

Management of radioactivity in drinking water including radon

Francesco Bochicchio

Italian National Institute of Health

Head of the National Center for Radiation Protection and Computational Physics

Head of the WHO Collaborative Center for Radiation and Health



**National Center for
Radiation Protection
and Computational Physics**

**WHO Collaborating Centre for
Radiation and Health**



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How does radon get into drinking-water?

(Q.1.6.1)

- Radon-222 (^{222}Rn , in the following referred to simply as “radon”), is produced by radioactive decay of radium-226 (^{226}Ra). The latter is in turn produced by uranium-238 (^{238}U), the most common isotope of uranium found in nature, especially in the ground.
- It dissolves in water and its **solubility increases with decreasing temperature**.
- Water can be radon-enriched in two different ways:
 - Emanation of radon into water-filled porous, or interstitial spaces, of rocks matrixes;
 - Radioactive decay of radium-226 dissolved in water.



Which are the patterns of exposure to radon in drinking water?

- The two main sources of drinking water are:
 - **Groundwater**;
 - **Surface water**.
- Drinking water is provided to consumers through:
 - wells that pump **groundwater** from the aquifer to the house;
 - water distribution systems whose source may be either **surface water** or **groundwater**.

- Groundwaters from **springs, wells and boreholes**, where there is a relatively short time between water extraction and its use, are more likely to result in an **increased exposure to radon**.
- Levels of **radon in surface waters** are **typically low** due to the natural degassing into the outdoor air.

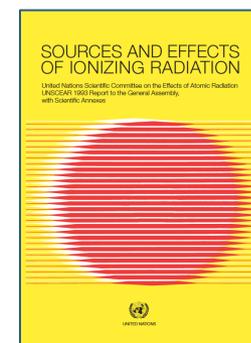


Radon concentrations measured in different water sources

The concentrations of radon in water may range over several orders of magnitude, generally being:

- highest in **well water**
- intermediate in **groundwater**
- lowest in **surface water**.

Type of supply	Typical percentage of usage (%)	Radon concentration (Bq L ⁻¹)	
		Typical	Up to
Well water	10%	100	80 000
Groundwater	30%	10	4 000
Surface water	60%	1	10



UNSCEAR 1993

A review of Rn concentration measured in different water sources

Water type	²²² Rn activity concentration (Bq L ⁻¹)	Country, region	Geology	References
Drinking water	<3	Serbia/Novi Sad		Todorovic et al., 2012
	0.3–24	Cyprus and Greece (Attica-Crete)	Not detailed	Nikolopoulos and Louizi, 2008
	1.46–644	Austria	granite bedrock	Wallner and Steininger, 2007
	<1.3–1800	Germany		Beyermann et al., 2010
	1.9–112.77	Portugal	tap	Lopes et al., 2005
Surface water	0.19–71.1	UK	tap	Henshaw et al., 1993
	<1	Slovenia		ISO 13164-3, 2013
	5.4			Kobal et al., 1990
Groundwater	1–1000	Spain, La Garrotxa region		ISO 13164-3, 2013
	0.2–26		volcanic	Moreno et al., 2014
	3043	Poland, the Sudety Mountains	Volcanic (e.g. crystalline rocks)	Przylibski et al., 2014
	3800	Finland	Soil (no detail)	Salonen, 1988
	1220	Germany, east Bavaria	granite, gneiss	Trautmannsheimer et al., 2002
	17–3856	Portugal, Nisa	Granites, sediments	Pereira et al., 2015
	5.8–36.6	UK, Northern Ireland	Sherwood Sandstone Aquifer	Gibbons and Kalin, 1997
Spring and non-bottled mineral waters	1.4–105	Spain, South Catalonia,	volcanic (granite) and sedimentary rocks (e.g. limestone, sandstone)	Fonollosa et al., 2016
	2.11–120	Hungary, Balaton Highland, South Transdanubia and The South Great Plain	Sedimentary rocks	Somlai et al., 2007
	Mineral water 0.91–1463	Serbia	various	Todorovic et al., 2012
	1595	Bulgaria, Momin prohod		Pressyanov et al., 2007
	1.4–43.7	Lithuania	crystalline and sediment rocks	Ladygiene et al., 1999
Well water	1.5–181	Italy, Padua, Euganean Thermal District	volcanic rocks	Cantaluppi et al., 2014
	1029	Spain, Galicia	granitic and slate rocks	Llerena et al., 2013
	10–300	Norway		ISO 13164-3, 2013
	32000		Not detailed	Strand et al., 1998
	4–63560	Sweden, Stockholm County	various, crystalline bedrock	Skeppström and Olofsson, 2006
	47–1600	Belgium, Visé		Bourgoignie et al., 1982
	77000	Finland,	mainly granitic bedrock	Salonen, 1988

Source:
Jobbagy et al. (2017)

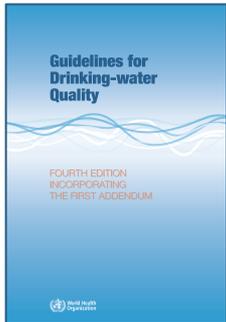


Do national standards for radon in drinking-water need to be established?

(Q.1.6.2)

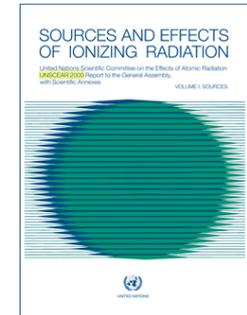
Not necessarily.

The **Guidelines for Drinking-water Quality** (Chapter 9) **does not provide guidance level for radon** because it is considered more appropriate to measure radon concentrations in indoor air rather than in drinking-water.



Relying on the review of available data performed by UNSCEAR in 2000, on average, **90% of the dose from radon in drinking-water comes from inhalation of radon released from water rather than ingestion of water.**

1000 Bq L⁻¹ in water ➔ 100 Bq m⁻³ in indoor air



If a country wants to set a national standard for radon in drinking-water, **screening levels for radon in drinking-water should be based on the national reference level for radon in indoor air.**

Some countries have set **national standards for radon in drinking-water** (required by Euratom directive on drinking-water)



Comparison between WHO and Euratom directive approaches

	WHO approach	Euratom directive approach
Radon in drinking water	<p>The WHO GDWQ does not provide guidance levels for radon.</p> <p>Controlling the inhalation pathway rather than the ingestion pathway is considered the most effective way to control doses from radon in drinking-water.</p>	<p>According to the Directive 2013/51/Euratom, Member States have to set a <i>parametric value</i> (a screening level) of 100 Bq L⁻¹, above which evaluate the risk and evaluate if remedial actions are needed.</p> <p>In addition, Member States may set a level for radon which is judged inappropriate to be exceeded and below which optimization of protection should be continued (<i>reference level</i>). The level set by a Member State may be higher than 100 Bq L⁻¹ (<i>parametric value</i>) but lower than 1000 Bq L⁻¹.</p>
Radon in indoor air	<p>The reference level for radon concentration in indoor air recommended by WHO is 100 Bq m⁻³ (WHO Handbook on Indoor Radon, 2009). If this level cannot be reached under prevailing country-specific conditions, the level should not exceed 300 Bq m⁻³.</p>	<p>According to the Directive 2013/59/Euratom, Member States shall establish a national reference level for indoor radon concentration, which shall not be higher than 300 Bq m⁻³.</p>



Measuring radon concentration in drinking water?

From GDWQ, par. 9.7.3 “Guidance on radon in drinking water supplies”:

“As the dose from radon present in drinking-water is normally received from inhalation rather than ingestion, it is more appropriate to measure the radon concentration in air than in drinking-water.

...

Nevertheless, in circumstances where high radon concentrations might be expected in drinking-water, it is prudent to measure for radon and, if high concentrations are identified, consider whether measures to reduce the concentrations present are justified.

The concentration of radon in groundwater supplies can vary considerably. Consequently, in situations where high radon concentrations have been identified or are suspected, the frequency of gross alpha and gross beta measurements may need to be increased so that the presence of radon progeny (in particular polonium-210), which can be major contributors to dose, can be assessed and monitored on an ongoing basis.”



At what points in the water supply chain should measurements of radon in drinking-water be made?

(Q.1.6.3)

Radon concentration can strongly decrease within the distribution system from the source to the consumption point due to:

- Radioactive decay taking place during transport and possible storage;
- Spontaneous degassing in pipes and stations;
- Degassing associated to treatment process leading to agitation of water.

According to GDWQ, samples should ideally be taken **at the point of consumption** in order to obtain the best estimate of the radon actually contained in the water being ingested.

Measurements performed **at the source** could overestimate the committed dose from ingestion of drinking water. **Monitoring at the source** should be considered as an **indicator of potential radon content** and could be useful for evaluating the need of remedial actions.



What methods can be used for **sampling** and measuring radon in drinking-water supplies (Q.1.6.4)

Reference publication for sampling procedure is ISO 13164 (2013).

Important recommendations regarding sampling procedure are:

- To adjust the water flow to avoid turbulence and air bubbles at the outlet of the tap and in the sampling container;
- The sampling container should be completely filled without air bubbles below the cap after closing the container;
- In case of flowing water, it can be necessary to purge the supply system before taking the sample;
- To consider the likely water layering in case of stagnant water;
- To direct the container towards the flux direction in case of flowing water.



What about container to be used for transport?

- **ISO 5667-3** recommends to use **glass bottles**;
- **ISO 13164** recommends also **other bottle materials**:
 - Container made from non-porous to radon (e.g. aluminium) material.
 - Non-hydrophobic materials in order to minimize the presence of gas bubbles on the walls of the container.
 - Container resistant to pressure and temperature shock.
- Jobbagy et al. (2019) showed that (in addition to the bottle material) **the bottle cap** also plays an important role to preserve sample stability and bottle integrity.
 - They observed that bottles with rigid caps tend to break due to temperature changes => the cap should be radon tight and made of flexible material.



A brief overview of not specialized containers that can be used

- **PET** (either petrol- or bio-based types) and **PLA** are much more suitable than **HDPE/LDPE**:
 - **PET** bottles daily lose 0.1–1.4%, **PLA** ~1%
 - **HDPE** bottles daily lose 15–22%, **LDPE** ~27%.
- Intermediate daily radon loss (2–5%) was observed for polypropylene containers.
- The most influencing parameter, after material, is the **surface/volume ratio**, because both diffusion and absorption are highly influenced by bottle surface.
- **Radon loss reduces with increasing thickness** when considering bottles having similar surface/volume ratios.



What methods can be used for sampling and measuring radon in drinking-water supplies (Q.1.6.4)

The main international standard dedicated to water radon measurements is:

- ISO 13164:2013, parts 1 to 4

The measurement methods fall into two categories:

- direct measurement of the water sample without any transfer of phase:
 - Gamma-spectrometry
- indirect measurement involving the transfer of ^{222}Rn from the aqueous phase to another phase, before performing the measurement:
 - Gamma-spectrometry (on radon adsorbed on charcoal)
 - Emanometry (involving transfer of Rn from the aqueous phase to gaseous phase)
 - Liquid Scintillation Counting



Gamma spectrometry

The activity concentration of the ^{222}Rn is determined by measuring the characteristic gamma lines of ^{214}Bi or ^{214}Pb obtained by a HPGe (quantitative) or NaI (qualitative or semi-quantitative) detectors.

Advantages	Disadvantages
No sample treatment required	HPGe detectors are highly expensive (about 100 k€)
Data analysis (i.e., peak-recognition and activity evaluation) is fully automatized.	High (if compared with those of other techniques) turnaround time, i.e., 4-13 h (few measurements/week)
No specific training is required for the operators: the analysis procedure is the same as any other gamma-spectrometry measurement.	If ^{226}Ra is also present, the measurement should be repeated after secular equilibrium between radium and radon is established (~20-30 d) => significant time delay.
Measurement uncertainty generally could be very low, up to 5%.	The sample density and homogeneity, both influencing the detection efficiency, depend on the water temperature, suspended materials and air bubbles
	The measurement results are influenced by indoor radon in the laboratory air, due to the well acknowledged day-by-day variations of radon concentration.



Emanometry

Radon is transferred from the liquid to the gaseous phase in a closed circuit by controlled sample degassing.

Alpha particles from radon and its daughters are detected by different types of detectors (mainly Lucas cells, ion chambers, semiconductors).

Advantages	Disadvantages
Different detectors coupled with the degassing circuit can be used, with low-to-moderate costs (1-20 k€).	Degassing circuit required
Indoor radon in laboratory air does not significantly influence measurement procedure and results.	Sub-sampling is required: a certain quantity of water should be transferred from transport container to the degassing circuit.
Measurement uncertainty can be very low (up to 5%) if the method is properly managed.	If ^{226}Ra is present, degassing circuit and detector itself could be contaminated.
Possibility to perform in-situ measurements.	The technique is sensitive to thoron when measurements are performed in-situ.
Very low turnaround time, compared with those of other techniques (<1 h) => many measurements per day.	Several uncertainty sources have to be managed, controlled and the corresponding impact on results evaluated.



Liquid scintillation counting

The principle of the radon measurement by LSC is based on the extraction of ^{222}Rn from the water to the immiscible scintillation cocktail.

Advantages	Disadvantages
Once prepared the vial, the procedure is fully automatized and the result is directly returned by the software.	Instruments for liquid scintillation counting are expensive (at least 70 k€)
Several vials can be analysed at the same time => many measurements per day .	The turnaround time is quite high (approximately the same as gamma-spectrometry), 3-8 hours => no rapid results .
The lowest detection limit (0.05 Bq L ⁻¹), obtained by choosing alpha/beta discrimination and plastic-made vial. The vial to be measured can be prepared on-site: such procedure avoids the need of sub-sampling .	Calibration is cocktail specific, so each scintillation cocktail should be studied separately. Discrimination settings are crucial and can lead to significant errors when choosing alpha/beta discrimination operating mode.
Indoor radon in laboratory air does not significantly influence measurement procedure and results.	In-situ measurements can not be performed.



How can radon in drinking-water be managed when radon concentrations in the source water are high?

(Q.1.6.5)

In addition to general management approaches (e.g. providing an alternative drinking-water supply), some easy methods exist to reduce radon concentration in drinking-water:

- High-performance **aeration** which can achieve up to 99.9% removal efficiency. The main drawback could be increased indoor air radon levels.
- **Adsorption** via granular activated carbon, with or without ion exchange. However, this method is less efficient and requires large amounts of granular activated carbon.
- **Mixing** the water with high radon content with one whose radon concentration has been measured to be very low.
- **Storing** the water in tanks or basin before the distribution to consumers.
- **Combination** of previous methods.

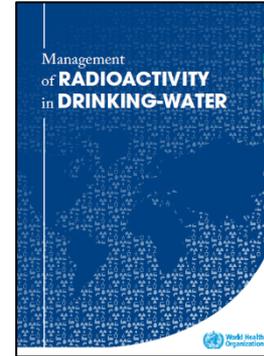
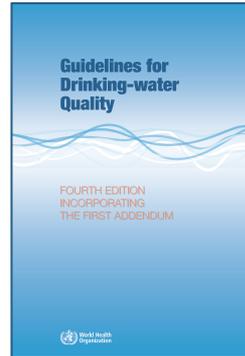
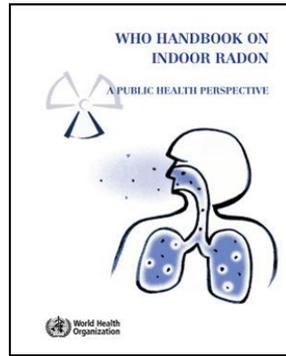
Some remedial measures to manage radon levels in indoor air will also reduce the concentration coming from the use of drinking-water



Managing very high radon concentration: a case study in northern Italy (Losana et al. 2006)

- Radon concentrations up to 25,000 Bq L⁻¹ have been measured in a spring water in Lurisia cave.
- As a consequence of such measurements, thermal and drinking use of such water have been forbidden by the Ministry for Health in 2000.
- In 2001, an aeration system was applied to the spring source and the resulting water was mixed with that from a near source ($C_{Rn} \approx 1,000$ Bq L⁻¹).
- Since September 2005, the mixing water was changed: the new one has been showed to have $C_{Rn} < 30$ Bq L⁻¹.
- As a result of these measures (aeration + mixing), radon concentration values <20 Bq L⁻¹ have finally been obtained in Lurisia water.





Thank you for your attention

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