ENVIRONMENTAL RADIATION MONITORING SURVEY IN LEBANON

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ABSTRACT

This work is part of a national environmental monitoring program. The objective is to have an Environmental Indicator by monitoring the long-term behavior of radionuclides and to provide a baseline in the event of radiological incident. The techniques cover the monitoring of radioactivity in land and costal areas, as well as, the collection and preparation of terrestrial samples such as soil, food, vegetables... and daily measurement of dose rates in air in different locations. The paper provides an overview of the current situation in respect to regulatory actions and technical development of radiation protection in relation to Natural Occurring Radionuclides Measurements (NORM) in Lebanon. Dose rates in air were carried out using portable beta and gamma detectors while for the other samples gamma spectroscopy methods were used. Concentrations of uranium, thorium series and potassium were quantified and the results presented consist of one-year monitoring.

Keywords: environment, NORM, Lebanon, monitoring, radioactivity

INTRODUCTION

Naturally occurring or industrially processed, the concentration of radionuclides in the environment must be continuously controlled, and data must be regularly collected and reviewed. The need for such environmental radioactivity measurements, in the frame of the national Lebanese monitoring program, appears at different levels. First, the collected data constitute the baseline, a reference level of radioactivity within the Lebanese territory and diet. Second, scientific measurements allow one to detect any abnormal level, any occurring contamination in accidental cases of radionuclides release and migration. Accordingly, valuable countermeasures and emergency strategies will be easily implemented and applied.

In fact, it is well known that the dose of radioactivity to which humans are exposed depends directly on the level of the radioactivity in the environment they live in and the food they consume. Such exposure dose originates from natural radionuclides whose primordial sources include uranium, thorium series and potassium; additionally to artificial radionuclides where the amount of cesium present in fallout is of particular interest. So, parallel to large amounts of worldwide environmental monitoring research results published, in the following sections, evaluation of the activity concentrations in Bq/kg of gamma emitting radionuclides calculated for food, water soil and sediment using high-resolution gamma spectrometer with HPGe Co-axial detectors are presented (Tables 3, 4 and 5). In this study, air was monitored daily using direct detection instruments. Gamma spectrometry analyses were performed on

soil samples covering four Lebanese provinces, on coastal samples (sediment and surface water samples) picked up from two of the largest Lebanese rivers, as well as, on typical food samples chosen from the Lebanese diet such as fresh milk, dairy products, some kind of fruits and vegetables and few kinds of conserved and industrial food.

MATERIALS AND METHODS

Sample collection and preparation

Soil samples were collected from 17 uncultivated sites in 4 Lebanese provinces (North, South, Mount Lebanon and Bekaa). The altitude, longitude and latitude parameters of the different locations were obtained using a Global Positioning System (GPS-2000XL) (see Table 1). All the samples were collected within a 25cm x 30cm surface using a stainless steel template, and at a depth of 3 cm.

TABLE 1
Soil Samples Locations and Identification

Location	Altitude (m)	Latitude	Longitude
North Lebanon			
Jabal Moustafa Ali	757	34 24.33 N	36 00.00 E
Sayyelet Al Raes	741	34 24.83 N	36 01.24 E
Assoun- Izal	1027	34 23.23 N	36 00.21 E
Akkar- the lac	368	34 36.10 N	36 12.18 E
South Lebanon			
Saida -Sharhabil	168	33 34.40 N	35 24.26 E
Adloun	229	33 23.87 N	35 16.59 E
Maaroub	230	33 17.10 N	35 20.10 E
Kana road	120	33 14.15 N	35 15.15 E
Mount Lebanon			
Ballouneh	504	33 57.28 N	35 39.79 E
Ayoun Al Siman	1228	33 59.56 N	35 44.97 E
Chehim - Marj Ali	780	33 37.52 N	35 29.57 E
Chweifat	304	33 48.03 N	35 31.56 E
Al Jiyeh	210	33 38.76 N	35 25.55 E
Bekaa			
Al - Kaa	665	34 24.41 N	36 27.11 E
Al - Hermel	760	34 22.41 N	36 25.07 E
Al- Fakha	1000	34 12.24 N	36 21.79 E

The monitoring program covers also 2 Lebanese rivers, one in Mount Lebanon, Ibrahim river and the second in the Bekaa, el Assi river where sediment and water samples were collected at different locations through the river flume (Table 2).

A total of 24 food samples were collected representing the main constituents of the Lebanese diet, such as fresh milk, dairy products, fruits and vegetables, preserves and industrial food.

All samples were prepared and filled in fixed counting geometry $500\,\mathrm{ml}$ polyethylene containers. The soil and sediment samples were grinded and homogenized then dried at $80\,\mathrm{^{0}C}$ for 24 hours. The dry-wet ratio was calculated for each sample. The water samples were acidified then 20 litres were evaporated to get $500\,\mathrm{ml}$ and the food samples were homogenized and measured as they are without any previous treatment.

To complete the national monitoring program, daily monitoring of air was conducted using portable beta and gamma detectors at four different locations in the Lebanese territory (two in Beirut, one in the North and one in the Bekaa).

TABLE 2

Location of Surface Water and Sediments Samples

Location	Altitude (m)	Latitude	Longitude		
Ibrahim river-Beirut					
Estuary	5	34 03.60 N	35 38.92 E		
Downstream	250	34 04.64 N	35 43.47 E		
Assi river- Bekaa					
Al-Shawagheer	602	34 25.28 N	36 27.44 E		
Nabea Al-Zarka	710	34 21.16 N	36 22.41 E		

Measurements

The measurements were carried out using two sets of gamma spectrometry systems from Canberra, equipped with two high purity P-type coaxial germanium detectors (HPGe) with high resolution (1.85 and 2.0 keV at 1332 keV respectively) and relative efficiency of 30% and 40% respectively. The detectors were surrounded with a 10 cm thick lead shield in order to reduce the background and by a 0.5 cm copper layer to attenuate X-rays emitted by the lead shield. The detectors were connected to standard electronics and the spectra were accumulated in 8K MCA (integrated data processor 1510 with S100 MCA band a desktop inspector from Canberra).

The detectors were energy calibrated using a multigamma standard source prepared in the same geometry as the analyzed samples. The efficiency calibration was performed and the curves were obtained by fitting the experimental efficiencies for each sample density. Efficiency curves were corrected for attenuation and absorption. The linearity of the detectors was checked using Eu-152 point source.

For quality assurance, certified reference materials milk powder, soil and water were used in the same counting geometry as the samples.

The background spectra were frequently measured under the same conditions as the samples measurements and were used to correct the calculated sample activities.

Due to the low activity concentrations in the measured samples especially for natural radionuclides, the average counting time ranged between 36 and 48 hours for each sample, to ensure good statistical significance.

The spectra were analyzed off-line using Genie 2000 software from Canberra including peak search, nuclide identification, and activity and uncertainty calculation modules.

Activity concentrations were expressed in Bq/kg dry or wet weight depending on the sample type.

The uncertainty reported is the combined uncertainty calculated using error propagation law and at 95% confidence level, based on the relative standard uncertainties of the sample mass, the net peak area, the full energy peak efficiency, the half-life of the radionuclide of interest and the emission probability.

RESULTS AND DISCUSSIONS

Soil samples

The results of the gamma spectrometry analysis for soil samples brought from different regions in the Lebanese provinces are presented in Table 3.

Table 3 shows the presence of Cs-137 in all the samples analysed. The Cs-137 is the only artificial radionuclide measured in the Lebanese soil. The activity concentration varies from 11 to 117 Bq/kg. These values are in accordance with previous work (El Samad *et al.* 2007) and lower than those reported in Syria (Al-Rayyes & Mamish, 1999).

Due to the short half-life of Cs-134 (2.06 years) its activity concentration was below the minimum detectable activity (MDA=0.2 Bq/kg) calculated using the Curie formula (Genie 2000). Values of U-series, Th-series and K-40 are comparable to the world average values reported in UNSCEAR 2000 which are respectively 33 Bq/kg, 45 Bq/kg and 420 Bq/kg.

K-40 is the most abundant natural radionuclide; its activity concentration in most soil samples varies from 110 to 513 Bq/kg.

The activity concentrations of Ra-226, Bi-214, Pb-214 were assumed to represent the activity of their parent U-238, while those of Ac-228, Bi-212 and Pb-212 were assumed to represent the activity of their parent Th-232 (Table 3). From these values, the secular equilibrium for the uranium and thorium series are respected, taking into account the branching ratio.

Sediment and surface water samples

Water has the ability to transport, dilute and disperse radioactivity. Hence part of the drift, contamination will go down the bottom and will affect sediments. The activity concentrations of K-40 and Cs-137 in surface water samples in the monitored rivers were below the minimum detectable activities 2 Bq/kg and 0.2 Bq/kg respectively. The peaks of the radionuclides representing the natural U-238 series and Th-232 series were identical to the

peaks present in a background spectrum; this can be clearly seen from comparison between the corresponding spectra. Concerning the activity concentration of the radionuclides resulting from the decay of uranium in the sediment samples collected from Assi River, it is less than the activity concentration obtained for Ibrahim River; and it is two times less for K-40. This can be explained according to the level of concentration of pollutants and wastes deposited or poured into the river. Comparable results are obtained for the Th-232 series (Table 4).

Food monitoring

In order to complete the monitoring program, some of the Lebanese diet's ingredients have been analyzed. The results of the activity concentration of K-40 and Cs-137 are reported in Table 5.

For Cs-137 most of food samples (vegetables, fruits, drinking water, and fresh milk) present an activity well below the minimum detectable activity (MDA) of 0.2 Bq/kg defined according to Curie's formula. In contrast, some manufactured food products exhibit a Cesium activity in the same order of magnitude as MDA value for example jams, nuts, coffee, tea and a sample of milk powder.

For jams, the observed 10 Bq/kg Cs-137 activity is essentially caused and due to the high absorption of radionuclides and chemicals present in the Lebanese soil and which characterizes what is commonly called "red fruits". The activity of $0.77~\mathrm{Bq/kg}$ for one of the two milk powder samples depends on the level of Cs-137 activity concentration in the country from which it is imported.

In nuts and coffee, the activity concentration of Cs-137 was very close to the MDA, 0.25 Bq/kg and 0.24 Bq/kg respectively.

The maximum permissible levels of Cs-137 measured in foodstuffs (with the exception of infants food) are 50 Bq/Kg as specified by the regulatory Authority thus the activity concentrations determined in this monitoring are negligible and do not implicate any public health risk.

The K-40 is the most abundant natural radionuclide present in food samples; its activity concentration varies from 5 Bq/kg to 686 Bq/kg depending on the kind and nature of the sample.

The activity concentration of K-40 detected in water samples was below the minimum detectable activity (MDA) estimated to 2.4 Bq/kg. The drinking water sample picked up from Saida has an activity of the order of 2.8 Bq/kg slightly higher than the water MDA value. The highest activity concentration of K-40 is recorded for the coffee and tea samples. This can be understood since the beans in coffee and tea plants are known to selectively take up and concentrate potassium within their cell tissues and therefore are more radioactive than other plants (Scheibel *et al.*, 2007).

The values of activity concentrations of Cs-137 and K-40 are in accordance with those reported previously by Nasreddine *et al.* (2008).

The analysis of vegetables samples (lettuce, parsley) present some traces of beryllium Be-7. These detected traces do not indicate any possible contamination since this is a natural radionuclide whose main source is the set of cosmic rays and hence does not contribute to exposure dose assessment.

Air monitoring

Since inhalation and digestion are the major exposure pathway to the large public (BSS No.115, IAEA), monitoring of air is of a particular importance. In this purpose, air was monitored daily at two sites in Beirut: at the Lebanese Atomic Energy Commission building and at Beirut port.

At the LAEC building, gamma count rate was measured using Novelec DG5A with NaI detector ($3^{"} \times 3^{"}$) and beta count rate was measured using portable Delta 5B with GM tube detector and the gamma dose rate was measured using GREATZ unit.

The readings were recorded over one year and presented according to their corresponding season in graphs 1 to 4 of Figure 1. The maximum recorded dose rate value was 130 nSv/h in spring and summer of 2007-2008. The minimum detectable dose rate value of 40 nSv/h has been noted in winter. Along the entire year, the mean gamma dose rate was around 90 nSv/h.

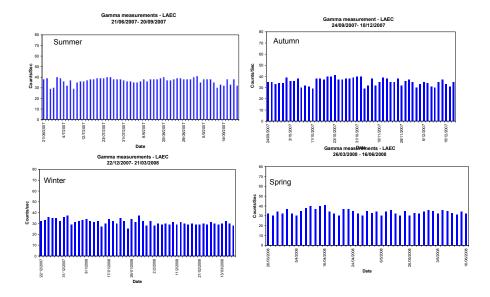


Figure 1. Air gamma measurement over one year.

TABLE 3

Activity Concentrations of K-40, Cs-137, U-235, U-238 Series and Th-232 Series in Soil Samples

Location	K-40 (Bq/kg)	Cs-137 (Bq/kg)	U-235 (Bq/kg)	U-238 series (Bq/kg)		Th-232 series (Bq/kg)			
				Ra-226	Bi-214	Pb-214	Ac-228	Bi-212	Pb-212
Jabal Moustafa Ali	224 ± 7	27 ± 0.6		129 ± 5.4	41 ± 1	44 ± 1	42.4 ±1.2	24.7 ± 1.3	40 ± 1
Sayyelet Al-Raes	237 ± 11	42.1±1.4	2.81 ± 0.74	122 ± 14	48 ± 2	48.5 ± 0.3	45 ± 2	27 ± 4	45 ± 2
Assoun- Izal	435 ± 14	21.1 ± 0.7	3.7 ± 0.9	165 ± 8	56 ± 2	61 ± 2	32 ± 1	19 ± 1	32 ± 1
Akkar-The lac	200 ± 7	23.4 ±0.8	2.1 ± 0.2	35 ± 3	11.1±0.5	11.8 ± 0.5	16.3 ±0.7	9.58±0.92	15.6±0.4
Saida- Sharhabil		23.0 ± 0.7	3 ± 0.1	50 ± 2	21.5±0.7	24 ± 1	10.0 ±0.3	6.0 ± 0.2	11.0±0.3
Adloun	182 ± 7	28 ± 1	2.1 ± 0.5		15 ± 1	16 ± 1	26 ± 1	15 ± 1	27 ± 1
Maaroub	110 ± 5	45 ± 1		40 ± 10	14.4±0.7	15 ± 2.0	14 ± 1	9.2 ± 0.5	14 ± 2
Kana Road	74 ± 3	20.7±0.7	2.0±0.5		10.4±3.0	11.6 ± 0.58	8.2 ± 0.5	7.6±0.4	8.3±0.4
Ballouneh	27.1±2.5	28±1		36.4±12.5	15.2±1.3	17 ± 1	4.7±0.4	3.2±0.4	3.5±0.6
Ayoun Al Siman	200.0±8.3	117 ± 4		108 ± 9	33 ± 6	37 ± 1	38.0 ± 1.5	30 ± 2	33 ± 1
Bhamdoun	513 ± 16	33 ± 1	4 ± 0.4	163 ± 6	64 ± 2	71.5 ± 2.2	16.5 ± 0.8	17.5 ± 0.5	21 ± 0.6
Chehim- Marj Ali	112 ± 5	69 ± 4		37.3 ± 5.6	11.8 ± 5.3	12.4 ± 5.4	14 ± 5	11.3 ± 1.5	15.5±1.3
Chweifat	52.3 ±2	42.1 ± 1.3		14 ± 1.6					
Aljiyeh	215 ± 19	14.4 ± 1.3	4 ± 0.7	173 ± 16	69 ± 6	70 ± 7	50.4 ± 4.4	31±3	50.0±4.3
Al-Kaa	400± 10	36 ± 1	1.4 ± 0.2	50.4 ± 1.7	20.2 ± 0.4	19± 1	29 ± 0.6	17 ± 0.5	20 ± 1
Al- Hermel	443 ± 10	11.0 ± 0.2		53 ±2	21.1 ± 0.4	22.3 ± 0.4	31.4±0.6	18.5 ± 0.5	29 ± 2
Al- Fakha	197 ± 5	54 ± 1	1.1 ± 0.2	42.1 ± 1.5	16.6 ± 0.4	17.5 ± 0.6	18.0 ± 0.4	10.7 ± 0.4	17.1±0.4

TABLE 4

Activity Concentrations of K-40, Cs-137, U-235, U-238 Series and Th-232 Series in Sediment Samples

Location	K-40 (Bq/kg)	Cs-137 (Bq/kg)	U-235 (Bq/kg)	U-238 series (Bq/kg)			Th-232 series (Bq/kg)		
				Ra-226	Bi-214	Pb-214	Ac-228	Bi-212	Pb-212
Ibrahim River									
Estuary	191±8	1.17±0.14		82.4 ± 6.2	38 ± 2	39 ± 1	13.5 ±1.0	10 ± 1	16 ± 1
Downstream	197 ± 7	1.4 ± 0.1	2.5 ± 0.5	69.0 ± 3.4	30 ± 1	32 ± 1	9.0 ± 0.4	7.0 ± 0.5	10.5±0.4
Assi river									
Al-shawagheer	101 ± 3.4	4.32±0.15			14.4 ± 0.5	15 ± 1	10.5 ± 0.4	5.8 ± 0.4	11.1±0.3
Nabeh Al Zarka	105 ± 8	1.4 ± 0.1	2.08 ± 0.33		15 ± 1	16.5 ± 1.4	12 ± 1	8 ± 1	12 ±1

 $\label{eq:table 5} \textbf{Activity Concentrations of K-40 and Cs-137 in Some Lebanese Diet's Ingredients}$

Sample kind	Sample Code	K-40	MDA	Cs-137	MDA
**		(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)
Vegetables	//			3.65.4	
Lettuce	#1820	135 ± 5	4.5	< MDA	0.24
Parsley	#1819	149 ± 6	4.8	< MDA	0.25
Fruits					
Avocado	# 2088	251 ± 7	3.7	< MDA	0.18
Bou sfeir	# 2089	89 ± 3	3.1	< MDA	0.15
Pumpkin	# 2178	42.3 ± 1.5	2.23	< MDA	0.08
Industrial					
products					
Pickle Cucumber	# 1816	63 ± 2	3	< MDA	0.20
Jams	# 1815	24.4 ± 1.2	1.4	9.7 ± 0.3	0.14
(strawberry,					
berries, grape)					
Nuts	# 1892	211 ± 6	3	0.25 ± 0.04	0.22
Coffee	# 1891	614 ± 17	4	0.24 ± 0.04	0.21
Beer	# 1902	4.5 ± 0.7	2.41	< MDA	0.11
Pepsi diet	# 2106	< MDA	5.54	< MDA	0.30
Tea	# 2163	686 ± 33	9.00	2.6 ± 0.2	0.48
Yogurt	# 2079	50 ± 30	2.43	< MDA	0.11
Drinking water					
Potable water- Saida	# 1905	2.8 ± 0.6	2.4	< MDA	0.11
Potable water - Beirut	# 2100	< MDA	2.7	< MDA	0.10
Mineral water	# 2144	< MDA	2.4	< MDA	0.10
Nabeh Al- sokkar	# 1920	< MDA	2.4	< MDA	0.13
Nabeh Al- zahlan	# 1921	< MDA	2.3	< MDA	0.10
Fresh milk					
Pasteurised milk	# 1943	45.3 ± 1.4	2.06	< MDA	0.10
Pasteurised milk	# 1944	45 ± 2	2.06	< MDA	0.10
Fresh cow milk	# 1948	55 ± 3	1.5	< MDA	0.15
Chicken		7,0			
Chicken nuggets	# 2014	52 ± 2	2.77	< MDA	0.13
Chicken nuggets	# 2079	91 ± 3	4	< MDA	0.16
Luncheon chicken	# 1898	45.0 ± 1.5	2.2	< MDA	0.10
Meat					
Luncheon beef	# 2122	66 ± 2	2.36	0.20 ± 0.02	0.10
Hamburger Milk Powder	# 2179	111 ± 15	7.14	< MDA	0.13
Milk powder	# 2436	559 ± 16	5.07	< MDA	0.34
Milk powder	# 2446	387 ± 12	4.51	0.77 ± 0.07	0.26

For the beta measurements, the minimum count rate recorded is 4.59 counts/sec. The maximum value is 7.1 counts/sec and the mean value is about 6.10 counts/sec. The beta readings were plotted in function of seasons in graphs 5 to 8 in Figure 2.

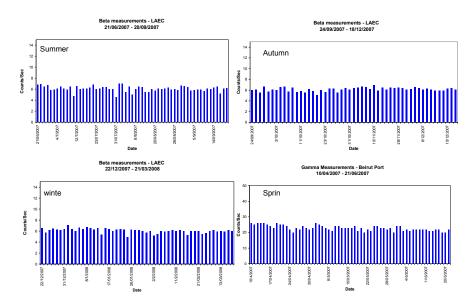


Figure 2. Air beta measurement over one year.

In the same manner, at Beirut port gamma count rate was measured daily using Exporanium GR 110 portable NaI detector (1.5" x 1.5" x 2"). The average value of the readings taken over one year was 22 counts/sec, the minimum value was 20 counts/sec and the maximum 30 counts/sec. Gamma count rate was measured also at Tripoli port in north Lebanon using Exploranium GR110 portable detector. The average value of the readings taken over one year was 39 counts/sec, the minimum value was 33 counts/sec and the maximum value was 42 counts /sec.

CONCLUSION

The global available results of this study indicate that the activity concentration of radionuclides detected are in the normal range. They do not represent neither a possible contamination nor a threat to the health of the population. This paper shows that building up the baseline of the picked analysed samples was a success. In the future and in order to have long-term build-up of radioactivity, sampling and monitoring the Lebanese environment, territory and diet must be continued and controlled regularly making emergency actions easier to implement in case of a radiation incident or major accident like Chernobyl 1986.

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