

Lifetime risk assessment of Radium-226 in drinking water samples

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ABSTRACT

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Background: One of the most significant radionuclides in natural drinking waters is ²²⁶Ra and its decay products. It is potential of health problems, including cancer risk. In this study, the effect of ²²⁶Ra concentration in 28 drinking water samples collected from the North Guilan province was investigated. **Materials and Methods:** The activity concentrations of ²²⁶Ra 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 ²²⁶Ra 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. ²²⁶Ra is a product of the ²³⁸U series with half-life 1600 y that alpha and beta particles are emitted from it. Whereas, ²²⁶Ra is one of the most important radionuclides in drinking water, may pose a health impact (1-4). ²²⁶Ra and ⁴⁰Ca metabolic path in the body are same and principally positioned in the through body bone (5).

Absorption of the ²²⁶Ra with the body

depends on various factors such age, body weight, sex, and metabolic activity. The health effects of ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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 km² 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 HNO₃ 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 (BaSO₄) for precipitation of ²²⁶Ra in the sample. The obtained precipitate was dissolved in EDTA solution. For equilibrium between ²²⁶Ra and ²²²Rn, the sample placed in a sealed bubbler and stored. After growth ²²²Rn gas in the bubbler, the ²²²Rn gas was evacuated into scintillation Lucas cells, by noble gas. The alpha particles emitted from ²²²Rn 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 ²²⁶Ra in water sample was calculated using equation (1):

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

Where A_{Ra} is the ²²⁶Ra activity concentration (pCi l⁻¹), and C_{net} , ϵ 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 ²²²Rn ($2.1 \times 10^{-6} \text{ s}^{-1}$), respectively. The conversion factor from dps/pCi is 133.2 constant.

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

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

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

Internal exposure

According to the world health organisation (WHO) the relevance between total ingestion dose $E(\text{Sv y}^{-1})$ 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 ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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).

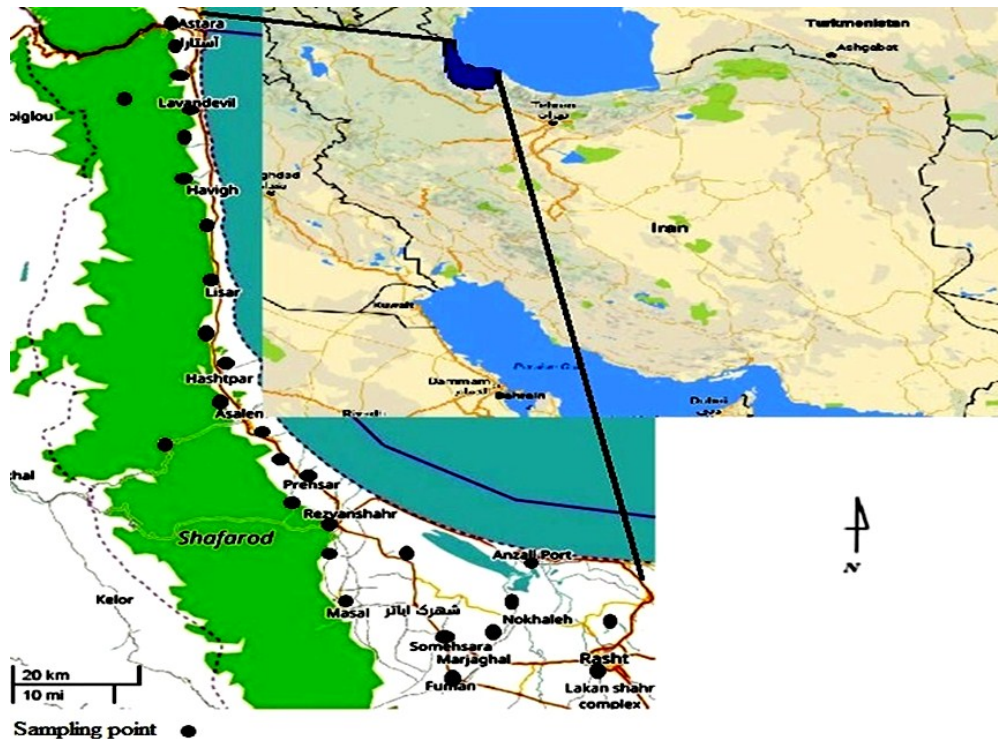


Figure 1. The map showing the study area and sampling sites.

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 ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra concentration, respectively. The mean value of the lifetime average daily dose was 2.52×10^{-2} .

The chemical toxicity risks of ²²⁶Ra 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 ²²⁶Ra in drinking water samples.

²²⁶ Ra concentration range	²²⁶ Ra concentration 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 ²²⁶Ra 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), ²²⁶Ra concentration in drinking water level should not exceed the value of 0.74 Bq l⁻¹. So, the measured levels of ²²⁶Ra 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.

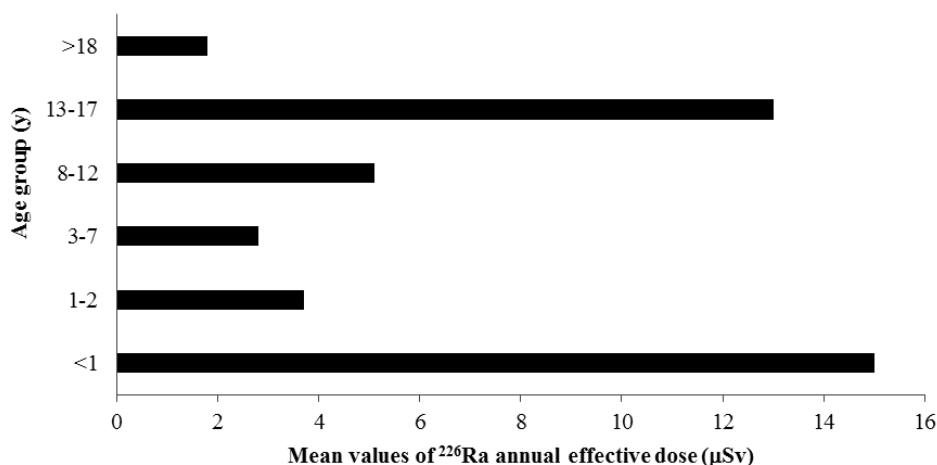
The annual effective dose magnitudes of ²²⁶Ra according to age groups were calculated by assuming the mean value of ²²⁶Ra concentration. The highest value of annual effective dose related to infants and teens groups, while, the lower value of this parameter was at >18 age groups, figure 2. The adult age group with a reduction of metabolic functions has much less sensitive to the existence of ²²⁶Ra. 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 ²²⁶Ra in drinking water is mainly concerned with the effective dose less than chemical radiotoxicity (25).

The radiological risk due to ²²⁶Ra 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 ²²⁶Ra 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 ²²⁶Ra 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.

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REFERENCES

1. Clark MW, Akhurst DJ, Fergusson L (2011) Removal of radium from groundwater using a modified bauxite refinery residue. *Journal of Environmental Quality*, **40(6)**: 1835-1843.
2. Kiro Y, Weinstein Y, Starinsky A, Yechieli Y (2015) Application of radon and radium isotopes to groundwater flow dynamics: An example from the Dead Sea. *Chemical Geology*, **411**: 155-171.
3. Kurttio P, Komulainen H, Leino A, Salonen L, Auvinen A, Saha H (2005) Bone as a possible target of chemical toxicity of natural uranium in drinking water. *Environmental Health Perspectives*, **113(1)**: 68.
4. Lauria DC, Almeida RMR, Sracek O (2004) Behavior of radium, thorium and uranium in groundwater near the Buena Lagoon in the Coastal Zone of the State of Rio de Janeiro,

- Brazil. *Environmental Geology*, **47(1)**: 11-19.
5. Schlenker RA, Keane, AT, Holtzman RB (1982) The retention of ²²⁶Ra in human soft tissue and bone; implications for the ICRP 20 alkaline earth model. *Health Physics*, **42(5)**: 671-693.
 6. Patra A C, Mohapatra S, Sahoo SK, Lenka P, Dubey JS, Tripathi RM, Puranik VD (2013) Age-dependent dose and health risk due to intake of uranium in drinking water from Jaduguda, India. *Radiation Protection Dosimetry*, **155(2)**: 210-216.
 7. ICRP, International Commission on Radiological Protection (1993). Age dependent doses to members of the public from intake of radionuclides: Part 2: Ingestion dose coefficients. *Publication 67, Oxford Pergamon Press: Oxford*; 23 (3/4).
 8. De Bortoli M and Gaglione P (1972) Radium-226 in environmental materials and foods. *Health Physics*, **22(1)**: 43-48.
 9. Kuo YC, Lai SY, Huang CC, Lin, YM (1997) Activity concentrations and population dose from radium-226 in food and drinking water in Taiwan. *Applied Radiation and Isotopes*, **48(9)**: 1245-1249.
 10. Muth H, Rajewsky B, Hantke HJ, Aurand K (1959) The normal radium content and the Ra-226/Ca ratio of various foods, drinking water and different organs and tissues of the human body. *Health Physics*, **2(3)**: 239-245.
 11. Zhuo W, Lida T, Yang X (2001) Occurrence of ²²²Rn, ²²⁶Ra, ²²⁸Ra and U in groundwater in Fujian Province, China. *Journal of Environmental Radioactivity*, **53(1)**: 111-120.
 12. Malakootian M and Nejhad YS (2017) Determination of radon concentration in drinking water of Bam villages and evaluation of the annual effective dose. *Int J Radiat Res*, **15(1)**: 81-89.
 13. Kavitha E, Chandrashekhara MS, Paramesh L (2015) ²²⁶Ra and ²¹⁰Po concentration in drinking water of Cauvery river basin south interior Karnataka State, India. *Journal of Radiation Research and Applied Sciences*, **10(1)**: 20-23.
 14. Bergamini G, Taddei M, Rosa M, Ferreira M, Cheberle L, Santos S, Ramos E (2016) Determination of Ra in drinking water samples by alpha spectrometry. *Journal of Radioanalytical & Nuclear Chemistry*, **307(1)**. Page numbers??
 15. Le CH, Huynh NPT, Le QB (2015) Radon and radium concentrations in drinkable water supplies of the Thu Duc region in Ho Chi Minh City, Vietnam. *Applied Radiation and Isotopes*, **105**: 219-224.
 16. Walsh S, Satkunam M, Su B, Festarini A, Bugden M, Peery H, Stuart M (2015) Health, growth and reproductive success of mice exposed to environmentally relevant levels of Ra-226 via drinking water over multiple generations. *International Journal of Radiation Biology*, **91(7)**: 576-584.
 17. Althoyaib S and El-Taher A (2015) Natural radioactivity measurements in groundwater from Al-Jawa, Saudi Arabia. *Journal of Radioanalytical & Nuclear Chemistry*, **304(2)**: 547-552.
 18. Shweikani R and Raja G (2015) Natural radionuclides monitoring in drinking water of Homs city. *Radiation Physics and Chemistry*, **106**: 333-336.
 19. Moldovan M, Benea V, Niță DC, Papp B, Burgele BD, Bican-Brișan N, Cosma C (2014) Radon and radium concentration in water from North-West of Romania and the estimated doses. *Radiation Protection Dosimetry*, **162(1-2)**: 96-100.
 20. Abbasi A, Mirekhtyari F (2017) Gross alpha and beta exposure assessment due to intake of drinking water in Guilan, Iran. *Journal of Radioanalytical and Nuclear Chemistry*, **314(2)**: 1075-1081.
 21. Tabar E and Yakut H (2014) Determination of Ra-226 concentration in bottled mineral water and assessment of effective doses, a survey in Turkey. *Int J Radiat Res*, **12(3)**: 193-201.
 22. Kolo MT, Amin YM, Khandaker MU, Abdullah WHB (2017) Radionuclide concentrations and excess lifetime cancer risk due to gamma radioactivity in tailing enriched soil around Maiganga coal mine, Northeast Nigeria. *Int J Radiat Res*, **15(1)**: 71-80.
 23. Dizman S, Görür FK, Keser R (2016) Determination of radioactivity levels of soil samples and the excess of lifetime cancer risk in Rize province, Turkey. *Int J Radiat Res*, **14(3)**: 237-244.
 24. Ferdous J, Begum, A, Islam A (2015) Radioactivity of soil at proposed Rooppur Nuclear Power Plant site in Bangladesh. *Int J Radiat Res*, **13(2)**: 135-142.
 25. World Health Organization (WHO) (2004) Guidelines for drinking-water quality (Vol. 1). World Health Organization. Geneva, Switzerland, WHO Library.
 26. Mohebbi M, Mahmoodi M, Wolfe R, Nourijelyani K, Mohammad K, Zeraati H, Fotouhi A (2008) Geographical spread of gastrointestinal tract cancer incidence in the Caspian Sea region of Iran: spatial analysis of cancer registry data. *BMC Cancer*, **8(1)**: 137.
 27. Sethy NK, Jha VN, Ravi PM, Tripathi RM (2014) A simple method for calibration of Lucas scintillation cell counting system for measurement of ²²⁶Ra and ²²²Rn. *Journal of Radiation Research and Applied Sciences*, **7(4)**: 472-477.
 28. Szabo Z, Vincent T, Fischer JM, Kraemer TF, Jacobsen E (2012) Occurrence and geochemistry of radium in water from principal drinking-water aquifer systems of the United States. *Applied Geochemistry*, **27(3)**: 729-752.
 29. Jeong DH, Kim MS, Noh HJ, Yoon YY, Kim DS, Lee YJ, Kim TS (2012) A Preliminary Study for the Analytical Method and Environmental Characteristics of Radium-226 in Groundwater. *Journal of Soil and Groundwater Environment*, **17(2)**: 22-27.
 30. Abbasi A and Bashiry V (2016) Measurement of radium-226 concentration and dose calculation of drinking water samples in Guilan province of Iran. *Int J Radiat Res*, **14(4)**: 361-366.
 31. Hamlat MS, Kadi H, Djeflal S, Brahimi H (2003) Radon concentrations in Algerian oil and gas industry. *Applied Radiation and Isotopes*, **58(1)**: 125-130.
 32. Smith CG, Cable JE, Martin JB, Roy M (2008) Evaluating the source and seasonality of submarine groundwater dis-

- charge using a radon-222 pore water transport model. *Earth and Planetary Science Letters*, **273(3)**: 312-322.
33. Mathieu GG, Biscaye PE, Lupton RA, Hammond DE (1988) System for measurement of ²²²Rn at low levels in natural waters. *Health Physics*, **55(6)**: 989-992.
 34. Papastefanou C (2007) Measuring radon in soil gas and groundwaters: a review. *Annals of geophysics*, **50(4)**: 569-578.
 35. Shashikumar TS, Chandrashekara MS, Paramesh L (2011) Studies on radon in soil gas and natural radionuclides in soil, rock and ground water samples around Mysore city. *International Journal of Environmental Sciences*, **1(5)**: 786-791.
 36. International Commission on Radiological Protection (1974) Report of the task group on Reference Man. Oxford: Pergamon Press; ICRP Publication 23.
 37. Hu Y, Yan S, Xia C, Dong Z, Liu G (2017) Distribution characteristics and radiotoxicity risks of radium-226 (²²⁶Ra) in groundwater from Wanbei Plain, China. *Journal of Radio-analytical and Nuclear Chemistry*, **3(311)**: 2079-2084.
 38. UNSCEAR (2000) Sources, effect and risks of ionising radiation. Report to the general assembly with scientific annexes. United Nations. New York.
 39. EPA (1999) Cancer risk coefficients for environmental exposure to radionuclides. United State Environmental Protection Agency. *Federal Guidance Report No-13 (EPA. 402 R-99-001)*.
 40. Bronzovic M, Marovic G, Vrtar M, Bituh T (2007) Life exposure to ²²⁶Ra and possible consequences. *Journal of Environmental Science and Health Part A*, **42(6)**: 817-823.
 41. Karahan G, Öztürk N, Bayülken A (2000) Natural radioactivity in various surface waters in Istanbul, Turkey. *Water Research*, **34(18)**: 4367-4370.
 42. Godoy JM, da S Amaral EC, Godoy MLD (2001) Natural radionuclides in Brazilian mineral water and consequent doses to the population. *Journal of Environmental Radioactivity*, **53(2)**: 175-182.
 43. World Health Organization. Radiological aspects (1993) In: Guidelines for drinking-water quality. Geneva: *World Health Organization*, 114 –121.
 44. Main KM, Schmidt IM, Skakkebaek NE (2000) A possible role for reproductive hormones in newborn boys: progressive hypogonadism without the postnatal testosterone peak. *The Journal of Clinical Endocrinology & Metabolism*, **85(12)**: 4905-4907.

