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A brief overview on radon measurements in drinking water

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ABSTRACT

The aim of this paper is to present information about currently used standard and routine methods for radon analysis in drinking waters. An overview is given about the current situation and the performance of different measurement methods based on literature data. The following parameters are compared and discussed: initial sample volume and sample preparation, detection systems, minimum detectable activity, counting efficiency, interferences, measurement uncertainty, sample capacity and overall turnaround time. Moreover, the parametric levels for radon in drinking water from the different legislations and directives/guidelines on radon are presented.

1. Introduction

On the basis of Articles 35–36 of the EURATOM Treaty (EURATOM, 2010) monitoring and reporting of environmental radioactivity is an obligation for the EU Member States. To check the quality and comparability of these measurement results the Institute for Reference Materials and Measurements (JRC-IRMM/JRC-Geel[1]) is requested by the EC DG ENER to organize interlaboratory comparisons (ILCs). In an upcoming ILC, the focus is on one of the most widespread monitoring methods, radon activity measurement in drinking water samples. In this context this paper gives a brief overview on the current situation and the performance of different radon measurement methods. This paper deals solely with radon (^{222}Rn) and not other more short-lived radon isotopes, like thoron (^{220}Rn) and actinon (^{219}Rn).

Surface and underground waters contain radionuclides as natural components in various concentrations depending on their origin. Radon is released into waters as a result of natural processes like decay of its parent nuclide ^{226}Ra and predominantly dissolution from the surrounding geological environment (rocks, soils) as discussed by (Moreno et al., 2014; Fonollosa et al., 2016). Radon in water may also origin from dissolution of airborne radon into water and other higher radon bearing water in-flows in the catchment area.

Radon solubility in water is relatively low, 0.01 mol kg^{-1} bar^{-1} at 293 K (Lerman, 1979). Solubility is commonly expressed by the partitioning coefficient (L) of ^{222}Rn between pure solvent and air (Clever, 1979; Schubert et al., 2007). The partitioning coefficient of ^{222}Rn in water is approximately 0.23–0.25 at 293 K. Radon affinity towards organic solvents and oils is higher, for example L ≅ 6 for ethanol. This behaviour can be used during the different analytical approaches where phase transfer is needed. The higher the carbon chain length the higher the radon solubility is. Despite the relatively low solubility of radon in water, its activity concentration in waters can be some orders of magnitude higher than that of other natural radionuclides (Fonollosa et al., 2016).

The relatively low cost and simplicity of many radon measurement techniques has made them common to apply in many laboratories. Numerous reliable radon measurement devices are available on the market with reasonably low detection limits, affordable price and simple operation. Furthermore, sample preparation for the water radon analysis is usually simple, rapid and does not need extensive chemical manipulations. This gives an advantage of much shorter turnaround time than other radioanalytical techniques involving radiochemical manipulations. In certain cases result can be delivered even within an hour

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[1] As of 1 July 2016 JRC-IRMM will change its name to JRC-Geel.

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(Todorovic et al., 2012; Lucchetti et al., 2016).

Water intended for drinking purposes has to be analysed for its radon content according to the new EURATOM Drinking Water Directive referred as E-DWD (EURATOM, 2013). Article 8 of the E-DWD stipulates the transposition by 28 November 2015.

Furthermore, this overview aims to contribute 1) finding reliable methods to determine reference values for radon in water samples and; 2) evaluating suitable radon measurements to support work in MS laboratories.

2. Radon in water: regulations and examples

To protect the health of citizens from radon in drinking water, different radon levels are introduced. For waters intended for human consumption the E-DWD establishes parametric values, WHO (WHO, 2008) uses guidance level while in the United States maximum contaminant levels are introduced.

Guidance levels and parametric values should not be regarded as limit or reference values as explained in the E-DWD and WHO publications. The country concerned establishes guidance levels and parametric values on the basis whether that value poses a risk to human health from a radiation protection point of view or not (i.e. if further remediation action is needed or not).

The guidance and parametric levels were reviewed for all MS and are summarized in Table 1. For waters intended for human consumption in the European Union countries guidance and parametric levels are in accordance with the E-DWD, i.e. between 100 and 1000 Bq L^{-1}. It has to be noted that mineral waters are still exempted from this directive despite their regular/preferred consumption. In the United States, two different levels are given for the maximum contaminant level (US-EPA, 1999). Exceeding the highest alternative maximum contaminant level might result in elevated health risks from indoor radon (i.e. radon escaping from water and into the indoor air) so mitigation is needed in the places concerned. The higher alternative maximum contaminant level could contribute approximately one tenth (14.8 Bq m^{-3}) of the total indoor radon which is equivalent to the average outdoors radon concentration in the US (Bartram, 2015). The World Health Organization set the guidance level to 100 Bq L^{-1} in the third edition of the WHO drinking water guidelines (WHO, 2008). However, in the current publication of the drinking water guidelines radon guidance level is missing and no other concrete guidance level is given (WHO, 2011). A selection of international radon guidance and parametric values for drinking waters are presented in Table 1.

Examples of radon concentration in different water sources including extreme high values are presented in Table 2.

Radon activity concentrations are highly variable not only among the different water types but even within the same type. This can be mainly explained by their origin, lithology of the aquifer host rock and the different processes on their way to the consumer (e.g. degassing, dilution, decay). Where drinking water has shorter way and time between the water source and consumer the reduction in radon activity concentration is lower. In general the consumption of water with respect to radon can be considered safe, since the typical radon concentrations of waters fall below the corresponding international and European directives, recommendations. However, there are waters with some extreme values above the parametric and guidance levels so monitoring will remain important. The applied radon measurement techniques should be capable of measuring radon activities with confidence and accuracy in activity concentrations from <1 and up to several thousands of Bq L^{-1}. According to radon activity concentration studies, the majority of the studied drinking and surface waters meet the directives and regulations therefore, no elevated radon risk is present due to water consumption. Those findings are mainly due to the fact that these waters either undergo water treatment (de-gassing) where radon is removed from the waters together with other dissolved gases like methane. Or simply that radon has lower chance to accumulate and higher chance to escape as it happens in the case of surface waters.

The situation is different in case of ground waters, since they are in contact with geological formations rich in uranium and sometimes in a closed or nearly closed system where radon can accumulate and reach activity concentration of several kBq L^{-1}. Waters tend to have higher radon concentration in vulcanite areas and the vicinity of tectonic faults (Fonollosa et al., 2016; Popit and Vaupotič, 2002).

3. Standard water radon measurement methods

There are international standards dedicated to water radon measurements like ISO 13164-3:2013, ISO 13164-4:2015 and ASTM D5072-09 (2016). Besides the technical part, they describe the principles of the methods, some of the sampling issues and approaches, transportation and storage conditions. It is not the aim of this paper to present the standards in detail but to highlight the applicability and pitfalls of these proposed methods to further improve the measurement accuracy.

The major international standards include the following:

a) ISO 13164-3:2013: Water quality - Radon-222- Part 1–3.
   Part 1: General principles
   Part 2: Test method using gamma-ray spectrometry.
   Part 3: Test method using emanometry
b) ISO 13164-4:2015: Water quality Radon-222 - Part 4.
   Test method using two-phase liquid scintillation counting.
c) ASTM D5072-09 (2016) Standard Test Method for Radon in Drinking Water based on LSC.

3.1. Methods

In principle there are three different water radon measurement approaches. The first one uses gamma-ray spectrometry, the

Table 1
International radon guidance and parametric values in drinking water.

Directive/recommendation	Activity concentration (Bq L^{-1})	Reference
EURATOM DWD (E-DWD)	100–1000[a]	EURATOM, 2013
24 EU Member States[b]	100	MS National law
Ireland, Portugal, Spain	500	MS National law
Finland	1000	MS National law
WHO guidance level	100	WHO, 2008
US-EPA maximum contaminant level	~11.1	US-EPA, 1999
US-EPA alternative higher maximum contaminant level	148	

[a] >1000 Bq L^{-1} remedial action without further consideration is justified in all EU countries.
[b] Austria, Belgium, Bulgaria, Croatia, Cyprus, Czech Republic, Denmark, Estonia, France, Germany, Greece, Hungary, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Romania, Slovakia, Slovenia, Sweden, United Kingdom.

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