RADIATION BASELINE LEVELS IN LEBANON: ENVIRONMENTAL SURVEY AND PUBLIC DOSE ASSESSMENT

Omar El Samad, Rola Alayan, Rana Baydoun and Wissam Zaidan
Lebanese Atomic Energy Commission, National Council for Scientific Research,
P.O.Box: 11-8281, Beirut, Lebanon
osamad@cnrs.edu.lb

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ABSTRACT

Establishing radioactivity baseline levels for the different components of the environment in Lebanon and as consequence the detection of any accidental releases are the main goals of this research work. The essential elements of the National Environmental Radiation Survey Program are presented. The data obtained from the radionuclides determination in fresh and drinking water, foodstuff, marine environment samples and daily air monitoring during the years 2009 and 2010 are shown. The yearly averaged radionuclides concentrations are within the range of the previously reported values and the recorded gamma dose rate in air are within the average worldwide values. While the annual effective dose from the intake of radionuclides by food ingestion was calculated and it was found to be lower than worldwide average value.

Keywords: environment, monitoring, radioactivity, radiation, annual effective dose, Lebanon

INTRODUCTION

Over the last decades, the nuclear industry was subjected to an increasingly importance. This importance is enforced by the multiple applications and areas in which it is included.

Nuclear weapons testing that took place between 1950 and 1960 were the main sources for the release of anthropogenic radionuclides to the environment. However these tests are banned.

During the last decades the peaceful use of nuclear technologies is widely spread all around the world guarding and carrying potential risks. The Chernobyl accident in 1986 caused the release of Caesium isotopes to the atmosphere.

Despite the safety performance of the nuclear industry, incidents and accidents may occur with or without significant impact on the environment and on public health. In Lebanon, there are no nuclear power plants but the country could be influenced by the releases caused from nuclear facilities existing in neighbouring countries. It is therefore essential to maintain vigilance and continuously improve the safety of the environment and living organisms' health.

In this paper, the results of the current national monitoring program applied for the year 2009 and 2010 are reported. This program is established to monitor radiation levels in the atmospheric, aquatic and terrestrial Lebanese environment, as well as foodstuffs in order to provide input of radionuclides concentrations and of public exposure under normal circumstances and to look for any abnormalities.

MATERIALS AND METHODS

Sampling and samples preparation

More than one hundred samples were analyzed during the years 2009 and 2010. Forty two samples, including thirty three food samples covering the entire 2009 year, were selected for the present study. The same items were purchased and analysis were conducted with an increase in the number of samples reaching sixty along the year 2010 including drinking water. This set of samples may be shared out, in 2009, into two predominant groups: thirty three food samples including dairy and industrial products with meat, vegetables and fruits… and nine marine samples composed of surface seawater, surface sediment and sand samples picked up from the main cities along the Lebanese coast: Tripoli the capital of the north of Lebanon, Saida the southern capital and Beirut, the capital of Lebanon. The altitude, longitude and latitude parameters of the different samples locations were obtained using Global Positioning system (GPS-2000XL). In 2010, the sample set was extended to include six drinking water samples that were chosen as representative samples of the main and populated Lebanese north, south and central regions.

The national program includes the monitoring of the Lebanese rivers. For this study sediment and fresh water samples were collected at the estuary of two rivers Awwali in south and Ibrahim in north Lebanon.

Concerning foodstuff, the analyzed items constitute integrated part of the Lebanese alimentation. Mixed diet was sampled as separate ingredients consisting of 33 samples taken from supermarkets and shops. The samples are divided into five groups: vegetables and fruit, milk, meat, industrial products and drinking water.

To complete the national monitoring program, air was monitored daily using portable beta and gamma detectors at 3 different locations on the Lebanese territory, 2 in Beirut and 1 in the north.

For gamma spectroscopy measurement, the totality of the selected samples (except for air) were prepared, homogenized and filled into fixed counting geometry 500 ml polyethylene containers (El Samad *et al.*, 2011). The wet samples were then weighed using an analytical calibrated balance.

The sand and sediment samples were grinded and homogenized then dried at 80^0C for 24 hours then the dry to wet weight ratio have been calculated for each (El Samad *et al.*, 2011).

Finally, fresh and drinking water samples were acidified then 20 litres were concentrated in order to collect about 500 ml for analysis (El Samad *et al.*, 2011).

The filled containers were trapped and placed on the top of the detector end-cap and measured for 36 to 48 hours each.

Caesium-137 in seawater samples (80 L sample) was precipitated using ammonium molybdophosphate (AMP) in presence of Cs-134 as tracer to determine the chemical yield. The solution was left overnight to settle down, and then the supernatant was removed. The precipitate was dried and finally counted by gamma spectroscopy (Aoyama *et al.*, 2000; Godoy *et al.*, 2003; Kim *et al.*, 2011).

For the determination of uranium isotopes, and polonium-210 by alpha spectroscopy, chemical treatment is needed to extract the analytes of interest. For Po-210 analysis, wet digestion of the sample was carried out in presence of Po-208 as tracer, followed by evaporation, dissolution of the moist residue and finally deposition on a rotating silver disk (El Samad *et al.,* 2010). Ascorbic acid was added during dissolution to reduce iron (III) and eliminate its interference during deposition (Suriyanarayanan et *al.,* 2008). For uranium isotopes analysis in seawater samples, the main steps are precipitation using $KMnO_4$ and $MnCl_2$ in alkaline medium after addition of U-232 as tracer, dissolution in acidic mixture, evaporation to near dryness. The moist residue obtained was then dissolved in HCl, and introduced to an anion exchange column to carry double extraction in order to separate uranium isotopes. The fraction obtained was evaporated and the resulting moist residue was dissolved in ammonium sulfate solution (Tasoula & Ioannis, 2010**).** Then electro-deposition on a stain less steel disc was carried out.

Measurements

Gamma spectrometry was used to assess gamma ray emitting radionuclides in the prepared samples without any prior chemical operation or other destructive treatment. Three sets of gamma spectrometers from Canberra were used, one of them is equipped with low level Extended Range High Purity Germanium Detector with beryllium window, resolution (2.0 KeV at 1332 KeV) and relative efficiency 50 %, while the two others are equipped with P-type coaxial high purity 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 housed in a 10 cm thick lead shield in order to reduce the ambient 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. 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 and the resolution of the detectors were checked using Eu-152 point source. For quality assurance, certified reference materials milk powder, soil and water were counted and analyzed 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.

The recorded 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 (Accredited standard method ISO 17025, CEI/IEC 1452: 1995)

Cesium-137 and potassium-40 were determined directly *via* their gamma lines at 662 KeV and 1461 KeV, while thorium-232 was analyzed *via* its daughter Ac-228 at 911 KeV. The low level Extended Range HPGe detector was used to measure the Pb-210 directly *via* its gamma line 46.5 KeV.

For alpha emitter's measurements, both silver and stain less steel discs are counted for 24 to 48 hours using alpha spectrometer with Passivated Implanted Planar Silicon detector of resolution 10.5 KeV at 5486 KeV, active area 450 mm^2, mounted in a vacuum chamber and connected to standard electronics to display spectra. For quality assurance purposes, energy calibration was carried out using multi-alpha source, and Pulser test was applied to control the resolution. Working procedures for chemical separation are applied to reference materials for quality control. The spectra of Po-210 measurements showed its peak at 5.15 MeV and tracer Po-208 peak at 5.3 MeV. While spectra of uranium measurements showed the presence of three peaks at 4.2 MeV, 4.8 MeV and 5.3 MeV corresponding to U-238, U-234 and tracer U-232 respectively.

The activity concentrations were calculated and corrected for recovery by comparison with the measured activity of the Po-208 yield tracer and for radioactive decay starting at the sampling time (El Samad *et al.,* 2010; Kanish, 2004)

Daily air monitoring is carried out at 3 locations 2 in Beirut (at the LAEC building and at Beirut port) and 1 at Tripoli port. Measurements are taken using portable NaI detector ($3'' \times 3''$) and Greatz dosimeters for gamma dose rate measurements.

Dose calculation

In order to assess the radiation risk, the annual effective dose from food ingestion was calculated, taking into account the conversion factor for a given radionuclide (r) and the annual consumption rate of a specific kind of food (f) (Nasreddine *et al.,* 2008). Equation (1) represents the formula used for dose calculation.

$$D_{rf} = C_r A_{rf} R_f \qquad (1)$$

where D_{rf} is the annual effective dose (Sv/year), C_r is the conversion factor for a given radionuclide (r) (Sv/Bq), A_{rf} is the activity concentration of radionuclide in food (Bq/Kg) and R_f is the annual consumption rate of food (Kg/year)

RESULTS AND DISCUSSIONS

Marine Monitoring

The parameters altitude and longitude of the sampling location along the Lebanese coast were obtained based on the Global Positioning System and

are presented in Table 1.

TABLE 1

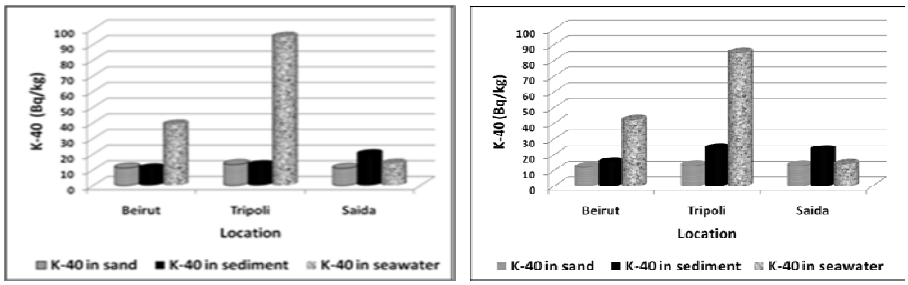
Coordinates of the Sampling Locations

Location	Latitude	Longitude
Beirut	33^0 52.72 N	35^0 28.80 E
Tripoli	34^0 25.92 N	35^0 48.80 E
Saida	33^0 33.35 N	35^0 21.99 E

The activity concentrations of Cs-137 in marine samples (sand, sediment and seawater) for 2009 and 2010 were found to lie below the minimum detectable activity (MDA) in the three locations for all analyzed samples. The MDA value for Cs-137, depending mainly on the counting statistics and variation in sample density, was estimated, using Curie Formula (Genie, 2000), to be approximately 0.13 Bq/Kg dry in sand and sediment, and 0.3 mBq/L in seawater. Ra-226 was not detected in 2009 as well as 2010 year in sand or sediment at any of the three locations since its activity concentration was below the MDA value of 3.2 Bq/Kg dry, except at Tripoli where the measured activity concentration in sediment was, in 2010, 18 Bq/Kg dry. This could be related to the sampling period and the local discharges. In all locations, sampling was carried out in spring, while in Tripoli it was done in November. This month is characterized by lack of rain, absence of runoff and increase of evaporation process. Th-232 was determined in sand and sediment samples, whereas Pb-210 and Po-210 were analyzed in sediment, and uranium isotopes in seawater only.

The activity concentrations of K-40 in analyzed marine samples were comparable at three locations along both years 2009 and 2010. It varies from 11 to 13 Bq/Kg dry in beach sand, from 11 to 24 Bq/Kg dry in coastal sediment and from 14 to 95 Bq/Kg in shallower seawater samples. Higher values in Beirut and Tripoli are attributed to the sampling locations near a sewage outlet and the local discharges. The results are illustrated in Figures 1.

Th-232 in sand and sediment samples was determined *via* Ac-228; the results are presented in Figure 2. The values are comparable at the three locations and along both years 2009 and 2010, and they are found to lay within the range of 1.6 to 3 Bq/Kg dry.



2009 **2010**

Figure 1. K40 in marine samples.

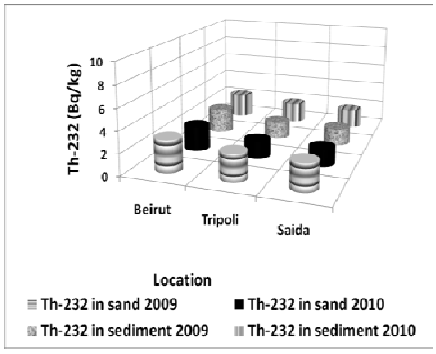


Figure 2. Th-232 in beach sand and sediment samples during 2009 and 2010.

The measured activity concentration in seawater along the years 2009 and 2010 at the three sampling sites for uranium isotopes are presented in Figure 3. The results vary in the ranges of 46 mBq/L to 68 mBq/L, and from 44 mBq/L to 57 mBq/L for U-234 and U-238 respectively. Whereas, the activity concentration of Po-210 in sediment samples during 2010 varied from 4 to 10 Bq/Kg dry, while that of Pb-210 determined during 2009 and 2010 was found to vary in the range of 8 to 19 Bq/Kg dry. Figure 4 illustrates the results of Pb-210 in sediment samples.

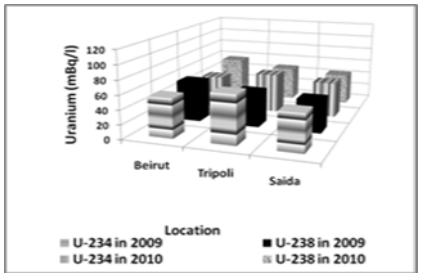


Figure 3. Uranium isotopes in seawater during 2009 and 2010.

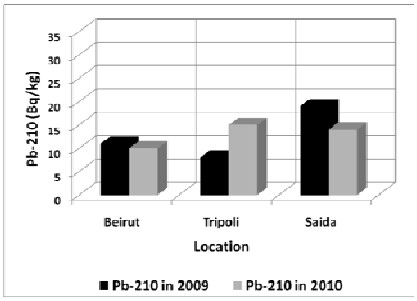


Figure 4. Pb-210 in sediment during 2009 and 2010.

In fish samples which constitute a macro-organism of the marine ecosystem, the results of twelve samples analyzed in 2009 are presented in Table 2. The activity concentration measured for K-40 varies from 51 Bq/Kg wet for Siganus rivulatus sample collected at Beirut to 180 Bq/Kg wet in Shellfish samples collected in Batroun. The values obtained for Cs-137 are close to MDA which is equal to 0.04 Bq/Kg wet, except at Tripoli the activity concentration of Cs-137 in Siganus rivulatus is found to be 0.2 Bq/Kg wet. These values are in agreement to those reported in previous studies (Nasreddine *et al*., 2008). Noting that as part of the national radiation monitoring program, Pb-210 and Polonium-210 as natural radionuclides and main contributors to dose *via* ingestion of fish were analyzed in same fish samples in 2009 (El Samad *et al.*, 2010).

TABLE 2

Gamma Emitters in Fish Samples

Location	Sample kind	Cs-137 (Bq/kg wet)	K-40 (Bq/kg wet)
Tripoli	Siganus rivulatus	0.2 ± 0.02	91 ± 2
	Mugil sp.	0.040 ± 0.007	93 ± 2
	Sparus auratus	0.052 ± 0.008	107 ± 2
Batroun	Pagellus erythrinus	< MDA	100 ± 2
	Diplodus sargus	0.048 ± 0.001	96 ± 3
	Shellfish	0.21 ± 0.01	180 ± 6
Beirut	Siganus rivulatus	< MDA	51 ± 1
	Pagellus erythrinus	< MDA	93 ± 2
	Diplodus sargus	0.05± 0.0008	97 ± 2
Saida	Mugil sp.	0.037± 0.008	92.4 ± 2.2
	Sparus auratus	0.072 ± 0.008	106 ± 3

In general, marine organisms accumulate radionuclides *via* three possible ways; direct uptake of dissolved species from seawater, ingestion of sediment (Topcuoglu, 2001) and ingestion of contaminated pray items. The variation of activity concentration in fish samples is due to the fact that the accumulation of radionuclides depends on the digestive physiology of individual organisms (Jeffre, 2006) as well as the fish size related to its age and weight, Fish from the older age classes are more contaminated than the young fish (Kryshev, 2004). Essentially for Cs-137, the activity concentration is higher for the predatory fish rather than the non-predatory one (Smith *et al*., 2000).

Sediment and surface fresh water samples

The national radiation program includes the monitoring of the Lebanese rivers. The results of gamma emitters in sediment and fresh water samples, collected at the estuary of Ibrahim River at north Lebanon and Awwali river at south, are presented in Table 3.

TABLE 3

Gamma Emitters in Sediment and Fresh Water Samples

Sample	K-40 (Bq/kg)	Cs-137 (Bq/kg)	U-235 (Bq/kg)	U-238 (*Via* Bi-214) (Bq/kg)	Th-232 (*Via* Ac-228) (Bq/kg)
Ibrahim river					
Fresh water	< MDA	< MDA	< MDA	< MDA	< MDA
Sediment	68.08 ± 0.8	0.54± 0.04	1.5 ± 0.08	11.9 ± 0.3	5.7 ± 0.2
Awwali river					
Fresh water	< MDA	< MDA	< MDA	< MDA	< MDA
Sediment	37.9 ± 1.5	= MDA	1.11 ±0.07	8.9 ± 0.2	5.9 ± 0.3

Along the river fume, water, in its course, dilute and transport radionuclides which sink into the bottom of the river and affect the sediments. The activity concentrations of K-40 and Cs-137 in surface water samples were below the minimum calculated detectable activities 2.5 Bq/Kg and 0.2 Bq/Kg respectively for both Ibrahim and Awwali rivers. For Ibrahim river, the activity concentration of Bi-214 and Pb-214 (daughters of U-238 series) recorded are 11.9 and 13 Bq/Kg respectively while the concentration values of Bi-212 and Pb-212 (daughters of Th-232 series) are 6.5 and 5.4 Bq/Kg respectively. Whereas for Awwali river's sediment sample, the activity concentration of Bi-214 and Pb-214 obtained are 8.9 and 10.3 Bq/Kg respectively and those for Bi-212 and Pb-212 are 6.3 and 5.2 Bq/Kg.

Food monitoring

The activity concentration of Cs-137 as artificial radionuclide in the analyzed samples (vegetables, fruits, drinking water, and fresh milk) had a value well below the minimum detectable activity (MDA) of 0.24 Bq/Kg defined according to Curie's Formula. Such result is expected since the migration of radionuclides along the food chain is generally done without concentrating in it. As a result the radionuclides concentration in foodstuff is poor with respect to other parameters in the environment.

It should be noted that the recorded cesium activity concentration in the same order of magnitude or slightly higher is observed for jam (2.5 Bq/Kg), tea (1.98 Bq/Kg), and two milk powder samples from the same brand name and origin. These values are within the permissible national levels 50 Bq/Kg for artificial radionuclides, and do not present any alarming situation or case. (Abbady, 2006; Duric & Popovic, 1997).

The natural radionuclide essentially found in food items is the K-40. It represents 90% of the radioactivity of the elements and its activity concentration varies from 6.9 to

868 Bq/Kg (recorded for Tobacco) according to the kind and nature of the analyzed sample. In contrast, the analyzed drinking water samples have for Potassium-40 concentration values less than the MDA with mean value of 2.28 Bq/Kg. Figure 5 and Figure 6 represent the activity concentration of K-40 in dairy products and industrial food respectively.

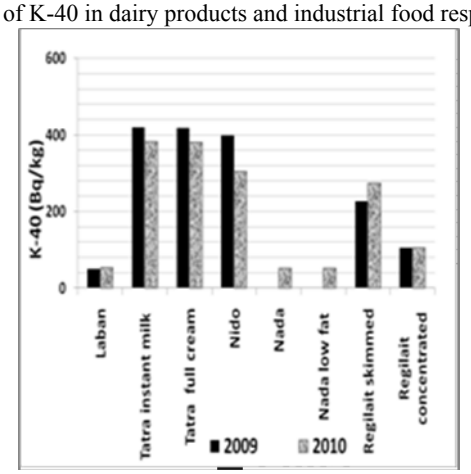


Figure 5. K-40 (Bq/Kg) in dairy products during 2009 and 2010.

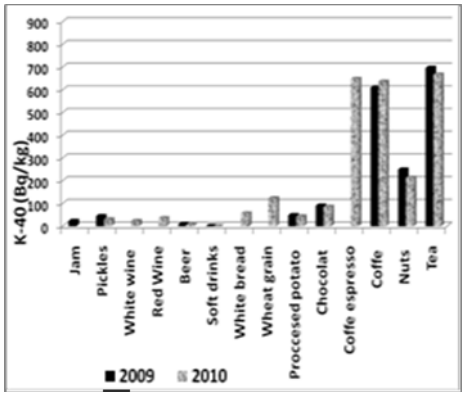


Figure 6. K-40 (Bq/Kg) in industrial food during 2009 and 2010.

The annual effective dose for adults from dietary intake of K-40 was calculated for each food category taking into account the annual consumption rate (Nasreddine *et al.*, 2006; 2008) and 6.2 x 10^{-9} the conversion factor of K-40 for adults (BSS. No 115, IAEA, 1996), the values were comparable to those reported in previous studies (Nasreddine *et al.*, 2008) and the total annual effective dose was found to be lower than the worldwide average value which is 0.3 mSv/year (UNSCEAR, 2000).

The activity concentration of K-40 in different vegetables and fruit species are represented in Figure 7.

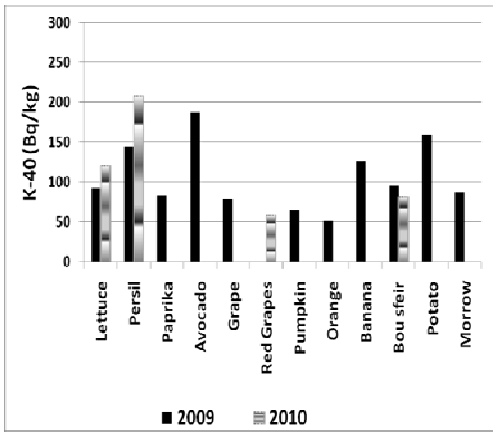


Figure 7. K-40 (Bq/Kg) in vegetables and fruits 2009 and 2010.

Air monitoring

Air monitoring represents almost the most important component of the monitoring program. The reason is that any radioactive pollution reaching the region would have been inevitably transported in the atmosphere and hence exposed people can be affected through inhalation and digestion processes, (BSS. No.115, IAEA, 1996). Thus the early warning is important to assess the direct impact of any accident. The monitoring is carried out daily at three locations, two in Beirut and one on Tripoli where recorded values in the year 2009 and 2010 were comparable to those obtained in 2008 (El Samad *et al.*, 2011).

At the LAEC location and along the entire years 2009 and 2010, the mean gamma dose rate was about 105 nSv/h, slightly higher than the value of 90 nSv/h recorded in 2008. Whereas the maximum recorded value was 112 nSv/h in January 2010, while the minimum value was observed in February and May 2009 at 97 nSv/h. This difference may be explained by the high percentage of rainfall during February 2009. While the average dose rate value at Beirut port location covering the year 2009 and 2010 was 52 nSv/h; with 50 nSv/h and 53 nSv/h as minimum and maximum recorded values covering the summer and winter season in 2009 and 2010 respectively.

At Tripoli port , the mean recorded dose rate value are higher than recorded in the either 2 Beirut sites this is due to the type of the portable detector used. This value is 133 nSv/h for 2009 and 137 nSv/h for 2010. The minimum recorded value is 130 nSv/h recorded during April and May 2009 and the maximum is 140 nSv/h recorded during February 2010. Figure 8 represents the values recorded at LAEC location and Tripoli port during the years 2009 and 2010.

The values obtained at three locations still within the average worldwide annual gamma dose rate (UNSCEAR, 2000).

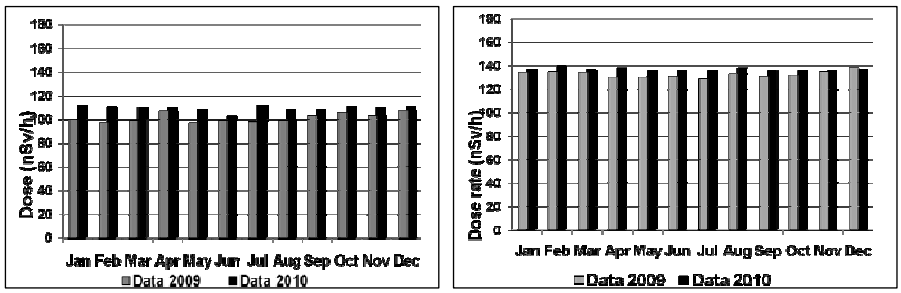


Figure 8. Air monitoring values at LAEC and Tripoli port locations.

CONCLUSION

The results of the radioactivity measurement in the Lebanese environment undergone in the frame of the national environmental radiation monitoring program, during the period 2009 and 2010 have been reported. The yearly averaged radionuclides concentrations are within the range of the previous years. The activity concentrations of artificial radionuclides mainly Cs-137 was below the minimum detectable activity except in jam and tea, where the values were slightly higher but still within the permissible national levels. While those for natural radionuclides mainly K-40 were comparable to those reported previously. The calculated annual effective dose from the intake of this radionuclide by food ingestion was found to be lower than the worldwide average value .

The data obtained from this study provide the building blocks to establish the environmental radioactivity baseline levels useful to detect and screen any suspected contamination or abnormal values resulting from any accidental situations.

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