Determination of Natural Radioactivity in Drinking Water and Consequent Dose to Public

W.M. Abdellah and H.M. Diab

National Center for Nuclear Safety and Radiation Control, Atomic Energy Authority of Egypt, P.O. 7551, Nasr City, Cairo, Egypt.
E-mail: wmsra@yahoo.com

Abstract: Tap potable water in Egypt is a necessity practice, rather than a choice. Radium as well as other heavy metals in drinking water can pose a health hazard to human. In this work, determination of natural radium isotopes (^{226}Ra and ^{228}Ra) activity concentrations (mBq/L) in potable water samples from various locations in Egypt were carried out. The effective dose (mSv/y) and the associated cancer risk to public were estimated. The activity concentrations of ^{226}Ra were found to be in the range of 0.5 ± 0.2 to 22.0 ± 1.3 mBq/L with an average 3.6 ± 0.4 mBq/L. The activity concentration of ^{228}Ra were in the range of 41.6 ± 5.19 to 116.8 ± 14.6 mBq/l with an average 57.97 ± 9.49 mBq/l. The average estimated effective doses due to water consumption were 0.73 μSv/y for ^{226}Ra and 38.26 μSv/y for ^{228}Ra. The cancer risks due to water consumption during the life time (70 y) estimated was found to be in the range 1.83 x 10^{-7} to 11.95 x 10^{-5} with an average value of 7.89 x 10^{-5}.
[W.M. Abdellah and H.M. Diab. **Determination of Natural Radioactivity in Drinking Water and Consequent Dose to Public.** Nature and Science 2012; 10(1): 137-142]. (ISSN: 1545-0740). http://www.sciencepub.net

Key words: Water; ^{226}Ra; ^{228}Ra; Dose equivalent; Cancer risk

INTRODUCTION

Determination of radium isotopes in water has become a matter of interest in public health. Also determination of long-lived ^{226}Ra and its progenies ^{210}Pb and ^{210}Po is of great interest in the field of radioprotection. These radionuclides preferentially accumulate in bones leading to rather long residence times in the human body. The radioactivity of the water intended for human consumption has been brought to public attention by recent international regulations such as EU council Directive 98/83/EC [1], Euratom [2,3] and WHO 1998 [4,5]. Upon ingestion of radium containing water, however, part of the radium is absorbed from the digestive tract and preferentially deposits in the bone. Radiation emitted by the radium isotopes and /or their progenies in the form of alpha or beta particles may then damage tissue and, over time, lead to the development of bone cancer.

The U.S. Environmental Protection Agency (USEPA) has established a Maximum Contaminant Level (MCL) for radium isotopes in public drinking-water supplies because of health risks associated with its ingestion [6]. The MCL is 5 pCi/L for combined radium, which is defined as the sum of ^{226}Ra and ^{228}Ra. The contribution of drinking water to total human exposure is growing due to the presence of naturally occurring radionuclides in both uranium and thorium decay series. An increasing concern presently exists in several countries for the determination of the radioactivity levels in drinking water from various resources by different routine control measurements. Four naturally occurring radium isotopes are among the nuclides pertaining to these decay chains. Of these, ^{226}Ra is an α-emitter with a long half-life of 1622 y and closely follows calcium metabolism in human body with eventual deposition in bones. This might lead to the buildup of ^{226}Ra and its daughters, causing potential health implications and high degree of radio-toxicity due to long exposure hazards. Following the same behavior, ^{228}Ra is a β-emitter with comparatively long half-life (5.77 y) with considerable significance in public health related studies [6]. Details of the dose assessment methodology are always implemented [7] and extensive estimates are referred to define recommended exposure levels for the population in several countries.

The metabolic behavior of radium on the human body is similar to calcium: around 20% of the consumed radium remains in the body, mainly in bones [8]. The behavior of radium in the human body is one of the reasons why radium has a high value of committed effective dose per unit intake [9]. Furthermore, the relative relevance of the radium isotopes regarding ingestion dose conversion factors has changed over time. While in 1992, the dose factor for ^{226}Ra and ^{228}Ra were similar, ^{228}Ra value established after 1996 is twice that of ^{226}Ra (6.9×10^{-7} and 2.8×10^{-7} $SvBq^{-1}$ respectively) and is similar to that of ^{210}Pb [9].

There is a growing interest in the development of improved methods for the detection of Ra isotopes in water. Because low levels of radium are ordinarily encountered in environmental water samples, the determination of radium initially requires one or more preliminary separation and/or pre-concentration steps, both to remove elements which may interfere with

counting and also to get rid of large quantities of inactive substances. Methods based on gamma spectrometry offer good sensitivity but they are time consuming and require the use of large water volumes to attain detection limits of some mBq/L. Therefore, even a very low background is not sufficient to reach the required detection limits. Here, radiochemical separation techniques which separate the ^{226}Ra and ^{228}Ra from the matrix can improve the detection limits. The procedure followed in this study for ^{228}Ra and ^{226}Ra determination was a pre-concentration of radium isotopes (and ^{133}Ba as a yield tracer) via adsorption onto MnO_2 precipitate, extraction of radium isotopes with diphonix resin followed by a second extraction of ^{228}Ac to measure by liquid scintillation counting, LSC and finally, ^{226}Ra co-precipitation with Barium sulphate to measure by an Alpha detector [10].

This study concerned with the determination of activity concentrations of ^{226}Ra and ^{228}Ra in eighteen water samples collected from nine Egyptian cities. A conservative approach used to assess total effective dose and the cancer risk was estimated based on USEPA approach.

EXPERIMENTAL

Apparatus: All samples were counted by alpha spectrometry and liquid scintillation counting, LSC. The α-counting was performed on an alpha spectrometr Model 7401/7401VR , while β-counting was carried out by The LSC using Packard TRI CARB 3170 TR/SL, which used pulse shape discrimination (PSD) for α/β separation. Additionally the background is reduced by the presence of a guard of bismuth germinate (BGO), which surrounds the counting chamber.

Samples: Eighteen tape water samples were collected from nine Egyptian towns. The samples were acidified and transferred to the laboratory. The water samples (one liter volume) were evaporated to a volume of 100 ml, then, it transferred into a Marinillie beaker for gamma spectrometric counting followed by the radiochemical separation of the investigated isotopes.

Procedures: Radiochemical separation of ^{226}Ra and ^{226}Ra isotopes was performed on all samples. MnO_2 co-precipitation at prepared using 0.015 g $KMnO_4$ and 0.5 M $MnCl_2$ the pH was adjusted between pH 8–9. MnO_2 used for the pre-concentration of radium isotopes and effectively separates some possible interfering radionuclides such as U isotopes and ^{90}Sr [10]. The Precipitate is dissolved in ~ 2ml concentrated HCl, which is then diluted to about 2M HCl by addition of 10 ml of water. An initial Diphonix resin has a strong affinity for actinides and lanthanides, and is used to decontaminate the sample of potential environmental interferences while allowing divalent cations such as the radium isotopes and barium to pass through. The decontaminated load and rinse solution containing the Ra (Ba) fraction is collected in a clean glass beaker and held for at least 30 h to allow for ^{228}Ac in-growth from the ^{228}Ra in the sample. The equilibrium activity of ^{228}Ac from ^{228}Ra in the sample is then loaded onto a second Diphonix resin column. The load and rinse solutions from the 2nd column is collected in a clean beaker and is processed for the yield determination by measuring the ^{133}Ba gamma peak at 356 keV via gamma spectrometry. The ^{228}Ac is then eluted from the second Diphonix column using (1 M 1-Hydroxyethane-1,1-diphosphonic acid) into a LSC vial and completed with UGAB liquid scintillation cocktail for measurement via LSC model TRI CARB 3170 TR/Sl.

^{226}Ra precipitated as $Ra(Ba)SO_4$ and determined by an alpha spectrometric model 7401/7401VR Alpha spectrometer system equipment with CANBERRA chamber of low background, high resolution PIPS detectors ion implanted detectors with 1200 mm^2 active area, with counting time not less than 80,000 seconds. The overall yield of this procedure is typically greater than 95%. Alpha-emitting radium isotopes (^{223}Ra, ^{224}Ra and ^{226}Ra), now also free of radioactive interferences, may be analyzed by alpha spectrometry after micro-precipitation with $BaSO_4$ from the final load and rinse solution of the 2nd Diphonix column.

For quality control, water reference materials IAEA-426 and IAEA-423 were analyzed during the same experiment run and the concentration of ^{226}Ra and ^{228}Ra were determined**.** The uncertainties of the results were evaluated considering counting statistics and calibration error only.

DOSE ESTIMATIONS

The Annual Effective dose estimations were calculated based on the radium isotopes activity concentration and the annual water consumption, using the following formula:

Annual Effective dose (μSv/y) = Activity Concentration (Bq/L) × Annual water consumption (L/y) × Dose coefficient factor (μSv/Bq)

Where:

The dose coefficient for ^{226}Ra and ^{228}Ra are 0.28μSvBq^{-1} and 0.69μSvBq^{-1} respectively correspond to a reference dose level (RDL) of 0.1mSv/year, annual water consumption assumed to be 730 liters/year, [11].

The Health effects subcommittee believes that the risks associated with all radium species should be combined so that the total risk is known. The assumed average ratio from the occurrence data is used to determine the concentration of each radium isotope that would meet an acceptable risk level [12].

The cancer risk due to radium isotopes intake was calculated as follows:

$$Risk = MCL \times RC \times TWI$$

Where:

Risk = Lifetime cancer risk corresponding to MCL (unit less)

MCL= Maximum contaminant level (Bq/L)

RC = Mortality risk coefficient for ^{226}Ra (7.17×10^{-9} Bq^{-1}), and for ^{228}Ra (2.0×10^{-8} Bq^{-1})

TWI = Total water intake (2 L/d × 365.4 d/y × 70 y).

RESULTS AND DISCUSSION

The activity concentration of ^{226}Ra and ^{228}Ra in mBq/L in water samples are given in Table 1. The activity concentrations range from 0.5±0.19 to 22±1.3 mBq/L with average 3.6 mBq/L for ^{226}Ra and from < DL to 116.8 ± 14.6 mBq/L with an average 75.9 mBq/L for ^{228}Ra. The mean activity concentrations of both ^{226}Ra and ^{228}Ra are within those in drinking water in several other countries such as united states [13,14,15,16], Pakistan [17,18], Finland [19,20], France [21,22], Germany [23,24,25] , Italy [26], Poland [27,28], Romania [29,30], Switzerland [31], Spain[32] and U.K. [33] as shown in Table 3 and Fig. 1 and Fig. 2. Water types originating from the Eastern Nile Delta area are characterized by low ^{226}Ra levels and relatively high ^{228}Ra activity, presumably due to the muddy agricultural nature of that area, [35] which is subject to water from several surface resources for irrigation, from this point of view, the slight increase of ^{228}Ra in few cities may be attributed to the same reason. The results showed that, in general, radium isotopes activity concentration in drinking water samples did not exceed the maximum contaminant level (5 pCi/L) recommended by USEPA [36]. The average committed effective doses µSv/y due to water consumption is given in Table 2. The highest values of the committed effective dose per year were 4.50 µSv/y for ^{226}Ra and 58.83 µSv/y for ^{228}Ra. The average committed effective doses per year were 0.73 µSv/y and 38.27 µSv/y for ^{226}Ra and ^{228}Ra respectively. The radium isotopes (and here especially ^{228}Ra) are responsible for the major part of the annual effective dose from ingestion water. The corresponding total committed effective dose obtained was 38.99 µSv/y. So, regular consumption of radium through drinking water, even at activity concentrations few times over the maximum recommended value, will not lead to an effective dose higher than recommended 0.1 mSv.

Risks associated with ingestion of radium isotopes (^{226}Ra and ^{228}Ra) for the concentrations and exposure periods discussed above are given in Table 2. The USEPA established a range of 1×10^{-4} to 1×10^{-6} as an acceptable cancer incidence risk in the Notice of data availability for radionuclides in drinking water that was published on April 21, 2000. It is noted that the resulting risks are the same order of magnitude as the EPA's target ceiling risk (10-4); however, the actual value is different for each radionuclide.

Table 1. The Estimated ^{226}Ra and ^{228}Ra Activities in the Collected Water Samples:

Cities / No.		Radioactivity level (mBq/L)		The mean of Radioactivity level (mBq/L)		Chemical Recovery (%)
		^{226}Ra	^{228}Ra	^{226}Ra	^{228}Ra	
Cairo	a	1.66 ±0.34	< DL	1.66 ±0.34	< DL	83
	b	1.66 ±0.34	< DL			
El Mansoura	a	1.17 ±0.27	< DL	1.17 ±0.27	< DL	98
	b	1.17 ±0.27	< DL			
Qualuab	a	1.69±0.29	72.53±9.06	1.69±0.29	72.53±9.06	98
	b	1.69±0.29	72.53±9.06			
Octobar	a	0.92 ±0.07	< DL	0.92 ±0.07	< DL	96
	b	0.92 ±0.07	< DL			
Alex	a	1.79 ±0.96	116.80±14.6	1.79 ±0.96	116.80±14.6	94
	b	1.79 ±0.96	116.80±14.6			
Tanta	a	0.53 ±0.17	< DL	0.53 ±0.17	< DL	78.9
	b	0.53 ±0.17	< DL			
Baniswif	a	0.50 ±0.19	41.55±5.19	0.50 ±0.19	41.55±5.19	94
	b	0.50 ±0.19	41.55±5.19			
Sinai	a	22.0±1.28	73.00±9.12	22.0±1.28	73.00±9.12	77
	b	22.0±1.28	73.00±9.12			
Siwa	a	1.79 ± 0.36	< DL	1.79± 0.36	< DL	83
	b	1.79 ± 0.36	< DL			

DL. Represent the detection limits for $^{228}Ra \simeq 20mBq/L$

Table 2. The Average Committed Effective Doses and Cancer Risk Associated Due to Consumption of Water:

Cities	Annual Effective Committed dose (μSv/a)		The Radiological Risk	
	^{226}Ra	^{228}Ra	^{226}Ra x 10^{-7}	^{228}Ra x 10^{-5}
Cairo	0.34	ND	6.09	ND
El Mansoura	0.24	ND	4.29	ND
Qualuab (Treatment factory)	0.35	36.50	6.20	7.42
Octobar	0.19	ND	3.38	ND
Alex	0.37	58.80	6.57	11.95
Tanta	0.11	ND	1.94	ND
Baniswif	0.10	20.93	1.83	4.25
Sinai	4.50	36.77	80.70	7.47
Siwa	0.37	ND	3.57	ND

ND. Not Determined

Table 3. Concentrations of ^{226}Ra and ^{228}Ra Isotopes in Drinking Water.

Region / country	Concentration (mBq/ L) in drinking water		Ref.
	Ra-226	Ra-228	
North America **United States**	0.4-1.8	0-0.5	Cothern, C.R. etal., 1983, Fisenne, I.M. et al ., 1987, Holtzman, R.B., 1964 and McCurdy, D.E. et al ., 1981[13,14,15,16]
Asia **Pakistan**	0.2-120		NEPA, 1990, Dang, H.S. et al., 1990 [17,18]
Europe **Finland** **France** **Germany** **Italy** **Poland** **Romania** **Switzerland** **Spain** **U.K.**	 10-49000 7-700 1-1800 0.2-1200 1.7-4.5 1.7-4.5 0-1500 0-1500 0-180	 18-570 0-200	 Asikainen, M.1982 and Salonen,L.1994[19,20] Descamps, B.et al., 1988 and Pellerin, P. et al., 1980[21,22] Bundesministeriumfür Umwelt 1994, Gans, I. 1985 and Glöbel, B. et al., 1980 [23,24,25] Sgorbati, G et al., 1997 [26] Pietrzak-Flis, Z. et al., 1997 and Pietrzak-Flis, Z. et al., 1997 [27,28] Botezatu, E., 1994 and RSRP., 1994 [29,30] SFOPH, 1997 [31] Soto, J. et al., 1988 [32] Bradley, E.J. 1993 [33]
Africa **Egypt**	0.5-22	41-117	This studies
Reference value **UNSCARE**	0.5	0.5	**UNSCARE** [34]

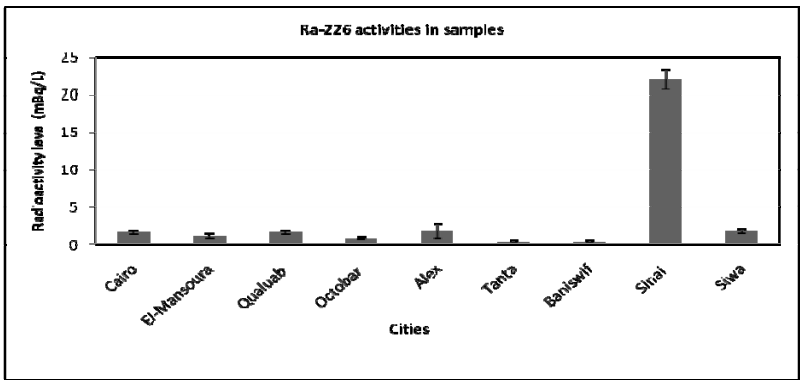

Fig. 1: The distribution of ^{226}Ra activity concentration found in samples collected from different cities in Egypt

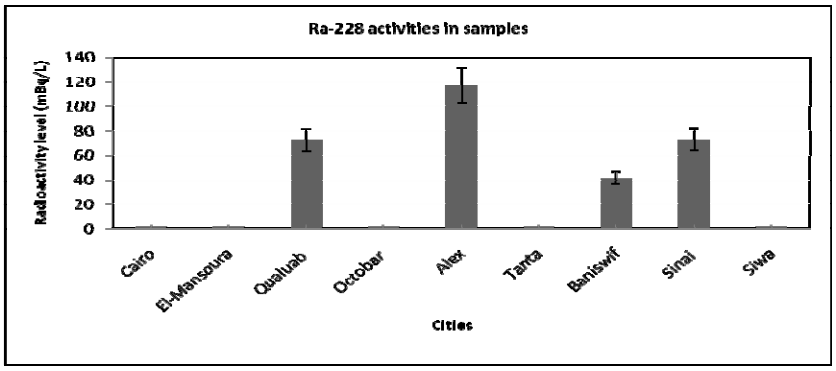

Fig. 2: The distribution of ^{228}Ra activity concentration found in samples collected from different cities in Egypt

CONCLUSION

Concentration of ^{226}Ra and ^{228}Ra in potable water collected from nine Egyptian towns vary over a large range. The highest concentration of Radium isotopes are found in water types originating from wells such as Sinai and Siwa. The radiation doses estimated from natural radium in water are low compared with the average total dose of 2.4 mSv/y from external and internal radiation in EPA and WHO regulation. The average doses for the population are safely below the total indicative dose of 0.1 mSv/y imposed by the EU drinking water directive and WHO recommendation [5]. The risk assessment data show that the radionuclides under this investigation do not pose any significant health risk to the public.

REFERENCES

[1] European Union, council directive 98/83/EC., 3 November 1998. The quality of water intended for human consumption, Official journal L330 (05/12/98).

[2] EURATOM, Commission recommendation, 20 December 2001. The protection of the public against exposures to radon in drinking water supplies, EURATOM 2001/928.

[3] EURATOM, Council Regulation, 18Th 1989. Amending Regulation No. 3954/89 laying down maximum radiological emergency, official journal L 3211 (22/07/89).

[4] WORLED HEALTH ORGANIZATION, 1998. Guidelines for drinking water quality, 2nd edition, addendum to volume 2: Health criteria and other supporting information, Geneva.

[5] WORLED HEALTH ORGANIZATION, 2008. Guidelines for Drinking Water Quality, 3rd edition, incorporating the first and second addenda Volume 1 Recommendations.

[6] Siegurd, M., John, N., Franz, S. 1999. Determination of ^{228}Ra, ^{226}Ra, and ^{210}Pb in drinking water using liquid scintillation counting. US Environmental Protection Agency EPA pages 269-274 federal register Vol. 64, 38, February 26, p 9560.

[7] US. Environmental Protection Agency (EPA), September 1993. External exposure to radionuclides in air, water and soil federal guidance report No. 12.

[8] UNSCEAR 2000."Sources and effects of ionizing radiation, report to the general assembly with scientific annexes". New York: United Nations.

[9] International commission on radiological protection (ICRP), 1996. Age-dependent dose to members of the public from intake of radionuclides: Part 5 Compilation of ingestion and inhalation dose coefficients. ICRP Publication 72, Oxford: Pergamon press.

[10] Nour, S., El-Sharkawy, A., Burnett, W.C., and Horwitz, E.P. Journal of applied radiation and isotopes 61, 1173–1178 (2004).

[11] STUK-A 213. September 2005. U-238 series radionuclide's in finnish ground water-based drinking water and effective doses.

[12] EPA., September 1999. Cancer risk coefficients for environmental exposure to radionuclides, federal guidance report No. 13.

[13] Cothern, C.R., and Lappenbusch, W.L. Journal of Health Phys. Vol. 45, No.1, 89-99 (1983).

[14] Fisenne, I., Perry, P.M. and Decker K.M. et al. The daily intake of $^{234, 235, 238}$U, $^{228, 230, 232}$Th and $^{226, 228}$Ra by New York City residents. Health Phys. 53(4): 357-363 (1987).

[15] Holtzman, R.B. Lead-210 (RaD) and polonium-210 (RaF) in potable waters in Illinois. p. 227-237 in the natural radiation environment (J.A.S. Adams and W.M. Lowder, Eds.). The University of Chicago press, Chicago, (1964).

[16] McCurdy, D.E. and Mellor. R.A. the concentration of ^{226}Ra and ^{228}Ra in domestic and imported bottled waters. Health Phys., 40: 250-253 (1981).

[17] National Environmental Protection Agency (NEPA). Nation wide survey of environmental radioactivity level in China (1983-1990). 90-S315-206. The people's republic of China (1990).

[18] Dang, H.S., Pullat, V.R. and Jaiswal, D.D. et al. Daily intake of uranium by the urban Indian population. J. Radioanal. Nucl. Chem. 138(1): 67-72 (1990).

[19] Asikainen, M., Natural radioactivity of ground water and drinking water in Finland. STL-A39 (1982).

[20] Salonen, L. ^{238}U series radionuclides as a source of increased radioactivity in groundwater originating from Finnish bedrock. p. 71-84 in: Future ground water resources at risk (J. Soukko, Ed.). IAHS Publication No. 222. IAHS Press, Oxford shire (1994).

[21] Descamps, B., and Foulquier. L. Natural radioactivity in the principal constituents of French river ecosystems. J. Radiat. Prot. Dosim. 24(1/4): 143-147 (1988).

[22] Pellerin, P., Gahinet, M.E., and Moroni, J.P. et al. Some observations on natural radioactivity in food in France. p. 331-348 in: Seminar on the radiological burden of man from natural radioactivity in the Countries of the European Communities. CEC Doc. No.V/2408/80 (1980).

[23] Bundesministeriumfür Umwelt, Naturschutz und Reaktorsicherheit. Umweltpolitik, Umweltradioaktivität und Strahlenbelastung. Jahresbericht (1994).

[24] Gans, I. Natural radionuclides in mineral waters. Sci. Total Environ. 45: 93-99 (1985).

[25] Glöbel, B. and Muth H. Natural radioactivity in drinking water, foodstuffs and man in Germany. p. 385-418 in: Seminar on the radiological burden of man from natural radioactivity in the Countries of the European Communities. CEC Doc. No. V/2408/80 (1980).

[26] Sgorbati, G. and Forte. M. Determination of ^{238}U and ^{226}Ra concentrations in drinking waters in Lombardia region, Italy. Communication to UNSCEAR Secretariat (1997).

[27] Pietrzak-Flis, Z., Chrzanowski, E., and Dembinska, S. Intake of ^{226}Ra, ^{210}Pb and ^{210}Po with food in Poland. Sci. Total Environ. 203: 157-165 (1997).

[28] Pietrzak-Flis, Z., Suplinska, M.M. and Rosiak, L. The dietary intake of ^{238}U, ^{234}U, ^{230}Th, ^{232}Th, ^{228}Th and ^{226}Ra from food and drinking water by inhabitants of the Walbrzych region. J. Radioanal. Nucl. Chem. 222(1-2): 189-193 (1997).

[29] Botezatu, E. Contribution of the dietary ingestion to the natural radiation exposure of Romanian population. J. Hyg. Public Health 44(1-2): 19-21 (1994).

[30] Romanian Society for Radiological Protection (RSRP). p. 109-111 in natural radioactivity in Romania, Bucharest. REG Project No. 852/1993 (1994).

[31] Swiss Federal Office of Public Health (SFOPH). Environmental radioactivity and radiation exposure in Switzerland. Bern (1997).

[32] Soto, J., Quindos, L.S. and Diaz-Caneja, N. et al. ^{226}Ra and ^{222}Rn in natural waters in two typical locations in Spain. Radiat. Prot. Dosim. 24 (1/4): 93-95 (1988).

[33] Bradley, E.J. Contract Report. Natural radionuclides in environmental media. NRPB-M 439 (1993).

[34] United Nations. Sources and effects of ionizing radiation. United Nations scientific committee on the effects of atomic radiation, 1993 report to the general assembly, with scientific annexes. United Nations sales publication E.94.IX.2. United Nations, New York, (1993).

[35] Lasheen, Y.F., Seliman, A.F. and Abdel Rassoul, A.A. Simultaneous measurement of ^{226}Ra and ^{228}Ra in natural water by liquid scintillation counting, J. Environ Radioact., 95 (2-3):86-97 (2007).

[36] Environmental Protection Agency (EPA), 1991. Final draft for the drinking water criteria document on radium, U.S. EPA, Washington, DC, TR-1241-85.

1/5/2012