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Measurement of radium-226 concentration and dose calculation of drinking water samples in Guilan province of Iran

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► ***Short report***

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ABSTRACT

Background: Radium-226 (^{226}Ra) is a product of the ^{238}U radionuclide decay series that significantly incorporated into the human body through water intake. It can also potentially cause a series of health problems including cancers of the digestive system. Radium-226 (^{226}Ra) is a product of the ^{238}U radionuclide decay series that significantly incorporated into the human body through water intake. It can also potentially cause a series of health problems including cancers of the digestive system. ***Materials and Methods:*** ^{226}Ra has been determined in drinking water samples collected from various locations of Guilan province of Iran. The water samples are taken from public water and urban tap water sources. For evolution of gross a and b exposure precipitation method and proportional scintillator system was used. The radon emanation method was used to measure the radium concentration in drinking water. ***Results:*** The measurements showed the gross α and β concentration ranges were between <38 mBql^{-1} to 92 mBql^{-1} and <41 mBql^{-1} to 328 mBql^{-1}, respectively. The radium concentration range was between 2 mBq l^{-1} to 38.2 mBq l^{-1}. The resulting contribution to the annual effective dose due to the digestion of ^{226}R in water was calculated to be between <0.4 µSv y^{-1} to 7.8 µSv y^{-1}, respectively. ***Conclusion:*** The average concentration of ^{226}Ra was found (7.6 mBq l^{-1}) in drinking water samples and the average annual effective dose, from the digestion of ^{226}Ra in water samples was calculated to be 1.5 µSv y^{-1}. According to UNSCEAR, the annual effective dose value by ingestion is report to be 0.12 mSv y^{-1}.

Keywords: *Radium-226, ingestion, drinking water, precipitation method.*

INTRODUCTION

The main sources of natural radioactivity are ^{238}U, ^{232}Th and their decay products and ^{40}K. Radium-226 is a product of the ^{238}U radionuclide decay series with half-life 1600 y that emits alpha and beta particles. Among radionuclides present in groundwater, the most radiotoxic and of concern to human health is radium [(1)]. The ^{226}Ra concentration in surface waters is commonly scant, but groundwater sources can contain a considerable concentration dependent upon the uranium and thorium content of the surrounding geology. Radium can enter groundwater by dissolution of aquifer materials that desorption from rock or sediment surfaces and ejection from minerals by radioactive decay.

Long term exposure to elevated levels of radium in drinking water has been associated with an increased risk of bone cancer development. Although the consumption of drinking water containing radium concentrations below the established MCL (Minimum Concentration Limit ≈1 Bq l^{-1}) does not alleviate all potential risk. Altogether the impact of public health at these concentrations is considered negligible [2].

Since the study of radioactivity concentration in drinking water has not already been done in this area and also because of the high percentage of various diseases, including cancer of the digestive system in the studied area, this study is performed to obtain an estimate of the activity concentration of ^{226}Ra in public water and urban tap water sources. Furthermore, the radiation dose from ingestion of drinking water by public in Guilan province of Iran was estimated.

MATERIALS AND METHODS

In this study, 28 drinking water samples from the waters which resource public waters (wells) and spring waters were collected. The study area was north section of Guilan province (figure 1). For sample preparation, the Environmental Protection Agency (EPA) protocol [3] was adopted, in which samples were collected in 4 Li container containing a solution with pH≈2. First of all, each sample was measured the gross a and b exposure by using a gas scintillation proportional counter (CANBERRA) to determine the ^{228}Ra level according EPA protocol. Commercial software Genie 2000 v2.1 was used for data analysis. The ^{226}Ra in the drinking water sample was concentrated and separated in solution by precipitation method. The precipitate obtained was dissolved in EDTA reagent, placed in a sealed bubbler and stored for ingrowth of ^{222}Rn. After ingrowth ^{222}Ra, the gas was purged into a scintillation Lucas cells, by noble gas. When the ^{222}Rn daughters are in equilibrium with the parent (approximately, 4h), the scintillation Lucas cell, was counted by using a Pylon AB-5 radon field-portable scintillation measurement system. Use of the Lucas cell for radon measurement is an established approach in field and laboratory settings [4-7]. The separation procedure and instrumentals' calibration was performed using a standard solution of this nuclide. The Lucas cell was evacuated and then attached to a sample cylinder with in-line valves, expansion tubing, and a pressure gage. The Lucas cell was filled with sample by opening the cylinder to the evacuated expansion tubing and then closing the cylinder valve and opening the valve to the Lucas cell. After the pressure stabilized, the process was repeated if necessary until the cell was filled with natural gas to approximately atmospheric pressure. The final pressure was recorded and the Lucas cell placed on the AB-5 photomultiplier tube to count the total alpha activity. The software SP-55 version 1.3 was used for data analysis. The counting interval was 90 minutes and was repeated as time p ermitted. Radon activities were calculated from the counting data after correcting for the efficiency of the Lucas cell, the cell volume, the recorded pressures, and the time interval between sample

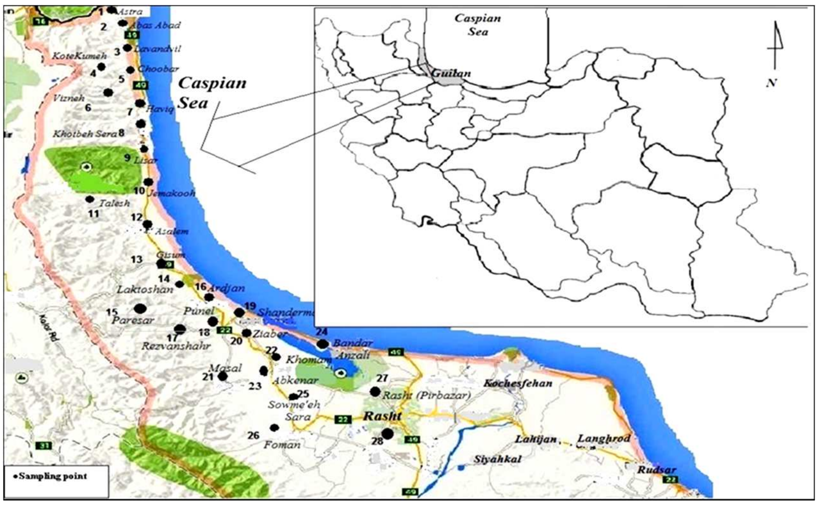

Figure 1. The study area in Guilan province of Iran.

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collections and counting.

Radium estimation in drinking water

The ^{226}Ra activity concentration was calculated in the pCi per liter using equation (1):

$$C_{Ra} = \frac{C\lambda t_3}{2.22\ EV(1-e^{-\lambda t_1})(e^{-\lambda t_2})(1-e^{-\lambda t_3})}, \qquad (1)$$

Where; C_{Ra} is the ^{226}Ra activity concentration (pCi l^{-1}), and C, E and V are the net count rate (cpm), calibration constant and sample volume (Li), respectively. The t_1, t_2, t_3 and λ parameters are the passed time between the first and second de-emanations (min), the time between the second de-emanation and counting, the counting time and the decay constant of ^{222}Rn (1.26×10^{-4} min^{-1}), respectively. The conversion factor constant from dpm/pCi is 2.22 [8].

The calibration constant (E), was calculated by using equation (2) as follow:

$$E = \frac{C}{A(1-e^{-\lambda t_1})(e^{-\lambda t_2})}, \qquad (2)$$

where; C is the net count rate (cpm), A is the activity of ^{226}Ra in the bubbler (dpm), t_1 and t_2 are ingrowth time of ^{222}Rn and the decay time of ^{222}Rn occurring between de-emanation and counting (min), respectively [3].

Radium dose calculation

The dose calculation from radium as a result of drinking water is calculated by equation (3) as follow:

$$D = D_{WI} \times D_{CF} \times Y \times C_R \qquad (3)$$

where; D is the annual effective dose (Sv y^{-1}) from the digestion of ^{226}Ra in water, C_R is the radium concentration in water (Bq l^{-1}), D_{WI} is the daily consumed of water (estimated as 2l d^{-1}), D_{CF} is the dose conversion factor for adults, (2.8×10^{-7}Sv Bq^{-1}) and Y is year (365 d) [9]. In all calculations the uncertainties are given within 1 standard deviation.

RESULTS

A total of 28 samples that were taken from the study area were included 14 tap water samples, 12 well water samples and 2 spring water samples. The results of gross a and b concentration in the drinking water samples are shown in table 1. The gross a and b range were between the low limit of detection LLD (<38 mBq l^{-1}) to 92 mBq l^{-1} and LLD (<41 mBq l^{-1}) to 328 mBq l^{-1}, respectively. Also the ^{226}Ra concentration and annual effective dose contribution due to the digestion of water samples used in North Guilan Iran are shown in table 1. The ^{226}Ra concentrations for 8 samples were below the LLD=2 mBq l^{-1} and maximum concentration was 38.2 mBql^{-1} in WS-8 (Khotbeh Sera). The annual effective dose range was calculated between to be <0.4 µSv y^{-1} to 7.8 µSv y^{-1}. The highest values related to tap water sample WS-8 (Khotbeh Sera) and spring water sample HSWS-4 (Kote Kumeh).

The correlation parameter between the radium-226 concentration and water pH are shown in figure 2. The result showed that high ^{226}Ra concentrations were in alkaline water samples.

In following the cause of high grossa, gross b and radium concentration area, with the geological map, the veins mass of metamorphic minerals and pyroxene andesitic contain were found.

DISCUSSION

The screening levels for gross α and gross β in drinking water samples were below 500 mBq l^{-1} and 1000 mBq l^{-1}, respectively, therefore, below the recommended World Health Organization (WHO) guideline level for drinking water (1.0 Bq l^{-1}) [10]. Also, according to the Environmental Protection Agency (EPA) the ^{226}Ra concentration in drinking water as safe limit is at the level of 0.74 Bql^{-1} [3]. This implies that all the water samples had values below the recommended levels.

In this study the average annual effective dose in water samples area was less than the WHO guideline level value of 0.1 mSy y^{-1}[10]. Also the ^{226}Ra concentrations measured in this study were compared with other published papers. These results were within reported ranges as shown in table 2.

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Table 1. The gross a, gross b, radium-226 concentration and annual effective dose from the digestion of ^{226}Ra in water samples used in North Guilan province, Iran.

No	Sample Code	Regain Name	Sample Resource	pH	Gross Alpha (mBq l^{-1})	Gross Beta (mBq l^{-1})	^{226}Ra Concentration (mBq l^{-1})	Annual dose due to Ingestion of ^{226}Ra (μSv y^{-1})
1	WS-1	Astara	Tap Water	7.6	>LLD*	>LLD**	>LLD***	0.4>
2	WS-2	Abas Abad	Tap Water	7.4	>LLD	4 ±54	0.03±7.4	1.5
3	WS-3	Lavandvil	Tap Water	7.1	>LLD	>LLD	0.02±2.1	0.4
4	HSWS-4	KoteKumeh	Spring Water	8.8	5±81	8±310	0.08±24.8	5.1
5	WS-5	Choobar	Well Water	7.2	>LLD	2±61	0.06±3.4	0.7
6	WS-6	Vizneh	Tap Water	6.9	>LLD	3± 42	0.02±2.7	0.6
7	WS-7	Haviq	Tap Water	7.2	>LLD	4±71	0.04±4.6	0.9
8	WS-8	Khotbeh Sera	Tap Water	7.6	6±92	12±328	0.08±38.2	7.8
9	WS-9	Lisar	Tap Water	7.2	>LLD	>LLD	>LLD	0.4>
10	WS-10	Jemakooh	Well Water	6.9	4±68	>LLD	0.07±8.2	1.7
11	WS-11	Talesh	Tap Water	7.1	>LLD	2±49	0.02±11.4	2.3
12	WS-12	Asalem	Tap Water	7.2	>LLD	>LLD	>LLD	0.4>
13	WS-13	Gisum	Well Water	7.4	>LLD	>LLD	>LLD	0.4>
14	HSWS-14	Laktoshan	Spring Water	7.7	3±78	4±75	0.09±21.5	4.4
15	WS-15	Paresar	Well Water	7.2	>LLD	>LLD	0.02±3.4	0.7
16	WS-16	Ardjan	Well Water	7.5	2±44	6±97	0.04±14.3	2.9
17	WS-17	Rezvanshahr	Tap Water	7.2	>LLD	>LLD	>LLD	0.4>
18	WS-18	Punel	Tap Water	7.5	>LLD	>LLD	>LLD	0.4>
19	WS-19	Shanderman	Well Water	7.5	>LLD	14±257	0.02±10.8	2.2
20	WS-20	Masal	Well Water	7.3	2±43	12±263	0.05±2.4	0.5
21	WS-21	Ziaber	Well Water	6.9	>LLD	4±88	0.03±5.2	1.1
22	WS-22	Bandar Anzali	Tap Water	7.2	>LLD	>LLD	0.04±5.3	1.1
23	WS-23	Abkenar	Well Water	7.6	>LLD	7±145	0.04±3.5	0.7
24	WS-24	Sowme'eh Sara	Well Water	7.5	3±51	4±57	0.03±21.3	4.4
25	WS-25	Fuman	Tap Water	7.4	>LLD	>LLD	>LLD	0.4>
26	WS-26	Khomam	Well Water	7.6	>LLD	3±48	>LLD	0.4>
27	WS-27	Rasht (Pirbazar)	Well Water	7.4	>LLD	12±281	0.05±2.8	0.6
28	WS-28	Rasht	Tap Water	7.2	>LLD	5±81	0.02±3.3	0.7
	Range			-6.9 8.8	>LLD- 92	>LLD-328	>LLD - 38.2	7.8 - 0.4>

*LLD= 38 mBq l^{-1}; **LLD=41 mBq l^{-1}; *** LLD=2 mBq l^{-1}

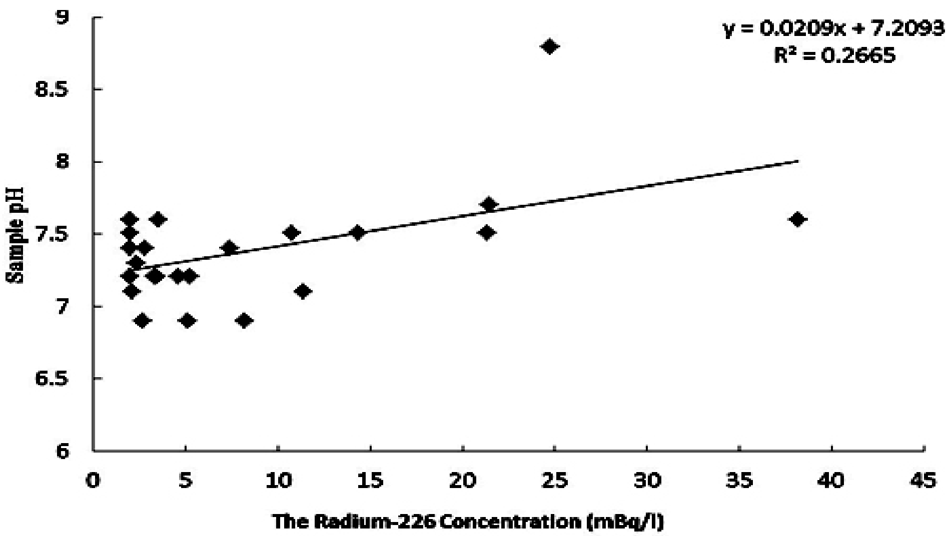

Figure 2. The correlation parameter between the radium-226 concentration and water pH.

Table 2. The radium-226 concentration in drinking water in the world.

Location	Sample resource	^{226}Ra concentration	Reference
Shandiz, Iran	Springs and rivers	MDA–2.2 Bq l^{-1}	Binesh *et al.* (11)
Extramadura, Spin	Springs, wells and spas	<0.2–142 mBq l^{-1}	Lopez *et al.* (12)
Austria	Mineral water	3.7–211 mBq l^{-1}	Wallner *et al.* (13)
Brazil	Groundwater aquifer	0.01–1.5 Bq l^{-1}	Bonotto *et al.* (14)
Milano. Italy	Tap water	0.4–7.2 mBq l^{-1}	Rusconi *et al.* (15)
Nigeria	Mineral water	2.22 – 15.5 Bq l^{-1}	Ajayi *et al.* (16)
Italy	Bottled mineral water	< 10–52.5 mBql^{-1}	Desideri *et al.* (17)
Kutahya, Turkey	Tap and spring waters	< 0.02–0.7+0.2 Bq l^{-1}	Sahin *et al.* (18)
Zahedan, Iran	Springs, wells and river	< 2-3 mBql^{-1} (well water) 5 mBql^{-1} (river) < 2 mBql^{-1} (spring water)	Hosseini (19)
Turkey	Mineral water	< 74 – 625 mBql^{-1}	Tabar *et al.* (20)
This work	Tap, spring and well waters	< 2- 38.2 m Bq l^{-1}	2014

CONCLUSION

Natural exposures, such as gross-α, gross-β and ^{226}Ra radionuclide were measured in the samples of Guilan province water supplies and springs. The results showed that the gross-α, gross-β and ^{226}Ra concentration in all water samples were lower than WHO and EPA recommendations considered safe for consumption in study area.

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Conflict of interest: *Declared none.*

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