gross- α	phosphate precipitation, liquid scintillation	0.3 l	ca. 20
Uran	kinetic phosphorescence analysis	0.3 l	0.7
^{238}U , ^{234}U , ^{228}Ra	extraction chromatography, alpha spectrometry	0.3 l	5 – 10
^{226}Ra	co-precipitation with barium sulphate and transfer into carbonate, extraction chromatography, low-co-precipitation with barium sulphate, emanation,	1.0 l	ca. 1
^{210}Pb , ^{210}Po	total- α -counting (radon and daughters, spontaneous deposition (Bi and Po) on Ni-disks, scintillation chamber)	4.0 l	ca. 2
^{222}Rn	low-level-beta-counting (^{210}Bi), alpha-counting liquid scintillation counting	0.015 l	ca. 1

The water samples were analysed for gross α -activity and uranium isotopes (chemical concentration by kinetic phosphorescence analysis; if the concentration exceeded $0.81 \mu\text{g/l} = 10 \text{ mBq/l } ^{238}\text{U}$, additionally the uranium isotopes ^{238}U , ^{234}U , ^{235}U were determined by the alpha spectrometric method). Radium isotopes (^{228}Ra , ^{226}Ra), ^{222}Rn and the radon long-lived decay products ^{210}Pb , ^{210}Po were determined by using German standard methods (BMU, 2006), Table 1.

RESULTS AND CONCLUSIONS

Radionuclide concentrations

A summary of the radionuclide concentration data of 582 drinking water from 564 water supplies is presented in Table 2 with their activity concentration ranges, median (50 %-quantile) and 95 %-quantile values. The median values for ^{226}Ra , ^{228}Ra and ^{238}U of 5.6 mBq/l, 4.6 mBq/l and 3.2 mBq/l, respectively, are lower than former investigation results (e.g. Gans *et al.*, 1987; Gans, 1992; Bunger, Ruhle, 1997). In other studies concerning ^{222}Rn , with

Table 2: Range of activity concentrations, median and 95 %-quantile values of 582 drinking water in Germany

radionuclide	concentration range	median	95 % quantile
^{226}Ra (mBq/l)	< 0.8 – 350	5.6	41
^{228}Ra (mBq/l)	< 0.7 – 120	4.6	26
^{238}U (mBq/l)	< 0.7 – 320	3.2	100

up to 1500 samples, medians had been found (Rühle *et al.*, 1995) which are about 20 % lower than the values of this study. The medians of ^{210}Pb and ^{210}Po of 2.3 mBq/l and 1.4 mBq/l, respectively, are much lower than those of the radium isotopes.

A classified overview on the regional distribution of the occurrence of natural radionuclides in drinking water is given in Figure 1. The figure shows the parameter gross- α -activity as a mean for each German county. Enhanced gross- α -activity concentrations in central Germany regions (predominant bed rock grit/sedimentary rocks) are mainly caused by the uranium isotopes.

Results in terms of dose

In order to permit an evaluation of the results, ingestion doses caused by the natural radionuclides ^{238}U , ^{234}U , ^{235}U , ^{228}Ra , ^{226}Ra , ^{222}Rn , ^{210}Pb and ^{210}Po as well as inhalation doses through ^{222}Rn were estimated for infants (age < 1 a), being the population group with the highest exposure, and for adults (age > 17 a). The base of estimation was an assumed drinking water consumption of 170 l/a and 350 l/a for infants and adults, respectively, and ingestion/inhalation dose coefficients according to the German Radiation Protection Ordinance (BMU, 2001) and the German Commission on Radiological Protection (SSK, 2004). If the values of the ^{234}U activity concentrations were absent (determination only in case of a ^{238}U activity concentration higher than 10 mBq/l) the estimated median of the $^{234}\text{U}/^{238}\text{U}$ -activity ratio of 1.65 was taken into account.

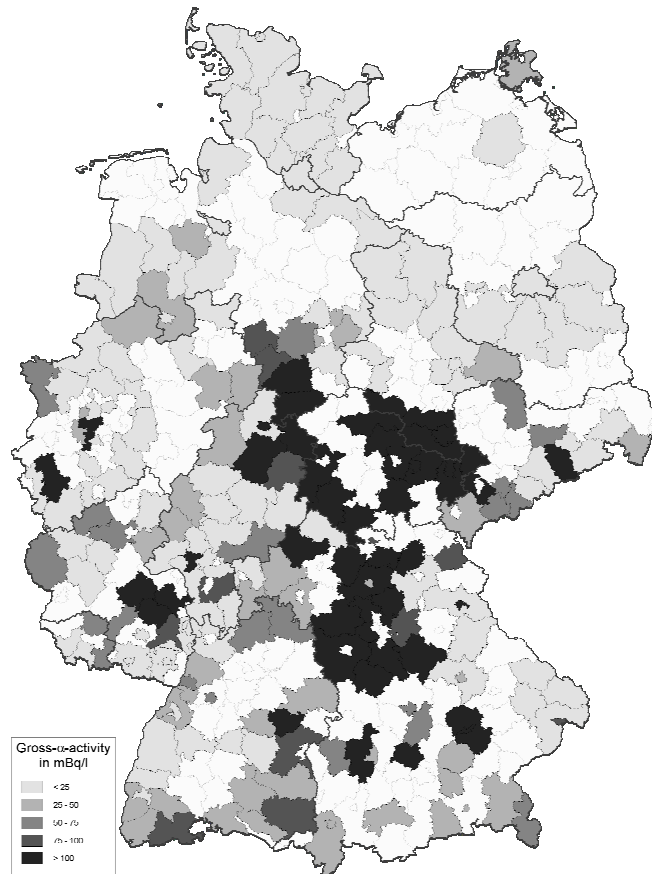


Figure 1: Regional distribution of the gross- α -activity concentrations of drinking water in Germany

Figure 2 shows the mean nuclide specific dose shares of the total ingestion doses of infants and adults within the dose range close to the medians. The specific part of the dose due to the uranium isotopes is negligible low for both age groups. For infants the predominant dose share is caused by ^{228}Ra , followed by the radionuclides ^{210}Po and ^{226}Ra . For adults the ^{222}Rn activities proved to be important with more than 50 % dose-responsibility. Figure 3 demonstrates the influence of the inhalation dose caused by ^{222}Rn released during the use of water in households. These inhalation dose shares are predominant for infants and adults.

If the radionuclides as a whole but not the inhalation shares are taken into account, the committed dose for infants is found to be in the range of < 9 $\mu\text{Sv/a}$ and 970 $\mu\text{Sv/a}$ with a

median of 50 $\mu\text{Sv/a}$ and a 95 %-quantile of 370 $\mu\text{Sv/a}$ whereas a range from <1.5 $\mu\text{Sv/a}$ up to 1140 $\mu\text{Sv/a}$ with a median of 8.6 $\mu\text{Sv/a}$ and a 95 %-percentile of 94 $\mu\text{Sv/a}$ is estimated for the age group of adults.

Calculating ingestion doses on the basis of the parameters given in the European Drinking Water Directive 98/83/EC (EU, 1998), the Total Indicative Dose (TID) for adults, based on an assumed drinking water consumption of 730 l/a and excluding radon and radon decay products, ranged from lower than 0.2 $\mu\text{Sv/a}$ up to 108 $\mu\text{Sv/a}$ with a median of 4.6 $\mu\text{Sv/a}$ and a 95 %-quantile of 27 $\mu\text{Sv/a}$.

The results of this project are meant to be a basis and an initiation for extension and clarification of the existing German regulations for natural radioactivity in drinking water.

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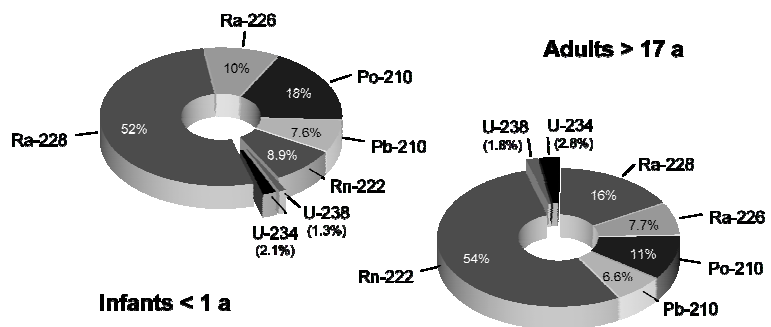


Figure 2: Nuclide specific ingestion dose shares of the total ingestion dose within the scope close to the medians

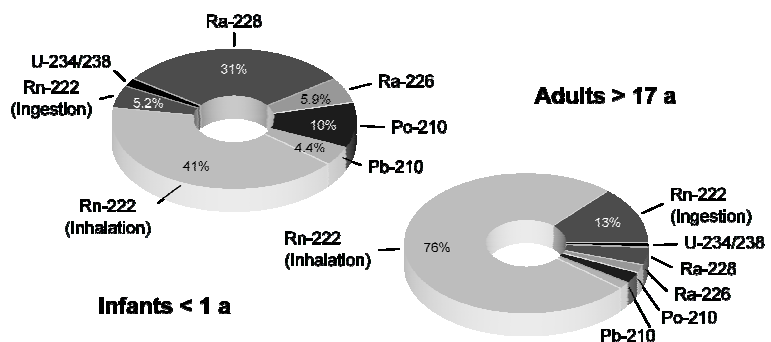


Figure 3: Nuclide specific dose shares (including inhalation dose) of the total internal radiation exposure within the scope close to the medians

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