Journal of Radiation and Nuclear Applications
An International Journal

http://dx.doi.org/10.18576/jrna/060112

^{222}Rn Concentrations in the Lebanese Drinking Water and its Associated Ingestion Committed Effective Dose to the Public

Maria Aoun[1], *Hamzeh El Jeaid*[1], *Dima Mansour*[2] *and Omar El Samad*[1*]

[1] Lebanese Atomic Energy Commission, National Council for Scientific Research, P. O. Box 11-8281, Riad El Solh, Beirut, Lebanon

[2] Lebanese University, Faculty of Sciences, Branch 1, P. O. Box 4, Beirut, Lebanon

Received: 13 Oct .2020, Revised: 21 Nov .2020, Accepted: 28 Dec. 2020.
Published online: 1 Jan. 2021.

Abstract: ^{222}Rn activity concentrations of the main drinking water resources in two important Lebanese provinces were measured for the first time, in order to assess the committed effective dose received by the Lebanese public due to the ingestion of ^{222}Rn from drinking water. Measurements were conducted using a professional radon monitor AlphaGUARD PQ 2000PRO. All values were below the United States Environmental Protection Agency and the World Health Organization recommended levels. From the obtained data, the committed effective doses were assessed, values varied from 0.58 to 15.09 µSv.y^{-1}, from 0.35 to 9.03 µSv.y^{-1} and from 0.29 to 7.65 µSv.y^{-1} for babies, children and adults respectively.

Keywords: Radon, drinking water, committed effective dose.

1 Introduction

Exposure to radon gas is one of the major worldwide public concerns related to naturally occurring radioactivity. Radon and its progenies contribute to around 50% of the total effective annual radiation dose received by human from natural sources [1]. Among the natural isotopes of radon, ^{222}Rn which is a radioactive decay product of ^{226}Ra; progeny of the ^{238}U natural decay chain; is the most important radionuclide due to its longest half-life [2]. ^{222}Rn gets into human body through inhaled air and drinking water. In fact, uranium occurs naturally at different levels in rocks and soils; fractions of radon gas emanate in the atmosphere and tend to concentrate in enclosed spaces such as underground mines and houses. Moreover, as radon is soluble in water, groundwater passing through these rocks and soils can contain this element [3]. The different types of waters may contain varying levels of ^{222}Rn activity concentrations which are influenced by several factors such as the geology of the area, the sediments, the streams and the temperature [4, 5]. In general, surface waters may contain low concentrations of radon; for groundwater; the higher concentrations are associated with granitic rocks while variably lower concentration areas are associated with the different types of sedimentary rocks [6, 7].
High levels of radon in drinking waters may lead to potential health risk through ingestion and inhalation; because radon and its progenies are alpha and beta emitters which may cause cancers in human organs [8].
Several authors have mainly assessed the doses and risks resulting from consuming water; on the other hand, countrywide surveys of radon activity in drinking water have received particular attention [9, 10]. However, in Lebanon, there has been no complete study that assesses radon activity in all the Lebanese drinking water. A national project aiming to evaluate the internal dose received by the Lebanese population through drinking water has been established in Lebanon by the Environmental Radiation Control Department at the Lebanese Atomic Energy Commission (LAEC). In this study, samples were collected from two important Lebanese provinces: the North and the Beqaa, in order to evaluate for the first time the radon activity concentrations in the main drinking water sources of these provinces in order to estimate the age-dependent annual effective doses due to ingestion of radon from drinking water and the contribution of radon to public exposure.

2 Materials and Methods

2.1 Study Area

Located on the eastern coast of the Mediterranean sea, Lebanon covers a total surface area of 10 452 km^2. In term of geology, Lebanon is mainly composed of carbonate platform sedimentary rocks deposited since Paleozoic

[*]Corresponding author e-mail: o.samad@laec-cnrs.gov.lb

times; the sedimentary sequence is made up predominantly of marine carbonate rocks ranging in age from early Jurassic to recent. General drinking water quality control has been taken into consideration in several studies while little assess the radiological aspect of drinking water in Lebanon. Springs and groundwater are by far the main sources for drinking water supply; the nature of bedrock may have a significant influence on the level of dissolved radon in these waters. Related to the climate, winter season in Lebanon is the wet period allowing groundwater recharges while the summer season is totally dry so discharged water may contribute to higher levels of dissolved radon in groundwater.

In our study, as springs and wells are the major sources of drinking water in the North and the Beqaa provinces of Lebanon, water samples were collected from the main wells and springs of these two areas during the period 2016-2017 (Figure 1). Sample locations of the analyzed water samples and their different lithologies are presented in Table 1.

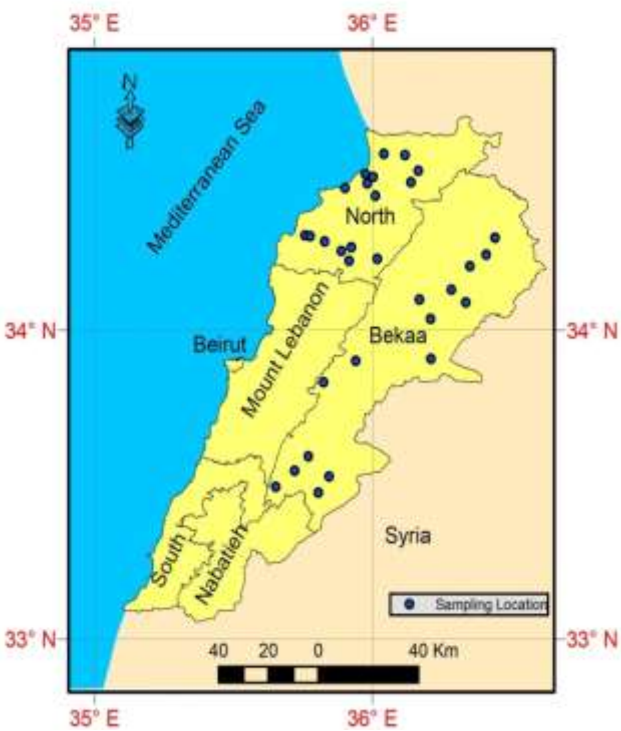

Fig. 1: Map of the sampling locations.

2.2 Sampling and Measurement Method

Springs and wells water samples were collected in 500 mL polyethylene bottles completely filled and tightly closed to prevent air entry and radon gas loss; the samples were transported to the laboratory of alpha- beta spectrometry of the Environmental Radiation Control Department at the Lebanese Atomic Energy Commission with a minimum delay in order to measure the radon activity concentrations at earliest possible time.

^{222}Rn activity concentrations were performed using a professional radon monitor AlphaGUARD PQ 2000PRO (Genitron-Saphymo, Germany); an ionization chamber designed for the measurement of radon in air, soil and water. A polyethylene–encapsulated ^{226}Ra/^{222}Rn emanation, NIST standard reference material SRM 4973, was used to check the calibration of the radon counting system. In fact, AlphaGUARD is suitable for continuous monitoring of radon concentrations with a measurement range of 2-20000000 Bq.m^{-3} with a sensitivity of 5 cpm for 100 Bq.m^{-3}. For radon in water samples, Aquakit equipment was added to the device. The measuring of radon in water samples consist of expelling radon from the water in a closed gas cycle, and collecting all drops into a security vessel during the degassing process [11]. Before every sample measurement, the background of empty set-up was measured. After water injection, AlphaGUARD and AlphaPUMP were switched on for 10 min, then the pump was switched off and the the AlphaGUARD remained switched on for another 20 min to continue the radon measurement. Three cycles of measurements were done for each sample in order to obtain better precision. The lower limit of detection is 0.1 Bq.L^{-1}. Taking into consideration the dilution of radon and its remaining quantity in the aqueous phase, the radon concentration in water samples was obtained from the following equation [11, 12]:

$$C_{water} = \frac{C_{air} \times \left(\frac{V_{system} - V_{sample}}{V_{sample}} + K(T)\right) - C_0}{1000} \quad (1)$$

Where C_{water} is the radon concentration in the water sample (Bq.L^{-1}), C_{air} is the radon concentration in the measurement system (Bq.m^{-3}), C_0 is the radon background concentration in the system, V_{system} is the inner volume of the measurement set-up (mL), V_{sample} is the volume of water sample (mL) and K(T) is partition coefficient which depends on the temperature of the water sample.

3 Results and Discussion

3.1 Radon in spring and Well Water

The activity concentrations of ^{222}Rn in the drinking water samples collected from the different locations of the present study are presented in Figure 2. Results showed that the dissolved radon activity concentrations varied over a wide range, from less than 0.1 to 4.37 Bq.L^{-1} for the 31 studied sites (wells and springs), with an average of 1.17 Bq.L^{-1}. For natural springs, the values of ^{222}Rn concentrations ranged from less than 0.1 to 3.55 Bq.L^{-1}, with an average activity of 1.14 Bq.L^{-1}, while they ranged from less than 0.1 to 4.37 Bq.L^{-1} with an average of 1.25 Bq.L^{-1} in the well water samples. One main reason for this variation in the studied samples could be attributed to the geology of the aquifers. In general, a high concentration of radon may occur due to uranium mineralization in granite and quartzite rocks, or due to the presence of faults and shears which

Table 1: Location and geology of the collected water samples.

Sample number	Water type	Coordinates	Rock Types
S1	Spring	34˚11'47 N 36˚21'04 E	Pudding stones- limestone
S2	Spring	34˚00'10 N 36˚13'19 E	Pudding stones- limestone- Marl
S3	Spring	34˚17'90 N 36˚42'06 E	Limestone
S4	Spring	34˚14'61 N 36˚47'14 E	Limestone
S5	Spring	34˚38'96 N 36˚37'09 E	Pudding stones
S6	Spring	34˚14'32 N 36˚24'17 E	Limestone- Marl
S7	Spring	33˚29'33 N 35˚48'43 E	Coastal sand, green marls-black dolomites-sandy clay soils, limestone
S8	Spring	33˚27'05 N 35˚46'54 E	Marls-Black dolomites
S9	Spring	33˚52'48 N 35˚52'03 E	Limestone
S10	Spring	33˚33'53 N 35˚43'27 E	Marl
S11	Spring	33˚29'53 N 35˚38'29 E	Marl-Black dolomites
S12	Spring	33˚36'05 N 35˚46'06 E	Marl-Dolomites
S13	Spring	34˚30'20 N 36˚08'37 E	Sandstone- Clay
S14	Spring	34˚53'04 N 36˚03'81 E	Arable land
S15	Spring	34˚14'57 N 36˚00'37 E	Limestone, little stones, flow molten lava
S16	Spring	34˚14'56 N 35˚53'47 E	Limestone
S17	Spring	34˚15'46 N 35˚52'42 E	Limestone
S18	Spring	34˚17'05 N 35˚52'52 E	Limestone, clay, sand, alluvium
S19	Spring	34˚38'85 N 35˚93'02 E	Limestone
S20	Spring	34˚39'94 N 35˚92'04 E	Limestone, clay, sand, alluvium
S21	Spring	34˚33'23 N 35˚83'88 E	Pudding stones, alluvium
S22	Spring	27˚94'10 N 34˚54'14 E	Pudding stones, clay, marl, limestones
W1	Well	33˚95'83 N 36˚16'65 E	Limestone
W2	Well	33˚54'02 N 35˚59'59 E	Limestone- marl pudding stones
W3	Well	33˚82'11 N 35˚88'24 E	Arable land
W4	Well	34˚27'37 N 36˚07'33 E	Little stone- flow molten lava
W5	Well	34˚25'41 N 35˚49'53 E	Arable land
W6	Well	34˚18'08 N 35˚64'45 E	Marl, limestone
W7	Well	34˚15'36 N 35˚52'42 E	Limestone
W8	Well	34˚17'07 N 35˚49'36 E	Limestone, marl
W9	Well	34˚27'49 N 35˚57'21 E	Arable land, clay, marl, limestone

facilitate the migration of radon gas [6]. In Table 1 is shown that the dominant rocks in the majority of the studied sampling site are of the limestones, green marl and black dolomite types, these sedimentary rocks are based on calcite and are generally low in uranium, which decays radon gas [13]. The geology of the sampling sites (S5 and S18) containing respectively pudding stones and alluvium may have influenced their radon concentrations, because these two types of rocks contain higher concentrations of uranium [13, 14] which causes the increase in emanation of radon. Likewise, several other parameters can influence the variation of radon activities, such as the depth of the well, the temperature and the porosity [15], this could be the case in the sampling site (W8) where the dominant rocks are low in uranium.

All the investigated water samples presented an activity concentration of radon below the conservative value of 11.1 Bq.L^{-1}, set by the United States Environmental Protection Agency (USEPA), as a safe limit of radon activity in water and also below 100 Bq.L^{-1}; limit recommended by the World health Organization (WHO) as a safe limit of radon activity in water for drinking purposes [16, 17].

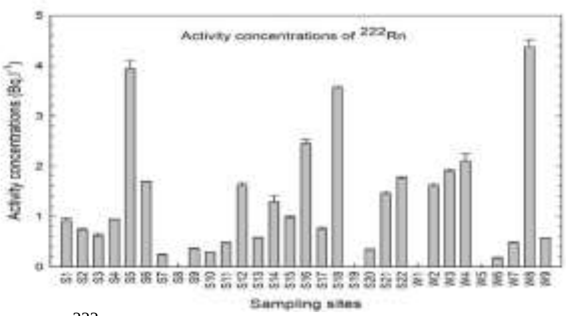

Fig.2: ^{222}Rn activity concentration of the different collected drinking water samples.

Turkey [4, 20] and lower than that reported in the groundwater of other places of the world [9, 18, 19, 21, 22, 24, 25, 26, 27], which may be due to the sedimentary rocks of their corresponding aquifers.

The overall values of measured radon concentrations in water are comparable to those obtained in a previous study performed in Lebanon by Abdallah et al. by having most of the locations at a very low concentrations [22]. However, areas which had hugely higher values of radon concentrations in Abdallah et al. study due to their locations near the convergence of active fault systems are absent in our areas of study. In fact, faults in tectonic plates constitute a pathway for radon emanation [27].

The radon concentrations of the collected spring water samples in the present study are similar to those reported in some regions of turkey [4, 20]. On the other hand spring well waters can have higher radon concentrations up to 129.3 Bq.L^{-1} (Romania) [27] and even up to 576 Bq.L^{-1} (Venezuala) [21].

The radon concentrations of well waters observed in the present study are compatible with those reported from Jordan [28], and Saudi Arabia [23]. On the other hand, radon concentrations were higher in Turkey 2 Bq.L^{-1} to 20.8 Bq.L^{-1}[6] and much more elevated in Syria and Mexico up to 28.4 Bq.L^{-1} and up to 39.75 Bq.L^{-1} respectively; [26] in his study in Sudan has found that radon concentration in well water was varying from 1.58 Bq.L^{-1} to 345.1 Bq.L^{-1} [26].

Table 2. ^{222}Rn activities in groundwater worldwide.

Water type	Concentration values (Bq.L^{-1})	Location	References
Spring	2.30 - 52.70 0.7 - 59.7 0.39 - 19.21 0.39-1.17 0.13 - 1.20 0.1-576 0.49-49.6	Italy Turkey Turkey Turkey Turkey Venezuela Lebanon	[18] [6] [19] [20] [4] [21] [22]
Well	2 - 20.8 0.34 - 7.21 41-127 1.78-39.75 7.5-28.4 1.58-345.1	Turkey Saudi Arabia China Mexico Syria Khartoum	[6] [23] [9] [24] [25] [26]
Spring well	2 - 129.3 0.6 - 112	Romania	[27]
Spring Well	3.3 - 10.7 3.1 - 5.7	Jordan	[28]
Well Spring	< 0.1 - 4.37 < 0.1 - 3.94	Lebanon-Beqaa province	Present study
Well Spring	< 0.1 - 2.10 < 0.1 - 3.55	Lebanon-North province	

3.2 Estimation of Ddoses by Ingestion

From the measured ^{222}Rn activity concentrations in the studied water samples, the radiological impact due to ingestion of dissolved radon in drinking water has been estimated by the calculation of the effective dose received by the population through the consumption of water in the areas of study. The annual effective dose for an individual due to intake of radon from drinking water is estimated using the following relationship [1]:

$$D_{ing}(\mu Sv/y) = C_{Rn} \times I_A \times D_f \qquad [1]$$

Where C_{Rn} is the radon concentration of ingested water in Bq.L^{-1}, I_A is the annual intake of drinking water and D_f is the dose conversion factor. For the age-dependent dose calculations I_A is considered 150, 350 and 500 $L.y^{-1}$ for babies (1–2 y old), children (8–12 y old) and adults (>17 y old) respectively and D_f for ^{222}Rn 23, 5.9 and 3.5 $nSv.y^{-1}$ for babies, children and adults, respectively [1].

The results presented in Figure 3 shows that the committed effective doses due to ingestion of ^{222}Rn in spring and well waters varied from 0.58 to 15.09 $\mu Sv.y^{-1}$for babies, from 0.35 to 9.03 $\mu Sv.y^{-1}$ for children and from 0.29 to 7.65 $\mu Sv.y^{-1}$ for adults. The effective doses from the consumption of randomly chosen water among the total of 31 sources, were 4.00 $\mu Sv.y^{-1}$ for babies, 2.42 $\mu Sv.y^{-1}$ for children and 2.05 $\mu Sv.y^{-1}$ for adults. On the other hand, the mean annual effective doses were 3.92, 2.35 and 1.99 $\mu Sv.y^{-1}$ in spring waters for babies, children and adults, respectively and 4.32, 2.58 and 2.19 $\mu Sv.y^{-1}$ in well waters.

Considering the Lebanese adult daily intake rate of drinking water 985.9 $g.d^{-1}$ in 2008 [29], the mean annual effective doses were 1.43 $\mu Sv.y^{-1}$ and 1.58 $\mu Sv.y^{-1}$ due to spring and well water consumption respectively.

As it is noticed from the obtained results that the annual committed effective doses due to ^{222}Rn in drinking water calculated for the different age groups are far below 100 $\mu Sv.y^{-1}$ the WHO recommended limit for drinking water.

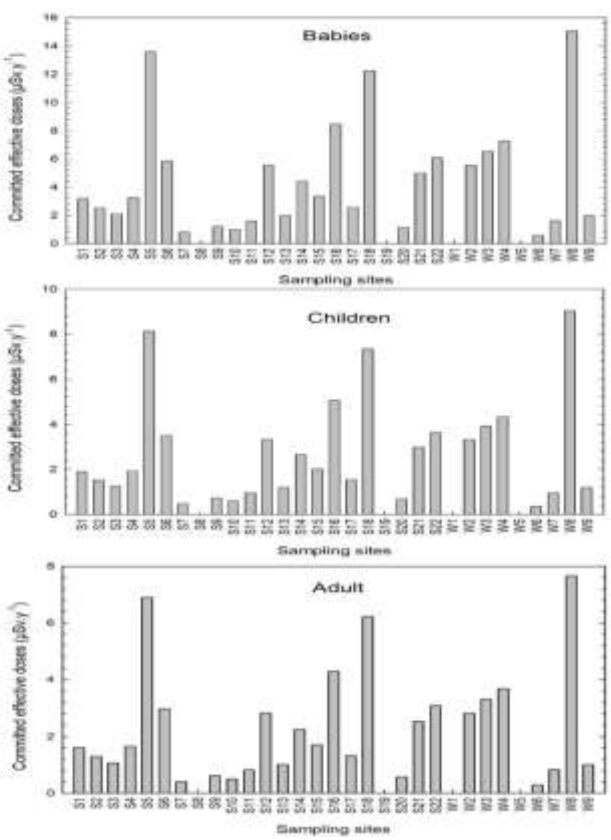

Fig. 3: Estimated age-dependent committed effective doses of spring and well waters.

4 Conclusions

In this study, the measurement of the radon concentration level of the main drinking water resources in the North and Bekaa governorates of Lebanon was carried out for the first time in order to estimate the annual effective dose resulting from the ingestion of water. The ^{222}Rn concentrations in all the groundwater samples were found to be lower or similar as compared with worldwide levels obtained in several studies, which may be attributed to the geology of the region. The age - dependent committed effective doses due to the ingestion of ^{222}Rn from the ingestion of drinking water were below 100 $\mu Sv.\ y^{-1}$, the WHO reference value for drinking water for radon, hence the groundwater of the areas investigated are safe for drinking.

Acknowledgements

This work was supported by the Grant Research program (GRP) of the Lebanese National Council for Scientific Research CNRSL. Project number: 02-05-16.

References

[1] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources, effects and risks of ionizing radiation. UNSCEAR Report. United Nations Publication., 2000.

[2] United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources and effects of ionizing radiation. UNSCEAR Report. United Nations Publication., 1993.

[3] Shilpa G.M., Anandaram B.N., Mohankumari T.L. Measurement of 222Rn concentration in drinking water in the environs of Thirthahalli taluk, Karnataka, India. Journal of Radiation Research and Applied Sciences., **10 (3)**, 262-368(2017).

[4] Yigitoglu I., Öner F., Yalim H.A., Akkurt A., Okur A., Ozkan A. Radon concentrations in water in the region of Tokat city in Turkey. Radiation Protection Dosimetry., **142**, 358-362(2010).

[5] Al-Masri M.S., Blackburn R. Radon-222 and related activities in surface waters of the English Lake District. Applied Radiation Isotopes., **50**, 1137-1143(1999).

[6] Yakut H., Tabar E., Zenginerler Z., Demirci N., Ertugral F. Measurement of (Rn)-222 concentration in drinking water in Sakarya, Turkey. Radiation Protection Dosimetry., **157**, 397-406(2013).

[7] Ali N., Khan E.U., Akhter P., Khan., Waheed A. Estimation of mean annual effective dose through radon concentration in the water and indoor air of Islamabad and Murree. Radiation Protection Dosimetry., **141**, 183-191(2010).

[8] Lee J., Kim G. A simple and rapid method for analyzing radon in coastal and ground waters using a radon-in-air monitor. Journal of Environmental Radioactivity., **89**, 219-228(2006).

[9] Xinwei L. Analysis of radon concentration in drinking water in Baoji (China) and the associated health effects. Radiation Protection Dosimetry., **121**, 452–455(2006).

[10] Todorovic N., Nikolov J., Forkapic S., Bikit I., Mrdja I., Krmar M., Veskovic M. Public exposure to radon in drinking water in Serbia. Applied Radiation and Isotopes .,**70**, 543–549(2012).

[11] Genitron Instruments GmbH, Aquakit manual., Germany 09/97,1999.

[12] Schubert M., Buerkin W., Pena P., Lopez A.E., Balcazar M. On-site determination of the radon concentration in water samples: methodical background and results from laboratory studies and a field-scale test. Radiation Measurements., **41**, 492–497(2006).

[13] Kenneth G.B. Uranium in carbonate rocks. Geological survey professional paper 474 –A, 1963.

[14] Taylor R.D., Andersson E.D. Quartz-Pebble-Conglomerate Gold Deposits, Chapter P of Mineral Deposit Models for Resource Assessment. USGS Scientific Investigations Report., 2010-5070, 2018

[15] A El-Taher and HA Madkour Environmental and radio-ecological studies on shallow marine sediments from harbour areas along the Red Sea coast of Egypt for identification of anthropogenic impacts Isotopes in Environmental and Health Studies., **50(1)**, 120-133(2014).

[16] World Health Organization (WHO). Guidelines for Third Edition Recommendations Drinking-water Quality, **1**, 2008.

[17] United States Environmental Protection Agency (USEPA), National primary drinking water regulations for radionuclides. EPA/570/9–91/700, (1991).

[18] D'Alessandro W., Vita F. Groundwater radon measurements in the Mt. Etna area. Journal of Environmental Radioactivity., **65**, 187–201(2003).

[19] Yalcin S., Gurler O., Akar U.T., Incirci F., Kaynak G., Gundogdu O. Measurements of radon concentration in drinking water sample from Kastamonu (Turkey). Isotopes in Environmental and Health Studies., **47(4)**, 438-445(2011).

[20] Oner F., Yalim H.A., Akkurt A., Orbay M. The measurements of radon concentrations in drinking water and the Yesılırmak river water in the area of Amasya in Turkey. Radiation Protection Dosimetry **133**, 223–226(2009).

[21] Horvath A., Bohus L.O., Urbani F., Marx G., Piroth A., Greaves E.D.Radon concentrations in hot spring waters in northern Venezuela. Journal of Environmental Radioactivity., **47**, 127–133(2000).

[22] Abdallah S.M., Habib R.R., Nuwayhida R.Y., Chatilac M., Katuld G. Radon measurements in well and springwater in Lebanon. Radiation Measurements., **42**, 298–303(2007).

[23] A El-Taher, F Alshahri, and R Elsaman Environmental impacts of heavy metals, rare earth elements and natural radionuclides in marine sediment from Ras Tanura, Saudi Arabia along the Arabian Gulf Applied Radiation and Isotopes., **132**, 95-104 (2018).

[24] A El-Taher, HMH Zakaly and R Elsaman Environmental implications and spatial distribution of natural radionuclides and heavy metals in sediments from four harbours in the Egyptian Red Sea coast Applied Radiation and Isotopes., **131**, 13-22(2018).

[25] WM Badawy, A El-Taher, MV Frontasyeva, HA Madkour and AEM Khater Assessment of anthropogenic and geogenic impacts on marine sediments along the coastal areas of Egyptian Red Sea Applied Radiation and Isotopes., **140**, 314-326(2018).

[26] A El-Taher, WM Badawy, AEM Khater, HA Madkour Distribution patterns of natural radionuclides and rare earth elements in marine sediments from the Red Sea, Egypt Applied Radiation and Isotopes., **151**, 171-181 (2019).

[27] Cosma C., Moldovan M., Dicu T. Radon in water from Transylvania (Romania). Radiation Measurements., **43**, 1423–1428(2008).

[28] Kullab M. Assessment of radon-222 concentrations in buildings, building materials, water and soil in Jordan. Applied Radiation and Isotopes., **62**, 765–773(2005)

[29] Nasreddine L., El Samad O., Hwalla N., Baydoun R., Hamzé M., Parent-Massin D. Activity concentrations and mean annual effective dose from gamma emitting radionuclides in the Lebanese diet. Radiation Protection Dosimetry., **131**, 545-550(2008).