J Radioanal Nucl Chem (2009) 282:467–471
DOI 10.1007/s10967-009-0178-4

Determination of ^{3}H, ^{226}Ra, ^{222}Rn and ^{238}U in Austrian ground- and drinking water

Claudia Landstetter · Christian Katzlberger

Received: 22 June 2009 / Published online: 16 July 2009

Abstract Screening measurements for ^{3}H, ^{226}Ra, ^{222}Rn and ^{238}U in ground water were performed within a ground- and drinking water project in Austria. The aim of this project is to get an overview of the distribution of natural radionuclide activity concentration levels in ground water bodies. In some cases this water is used for drinking water abstraction. In this paper methods and results of the screening measurements are presented. Regions with high activity concentrations were identified and in these regions further investigation for ^{228}Ra, ^{210}Pb and ^{210}Po will be conducted.

Keywords Ground water · Drinking water · ICP-MS · Liquid scintillation counting · Uranium · Radon · Radium · Tritium · Environmental · Austria

Introduction

Council Directive 98/83/EC [1] on the quality of water intended for human consumption was implemented into Austrian law by the drinking water ordinance. It recommends a total indicative dose (TID) as an indicator parametric value of 0.1 mSv/year. For the calculation of the TID in compliance with the Austrian Standard for the determination and evaluation of the total dose due to radionuclides in drinking water ^{226}Ra and ^{228}Ra are taken into account [2]. Other radionuclides with a minor contribution to the effective dose in Austria like ^{238}U and ^{234}U or other natural radionuclides with low solubility in water like ^{232}Th and ^{234}Th are generally disregarded unless considerable concentrations can be assumed. Artificial radionuclides and other natural radionuclides (with the exception of Tritium, ^{40}K, ^{222}Rn and its progenies) are taken into account if dose-relevant concentrations can be suspected. But reference values for ^{222}Rn, ^{210}Pb and ^{210}Po are proposed in the EU-Recommendation 2001/928/Euratom [3]. Monitoring should be arranged if significant concentrations are suspected. Above a concentration of 100 Bq/L of ^{222}Rn, member states should set a reference level for radon to be used for consideration whether remedial action is needed to protect human health.

In 2008 a project was started in Austria to get an overview of the distribution of natural radionuclide activity concentration levels in ground water bodies. Samples from public drinking water supplies have been measured previously [4] but the main focus in this project was put on groundwater samples before drinking water treatment. The location and boundary of these ground water bodies are defined by hydrological and geological differentiated areas.

The aim of this project is to identify regions with high activity concentrations and to identify regions with activity concentrations below the lower limit of detection. Due to the fact that water from ground water bodies is used for drinking water abstraction these data can be used for risk assessment within drinking water analyses. It gives the possibility to detect natural radionuclides with dose relevant concentrations and to concentrate on regions with high activity concentrations. On the basis of this project a more risk based monitoring program can be established within regions are monitored area-wide every year or otherwise just measured in a 5–10 year period.

C. Landstetter (✉) · C. Katzlberger
Austrian Agency for Health and Food Safety, CC Radiation Protection and Radiochemistry, Spargelfeldstr. 191, 1226 Vienna, Austria
e-mail: claudia.landstetter@ages.at

C. Katzlberger
e-mail: christian.katzlberger@ages.at

A total of 2500 sampling and measuring points for the monitoring of physical and chemical parameters of ground water bodies exist all over Austria. For this project former scientific studies for ^{222}Rn and ^{226}Ra of the federal institute for food analysis and research of Vienna were used to select 315 measuring points [5]. The number of chosen measuring points within one specified ground water body was based upon the expected activity concentrations from the above mentioned studies. First the sampling and measuring points used for drinking water abstraction within a ground water body were selected. If low activity concentrations were expected not all of the sampling points from one specified ground water body were used. Just a smaller but defined number was proposed. Otherwise all measuring points within a ground water body were chosen. In addition to this some measuring points of ground water not used for drinking water abstraction were chosen on the basis of the above mentioned data. These samples were analysed for ^{3}H, ^{226}Ra, ^{222}Rn and ^{238}U.

Experimental

^{222}Rn and ^{226}Ra analysis

Two 1 L gastight glass bottles were used for the sample collection. One bottle was acidified with 8 mL concentrated HNO_3 s.p. for stabilization shortly after the sample was filled into the bottle.

For ^{222}Rn determination the second bottle was filled carefully to avoid air bubbles in the sample and prevent the outgassing of ^{222}Rn. It is necessary to use bottles of appropriate size. They were filled completely to avoid ^{222}Rn diffusion into the free space between the sample and the cup of the bottle. After sample collection these bottles were capped immediately.

For ^{222}Rn determination 10 mL of water sample were mixed with 10 mL of Perkin Elmer high efficiency mineral oil scintillator^{TM} agitated and measured in equilibrium with ^{218}Po and ^{214}Po by liquid scintillation counting (LSC) [6]. Measurements were performed with Quantulus 1220^{TM} with pulse shape analysis. Pulse shape analysis allows simultaneous recording of alpha and beta spectra and counting of very small alpha activity in the presence of high beta activity. The Lower Limit of Detection (LLD) for ^{222}Rn with this method was 0.25 Bq/L.

Determination of ^{226}Ra was conducted by mixing 10 mL of the acidified water sample with 10 mL of Perkin Elmer high efficiency mineral oil scintillator^{TM} agitated and the ^{222}Rn was measured in equilibrium with ^{218}Po and ^{214}Po by LSC. The sample is measured after the original ^{222}Rn decayed below the LLD of ^{226}Ra of 0.046 Bq/L and the ^{226}Ra was in equilibrium with ^{222}Rn. If ^{222}Rn activity concentration in the sample was below 6 Bq/L the sample was stored for 4 weeks before measurement. If ^{222}Rn activity concentration in the sample was above 6 Bq/L the 10 mL sample was agitated extensively 100 times and after that ventilated to remove about 90% of the original ^{222}Rn before adding 10 mL of Perkin Elmer high efficiency mineral oil scintillator^{TM}. If the ^{222}Rn concentration in the sample was above 50 Bq/L the 10 mL sample was twice agitated extensively 100 times and ventilated to remove most of the ^{222}Rn (Table 1).

Table 1 Measured ^{222}Rn activity concentration of three different water samples before and after agitation

	^{222}Rn [Bq/L]		
	Sample 1	Sample 2	Sample 3
Original activity concentration in water	192.8	43.86	6.10
After first agitation and ventilation	20.53	1.75	0.550
After second agitation and ventilation	3.23	0.201	0.259

Methods used for these determinations are in scope of the accreditation and meet the requirements of the ÖVE/ÖNORM EN ISO/IEC 17025. For quality assurance repeat determinations from each sample are conducted and the method is approved by periodical participation in proficiency tests. At the beginning of each measuring cycle a ^{226}Ra standard solution and a background sample were measured for the determination of the efficiency and the blank value in the region of interest. Therefore 100 µL of a ^{226}Ra standard solution from PTB were added to 9.9 mL of water and extracted with the scintillation cocktail. This Standard solution was stored until full equilibrium between ^{226}Ra and ^{222}Rn was reached before it was used for the measurements. After 6 months the background and standard samples were renewed.

Tritium analysis

Tritium was determined by mixing 10 mL of not acidified water with 10 mL of Zinsser Analytic Quicksafe 400^{TM} and measured with a Quantulus 1220^{TM}. The LLD for Tritium with this method was 1.3 Bq/L. For the ^{3}H standard solution 10 mL of a NIST standard reference material 4361C was used and mixed with 10 mL of Zinsser Analytic Quicksafe 400^{TM}.

Determination of Uranium using ICP-MS

For the determination of Uranium at the mass number 238 a Perkin Elmer Elan DRC II was used. The method was

based on the direct introduction of the acidified samples (0.12 M HNO_3), without any chemical pre-treatment, into an inductively coupled plasma mass spectrometer (ICP-MS). Rhodium was used as internal standard. The method provides a limit of detection of 0.05 µg/L and a good repeatability tested at three different concentrations. Recovery percentage values found for the determination of uranium in synthetic water samples varied less than 2% (98–102%). Results obtained for the certified content of a reference material (TM-DWS from NRWI, Canada) are within the same range, thus indicating the accuracy of the ICP-MS procedure without the need of using isotope dilution.

Results and discussion

Screening measurements

The arithmetic mean ^{238}U concentration in Austrian ground water samples is 2.1 µg/L and the median value is 0.66 µg/L. Values vary between the LLD of 0.05 µg/L and 160 µg/L.

The provisional guideline value (reference value) for Uranium in drinking water according to the World Health Organisation is 15 µg/L [7]. In Fig. 1, samples with an ^{238}U concentration higher than 3 µg/L are plotted. Six samples are above the provisional guideline value of 15 µg/L uranium.

The sample with the highest ^{238}U concentration is in the federal state Lower Austria and was collected from ground water which is not used for drinking water abstraction. The second sample with a concentration exceeding 100 µg/L from Tyrol is taken from a sampling station where the water is used for drinking water abstraction. Most of the samples with higher ^{238}U activity concentration are located in the federal states Lower Austria (LA), Burgenland (B) and Tyrol (T).

The values of the ^{222}Rn activity concentration in Austrian ground water samples are within a range from below the LLD of 0.25 Bq/L and 910 Bq/L. The arithmetic mean of the activity concentration is 39 Bq/L and the median value is 16 Bq/L.

Samples with a ^{222}Rn activity concentration exceeding 100 Bq/L are plotted in Fig. 2. Most samples plotted in this figure are located in Upper Austria (UA) and in Lower Austria (LA). The sample with the highest ^{222}Rn activity concentration is in Tyrol (T).

The ^{226}Ra activity concentration values in Austrian ground water samples are within a range of the LLD and 0.19 Bq/L. Just 3% of the samples have a ^{226}Ra activity concentration above the LLD of 0.046 Bq/L. These samples are plotted in Fig. 3. A total of 60% of them are located in Lower Austria (LA).

The values of the ^{3}H activity concentration in Austrian ground water samples are within a range of the LLD of 1.3 Bq/L and 2.8 Bq/L which corresponds to the natural activity concentration and are far below the 100 Bq/L limit.

Future prospects

Based on these results 100 samples were chosen for further ^{228}Ra, ^{210}Pb and ^{210}Po measurements [8]. The criteria therefore were a ^{222}Rn activity concentration exceeding 100 Bq/L (Fig. 2) and further a ^{226}Ra activity concentration above the LLD (Fig. 3) and samples with ^{238}U concentrations exceeding 3 µg/L. Moreover, attention was paid to an equal distribution to the federal states.

Figure 4 shows a map of Austria and the regions where further determination of ^{228}Ra, ^{210}Pb and ^{210}Po will be

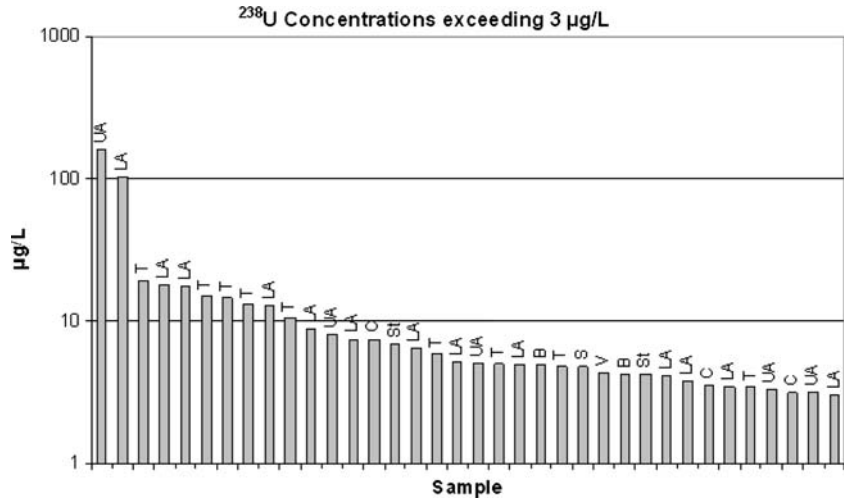

Fig. 1 ^{238}U concentrations exceeding 3 µg/L in samples from the federal states Lower Austria (LA), Tyrol (T), Burgenland (B), Styria (St), Upper Austria (UA), Carinthia (C), Salzburg (S) and Vorarlberg (V)

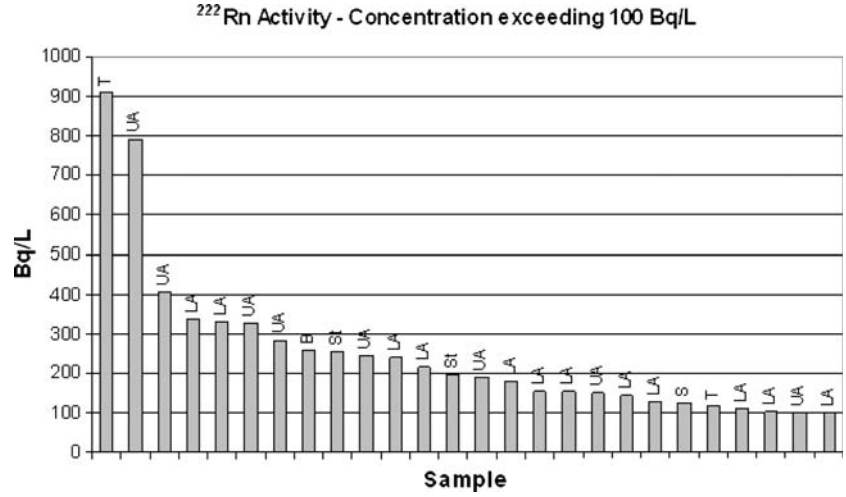

Fig. 2 ^{222}Rn activity concentrations exceeding 100 Bq/L in samples from the federal states Lower Austria (LA), Tyrol (T), Burgenland (B), Styria (St), Upper Austria (UA), Carinthia (C), Salzburg (S) and Vorarlberg (V)

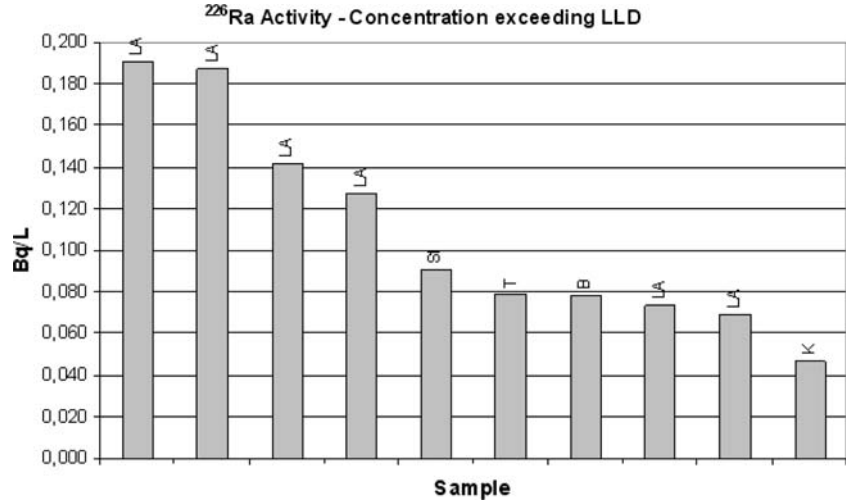

Fig. 3 ^{226}Ra activity concentrations exceeding the LLD in samples from the federal states Lower Austria (LA), Tyrol (T), Burgenland (B), Styria (St), Upper Austria (UA), Carinthia (C), Salzburg (S) and Vorarlberg (V)

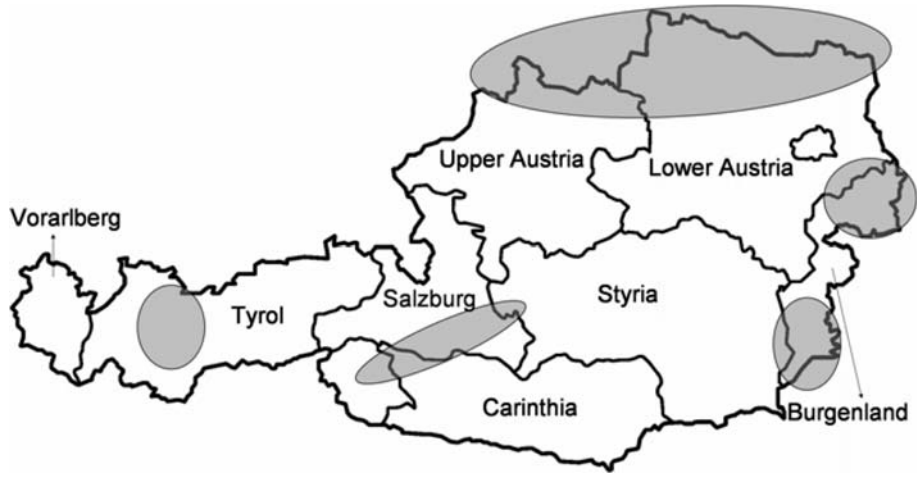

Fig. 4 Samples from these regions have ^{222}Rn activity concentrations above 100 Bq/L or ^{238}U concentrations above 3 μg/L or ^{226}Ra activity concentrations above the LLD and will be analysed for ^{228}Ra, ^{210}Pb and ^{210}Po

conducted. In the year 2009 area-wide investigations will be conducted in these regions. Overall in 150 water samples ^{3}H, ^{226}Ra, ^{222}Rn ^{238}U, ^{228}Ra, ^{210}Pb and ^{210}Po will be determined.

Conclusion

Methods used for these determinations require little sample preparation and are suitable for screening measurements

with high sample throughput. Regions in Austria could be determined, where area-wide sampling and further measurements for ^{3}H, ^{226}Ra, ^{222}Rn ^{238}U, ^{228}Ra, ^{210}Pb and ^{210}Po of ground water samples will be conducted. Fundamental data was obtained to establish a more risk based monitoring program for further drinking water analyses.

Acknowledgements This project is funded by the Federal Ministry of Health and the Federal Ministry of Agriculture, Forestry, Environment and Water Management. We like to thank the Federal Environmental Agency and the Federal State Governments of the eight federal states for their assistance with sample selection and sampling.

References

1. Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption. Draft of 20 April 2005 to Annex II (2005)
2. OENORM (2005) ON-S5251: determination and evaluation of the total dose due to radionuclides in drinking water, Austrian Standards Institute, Vienna (in German)
3. Commission Recommendation 2001/928/Euratom (2001) Commission recommendation on the protection of public against exposure to radon in drinking water supplies
4. Wallner G, Steininger G (2007) J Radioanal Nuclear Chem 274(3):511–516
5. Ditto M, Fimml W, Karg V, Korner M, Weisz J (1999) Report: Radon-222 in groundwater: survey throughout Austria. Federal Institute for Food Analysis and Research of Vienna (in German)
6. Schoenhofer F (1992) Radiat Prot Dosimetry 45(¼):123
7. WHO (2004) World Health Organization, guidelines for drinking-water quality, 3rd edn. WHO, Geneva, Switzerland
8. Landstetter C, Katzlberger C (2005) Advances in liquid scintillation spectrometry. International conference, Katowice, Poland: LSC 2005. p 181