IOSR Journal of Applied Physics (IOSR-JAP)
e-ISSN: 2278-4861.Volume 9, Issue 1 Ver. I (Jan. – Feb. 2017), PP 17-21
www.iosrjournals.org

Natural radioactivity in drinking water and associated age-dependent dose in Luxor, Upper Egypt

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Abstract: *The presence of Ra in drinking water may sometimes make important contribution to natural background radiation exposures. The paper describes the study of ^{226}Ra and ^{228}Ra content in drinking water of Luxor, a famous tourist city in Egypt. A total of thirty-five water samples were analyzed for ^{226}Ra and ^{228}Ra by gamma ray spectrometry with HPGe detector setup, coaxial type and 8192 channels MCA. The concentration of ^{226}Ra was found in the range from 16 to 181 with arithmetic mean 85.6 mBq·L^{-1}and the concentration of ^{228}Ra ranged from 8 to 98.4 mBq·L^{-1} with arithmetic mean 48.6 mBq·L^{-1}.The committed effective dose for the different age groups has been estimated and found to be not more than 0.038 mSv·yr^{-1} for adults and 0.149 mSv·yr^{-1} for critical age group (12-17 years old), for ^{226}Ra. While it is not more than 0.05 mSv·yr^{-1} for adults and 0.29 mSv·yr^{-1} for critical age group, for ^{228}Ra. The values obtained were compared with the reference values accepted for drinking water and doses resulting from consumption of these waters, due to their ^{226}Ra and ^{228}Ra contents, were calculated. The study showed that ^{226}Ra content for investigated categories of waters is below the levels at which any unacceptable dose due to ingestion would arise.*

Keywords: *^{226} Ra, ^{228}Ra, drinking water, radioactivity, effective doses.*

I. Introduction

The water has an importance in environmental studies because of its daily use for human consumption and its ability to transport pollutants. Radionuclides in drinking water causes human internal exposure, caused by the decay of radionuclides taken into the body through ingestion and inhalation indirectly when they are incorporated as part of the human food chain. Measurements of natural radioactivity in drinking water have been performed in many parts of the world, mostly for assessment of the doses and risk resulting from consuming water (1). Radium-226 and radium-228 radioisotopes present in water beyond the recommended level are considered to have potential risks to man from their consumption at a regular rate. This is because of their long environmental half-life, high radiotoxicity and high affinity to biota. When groundwater moves from one place to another, it takes away the soluble radionuclides that come in contact with the water. Groundwater deposits display a diverse range of quality and chemistry (2). The quality depends on the mineralogy and reactivity of the drift material and the degree of equilibrium that has been attained between water and rock. Elevated levels of radium in ground water are associated with low-grade radium deposits (3).

The radioactivity of drinking water is an environmental factor which contributes to the population exposure to ionizing radiations and the activity of monitoring the water radioactive content is the responsibility of the national public health systems by ensuring the maintaining of the effective dose by ingestion in the provided limits. The population usage of drinking water represents a way of the population exposure to the ionizing radiations by ingestion the radionuclides existing in it. UNSCEAR estimates that the natural sources contribution to the effective dose is 2.4 mSv y^{-1} (in this dose value being contained the value of 0.3 mSv y^{-1} due to the usage of food and water) (4,5). Natural radium is classified as a radiological toxic agent. ^{226}Ra and ^{228}Ra accumulate in the human body primarily through the intake of food and water. The contribution of drinking water to the total intake is important when the drinking-water supplies are drawn from ground water supplies, as the radium concentrations vary widely and levels in excess of 200 Bq·m^{-3} are not uncommon. When radium is taken into the body, its metabolic behavior is similar to that of calcium, and an appreciable fraction is deposited in bone. More than 70 % of the radium in the body is contained in bone, the remaining fraction being distributed rather uniformly in soft tissues (4). The elevated ingestion of ^{226}Ra might provide an annual internal dose near to the 0.1 mSv.yr^{-1} reference level recommended by the World Health Organization (6).

The primary route for transfer of ^{226}Ra to the human body is ingestion. More recent data indicate that the ^{226}Ra content in public water supplies is highly variable. The occurrence, natural distribution and concentration of radium in drinking water are strongly influenced by geological setting and the interactions of groundwater with radium bearing materials such as rocks, soil, ore bodies, etc.(7). Radium intake by drinking water originating from surface water sources is generally small. However, as drinking water supplies mostly use groundwater sources, the intake may be more significant, although in most cases the groundwater is subjected to water treatment processes before reaching the public (8). Determination of ^{226}Ra content in drinking water is useful for the purpose of the prevention of unnecessary exposure of humans to natural radiation. It is also of great importance to monitor for ^{226}Ra presence in water in order to prevent undesirable deposition of that hazardous element into the bone during childhood and to reduce the risk of occurrence of bone sarcomas later in life.

The aim of this study is to obtain a representative estimate of the concentration levels of natural radionuclides in drinking water and the corresponding radiation doses for people consuming this water. The data generated in this paper may contribute to determine the base-line levels of natural radioactivity in drinking water and help in the development of future guidelines in the country for radiological protection of the population.

II. Materials and methods

A total of 35 samples of drinking water were collected from water supply systems located in various parts of Luxor city. The systems drew their supply from surface water from rivers or lakes (20 water pipes) and from deep underground wells (15 water pipes).

A polyethylene marinelli beaker 1.4 liter was used as sampling and measuring containers. Before use, containers were washed with dilute hydrochloric acid and rinsed with distilled water. The beaker is filled up to the brim and a tight cap is pressed on so that the air is completely removed from it. The samples were taken to the laboratory and stored for a minimum period of one month to allow daughter products to come into radioactive equilibrium with their parents ^{226}Ra and ^{232}Th. Every sample was counted for 600 to 900 min depending on the level of concentrations of the radionuclides.

Each sample was subject to a gamma ray spectrometer with HPGe setup and multichannel analyzer 8192 channels. The detector is coaxial closed facing window geometry with vertical dipstick (500-800 micron). The HPGe detector is p-type with the following specifications: Resolution (FWHM) at 122 keV ^{57}Co is 1100 eV and at 1.33 MeV ^{60}Co is 2.00 keV, Relative efficiency at 1.33 MeV ^{60}Co is 30 % .

For background a 1.4 liter Marinelli beaker filled with distilled water was used. The background was measured frequently usually every week under the same conditions of the samples measuring. The spectra were either evaluated with the computer software programme Maestro (EG& G ORTIC), or manually with the use of a spreadsheet (Microsoft Excel).

The ^{226}Ra concentration was determined from the gamma lines of 295 keV, 352 keV, 609 keV and 1765 keV from ^{214}Pb and ^{214}Bi. Radium concentration was calculated as the mean value of the results of these gamma lines. The 186-keV gamma line from ^{226}Ra itself was not used because the intense gamma line of ^{235}U, with an energy of 185.7 key, will be mixed in the same peak. The use of ^{214}Pb and ^{214}Bi in the determination of radium concentration calls for radioactive equilibrium between the radium and radon in the sample. The ^{228}Ra concentration was determined from the gamma lines of 583 keV and 911 keV from ^{208}T1 and ^{228}Ac, respectively.

III. Results and discussion

1.1. Radioactivity of ^{226}Ra and ^{228}Ra in drinking water

The activity concentrations of ^{226}Ra and ^{228}Ra determined of drinking water samples from Luxor City, Egypt are presented in Table 1. The highest values of ^{226}Ra concentration were determined in ground waters with the mean value of 95.1 mBq L^{-1}. Tab waters show lower content of ^{226}Ra, ranging from 16 to max. 157.6 mBq L^{-1}, with the mean value of 77.9 mBq L^{-1}. With respect to ^{228}Ra, the lower and the max. values (8.1 and 98.4 mBq L^{-1}) were found in ground and tab waters ,respectively. The concentrations range were lower than those estimated by Godoy (9) for ingestion of groundwater in Brazilian groundwater, and those predicted from the ingestion of drinking water in Finland, Slovenia and Syria (10,11).

Table 1. Arithmetic mean (AM), Arithmetic stander deviation (AM.SD), Geometric mean (GM), Geometric stander deviation (GM.SD) and uncertainty for ^{226}Ra and ^{228}Ra concentration (mBq/L) in samples of drinking water from Luxor city, Upper Egypt.

Parameters	Tap water		Groundwater	
	Ra-226	Ra-228	Ra-226	Ra-228
AM	77.9	49.7	95.1	47.2
AM.SD	45.7	19.6	42.4	27.9
GM	61.1	45.0	84.2	37.7
GM.SD	2.2	1.6	1.7	2.1
Uncertainty	10.1	4.4	11	7.2
Range	16 -157.6	11.7 - 84.3	21.6 -181	8.1- 98.4

The concentrations range and expected doses estimated for the ingestion of ^{226}Ra radionuclide by the population of Luxor were lower than those obtained by UNSCEAR 2000 (France, Germany, Italy, Romania and Switzerland) (see Table 2). These doses were also lower than those estimated by Gans, Bettencourt, and Marovic (12 ,13).

Radionuclides from the aquatic environment may be incorporated into organisms or man by the consumption rate of water. The estimation of consumption rates is a complex matter and depends on a number of social, cultural, religious and economical factors. Environmental temperature and body activity also influence fluid requirements. At low temperatures fluid intake and water loss is scarcely affected by ambient temperature, but at temperature greater than 25 °C, there is a sharp increase in water intake (14). Egypt, as many countries such as Pakistan, characterized by two distinct seasons (summer and winter), where is a large variation in the temperature throughout the year. During winter (November-February) temperature drops to as low as 5 °C, while in summer (June-August) a sharp increase in the ambient temperature is observed and in some areas (Luxor and Aswan, Upper Egypt) it reaches a maximum value of 55 °C. Moreover, in the Holy Month of Fasting (Ramadan), the consumption rate of water also decreases to a large extent. Thus, the fluid intake requirements are not the same throughout the year.

Therefore, the exact rate of intake of water may not be easily calculated. According to NCRP, an average person consumes about 3L.d^{-1} of fresh water. This includes 1.2 L.d^{-1} from drinking water, 0.2 L. d^{-1} by inhalation and passage through the skin of atmospheric water, 1.3 L. d^{-1} from food and 0.3 L. d^{-1} is associated with seafood consumption. There is evidence from both human and animal studies that radiation exposure at low to moderate doses may increase the long-term incidence of cancer. The magnitude of natural exposure depends on geographical location and on human activities (15).

Table 2. Summarized published data on ^{226}Ra (mBq.L^{-1}) in drinking waters from various countries.

Country	Numbers of samples	Concentration range	Geometric mean	References
Serbia	117	120 - 1480	360	20
Yemen	9	2000- 6055	n.c.	21
Niger Delta	20	8 - 20	12	22
Italy	17	0.206–103	n.c.	23
Austria	197	<20-225	37	24
	34	<19-230	25	11
	16	3-140	30	25
France	11	<37-960	44	11
	n.c.	7- 700	n.c.	4
Italy	n.c.	0.2 -1200	n.c.	4
	234	<3-700	6.8	11
Brazil	39	2.2-235	5.7	26
Brazil	36	<10-220	27^{b}	27
Slovenia	34	7–614	35	9
Portugal	50	<3-2185	26.7	12
Spain	n.c.	20 - 4000	n.c.	4
U.K	n.c.	1 – 80	n.c.	4
China	n.c.	0.2 - 120	n.c.	4
Pakistan,	50	<31- n.c.	27^{b}	14
	50	<44- n.c.	42^{b}	14
France	11	<37-960	44	11
	n.c.	7- 700	n.c.	4
Switzerland	n.c.	< 20->500	n.c.	24
Thailand	13	2.5 – 8.5	n.c.	28
Germany	264	<1-1800	25	11
	17	<5-510	81	24
Croatia	2	68-303	151	13
Luxor , Upper Egypt	35	16 -181	70.2	Present study

n.c.: not cited, ***b*** Production weighed mean.

1.2. Estimation of annual effective doses

Dose from radioactivity in drinking water depends on intake and metabolic bodily reactions. Therefore, the effective dose was assessed for different male age groups as follows (6):

$$D_{Ra} (g) = C_{Ra} (i) \times WI (g) \times DCF_{Ra} (g) \qquad (1)$$

where

D Ra effective dose per year for specific age group from ingestion of Ra in water (Sv.y^{-1});

C Ra specific activity in sample *i* (Bq .L^{-1});

WI daily water intake for specific age group (L.y^{-1}); and

DCF dose conversion factor for Ra for specific age group (Sv.Bq^{-1}).

Table 3. Average, minimum and maximum values of ^{226}Ra and ^{228}Ra effective doses per year (μSv.y^{-1}) for different age groups.

Type Water	Parameters	^{226}Ra effective doses per year (μSv.y^{-1})					
		≤ 1 y	1-2 y	2-7 y	*7-12 y*	*12-17 y*	>17 y
Tap Water	Mean	91.5	26.2	16.9	21.8	64.2	16.4
	min	18.8	5.4	3.5	4.5	13.2	3.4
	max	185.1	52.9	34.2	44.1	130.0	33.1
Ground Water	Mean	111.7	32.0	20.6	26.6	78.5	20.0
	min	25.3	7.2	4.7	6.0	17.8	4.5
	max	212.4	60.8	39.2	50.6	149.2	38.0
Dose Coeff.(×10^{-7} Sv/Bq)		47	9.6	6.2	8	15	2.8
^{228}Ra effective doses per year (μSv.y^{-1})							
Tap Water	Mean	372.6	99.1	59.1	67.8	144.8	25.7
	min	87.6	23.3	13.9	15.9	34.1	6.0
	max	632.0	168.1	100.3	115.0	245.7	43.6
Ground Water	Mean	353.6	94.1	56.1	64.4	137.4	24.4
	min	60.71	16.15	9.63	11.05	23.59	4.19
	max	737.9	196.3	117.1	134.3	286.8	50.9
Dose Coeff.(×10^{-7} Sv/Bq)		300	5.7	34	39	53	6.9

The results were obtained by dose conversion factor for individual age groups (16, 17) . All calculations were carried out assuming an annual intake of drinking water of 250 L y^{-1} for age < 1 year, 350 L y^{-1} for ages between 1–10 years, 750 L y^{-1} for age >17 years For the age group 10–12 years the same consumption as the 1–10 year group was assumed; for the 12–17 year group, the interpolated value of 550 L y^{-1} was used. Table 3 shows assessed ^{226}Ra effective dose per year for infants and children between 2 and 17 y of age and adults. These age groups are supposed to consume all types of investigated waters. Figures 1 and 2 illustrated the ^{226}Ra and ^{228}Ra effective doses per year for different ages in drinking water samples.

As it is noticed from the data in Table 3 the assessed ^{226}Ra and ^{228}Ra effective doses per year for children 2-7 year of age from consumption of drinking waters show low values. The values of annual effective doses range between 0.0035 - 0.039 mSv.y^{-1} for ^{226}Ra and between 0.0096 - 0.117 mSv.y^{-1} for ^{228}Ra. The most exposure groups to ^{226}Ra in drinking water are infants and boys between 12 and 17. These are life periods of intensive production of sexual hormone, testosterone, which helps Ca deposition into the bone (18). For that reason, rapid growth accompanied with impact of testosterone could intensify ^{226}Ra deposition into the bone instead[6] of Ca. According to Table 3, the assessed ^{226}Ra effective dose per year reaches the maximum in infants, being about six times higher than that in adults. This value is followed by a dramatic fall in the second year of life and a very slow rise over the next few years. High values of assessed ^{226}Ra effective doses per year occur again between 12 and 17 y of life.

Under normal environmental conditions, the relative contribution of drinking water to the total ^{226}Ra intake by a standard man is minor; only about 10 % of radium intake occurs from drinking water. This, of course, may not be true for the sites with a high ^{226}Ra concentration determined in the water (19). According to ICRP recommendations, the limit for public exposure is 1 mSv per year (4). It may be concluded that the doses resulting from drinking waters in Luxor, Upper Egypt are not above the prescribed dose limits. Since the doses considered in our study are well below the permissible dose of 1 mSv.

According to the results of our study, it is evident that the activity concentration of ^{226}Ra measured in drinking water as well as the relative contribution of ^{226}Ra from tap and underground waters in the assumed ratio presents no significant risk for the Luxor city, Upper Egypt population.

IV. Conclusion

Natural radionuclides content in tap and groundwater samples were presented, including different sites from Luxor city, Upper Egypt. The geometric mean of ^{226}Ra and ^{228}Ra radionuclides in drinking water (tap water) were found to be 63.1±2.2 mBq.L^{-1} and 45.0±1.6 mBq.L^{-1}, while that of groundwater were 84.2 ± 1.7 mBq.L^{-1}and 37.7±2.1 mBq.L^{-1}. The effective doses from ingestion of ^{226}Ra in drinking water are age-dependent and they are considerable high in infants and children between 12 and 17. These are life periods of intensive production of testosterone accompanied with rapid growth that could amplify deposition of ^{226}Ra (as Ca homologue) into the bone. For the children of these age groups, ^{226}Ra concentration should be under 100 mBq L^{-1} in order to prevent increased ^{226}Ra deposition into the bone. Water consumption and water quality are necessary for health maintaining. Since exposure to low doses may increase the long-term incident of cancer, constant monitoring of ^{226}Ra presence in drinking water is required. According to our results, all analyzed water samples are acceptable for drinking with the levels of ^{226}Ra concentrations under the value as recommended by

WHO (5). The latest guideline activity concentration for ^{226}Ra recommended by the World Health Organization, assuming the intake of 2 litres of water per day for one year, is 1 Bq L^{-1} WHO. For a more accurate risk assessment for drinking water, more measurements of natural radioactivity in drinking water are still needed.

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