HIROSAKI UNIVERSITY

Radiation Environment and Medicine 2018 Vol.7, No.2 117–120

REM
Radiat Environ Med

Note

On the Importance of ^{228}Ra in Radiation Dose from Drinking Water Intake

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Received 31 January 2018; revised 4 April 2018; accepted 25 April 2018

A recent review of radioactivity in Canadian public water supply systems estimated that the average effective dose resulting from drinking water intake would be 4.2 μSv per year. That review did not consider the presence of ^{228}Ra, because data for ^{228}Ra in drinking water are limited in Canada. Using available data from the environmental monitoring program conducted by the Radiation Protection Bureau, Health Canada, in Regina, Elliot Lake and Port Hope, the average ^{228}Ra concentration was found to be 7 mBq/L. If the corresponding dose is included in the calculation of the national average annual dose from natural radionuclides in drinking water, it nearly doubles. The average effective dose resulting from drinking water intake should be revised to 7.5 μSv per year to include ^{228}Ra. This is still well below the Canadian recommended reference level of 100 μSv/year (0.1 mSv/year).

Key words: natural radionuclides, drinking water

1. Introduction

Radionuclides are naturally present in the environment. Natural sources of radiation are responsible for the large majority of radiation exposure to the public (greater than 98%, excluding medical exposure)[1]. The occurrence of natural radionuclides in drinking water is associated most commonly with groundwater. The concentrations are highly variable and are determined by the composition of the underlying bedrock as well as the physical and chemical conditions prevailing in the aquifer. Increased levels of natural radionuclides in surface waters may be linked to industrial processes, particularly uranium mining and milling operations[1]. In an effort to better characterize natural radioactivity levels, including total uranium in public water supply systems, drinking water quality data were recently reviewed for 24 metropolitan areas and cities which cover more than 64% of the total Canadian population[2]. The average effective dose resulting from natural radionuclides (^{226}Ra, ^{210}Pb and uranium isotopes) in drinking water was assessed to be 4.2 μSv per year.

It has since been recognized that the previous review did not consider naturally occurring radionuclides from the thorium decay chain, notably ^{228}Ra. This is because comparatively little data for ^{228}Ra is available, possibly because the Canadian Guidelines for Drinking Water Quality (GCDWQ) – Summary Table[3] and the GCDWQ: Guideline Technical Document – Radiological Parameters[4] do not include explicit direction to sample for ^{228}Ra, nor do they list the radionuclide in the summary table for the most commonly detected natural and artificial radionuclides in Canadian drinking water sources. Therefore, ^{228}Ra concentration is rarely

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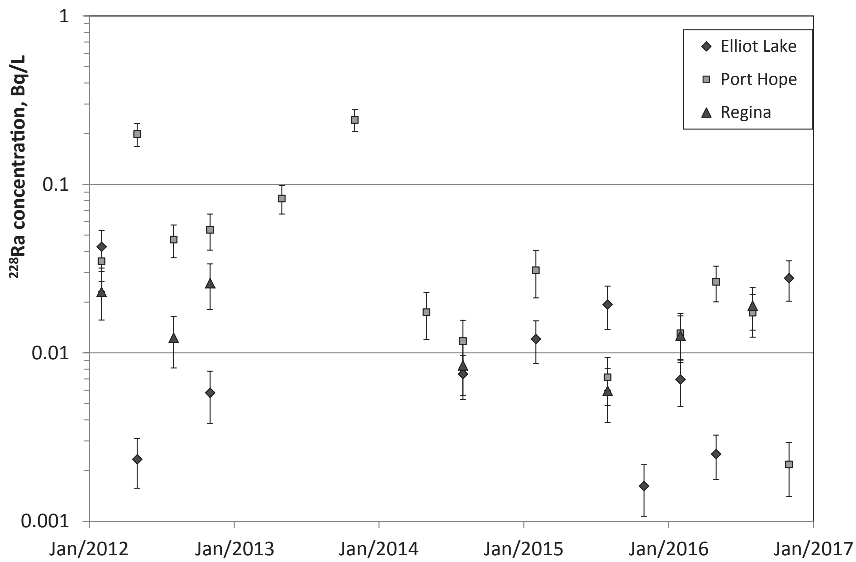

Fig. 1. Measured ^{228}Ra concentrations in drinking water of Elliot Lake, Port Hope and Regina.

monitored by operators of public drinking water systems although, when it is present, it can be a significant contributor to total dose.

It is well known that naturally occurring radionuclides of terrestrial origin are present to various degrees in all media in the environment[1)]. The main sources of natural radionuclides are from radionuclides in the Uranium-238 and Thorium-232 decay series. Canada's uranium resources are the fourth largest in the world, after those of Australia, Kazakhstan and Russia[5)]. Thorium is about as common as lead in the Earth's crust and several times more abundant in Earth's crust than all isotopes of uranium combined. In international studies of drinking water, commonly found natural radionuclides are ^{238}U, ^{234}U, ^{226}Ra and ^{210}Pb from the uranium decay series, and ^{232}Th and ^{228}Ra from the thorium decay series, as listed in the summary table of guidance levels (GL) for common natural and artificial radionuclides in the World Health Organization's guidelines for drinking water quality (the upper portion of the Table 9.2)[6)]. In order to estimate the contribution of ^{228}Ra to the total dose from natural radionuclides in drinking water, available monitoring data of ^{228}Ra in drinking water were analysed and reported here.

2. Data Sources

Managed by the Radiation Protection Bureau of Health Canada, the Canadian Radiological Monitoring Network (CRMN) is a national network that routinely collects air particulate, precipitation, external gamma dose, drinking water, atmospheric water vapor, milk, food and other biota samples for radioactivity analysis. In an effort to better characterize natural radioactivity levels in municipal drinking water supplies, routine measurements were initiated in 1975 for 17 municipalities across Canada. Over time, many measurements were discontinued, since all showed consistently low levels of radioactivity. Since 1986, only three communities (Regina, Elliot Lake and Port Hope) have been continuously monitored for natural radioactivity in drinking water. In the case of Regina, surveillance has been maintained due to high uranium concentrations in the sedimentary bedrock and groundwater was the main source of drinking water before 1980s. In the cases of Elliot Lake and Port Hope, surveillance has been maintained because of, respectively, past uranium mining operations and activities associated with historical waste management and refining operations[7)]. In recent decades, measurement data have shown that activity concentrations of naturally occurring radionuclides in the water supply systems of Regina, Elliot Lake and Port Hope are within normal variation range of background radiation in drinking water, comparable to the levels in water systems of many other Canadian communities[2, 7, 8)].

The data used in this analysis is from drinking water samples that were collected on a monthly basis from Elliot Lake, Port Hope and Regina from 2012-2016. The monthly samples were tested for Uranium-234/235/238. They were then composited and analyzed every quarter for ^{226}Ra and ^{228}Ra. Data of uranium isotopes and ^{226}Ra were reported in a previous publication[8)].Activity concentrations of ^{228}Ra in water samples from Elliot Lake, Port Hope and Regina are given in Figure 1 where data below the respective detection limits are not show. ^{228}Ra is a beta emitter. The detection limit (DL) depends on

Table 1. Annual average ^{228}Ra activity concentration (mBq/L) and variation range in drinking water samples from Regina, Elliot Lake and Port Hope

Location	2012	2013	2014	2015	2016
Regina	15 (< DL, 26)	< DL	2 (< DL, 8)	2 (< DL, 6)	8 (< DL, 19)
Elliot Lake	13 (< DL, 43)	< DL	2 (< DL, 8)	8 (< DL, 19)	9 (< DL, 28)
Port Hope	84 (35, **199**)	81 (< DL, **242**)	7 (< DL, 17)	10 (< DL, 31)	15 (2, 26)
Mean ± SD	37 ± 40	27 ± 47	4 ± 3	6 ± 4	11 ± 4
Population-weighted mean	18	4	2	2	9
%(<DL)	17%	83%	67%	50%	17%

Table 2. Annual radiation dose and percentage contribution for natural radionuclides commonly found in drinking water

Natural radionuclides	Dose coefficient Sv/Bq	Concentration	Annual Dose μSv	Contribution %
Total uranium (U)		0.23 μg/L		2.7%
Uranium-238 (U-238)	4.5×10^{-8}	2.84 mBq/L	0.093	
Uranium-235 (U-235)	4.7×10^{-8}	0.12 mBq/L	0.0041	
Uranium-234 (U-234)	4.9×10^{-8}	2.84 mBq/L	0.102	
Lead-210 (Pb-210)	6.9×10^{-7}	6.8 mBq/L	3.43	45.7%
Radium-226 (Ra-226)	2.8×10^{-7}	1.7 mBq/L	0.347	4.6%
Radium-228 (Ra-228)	6.9×10^{-7}	7 mBq/L	3.53	47.0%

sample collected, can vary widely. On average, the DL is around 0.018 ± 0.005 Bq/L.

3. Results and Discussion

In the World Health Organization's guidelines for drinking water quality [6], the guidance level (GL) for ^{228}Ra is 0.1 Bq/L, based on a reference dose level of 0.1 mSv for one year's consumption of drinking water, assuming a consumption of 2 L/day at the GL. Table 1 summarizes activity concentrations of ^{228}Ra in drinking water of Regina, Elliot Lake and Port Hope for the most recent 5 years for which data is available. One can see from Table 1 that many results were below or around detection limit, although at least two individual measurements were above the GL for ^{228}Ra, as highlighted in bold in Table 1. The annual average activity concentration was well below the GL for all locations in all years assessed.

Based on 2016 Canadian population statistics[9], the populations are 10740, 12590 and 247200 in Elliot Lake, Port Hope and Regina, respectively. Weighted by the populations, the 5-year mean ^{228}Ra concentration averaged over data from Regina, Elliot Lake and Port Hope is 7 mBq/L. When calculating the averages, zero value is assigned to all samples with non-detectable activity or reported as "<DL".

In the recent review of natural radioactivity levels in public water supply systems across Canada[2], levels of radioactivity were generally found to be low compared to national and international guideline levels. The population-weighted average levels are 1.7 mBq/L for ^{226}Ra, 6.8 mBq/L for ^{210}Pb, and 0.23 μg/L (5.8 mBq/L) for total uranium. The average effective dose resulting from drinking water intake at these levels would be 4.2 μSv per year.

In order to demonstrate the contribution of ^{228}Ra to the total ingestion dose resulting from natural radionuclides in drinking water, the average ^{228}Ra concentration of 7 mBq/L as summarized in Table 1 was added to the dose calculation from the previous review[2], as shown in Table 2, assuming drinking water intake of 2 L per day.

Radiation dose from total natural uranium is the combination of doses from ^{238}U, ^{235}U and ^{234}U. It contributes only 2.7% of the total dose from natural radionuclides in drinking water, as calculated based on national average concentrations, as shown in Table 2. Assuming 7 mBq/L of ^{228}Ra in drinking water, ^{228}Ra contributes about half of the annual radiation dose resulting from natural radionuclides in drinking water. The average effective dose resulting from drinking water intake at these levels, including ^{228}Ra, would be 7.5 μSv per year, significantly higher than the previous estimate of 4.2 μSv per year.

Data for ^{228}Ra in drinking water are very limited in Canada. The above estimate may not be representative for the whole country. Even so, the assessment conducted in this paper demonstrates that ^{228}Ra can be an important contributor to the ingestion dose from drinking water. It is important to note, however, that, the total annual effective dose due to natural radionuclides in drinking water, even with ^{228}Ra included, is still a relatively small fraction of the typical annual effective dose due to natural background radiation, which is in the range of 1 – 10 mSv with 2.4 mSv (2400 μSv) being the central estimate[1].

Radium-228 is a beta emitter. Beta emitters have wide energy spectra, from a few eV to several MeV. A gross beta measurement counts all beta emitters to a certain degree depending on the energy response range of the techniques used. A standard gross beta measurement will detect most radionuclides that emit beta particles. However, there are a few radionuclides that will not be detected by standard gross beta screening methods or where their concentrations will be underestimated because the energy of the beta particle emission is too low to be efficiently detected by the method, such as tritium (^{3}H). There are a few naturally occurring radionuclides (notably ^{228}Ra and ^{210}Po) where the 0.1 mSv per year could be exceeded, even if the gross beta screening levels are not exceeded in the situation (most likely in well-water) where these radionuclides are the main contributors to the total gross beta activity concentration. If the local geology and hydrology indicate that beta emitting radionuclides, such as ^{228}Ra, may be present, the individual radionuclides should be measured instead of gross beta screening.

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