National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Radon-222 in ground water and finished drinking water in the Dutch provinces Overijssel and Limburg

Measuring campaign 2015

RIVM Letter report 2016-0048

P.J.M. Kwakman | J.F.M. Versteegh

National Institute for Public Health
and the Environment
Ministry of Health, Welfare and Sport

Radon-222 in ground water and finished drinking water in the Dutch provinces Overijssel and Limburg

Measuring campaign 2015

RIVM Letter report 2016-0048
P.J.M. Kwakman | J.F.M. Versteegh

Colophon

P.J.M. Kwakman, (author), RIVM
J.F.M. Versteegh, (author), RIVM

Contact:
Pieter Kwakman
Centrum Veiligheid
pieter.kwakman@rivm.nl

This investigation was performed by order and for the account of Min I&M, inspectie IL&T, within the framework of project M/300001/16/DW.

This is a publication of:
National Institute for Public Health and the Environment
P.O. Box 1vironmentBilthoven
The Netherlands
www.rivm.nl/en

Publiekssamenvatting

Radon-222 in grondwater en drinkwater in de Nederlandse provincies Overijssel en Limburg

De metingen van radon-222 in een aantal grondwater- en drinkwatermonsters uit Overijssel en Limburg laten duidelijk zien dat de grenswaarde van 100 Becquerel per liter (Bq.l^{-1}), de maat voor radioactiviteit per liter watermonster, niet wordt overschreden. In grondwater variëren de meetwaarden van 1,6 – 16,7 Bq.l^{-1} en in het daaruit geproduceerd drinkwater van 0,2 – 9,5 Bq.l^{-1}.

Een vergelijking met de resultaten uit de vorige meetcampagne in 1995 laat zien dat het radon-222 gehalte in Nederland zich op een laag en constant niveau bevindt.

Volgens de Europese Euratom Richtlijn (2013/51/EC) is het niet nodig om de bepaling van radon-222 in het nationale drinkwatermonitoringprogramma op te nemen als kan worden aangetoond dat de waarden ver onder de norm zitten. De huidige routinematige bepaling van totaal-alfa, totaal-bèta en tritium is voldoende voor de meeste radioactiviteitsparameters en geeft een betrouwbare schatting van de dosis (indicatieve dosis).

Kernwoorden: ruwwater, reinwater, radon-222, meetcampagne 2015

Synopsis

Radon-222 in groundwater and finished drinking water in the Dutch provinces Overijssel and Limburg
Measuring campaign 2015

The determination of radon-222 in a number of groundwater and finished drinking water samples from the Dutch provinces Overijssel and Limburg has clearly shown that the parametric value of 100 Becquerel per liter ($Bq.l^{-1}$), the measure of radioactivity per liter water sample, is not exceeded. In groundwater the radon-222 activity concentration ranges from 1.6 – 16.7 $Bq.l^{-1}$, and in finished drinking water from 0.2 – 9.5 $Bq.l^{-1}$.

A comparison of the data with an earlier campaign in 1995 shows that radon-222 activity concentrations in water samples in the Netherlands are at a constant and low level.

Following Euratom Directive (2013/51/EC) it is not necessary to imply radon-222 in the Dutch drinking water monitoring programme if it is clearly demonstrated that all data are well below accepted levels. The routine determination of gross-alpha, gross-beta and tritium covers most radioactivity parameters and gives an accurate estimation of the dose (indicative dose).

Keywords: raw waters, purified waters, radon-222, campaign 2015

Contents

1 Introduction

The Euratom Directive 2013/51/EC [1] contains requirements for the determination of radiological parameters in drinking water. See Annex 1 for the detailed parametric values.
These parametric values are 100 $Bq.l^{-1}$ for tritium, 100 $Bq.l^{-1}$ for radon and 0.1 $mSv.a^{-1}$ for the Indicative Dose (ID). The Indicative Dose can not be measured but is calculated from all individual nuclides present in the sample. This is a complicated and time-consuming task. Therefore, for the sake of analytical simplicity, the parameters gross-alpha and gross-beta activity are used for compliance to the Indicative Dose. The gross-alpha activity should be lower than 0.1 $Bq.l^{-1}$ and the gross-beta activity lower than 1.0 $Bq.l^{-1}$.

In the Netherlands, the drinking water suppliers have a long history in measuring gross-alpha, gross-beta and tritium. The reported activity concentrations are usually lower than the parametric values above. In the Netherlands, the radon-222 content in water samples is generally very low. The measurement of radon-222 in water samples is therefore not routinely carried out by drinking water laboratories. As the last measurement campaign on radon-222 dates from 1995 [2] the new Directive urged an update of radon-222 measurements in drinking water samples.

The aim of this Dutch measurement campaign is to investigate if the activity concentration of radon-222 are below the parameter value of 100 $Bq.l^{-1}$. The sampling locations which gave somewhat higher radon-222 levels in 1995 were chosen for this campaign in 2015.

2 Methods

2.1 Sampling

Radon-222 is a noble gas resulting from the decay of radium-226, a member of the uranium-238 decay chain. As uranium-238 is present everywhere in the earth's crust its decay product radon-222 builds up and often diffuses into ground water.
Radon gas does not dissolve very well in water and it will escape from a water sample simply by stirring or shaking. Transportation of water samples in bottles in the trunk of a car would inevitably lead to the loss of radon. It was therefore chosen to carry out sampling at the water entry point in such a way that radon can not escape from the sample.
The procedure was as follows :

- Flush the system first with a fast water flow for at least one minute.
- Fill a Teflon beaker (about 100 mL) gently with water using a very slow flow. Avoid stirring or shaking of the sample. The water flow should be along the side of the beaker.
- Using a 10 mL syringe, take a 10 mL sample and transfer this gently into a 20 mL vial ***under*** the organic layer. This organic layer remains on top of the water layer, thus preventing the radon gas to escape.
- All vials contain 10 ml non-water miscible liquid scintillation cocktail, such as Ultima Gold F.
- Sampling is carried out in triplicate in order to demonstrate the repeatability of the sampling procedure.
- Radon gas will diffuse into the organic layer during transport back to the laboratory.

This method is an adapted version of ISO 13164-4:2015 [3].

2.2 Determination of radon-222 in water

All vials are counted with a liquid scintillation counter, a Packard 3100 TR, using alfa-beta discrimination. The radon extraction + counting efficiency is determined by weighing a Ra-226 standard into a 20 mL vial. After about 4 weeks the radiochemical equilibrium between Ra-226 and Rn-222 is established. The counting efficiency is then calculated using the weighed activity of Ra-226 and the number of alpha counts in the alpha window. See for analytical details ISO 13164-4 [3].

Radon-222 has a half-life of 3.8 days and decays to the short-lived alpha emitters Po-218 (3.05 min) and Po-214 (0.16 ms). Thus an alpha decay of Radon-222 results in a total of three consecutive alpha emissions.
Using liquid scintillation counting an alpha can be detected with almost 100 % efficiency. So theoretically, for the three alphas in the decay of radon-222 a counting efficiency of 300 % can be obtained. In practice the counting efficiency is somewhat lower due to the extraction into the organic layer and the alpha-beta discrimination of the counter.

Due to the selectivity of the counting technique a very low background is obtained. In combination with the very high counting efficiency this results in a detection limit of 0.5 $Bq.l^{-1}$ with a 60 min counting time.

3 Results and discussion

3.1 Results for the radon-222 activity concentrations

In Tabel 1 and 2 the results are given for the radon-222 analyses in water samples from the eastern and south-eastern parts of the Netherlands. Both tables demonstrate the presence of low activity concentrations of radon-222. For clarity the data from 1995 are also given in these tables.

Table 1 Radon-222 activity concentration in ground water (used for the production of drinking water) and finished drinking water from eastern part of the Netherlands (province Overijssel); sampling date 13 November, 2015.

Location East-NL Overijssel	Watersample Ground w Finished w	2015 Rn-222 (Bq/L)	±	2015 uncertainty (Bq/L)	1995 Rn-222 (Bq/L)	±	1995 uncertainty (Bq/L)
Lochem	Ground w	13.4	±	0.8	12.8	±	0.3
Lochem	Finished w	9.5	±	0.6	8.6	±	0.2
Hengelo Klooster	Ground w	10.6	±	0.6	8.0	±	0.3
Hengelo Klooster	Finished w	5.1	±	0.3	3.7	±	0.1
Losser	Ground w	6.2	±	0.4	not analyzed		
Losser	Finished w	0.24	±	0.04	not analyzed		
Weerselo	Ground w	5.2	±	0.3	not analyzed		
Weerselo	Finished w	0.40	±	0.05	not analyzed		
Manderveen	Ground w	8.7	±	0.5	8.4	±	0.2
Manderveen	Finished w	4.3	±	0.3	0.2	±	0.1
Wierden	Ground w	6.2	±	0.4	7.5	±	0.3
Wierden	Finished w	2.63	±	0.19	0.8	±	0.1
Nijverdal	Ground w	n.a.			10.0	±	0.2
Nijverdal	Finished w	6.5	±	0.4	5.6	±	0.2
Archemerberg	Ground w	6.9	±	0.4	6.3	±	0.2
Archemerberg	Finished w	2.33	±	0.18	3.2	±	0.1

Table 2 Radon-222 activity concentration in ground water (used for the production of drinking water) and finished drinking water from South-East Netherlands (province Limburg); sampling date 21 December, 2015. Airated ground water is a half product which will be softened before its distribution as drinking water.

Location South-east NL Limburg	Watersample Ground w Finished w	2015 Rn-222 (Bq/L)	±	2015 Uncertainty (Bq/L)	1995 Rn-222 (Bq/L)	±	1995 Uncertainty (Bq/L)
Sittard	Ground w	4.4	±	0.3	6.0	±	0.2
Sittard	Finished w	0.21	±	0.04	1.2	±	0.1
Schinveld	Ground w	7.6	±	0.5	9.5	±	0.2
Schinveld	Finished w	2.43	±	0.17	3.5	±	0.2
Roodborn	Ground w	16.7	±	1.0	19.8	±	0.3
Roodborn	Aer. Ground w	12.6	±	0.7	7.6	±	0.2
Waterval	Ground w	1.58	±	0.12	2.1	±	0.1
Waterval	Aer. Ground w	2.13	±	0.15	1.6	±	0.1

Aer. = aerated

3.2 Discussion

Compliance to the parametric value of 100 Bq.l^{-1}
As can be easily seen from all values in tables 1 and 2 all radon-222 activity concentrations are far below the parametric value of 100 Bq.l^{-1}.

Measurements of groundwater against finished drinking water
In all samples it is clear that ground waters used for drinking water production contain higher amounts of radon-222 than finished drinking waters. This can easily be explained by the application of treatment processes, such as flocculation and aeration, where large parts of the radon-222 gas will escape from the water.

Triplicate results – repeatability.
Considering the difficulty of sampling a gas in water the triplicate results were a strong indication for the repeatability. For all triplicates the relative standard deviation was calculated against the average of the triplicate measurement; this relative standard deviation of ~9 % (k=1) equals the repeatability. Next to the usual counting and experimental errors this repeatability was part of the total uncertainty budget.

Determination of radium-226 by ingrowth of radon-222
In a groundwater sample radon-222 may be present as is shown in this measurement campaign. The origin of radon-222 is radium-226 in deep rock formations: the radon-gas escapes from the rocks and slowly dissolves into groundwater aquifers. Usually, these groundwaters contain high radon-222 contents and low radium-226 contents. It is, therefore, necessary to determine which part of this radon-222 originates from deep rock formations and which part comes from radium-226 present in the water sample.

The easiest way to do this is to measure the liquid scintillation vial again after 4 weeks. In these 4 weeks, the excess of radon decays to less than 1 % of its original activity. And radium-226 in the water phase will reach radiochemical equilibrium with its daughter nuclide radon-222. Basically all radon-222 present after 4 weeks is equivalent to radium-226 in the sample.

Results for radium-226
In all samples re-counting the samples after 4 weeks only detection limits (< 0.5 Bq.l^{-1}) for radium-226 were obtained. This means that all observed radon-222 activities originated from 'unsupported' radon-222 without any radium-226 in the water phase.

3.3 Comparison of 2015 results with 1995 results

In Annex 2 all radon-222 results of the former campaign in 1995 are given. For the sampling locations in the 2015 campaign the radon-222 data are given next to data from 1995 in table 1 and 2. Both the activity concentrations and the differences between groundwaters and finished drinking waters are fully confirmed by the findings of the 2015 campaign.
The conclusion is justified that the activity concentration of radon-222 in (drinking) water samples in the Netherlands is at a very constant and low level.

4 Conclusions

The determination of radon-222 in a number of ground water and finished water samples from the Dutch provinces Overijssel and Limburg has clearly shown that the parametric value of 100 $Bq.l^{-1}$ is not exceeded. In a number of ground water samples and finished drinking water radon-222 activity concentrations range from 1.6 – 16.7 $Bq.l^{-1}$, and in finished waters from 0.2 – 9.5 $Bq.l^{-1}$.

Comparing the activity concentrations in 1995 and 2015, is justified to state that radon-222 in water samples in the Netherlands is at a constant and low level.

Following Euratom Directive 2013/51/EC it is not necessary to imply radon-222 in the Dutch drinking water monitoring programme. The routine determination of gross-alpha, gross-beta and tritium is sufficient in the Netherlands to cover most radioactivity parameters and give an accurate estimation of the Indicative Dose.

5 Annex 1 Requirements in Drinking Water Directive 2013/51/EC.

ANNEX I

PARAMETRIC VALUES FOR RADON, TRITIUM AND ID OF WATER INTENDED FOR HUMAN CONSUMPTION

Parameter	Parametric value	Unit	Notes
Radon	100	Bq/l	(Note 1)
Tritium	100	Bq/l	(Note 2)
ID	0,10	mSv	

Note 1:

(a) Member States may set a level for radon which is judged inappropriate to be exceeded and below which optimisation of protection should be continued, without compromising water supply on a national or regional scale. The level set by a Member State may be higher than 100 Bq/l but lower than 1 000 Bq/l. In order to simplify national legislation, Member States may choose to adjust the parametric value to this level.

(b) Remedial action is deemed to be justified on radiological protection grounds, without further consideration, where radon concentrations exceed 1 000 Bq/l.

Note 2: Elevated levels of tritium may indicate the presence of other artificial radionuclides. If the tritium concentration exceeds its parametric value, an analysis of the presence of other artificial radionuclides shall be required.

ID = Indicative Dose

6 Annex 2 Radon-222 in water samples in Bq.l^{-1}. Measurement results from 1995 [2].

pompstationnr. (RIVM)	bedrijf	pompstationnaam	ruw	stdev.	rein	stdev.
Limburg						
1022	WML	Schinveld/Schuttersveld	9,5	0,2	3,5	0,1
144	WML	Waterval	2,1	0,1	1,6	0,1
104	WML	Barrier	5,2	0,1	1,4	0,1
267	WML	Sittard-Hoogveld	6,0	0,2	1,2	0,1
139	WML	Roodborn	19,8	0,3	7,6	0,2
153	Maastr.	Maastricht-Caberg	4,6	0,1	3,7	0,1
Centrale slenk						
48	NRE	Eindhoven (Aalsterweg)	3,9	0,2	1,2	0,1
114	WOB	Helmond-Bakelsedijk[1)]	8,7 4,5	0,3 0,2	1,5	0,1
203	WOB	Haaren	3,3	0,2	1,0	0,1
206	WOB	Veghel	4,2	0,2	0,2	0,1
205	WOB	Macharen	6,0	0,2	1,0	0,1
145	WML	Ospel	3,2	0,2	0,7	0,1
273	Tilburg	Gilzerbaan	3,5	0,2	0,6	0,1
Utrecht						
4	WZHO	Lexmond	3,5	0,1	1,5	0,1
50	WZHO	Kamerik	3,6	0,1	0,1	0,1
170	WMN	Linschoten[1)]	5,2 7,3 4,2	0,2 0,3 0,2	2,3	0,1
176	WMN	Tull en 't Waal	4,2	0,2	2,4	0,1
292	WMN	IJsselstein	4,4	0,2	0,4	0,1
173	WMN	De Meern	4,8	0,2	1,4	0,1
Oost-Nederland						
8	COGAS	Almelo-Wierden[1)]	8,4 8,5 5,7	0,3 0,3 0,2	0,8	0,1
223	WOG	Lochem[1)]	12,9 13,4 12,0	0,3 0,3 0,4	8,6	0,2
220	WOG	Hengelo 't Klooster[1)]	7,9 8,0	0,3 0,3	3,7	0,1
243	WMO	Manderveen	8,4	0,2	0,2	0,1
245	WMO	Nijverdal	10,0	0,2	5,6	0,2
232	WMO	Archemerberg	6,3	0,2	3,2	0,1

7 Acknowledgements

Bert van Dijk is kindly acknowledged for carrying out all water sampling on location in Overijssel (east) and Limburg (south-east parts of the Netherlands).

8 References

[1] Council Directive 2013/51/EC Euratom; 22 October 2013.

[2] Resultaten van het meetprogramma drinkwater, 1994 voor parameters uit het Waterleidingbesluit en enkele aanvullende parameters. JFM Versteegh, FW van Gaalen, BA Baumann, E Smit en L Vaas, RIVM rapport 731011009, december 1995.

[3] ISO 13164: 2015. Water quality - Radon-222 - Part 4: Test method using two-phase liquid scintillation counting

RIVM
Committed to *health and sustainability*