Volume 17, No 1 | International Journal of Radiation Research, January 2019

Lifetime risk assessment of Radium-226 in drinking water samples

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▶ ***Original article***

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Revised: September 2017
Accepted: October 2017

Int. J. Radiat. Res., January 2019; 17(1): 163-169

DOI: 10.18869/acadpub.ijrr.17.1.163

ABSTRACT

Background: One of the most significant radionuclides in natural drinking waters is ^{226}Ra and its decay products. It is potential of health problems, including cancer risk. In this study, the effect of ^{226}Ra concentration in 28 drinking water samples collected from the North Guilan province was investigated. ***Materials and Methods:*** The activity concentrations of ^{226}Ra were measured by using of radon emanation method and Pylon AB-5 radon scintillation detector. The annual effective dose distribution by age groups, radiological risk and chemical toxicity risk were calculated in drinking water. ***Results:*** The activity concentration results range from a low limit of detection (LLD) 2.0±0.1 mBq l^{-1} to 38.2±2.4 mBq l^{-1}. Also, the annual effective dose distribution by age groups estimated results were from 1.8×10^{-6} Sv y^{-1} for adults to 1.5×10^{-5} Sv y^{-1} for infants. The radiological risk assessment results were 1.06×10^{-6} to 2.03×10^{-5} for morbidity risk, 7.32×10^{-7} to 1.40×10^{-5} for mortality risk. The chemical toxicity risk results obtained from 1.08×10^{-1} to 5.63×10^{-3}. ***Conclusion:*** The activity concentration level of ^{226}Ra in all drinking water samples were less than the recommended level WHO for drinking water 1000 mBq l^{-1}. Meanwhile, annual effective dose level, cancer morbidity, mortality risk and life annual daily dose due to consumption of selected drink water samples were less than the standard limit.

Keywords: *Ra-226 radionuclide, drinking waters, radiological risk, chemical toxicity, cancer.*

INTRODUCTION

Drinking water, as a necessary part of people diet may contain natural radioactivity. This natural radioactivity can be as prolonged source of human exposure. The main exposure from natural radionuclides is due to U and Th series. ^{226}Ra is a product of the ^{238}U series with half-life 1600 y that alpha and beta particles are emitted from it. Whereas, ^{226}Ra is one of the most important radionuclides in drinking water, may pose a health impact (1-4). ^{226}Ra and ^{40}Ca metabolic path in the body are same and principally positioned in the through body bone (5).

Absorption of the ^{226}Ra with the body depends on various factors such age, body weight, sex, and metabolic activity. The health effects of ^{226}Ra are divided into radiological risk posed by the radiation due to radium and the chemical risk posed by radium as a heavy metal (6). The important pathway of absorption of ^{226}Ra in the body is the digestive system. The value of absorption in the digestive system is about 15-21% of the ingested amount called ρ_i absorption coefficient value (fractional absorption of ^{226}Ra in the digestive system) which this coefficient for infants is 1.0 and for adults is 0.2 values. Whereas, the absorption coefficient for age groups of 1 to 15 y is from 0.6 to 0.3 values (7).

Continual exposure to ^{226}Ra causes of investigation this radionuclide concentration in food [8-10], drinking waters [11-21] and soil [22-24]. In some of these investigations, the annual effective dose and radiological risk were estimated from water consumption. In all of the reports, the obtained results were compared with WHO or other international and national recommended levels.

The purpose of this study is the annual effective dose estimation, radiological risk and chemical risk probability value due to ^{226}Ra concentration in drinking water samples of the study area. Hence, according to WHO guideline the annual effective dose distribution along with drinking water was calculated by age groups [25].

MATERIALS AND METHODS

Study area

The North Guilan province (37° 13' ~ 38° 24' N, 48° 36' ~ 49° 38' E) is located in the north of Iran and South of Caspian sea with a total area of 7200 km^2 and nearly 0.6% of Iran's total area. The spring and well water in the study area is the main resource as drinking water and consumed by the approximately 700,000 people living in this area figure 1. High incidence rates of gastrointestinal cancers have been reported in the Caspian region of Iran. In this case, the oesophagus and stomach is ranked first and second [26].

Sampling collection and preparation

In order to investigate the activity concentration in drinking water samples of the North Guilan province, 28 drinking water samples with groundwater resources were collected according to the population distribution. The taken samples were stored in 4 L volume polyethylene containers and diluted with HNO_3 acid to pH 2 to avoid any loss by sorption of the radioelements inside the container walls and reduce the microorganism activity.

Determination of Ra-226 concentration

According to the standard methods illustrated in some reports, the selected drinking water samples were analysed [27-30]. Each water sample was transferred to a 4-liter lab container and added barium sulfate ($BaSO_4$) for precipitation of ^{226}Ra in the sample. The obtained precipitate was dissolved in EDTA solution. For equilibrium between ^{226}Ra and ^{222}Rn, the sample placed in a sealed bubbler and stored. After growth ^{222}Rn gas in the bubbler, the ^{222}Rn gas was evacuated into scintillation Lucas cells, by noble gas. The alpha particles emitted from ^{222}Rn in scintillation Lucas cells was measured by using a Pylon AB-5 radon scintillation measurement system. Application of Lucas cells as a luminescence instrument is an established approach in field and laboratory settings [31-34]. The activity concentration of ^{226}Ra in water sample was calculated using equation (1):

$$A_{Ra} = \frac{C_{net}\lambda}{133.2\ \varepsilon V(1-e^{-\lambda t_1})} \times \frac{1}{e^{-\lambda t_2}} \times \frac{t_3}{1-e^{-\lambda t_3}} \qquad (1)$$

Where A_{Ra} is the ^{226}Ra activity concentration (pCi l^{-1}), and C_{net}, Ɛ and V are the net count rate (cps), the calibration constant and sample volume (l), respectively. The t_1, t_2, t_3 and λ parameters are the passed time between the first and second de-emanations (second), the time between the second de-emanation and counting, the counting time and the decay constant of ^{222}Rn (2.1×10^{-6} s^{-1}), respectively. The conversion factor from dps/pCi is 133.2 constant.

The calibration constant (Ɛ), was calculated according equation (2):

$$\varepsilon = \frac{C_{net}}{A(1-e^{-\lambda t_1})} \times \frac{1}{e^{-\lambda t_2}} \qquad (2)$$

In this equation, C_{net} is the net count rate (cps), A is the activity of ^{226}Ra in the bubbler (dps), t_1 and t_2 are growth time of ^{222}Rn, the decay time of ^{222}Rn occurring between emanation and counting (s), respectively [35].

Internal exposure

According to the world health organisation (WHO) the relevance between total ingestion dose E(Sv y^{-1}) and activity concentration

A(Bq l^{-1}) in drinking water is expressed by equation 3 [25]:

$$E = A \times DC \times WI \quad (3)$$

Where DC is the dose coefficient for ^{226}Ra for various age group (Sv Bq^{-1}) and WI is annual water intake (l y^{-1}).

Radiological risk assessment

The radiological risk assessment was calculated to obtain excess lifetime cancer risk (ELCR) with the mean intake of ^{226}Ra radioelement in drinking water samples. The excess lifetime cancer risk could be estimated by the equation 4 [37]:

$$ELCR = r \times I \quad (4)$$

Where r is the risk coefficient factor (Bq^{-1}) and I is per capita activity concentration intake during the lifetime (Bq).

Chemical toxicity risk

The chemical toxicity risk of ^{226}Ra and its aftermath as carcinogenic risk were also calculated. The carcinogenic risk expressed by lifetime average daily dose (LADD) of radioelements through the intake. The lifetime average daily dose (mg kg^{-1} day^{-1}) in drinking water was calculated as equation 5 [37]:

$$LADD = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \quad (5)$$

Where EPC is the exposure point concentration (mg l^{-1}), IR is the daily water intake (~2 l day^{-1}), EF is the exposure frequency (days y^{-1}), ED is the exposure duration (y), AT is the average time (days) and BW is the body weight (~70 kg).

Figure 1. The map showing the study area and sampling sites.

RESULTS

In table 1 the age- dependent water intake, dose coefficient and ingestion dose for different age groups were reported. As presented results with the mean value of ^{226}Ra concentration, the infant's age group receive the highest annual dose value of 1.5×10^{-5} Sv y^{-1} and adults age groups have lowest annual dose value of 1.8×10^{-6} Sv y^{-1}.

The activity concentration of ^{226}Ra in 28 drinking water samples collected from different locations was ranged from the lower limit of detection LLD (2.0±0.1 mBq l^{-1}) to 38.2±2.4 mBq l^{-1} with a mean of 7.6 mBq l^{-1} table 2.

The cancer risk coefficient of ^{226}Ra for morbidity and mortality cases are 1.04×10^{-8} and 7.17×10^{-9} Bq^{-1}, respectively [38,39]. So, the cancer morbidity and mortality risk due to ^{226}Ra in drinking water during the life (70 y) could be estimated and presented in Table 2. The results show that the cancer morbidity risk ranged from 1.06×10^{-6} to 2.03×10^{-5} with a mean value of 4.75×10^{-6}, while the cancer mortality risk ranged from 7.32×10^{-7} to 1.40×10^{-5} with an average of 3.28×10^{-6}.

The results of lifetime average daily dose calculation were shown in table 2. The obtained results were ranged 1.08×10^{-1} to 5.63×10^{-3} according to a maximum and minimum value of ^{226}Ra concentration, respectively. The mean value of the lifetime average daily dose was 2.52×10^{-2}.

The chemical toxicity risks of ^{226}Ra were evaluated from 5.63×10^{-3} to 1.08×10^{-1} mg/kg day with a mean value of 2.52×10^{-2} mg/kg day. The calculation results were shown in table 2.

Table 1. Age-dependent water intake, dose coefficient and ingestion dose for different age groups.

Age (y)	WI (l) [a]	DC (Sv Bq^{-1}) [b]	E (Sv y^{-1})
<1	365	4.7×10^{-6}	1.5×10^{-5}
1-2	427	9.6×10^{-7}	3.7×10^{-6}
3-7	500	6.2×10^{-7}	2.8×10^{-6}
8-12	715	8.0×10^{-7}	5.1×10^{-6}
13-17	985	1.5×10^{-6}	1.3×10^{-5}
>18	730	2.8×10^{-7}	1.8×10^{-6}

[a] ICRP 1974 [36]
[b] ICRP 1993 [7]

Table 2. The activity concentration range, the cancer morbidity risk, cancer mortality risk and lifetime average daily dose of ^{226}Ra in drinking water samples.

^{226}Ra concent ration range	^{226}Ra concen-tration value (mBq l^{-1})[a]	Cancer morbidity risk	Cancer mortality risk	LADD (mg/kg day)
Maximum	38.2±2.4	2.03×10^{-5}	1.40×10^{-5}	1.08×10^{-1}
Minimum	2.0±0.1	1.06×10^{-6}	7.32×10^{-7}	5.63×10^{-3}
Average	7.6±0.6	4.75×10^{-6}	3.28×10^{-6}	2.52×10^{-2}

[a] For 28 water samples

DISCUSSION

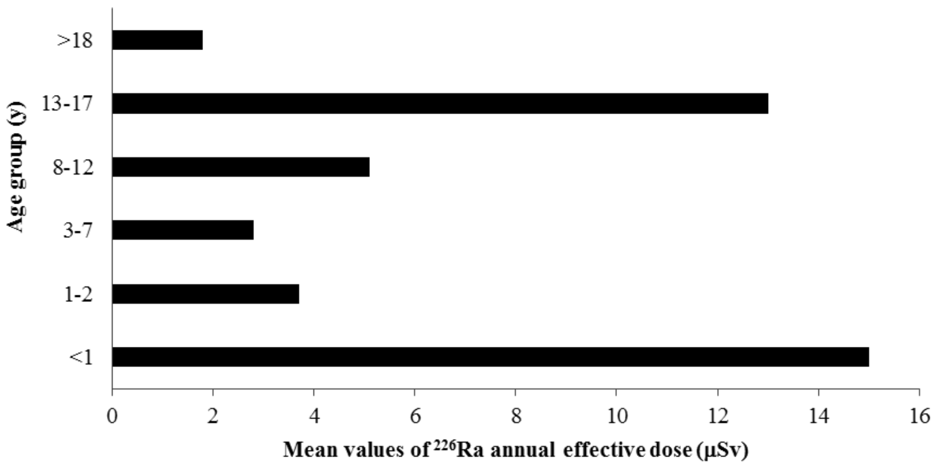
The concentration range of ^{226}Ra in investigated drinking water samples were 2.0±0.1 to 38.2±2.4 mBq l^{-1}. These results were compared with 1.27 to 27.46 mBq l^{-1} in China [37], 8.42 to 40.57 mBq l^{-1} in Croatia [40], 11 to 36 mBq l^{-1} in Turkey [36] and 8 to 83 mBq l^{-1} in Brazil [42]. According to the guidelines for drinking water of the Environmental Protection Agency (EPA) [39], ^{226}Ra concentration in drinking water level should not exceed the value of 0.74 Bq l^{-1}. So, the measured levels of ^{226}Ra in this research are below of the recommended level. Also, recommended level by WHO in drinking water 1000 mBq l^{-1} [43], is higher than our maximum concentration value.

The annual effective dose magnitudes of ^{226}Ra according to age groups were calculated by assuming the mean value of ^{226}Ra concentration. The highest value of annual effective dose related to infants and teens groups, while, the lower value of this parameter was at >18 age groups, figure 2. The adult age group with a reduction of metabolic functions has much less sensitive to the existence of ^{226}Ra. In the comparison of these results with the international guideline level of annual effective dose 100 mSv, our result was lower than the recommended level [43].

According to the WHO's suggestion, the health risk of ^{226}Ra in drinking water is mainly concerned with the effective dose less than chemical radiotoxicity (25).

The radiological risk due to ^{226}Ra in drinking water was calculated and obtained results were compared with the standard level of 10^{-3} for the radiological risk recommended by EPA (39), the cancer mortality and morbidity risk for the study area were regarded to be negligible.

In the comparison of the chemical toxicity risks parameter in studied area with the referenced level dose recommended by EPA 0.6 mg/kg day (39), the obtained results were lower than the recommended level. As can be observed the maximum value of lifetime average daily dose parameter in the study area was 6 times lower than the reference level.



CONCLUSION

The values of ^{226}Ra activity concentration in drinking water samples of the study area ranged from 2.0 to 38.2 mBq l^{-1}. The annual effective dose evaluations according to age groups were shown that infants and adult groups have the highest and lowest sensitivity to the presence of ^{226}Ra in drinking water. For these groups, the high value of dose absorption could be related to the procedure of testosterone, which has significant rules to bone calcification during these life periods (44). The ranges of mortality and morbidity risks were from 7.32×10^{-7} to 1.40×10^{-5} and 1.06×10^{-6} to 2.03×10^{-5}, with a mean of 3.28×10^{-6} and 4.75×10^{-6}, respectively. These results were lower than the recommended level of WHO, 10^{-3}. The chemical risk, as estimated by the LADD, has a mean value of 2.52×10^{-2} mg/kg day, which this value is below the standard level (0.6 mg/kg day) of LADD by EPA. Finally, the results demonstrated that on adverse health risk is posed to the public by ingestion of drinking water from the selected area.

ACKNOWLEDGEMENT

The authors would like to thank the personnel of the radioecology labs in the National Radiation Protection Institute (NRPI) of Iran.

Conflicts of interest: *Declared none.*

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