

Public exposure to radioactivity levels in the Lebanese environment

O. El Samad¹ · R. Baydoun¹ · M. Aoun¹ · W. Zaidan¹ · H. El Jaïd¹

Received: 11 May 2016 / Accepted: 13 October 2016 / Published online: 2 November 2016
© Springer-Verlag Berlin Heidelberg 2016

Abstract In the framework of a National Environmental Radiation Survey Program, a total of 950 samples were collected and analyzed over 7 years. The program covers different compartment of Lebanese environment, rivers, wells, marine samples, soil, and foodstuff. Air was monitored continuously through a radiation early warning network system. Data collected from 2009 to 2015 are presented in this work. Gross alpha/gross beta values in well samples were below the guidance levels stated by the WHO. Cesium-137 was detected in milk samples, whey, and jam. However, its content was much lower than the national permissible level, while its activity concentration in marine samples was lower than the values reported in studies carried out in the Mediterranean Sea. The activity concentration of ^{40}K in food samples was comparable to studies carried out in neighboring countries and higher than the content determined in non Mediterranean countries. Concerning marine environment, the activity concentrations of natural radionuclides ^{238}U , ^{232}Th , and ^{40}K were comparable to those reported in other studies carried out in different countries. As well as their content in rivers and sediments was lower than those reported in neighboring region. Potassium-40 in food and ^{210}Po in fish were the main contributors to the internal dose. The average annual effective dose due to external exposure and internal, excluding radon gas which constitutes around 43 % of the total dose, was found to be lower than the total worldwide value, $2.4 \text{ mSv year}^{-1}$. Iodine-131 was detected in grass samples, collected in 2011 during

Fukushima accident; its content ranged from 0.40 ± 0.06 to $0.9 \pm 0.1 \text{ Bq kg}^{-1}$, as well as ^{137}Cs was detected in some seafood samples imported from Japan and neighboring countries. Its activity concentration varied between 0.15 ± 0.04 and $0.40 \pm 0.02 \text{ Bq kg}^{-1}$.

Keywords Environmental radiation survey program · Gamma spectroscopy · Alpha spectroscopy · Liquid scintillation counter · Radiation baseline levels · Annual effective dose

Introduction

In the framework of the main mandates of the Lebanese Atomic Energy Commission (LAEC), the Environmental Radiation Control Department has established in 2007 a National Environmental Radiation Survey Program to control natural and artificial radioactivity levels, in the different components of the Lebanese environment in order to assess population exposure, set radiation baseline levels, and detect any accidental radioactivity due to the release that could reach the country from any abroad nuclear accident, as well as to plot the trend of radioactivity levels over time. All living organisms are continuously exposed to ionizing radiation coming from both natural and artificial radionuclides. The received dose could be internal or external. About 85 % of this dose is due to natural radiation (Dołhańczuk-Śródka 2011) originating mainly from the primordial radionuclides, such as ^{238}U and ^{232}Th and their decay products, as well as ^{40}K . These are present at trace levels in all ground formations (Awudu et al. 2012). However, their concentration in soil, sands, and rocks varies from region to another depending on local geographical and geological factors. Their existence in biotic system of plants,

Responsible editor: Philippe Garrigues

✉ O. El Samad
osamad@cnsr.edu.lb

¹ Lebanese Atomic Energy Commission, National Council for Scientific Research, P.O. Box 11-828, Beirut, Lebanon

animals, water, and air could be a significant contributor to the ingestion dose. Food crops could be subjected to elevated activity concentration of naturally occurring radionuclides (NORM) due to the use of fertilizers (Awudu et al. 2012). Man-made or anthropogenic radionuclides have been released to the environment by different nuclear activities. In the period 1950s–1960s, they were produced from nuclear weapons testing as global and local fallout, and then they were emitted from nuclear accidents, such as Chernobyl and Fukushima accidents (Andoh et al. 2015). Cesium-137, ¹³⁴Cs, ⁹⁰Sr, ⁸⁹Sr, and ¹³¹I are artificial radionuclides that may be transported with the radioactive plume for long distances across boundaries away from the accident zone. They could be deposited by fallout over soil and plants and then transferred to the food chain. Therefore, with the widespread use of radiation, measurement of natural and artificial radioactivity content in different environmental components is very important to maintain control of dominating radiation levels, and many countries had conducted environmental radiation survey programs. In Lebanon, many studies have been carried out to determine the radioactivity levels. In the period 1998–2000, cesium-137 was analyzed in soil samples in order to study the fallout one decade after Chernobyl accident (El Samad et al. 2007). Natural and artificial radioactivity was determined in Lebanese soil and radioactive carts were established (El Samad et al. 2013). Natural radioactivity was determined in building materials (Kobeissi et al. 2008; Kobeissi et al. 2013). Gamma emitters were analyzed in food samples representing the market basket of an adult urban population in Beirut (Nasreddine et al. 2008). Polonium-210 and ²¹⁰Pb were analyzed in fish (El Samad et al. 2010). Recently, an investigation of radiological impact on coastal area around a fertilizer industry was carried out (El Samad et al. 2014; Aoun et al. 2015). Hence, it was essential to establish a national environmental radiation monitoring program that covers various compartments of the Lebanese environment, in order to complete the work carried out and to establish radiological database over years. This should cover food stuff to assess public ingestion dose due to food intake, as well as marine environment and rivers in order to set radioactivity baseline levels. As well as air monitoring and radioactivity in soils should be determined for external public dose assessment. For this purpose, gamma emitters were determined in various foodstuff samples representing the main constituents of the Lebanese diet, also the surveys covers two rivers, marine samples from three coastal cities, soil, and air monitoring. Six wells were monitored. Gross alpha/gross beta, radium, and uranium isotopes were determined. In addition to gamma emitters, ²¹⁰Po and uranium isotopes are analyzed in some selected

samples. Results obtained during 7 years were discussed in this work.

Materials and methods

Sampling

The sampling campaign executed in 2009, covered 65 samples. Sampling was carried out periodically and extended with years to cover more commodities and species. In 2015, a total of 950 samples were analyzed over 7 years. Sampling plan is presented in Table 1.

Most of the foodstuff samples were collected from local markets taking into account the main constituents of the Lebanese diet (Nasreddine et al. 2008). These include vegetables and fruits, milk and dairy products, meat, and processed goods. No concern was given to the provenance or country of origin, except for wheat grain where samples imported from different countries were analyzed in addition to local origin wheat.

Marine environment samples, beach sand, and sediment and seawater samples were collected twice a year from three main coastal cities, Saida, Beirut, and Tripoli. Benthic and pelagic marine fishes were collected from the fresh catch sold in the local markets. The chosen species were mostly consumed by the majority of the Lebanese population. Two rivers, situated at south Lebanon (river 1) and north Lebanon (river 2) were selected for monitoring; fresh water and sediment samples were collected. Soil samples were collected from uncultivated areas. While for groundwater, six wells situated at the coastal area south the Capital Beirut were monitored.

Table 1 Sampling plan

Matrix	Radionuclide	Frequency
Vegetables and fruits	Gamma emitters	1/year
Processed foodstuff	Gamma emitters	1/year
Milk and dairy products	Gamma emitters	1/year
Wheat and grains	Gamma emitters	1/year
Meat and chicken	Gamma emitters	1/year
Groundwater	Gamma emitters, gross alpha/gross beta, uranium isotopes, ²²⁶ Ra	1/year
Fresh water and river sediment	Gamma emitters	1/year
Seawater	Gamma emitters, uranium isotopes	2/year
Beach sand	Gamma emitters, ²¹⁰ Pb	2/year
Sea sediment	Gamma emitters, ²¹⁰ Pb, ²¹⁰ Po	2/year
Fish	Gamma emitters, ²¹⁰ Pb, ²¹⁰ Po	2/year

Concerning air monitoring, three locations were selected for daily ambient dose determination; by the mid of 2013, 20 locations were monitored continuously through the establishment of radiation early warning network system. The distribution of the stations along the Lebanese territory is presented in Fig. 1. This network system is consisted of 20 dose rate meters with Geiger Muller tubes that record dose rate continuously and transmit data to the central station via SMS or binary messages that are converted to digital quantities. These data are checked, controlled, and managed permanently to ensure proper function of the system.

Sample preparation

The standard sample preparation procedures were applied (IAEA 295, 1989) depending on required radionuclide and the technique used for analysis. For gamma spectroscopy measurements, solid samples were crushed and homogenized. Sand and sediment samples were dried at 85 °C for minimum of 24 h, and then dry/wet ratio was calculated (El Samad et al. 2013). All samples were prepared in the adopted counting geometry, 500 ml polyethylene cylindrical container. For the determination of ^{137}Cs in seawater, precipitation was carried out using ammonium molybdophosphate (AMP). ^{134}Cs was added as tracer in order to determine the chemical yield. The obtained precipitate was dried before the measurement by gamma spectroscopy (Kim et al. 2012).

Radiochemical separation was applied to extract the alpha emitters of interest. For ^{210}Po analysis, wet digestion of the sample was carried out in presence of ^{208}Po as tracer, followed by evaporation, dissolution of the moist residue, and finally deposition on a rotating silver disk. Ascorbic acid was added during dissolution to reduce iron (III) and eliminate its interference during deposition (Suriyanarayanan et al. 2008). Uranium isotopes are chemically separated by precipitation using KMnO_4 and MnCl_2 in alkaline medium, after addition of ^{232}U as tracer. Then, dissolution in acidic mixture and evaporation to near dryness was applied. The moist residue obtained was dissolved in HCl and introduced to an anion exchange column. The fraction obtained was evaporated and the resulting moist residue was dissolved in ammonium sulfate solution. Then electrodeposition on a stainless steel disk was carried out.

For gross alpha/gross beta determination in groundwater, samples were acidified, evaporated (Fons et al. 2013), then an aliquot of 8 ml was mixed with 12 ml Hisafe 3 scintillation cocktail in a plastic vial.

Measurement

A standard method (IEC 1452: 1995) was applied for the measurement and analysis of gamma emitter's radionuclides, using three sets of gamma spectrometers with high-purity (P-

type) coaxial germanium detectors (HPGe) of relative efficiency 30, 40, and 50 %. The gamma spectroscopy laboratory is accredited according to ISO/IEC 17025 standard for testing and calibration laboratories. The detectors were connected to standard electronics and the spectra were accumulated in an 8K multichannel analyzer. HPGe detectors were housed in 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 energy calibration of the spectrum was done using a standard multigamma source; the efficiency calibration was performed. Efficiency curves were corrected for attenuation and absorption (Kobeissi et al. 2008). Quality control procedure was applied through the measurement and analysis of different reference materials representing the routinely analyzed samples. The background spectra were measured periodically under the same conditions of sample measurements and were used to correct the calculated sample activities. The sample counting time ranged from 36 to 48 h. The spectra were analyzed offline using the Genie 2000 software from Canberra Version V3.1a. The uncertainty was calculated using error propagation law taking into account all sources of uncertainties. The ^{238}U was determined from the gamma line of its daughter $^{234\text{m}}\text{Pa}$ at 1001.03 keV, as well as ^{232}Th via ^{228}Ac at 911.2 keV. The ^{226}Ra was determined from its gamma line at 186.2 keV after correction for ^{235}U interference. The total count of the peak 186 keV is built up from counts of ^{235}U and ^{226}Ra . The correction method applied consists to calculate the activity concentration of ^{235}U from the gamma line 163.3 keV, and then this activity is used to calculate the net peak area corresponding to ^{235}U at the gamma line 185.7 keV, that is subtracted from the total net peak area at 186 keV. The remaining net peak area is used to calculate the activity concentration of ^{226}Ra (Völgyesi et al. 2014). The ^{137}Cs and ^{40}K were determined directly from their corresponding gamma lines at 661.6 and 1460.8 keV, respectively (El Samad et al. 2007), as well as ^{210}Pb was determined directly from its gamma ray energy 46.5 keV using low-level extended range HPGe detector with Beryllium window.

For the measurement of alpha emitters, silver and stainless steel disks were counted using alpha spectrometer with passivated implanted planar silicon detectors of resolution 10.5 at 5486 keV, active area 450 mm², mounted in a vacuum chamber, and connected to standard electronics to display spectra. Energy calibration was carried out using multialpha source. For quality control purposes, adopted working procedures for chemical separation were applied to reference materials. 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, 4.8, and 5.3 MeV corresponding to ^{238}U , ^{234}U , and tracer ^{232}U , respectively. The activity concentrations were calculated and corrected for chemical recovery and for radioactive decay starting from sampling time.

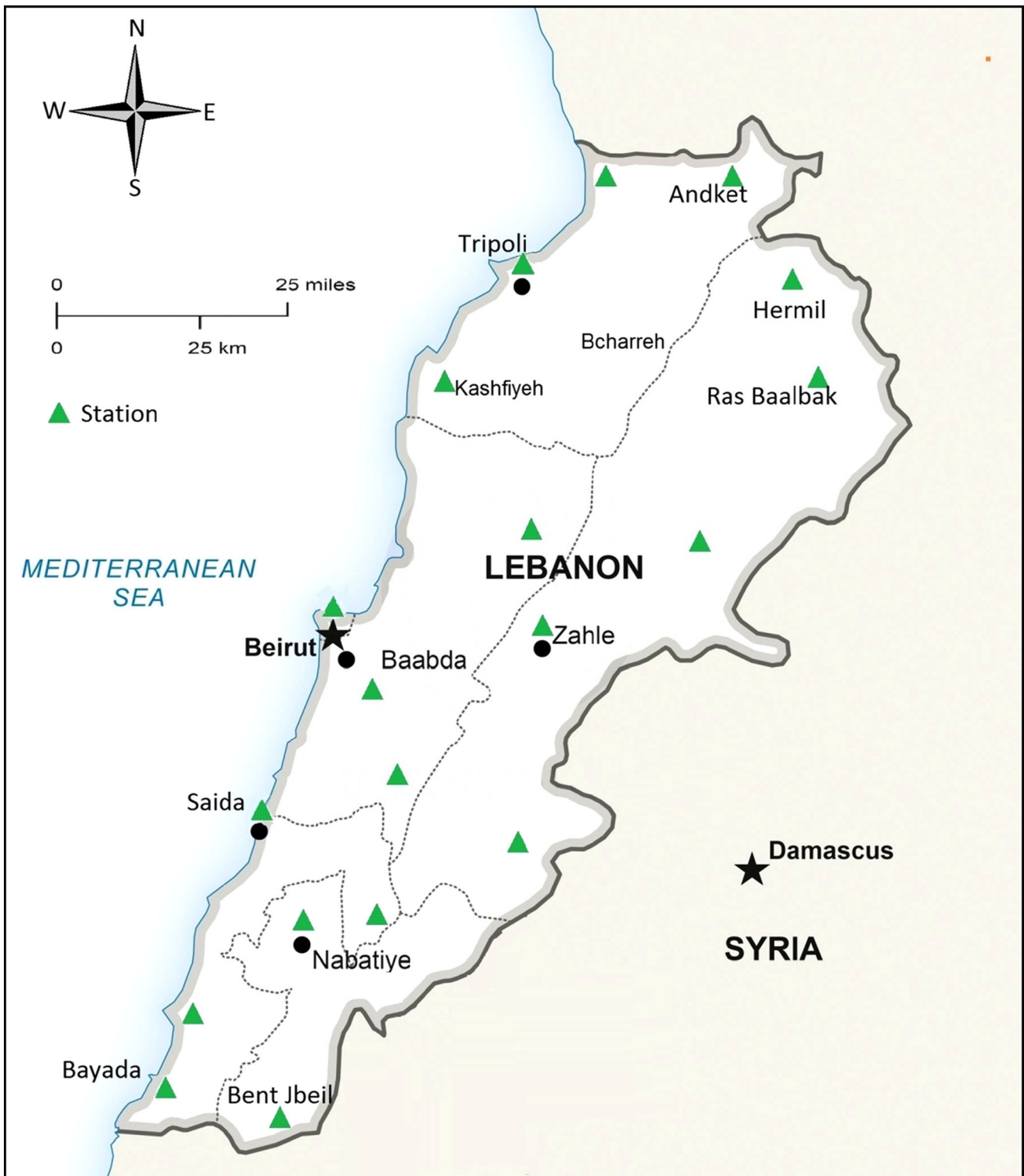


Fig. 1 Distribution of the radiation early system along Lebanese territory

A low-level liquid scintillation counter, Tri-Carb 3180 TR/SL, was used for the determination of gross alpha/gross beta and ^{226}Ra in water samples; the counting system is composed of two photomultiplier tubes (PMT), amplifier, analog to digital convertor (ADC), multichannel analyzer (MCA), and a PC

to display spectra via Quanta Smart program. The whole electronic components are surrounded by a lead shield. The Tri-Carb 3180 incorporates a programmable time resolved electronics that, in addition to the incorporated Bismuth Germanate detector $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ and the pulse shape analyzer

(PSA), reduce to the minimum the background interference and discriminate the true beta events.

In 2013, a radiation early warning network system, from Saphymo Company, was installed to monitor continuously the dose rate in air at 20 locations distributed along the Lebanese territory and to detect any abnormal radiation levels.

Dose estimation

Dose estimation was carried out taking into account the external exposure from air and terrestrial radiation, as well as internal exposure via ingestion, excluding inhalation due to radon exposure which constitutes 43 % of the total dose (UNSCEAR 2000). The absorbed dose, D (nGy h^{-1}), from natural and artificial radionuclides in soil was estimated as described in UNSCEAR 2000, then the annual effective dose ($\mu\text{Sv year}^{-1}$) was calculated taking into account the conversion factor 0.7 Sv Gy^{-1} and the occupancy factor 0.2 that specifies the fraction of total time spent outdoors (El Samad et al. 2013), while the exposure from air was measured at 1 m above ground, through the radiation early network warning system.

In order to assess the radiation risk from food ingestion, the annual effective dose was calculated according to Eq. (1), 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).

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

where D_{rf} is the annual effective dose (Sv year^{-1}), C_r is the conversion factor for a given radionuclide (r) (Sv Bq^{-1}), A_{rf} is the massic activity of radionuclide in food (Bq kg^{-1}), and R_f is the annual consumption rate of food (kg year^{-1}). The main contributors in the internal radiation dose due to food ingestion were ^{40}K in foodstuff samples and ^{210}Po in fish, having conversion factors for adults of 6.2×10^{-9} and 1.2×10^{-6} (GSR 2014).

Monitoring Fukushima accident

During the Fukushima accident, grass and rain water samples were collected at the end of March 2011, from different locations along the Lebanese territory. This was carried out upon live tracking of the radioactive plume movement through the international stations of the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) that simulates the path of the plume taking in consideration the weather conditions. At the end of March 2011, ^{131}I was detected in Greece and Lebanon. Analysis was carried out in order to detect any artificial radionuclides resulting from the plume. All seafood goods imported from Japan and its neighboring countries were

subjected to radiation survey, under a circumstantial national decree.

Statistical analysis

Analysis of variance test, two-way ANOVA was applied in order to study the variation between the data obtained. This is based on comparison of means between groups and determines whether any of those means significantly differs from each other. The statistical test was carried out to study the variation of data between species, years, and locations.

Results

Food samples

Among artificial radionuclides, only ^{137}Cs was detected in foodstuffs; however, its content in the majority of samples, analyzed over 7 years, was below the minimum detectable activity ($\text{MDA} = 0.2 \text{ Bq kg}^{-1}$), and hence, its content in vegetables and fruits is lower than that reported in neighboring countries (Canbazoğlu and Dođlu 2013; Alhadj et al. 2014). As well as the activity concentrations of ^{137}Cs was lower than values stated in studies carried out in non Mediterranean countries such as India and Nigeria (Rao et al. 2010; Tchokossa et al. 2013) and lower than pre-Fukushima ^{137}Cs levels detected in food samples in Japan (Merz et al. 2015). The activity concentration in milk powder samples ranged between 0.40 ± 0.06 and $1.7 \pm 0.08 \text{ Bq kg}^{-1}$; these samples are imported from the European countries mainly affected by Chernobyl accident in 1986. Values were comparable to those stated in other studies (Ababneh et al. 2010; Pearson et al. 2016). Cesium-137 was detected in one coffee sample with activity concentration of $0.5 \pm 0.07 \text{ Bq kg}^{-1}$. Higher ^{137}Cs content was found in a brand jam and a whey sample, 2.5 ± 0.1 and $5.24 \pm 0.13 \text{ Bq kg}^{-1}$, respectively. However, all determined values were much lower than the maximum national permissible levels, 50 Bq kg^{-1} for milk and 150 Bq kg^{-1} for other products. In 2014, two samples of a local jam brand showed unexpected ^{137}Cs activity of concentration of 23.17 ± 0.43 and $42.77 \pm 0.84 \text{ Bq kg}^{-1}$. After investigation, it was found that these values are attributed to the high cesium content in main raw materials used, the berry, imported from Poland. The detected ^{137}Cs values were comparable to those found in berry samples analyzed in Poland (Gwynn et al. 2013). However, values are below the defined national permissible levels for artificial radionuclides. The activity concentration of ^{137}Cs in some species is presented in Fig. 2.

The significant natural radionuclide determined in food samples was ^{40}K . ANOVA test showed that its activity concentration varied significantly between matrices but still comparable over years within same species, and no significant variations were found, as well as values obtained were comparable to those reported by previous studies and to values found in other countries (Nasreddine et al. 2008; Basu et al. 2015). The lowest content was found in juice and beer; values ranged between 2.25 ± 0.66 and $9.46 \pm 1.00 \text{ Bq kg}^{-1}$. While its activity concentration in vegetables ranged from $48 \pm 2 \text{ Bq kg}^{-1}$ in lettuce to $254 \pm 6 \text{ Bq kg}^{-1}$ in parsley, values were comparable to those determined in neighboring country (Al-Masri et al. 2004a) and lower than those reported in other studies carried out in the Middle East region (Al-Absi et al. 2015; Harb 2015). However, it was found that this content was higher than that determined in non Mediterranean countries (Tchokossa et al. 2013). Its activity concentration in fruits varied between $30 \pm 2 \text{ Bq kg}^{-1}$ in apple and $217 \pm 8 \text{ Bq kg}^{-1}$ in avocado, values comparable to the range stated in other researches carried out in the region (Al-Masri et al. 2004a; Harb 2015) and higher than those reported by other regions in the world (Tchokossa et al. 2013). The determined activity concentrations of ^{40}K in fruits and vegetables were presented in Fig. 3. In milk powder samples, the activity concentration of ^{40}K ranged from 100 ± 2 to $650 \pm 20 \text{ Bq kg}^{-1}$ depending on the country of origin. Values were comparable to those reported in other countries (Ababneh et al. 2010). The average activity concentration of ^{40}K in coffee was found to be $640 \pm 18 \text{ Bq kg}^{-1}$, a value lower than that reported in other countries (Roselli et al. 2013) while that in tea was $700 \pm 20 \text{ Bq kg}^{-1}$ slightly higher than that found in other studies (Harb 2007). However, these values were comparable to those determined in neighboring countries (Al-Masri et al. 2004a). Potassium-40 content in grains varied from $26 \pm 1 \text{ Bq kg}^{-1}$ in rice to $460 \pm 13 \text{ Bq kg}^{-1}$ in beans, a range slightly wider than the range reported in other countries (Al-Masri et al. 2004a).

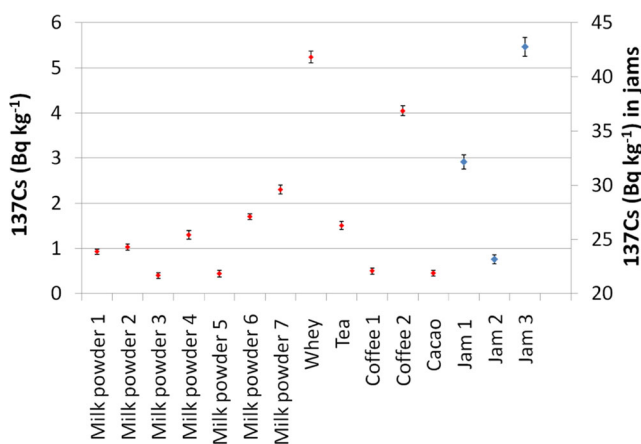


Fig. 2 Activity concentration of detected ^{137}Cs in foodstuff samples (2009–2014)

Marine samples

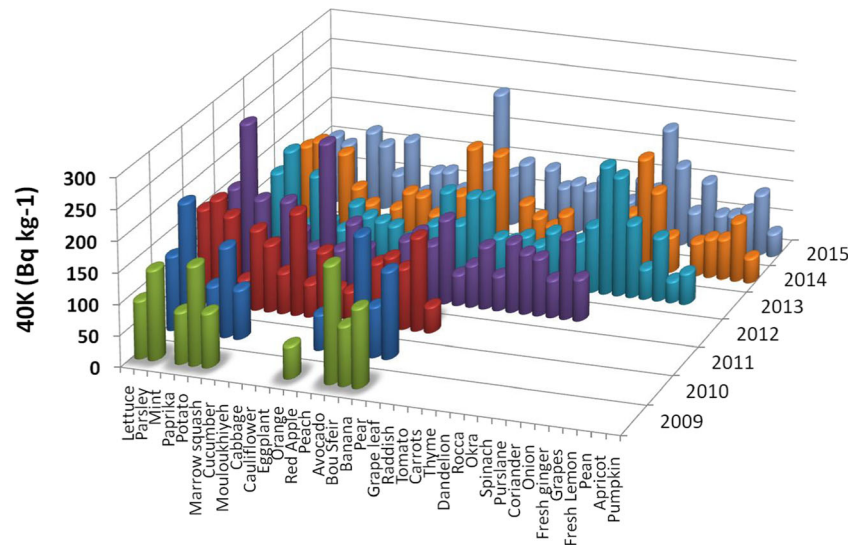
The activity concentration of ^{137}Cs in all seawater samples collected from the three locations over 7 years was lower than 0.3 mBq l^{-1} , and hence lower than values reported by other studies carried out at the Mediterranean Sea (Livingston and Povinec 2000; Alhadj et al. 2014). As well as its activity concentration in sediment and beach sand was lower than $\text{MDA} = 0.2 \text{ Bq kg}^{-1}$.

Among natural radionuclides, the content of ^{238}U , ^{232}Th , and ^{40}K was comparable in sediment and sand samples and no significant variation was detected. As shown by ANOVA test, values were comparable for the three sampling sites, as well as over years. The activity concentration of ^{238}U ranged from 25 to 35 Bq kg^{-1} , a value comparable to worldwide average, 35 Bq kg^{-1} (UNSCEAR 2000). The content of ^{232}Th was lower than worldwide average value, 30 Bq kg^{-1} , and comparable to those reported in other countries on the Mediterranean Sea (Kut and Berker 2014). It varied between 2 ± 0.1 and $4.0 \pm 0.2 \text{ Bq kg}^{-1}$, while activity concentration of ^{40}K ranged from 8.0 ± 0.7 to $37 \pm 2 \text{ Bq kg}^{-1}$, values lower than worldwide average, 400 Bq kg^{-1} , and comparable to other studies (El-Saharty 2013). Among natural radionuclides in seawater, ^{40}K content was comparable over years within the same location and no significant variation was found. The lowest values were found in Saida in a range of 12 ± 1 to $15 \pm 1 \text{ Bq kg}^{-1}$. These are comparable to those found in other countries (Petrinec et al. 2012; El-Saharty 2013). Higher values were determined in Beirut within a range of 38 ± 2 to $42 \pm 2 \text{ Bq kg}^{-1}$. Samples collected from Tripoli showed maximum activity concentration of ^{40}K , which varied between 85 ± 3 and $96 \pm 3 \text{ Bq kg}^{-1}$. As shown by ANOVA test, there was significant variation between sampling locations. This could be attributed to the presence of sewage outlet or local discharges. Uranium isotopes were determined in seawater; applied statistical test showed that the values were comparable between locations as well as over years and no significant variation was detected. Uranium-238 content ranged from 32 ± 2 to $57 \pm 4 \text{ mBq l}^{-1}$, while that of ^{234}U varied between 37 ± 2 and $68 \pm 5 \text{ mBq l}^{-1}$. These values were comparable to the data reported in previous studies (El Samad et al. 2014).

The activity concentration of ^{137}Cs in fish samples was found to lie below MDA equal to 0.13 Bq kg^{-1} dry weights in most species. For comparison, its content was lower than values detected in neighboring countries (Alhadj et al. 2014) and in New Zealand (Pearson et al. 2016), as well as in North Atlantic ocean (Carvalho et al. 2011). However, one sample of *Siganus rivulatus* collected from Tripoli showed a value of $1 \pm 0.13 \text{ Bq kg}^{-1}$ dry weights while the same species collected from other areas showed activity concentration below MDA .

Concerning natural radioactivity in fish samples, ANOVA test showed that the activity concentration of ^{40}K was comparable between sampling locations, over years, as well as

Fig. 3 Activity concentration of ^{40}K in fruits and vegetables



between species, and no significant variation was found. The lowest values were $208 \pm 7 \text{ Bq kg}^{-1}$ dry weights detected in *Diplodus sargus*, while the highest values were $477 \pm 26 \text{ Bq kg}^{-1}$ weights detected in *S. rivulatus*, values higher than those reported in other countries (Carvalho et al. 2011; Khandaker et al. 2015).

The activity concentration of ^{210}Po was comparable to that reported in other countries (çatal et al. 2012) and varied between 23.3 ± 0.3 and $78 \pm 3 \text{ Bq kg}^{-1}$ dry weights according to species and location. Average values are represented in Table 2. Statistical test showed that there is no significant variation between sampling locations, and species, except on 2015, the content of ^{210}Po varied significantly between species. This was reflected in a significant variation over year in one species, *D. sargus*. This could be attributed to the differences in metabolism and food intake pattern for various species, as well as the existing anthropogenic activities in surrounding areas that could influence the radionuclides contents in biota.

River and well monitoring

The activity concentration of ^{137}Cs in surface fresh water and in sediment samples collected from two rivers was

Table 2 Average activity concentration (A) of ^{210}Po (Bq kg^{-1}) in dry weight (dw)

Species	A (Bq kg^{-1}) in dw at Saida	A (Bq kg^{-1}) in dw at Beirut	A (Bq kg^{-1}) in dw at Tripoli
<i>Diplodus</i>	40 ± 2	20 ± 1	36 ± 2
<i>Siganus rivulatus</i>	22 ± 1	25 ± 1	29 ± 1
<i>Pagellus erythrinus</i>	32 ± 1	30 ± 1	33 ± 2
Mugil p.	14 ± 1	20 ± 1	17 ± 1

below 0.3 mBq l^{-1} and lower than that reported by neighboring countries (Alhadj et al. 2014). The content of ^{40}K in fresh water was below MDA, while its activity concentration in sediment samples was comparable over years and no significant variation was recognized. The average values were $40 \pm 2 \text{ Bq kg}^{-1}$ in river 1 located in south Lebanon and $68 \pm 2 \text{ Bq kg}^{-1}$ in river 2 situated at the north. As shown by ANOVA test, the activity concentrations of ^{232}Th and ^{226}Ra were comparable for the two rivers, as well as no significant change over years was detected. Their average values were 12 ± 1 and $6.0 \pm 0.4 \text{ Bq kg}^{-1}$, respectively. The content of the mentioned natural radionuclides is lower than that reported in other studies (Al-Masri et al. 2004a).

Gross alpha in monitored wells was found below $\text{MDA} = 0.001 \text{ Bq l}^{-1}$ in all samples except in one well which was $0.070 \pm 0.006 \text{ Bq l}^{-1}$. The values of total beta ranged from 0.230 ± 0.002 to $0.78 \pm 0.03 \text{ Bq l}^{-1}$, values lower than the maximum acceptable limits stated in WHO guidelines (WHO 2011), 0.5 and 1 Bq l^{-1} , respectively. The activity concentration of ^{226}Ra was comparable to $\text{MDA} = 0.04 \text{ Bq l}^{-1}$ in all samples, with an average value of 0.048 Bq l^{-1} , which is lower than the guidance level of 1 Bq l^{-1} settled by WHO, while the activity concentrations of ^{238}U and ^{234}U were comparable and varied between 10.0 ± 0.5 and $17 \pm 1 \text{ Bq l}^{-1}$.

Dosimetry

The average annual effective doses, at 20 locations distributed along the Lebanese territory, ranged from 0.48 to $0.77 \text{ mSv year}^{-1}$. Statistical test showed significant variation of the dose rate between stations that could be attributed to the variation in the altitude of the location. However, data did not vary significantly over time.

The average external dose due to terrestrial radiation, calculated in areas where 70–80 % of the Lebanese population lived, was found to be 0.066 mSv year⁻¹, a value lower than the worldwide average value, 0.5 mSv year⁻¹ (UNSCEAR 2000).

As the activity concentration of ¹³⁷Cs was negligible in nearly all food samples, the total annual effective internal doses from ingestion were based on ⁴⁰K intake in food and ²¹⁰Po in fish samples; their values from 2009 to 2015 were 0.060, 0.074, 0.075, 0.084, 0.080, 0.086, and 0.082 mSv year⁻¹, respectively. The average value was found to be 0.077 mSv year⁻¹ which is lower than the worldwide average value of internal dose due to ingestion, 0.3 mSv year⁻¹ (UNSCEAR 2000).

The total annual effective doses due to both external and internal exposure over 7 years, excluding radon, were found to be respectively 0.66, 0.67, 0.67, 0.68, 0.67, 0.68, and 0.67 mSv year⁻¹ with an average of 0.67 mSv year⁻¹, a value lower than the worldwide average value, 2.4 mSv year⁻¹.

Monitoring Fukushima accident

As a result of wet deposition, iodine-131 was detected in grass samples collected in the end of March 2011 during Fukushima accident; its content ranged from 0.40 ± 0.06 to 0.9 ± 0.1 Bq kg⁻¹ wet, with an MDA of 0.2 Bq kg⁻¹. However, no cesium isotopes were detected in rain water, while in some grass samples, the activity concentration of ¹³⁷Cs was below the MDA = 0.2 Bq kg⁻¹ wet. As well as cesium-137 was detected in some seafood samples imported from Japan and neighboring countries. Its content varied between 0.15 ± 0.04 and 0.40 ± 0.02 Bq kg⁻¹.

Conclusion

A total of 950 samples, from different compartments of the Lebanese environment, were collected and analyzed over 7 years, in the framework of the National Environmental Radiation Survey Program. Generally, the results are low and within the range of internationally recommended limits and no significant radiological hazard was found. The activity concentration of ¹³⁷Cs was below the minimum detectable activity in the majority of analyzed samples. However, its content in milk powder samples was comparable to that reported in other countries. Potassium-40 was the main natural radionuclide detected in food stuff samples; its activity concentration was comparable to that reported by other studies. Concerning marine environment, the activity concentration of ¹³⁷Cs was lower than that reported by other countries on the Mediterranean Sea, while that of natural radionuclides, ²³⁸U, ²³²Th, and ⁴⁰K, was comparable to other studies. As well as their activity concentration in rivers and sediments was lower

than that reported by neighboring countries. Potassium-40 in food and ²¹⁰Po in fish are the main contributors to internal dose. Public dose was assessed based on the external and internal exposure, excluding radon. The average annual effective dose was found to be lower than the total worldwide value, 2.4 mSv year⁻¹. The sustainability of the program will be maintained and more commodities and species will be analyzed.

References

Ababneh ZQ, Alayassin AM, Aljarrah KM, Ababneh AM (2010) Measurement of natural and artificial radioactivity in powdered milk consumed in Jordan and estimates of the corresponding annual effective dose. *Radiat Prot Dosim* 138(2):278–283

Al-Absi E, Al-Abdullah T, Shehadeh H, Al-Jundi J (2015) ²²⁶Ra, ²²⁸Ra, and ⁴⁰K activity concentration in some vegetables consumed in Jordan, and resultant annual ingestion effective dose. *Radiat Prot Environ* 38:29–34

Alhaji E, Al-Masri MS, Al-Hamwi A, Al-Haleem MA, Hassan M, Mamish S, Al-Kharfan K, Khalily H, Alkhatib Y, Sheib Z (2014) Monitoring of ¹³⁷Cs fallout in Syrian environment. *Bull Environ Toxicol* 93:370–374

Al-Masri MS, Byrakdar ME, Mamish S, Al-Haleem MA (2004a) Determination of natural radioactivity in Euphrates river. *J Radioanal Nucl Chem* 261(2):349–355

Al-Masri MS, Mukallati H, Al-Hamwi A, Khalili H, Hassan M, Assaf H, Amin Y, Nashawati A (2004b) Natural radionuclides in Syrian diet and their daily intake. *J Radioanal Nucl Chem* 260(2):405–412

Andoh M, Nakahara Y, Tsudu S, Yoshida T, Matsuda N, Takahashi F, Mikami S, Kinouchi N, Sato T, Tanigaki M, Takamiya K, Sato N, Okumura R, Uchichori Y, Saito K (2015) Measurement of air dose rates over a wide area around the Fukushima Dai-Ichi nuclear power plant through a series of ca-borne surveys. *J Environ. Radioactivity* 139:266–280

Aoun M, El Samad O, Bou Khouzam R, Lobenski R (2015) Assessment of committed effective dose due to the investigation of ²¹⁰Po and ²¹⁰Pb in consumed Lebanese fish affected by a phosphate fertilizer plant. *J Environ. Radioactivity* 140:25–29

Awudu AR, Faanu A, Darko EO, Emi-Reynolds G, Aduko OK, Kpeglo DO, Lawluvi H, Kpodzro R, Ali ID, Obeng MK, Agyeman B (2012) Preliminary studies on ²²⁶Ra, ²²⁸Ra, ²²⁸Th and ⁴⁰K concentrations in foodstuffs consumed by inhabitants of Accra metropolitan area. *Ghana J Radioanal Nucl Chem* 291:635–641

Basu P, Sarangapani R, Sivasubramanian K, Venkatraman B (2015) Estimation of annual effective dose rate due to the ingestion of the primordial radionuclide ⁴⁰K for the population around the Kalpakkam nuclear site, Tamil Nadu, India. *Radiat Prot Environ* 38(2):14–22

Canbazoglu C, Doglu M (2013) A preliminary study on ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs activity concentrations in vegetables and fruits frequently consumed by inhabitants of Elazig region. *Turkey J Radioanal Nucl Chem* 295(2):1245–1249

Carvalho FP, Oliveira JM, Malta M (2011) Radionuclides in deep-sea fish and other organisms from the North Atlantic Ocean. *ICES J Mar Sci* 68(2):333–340

Catal EM, Ugrur A, Ozden B, Filizok I (2012) ²¹⁰Po and ²¹⁰Pb variations in fish species from the Aegean Sea and the contribution of ²¹⁰Po to the radiation dose. *Mar Pollut Bull* 64:801–806

- Dolhańczuk-Śródka A (2011) Estimation of external gamma radiation dose in the area of Bory Stobrawskie forests (PL). *Environ Monit Assess* 238:1–7
- El Samad O, Zahraman K, Baydoun R, Nasreddine M (2007) Analysis of radiocaesium in the Lebanese soil one decade after the Chernobyl accident. *J. Environ. Radioactivity* 92:72–79
- El Samad O, Baydoun R, El jeaid H (2010) Activity concentrations of polonium-210 and lead-210 in Lebanese fish. *Lebanese Science Journal* 11(2):39–45
- El Samad O, Baydoun R, Nsouli B, Darwish T (2013) Determination of natural and artificial radioactivity in soil at North Lebanon province. *J. Environ. Radioactivity* 125:36–39
- El Samad O, Aoun M, Nsouli B, Khalaf G, Hamze M (2014) Investigation of the radiological impact on the coastal environment surrounding a fertilizer plant. *J. Environ. Radioactivity* 133:69–74
- EL-Saharty AA (2013) Radioactive survey of coastal water and sediments across Alexandria and Rashid coasts. *Egypt J Aquat Res* 39:21–30
- Fons J, Zapata-Garcia D, Tent J, Llauro M (2013) Simultaneous determination of gross alpha, gross beta and ^{226}Ra in natural water by liquid scintillation counting. *J. Environ. Radioactivity* 125:56–60
- General Safety Requirements Part 3 (2014) Radiation protection and safety of radiation sources: international basic safety standards. IAEA Safety Standards for Protecting people and the environment. IAEA, Vienna
- Gwynn JP, Nalbandyan A, Rudolfsen G (2013) ^{210}Po , ^{210}Pb , ^{40}K and ^{137}Cs in edible wild berries and mushrooms and ingestion doses to man from high consumption rates of these wild foods. *J. Environ. Radioactivity* 116:34–41
- Harb S (2007) Measurement of the radioactivity of ^{238}U , ^{226}Ra , ^{210}Pb , ^{228}Th , ^{232}Th , ^{228}Ra , ^{137}Cs and ^{40}K in tea using gamma-spectrometry. *Radioanal Nucl Chem* 274(1):63–66
- Harb S (2015) Natural radioactivity concentration and annual effective dose in selected vegetables and fruits. *J Nucl Part Phys* 5(3):70–73
- IEC 1452:1995 – International Standard. Nuclear instrumentation—Measurement of gamma-ray emission rates of radionuclides – calibration and use of germanium spectrometers. Geneva, 1995
- Khandaker MU, Olatunji MA, Shuib KSK, Hakimi NA, Nasir NLM, Asaduzzaman KH, Amin YM, Kassim HA (2015) Natural radioactivity and effective dose due to the bottom sea and estuaries marine animals in the coastal waters around Peninsular Malaysia. *Radiat Prot Dosim* 167:1–5
- Kim CK, Chae JS, Choi HY, Choi SW, Kim DJ, Kim YJ, Lee DM, Park WJ, Yim SA, Yum JY (2012) Radiological impact in Korea following the Fukushima nuclear accident. *J. Environ. Radioactivity* 111: 70–82
- Kobeissi MA, El Samad O, Zahraman K, Milky S, Bahsoun F, Abumurad KM (2008) Natural radioactivity measurements in building materials in southern Lebanon. *J Environ Radioactivity* 99:1279–1288
- Kobeissi MA, El Samad O, Rachidi I (2013) Health assessment of natural radioactivity and radon exhalation rate in granites used as building materials in Lebanon. *Radiat Prot Dosim* 153(3):342–351
- Kut K, Berker S (2014) Measurement of natural radioactivity in beach sand of Akkuyu Mersin, Turkey. *Journal of Natural Sciences Research* 4(17):83–89
- Livingston HD, Povinec PP (2000) Anthropogenic marine radioactivity. *Ocean Coast Manag* 43:689–712
- Merz S, Shozugawa K, Steinhäuser G (2015) Analysis of Japanese radionuclide monitoring data of food before and after the Fukushima nuclear accident. *Environ Sci Technol* 49:2875–2885
- Nasreddine L, El Samad O, Hwalla N, Baydoun R, Hamzeh M, Parent-Massin D (2008) Activity concentrations and mean annual effective dose from gamma-emitting radionuclides in the Lebanese diet. *Radiat Prot Dosim* 131(4):545–550
- Pearson AV, Gaw S, Hermanspahn N, Glover CN (2016) Natural and anthropogenic radionuclide activity concentration in the New Zealand diet. *J. Environ. Radioactivity* 151:601–608
- Petrinec B, Strok M, Franic Z, Smodis B, Hamer DP (2012) Radionuclides in the Adriatic Sea and related dose-rate assessment for marine biota. *Radiat Prot Dosim* 1-11
- Rao DD, Baburajan A, Sudheendran V, Verma PC, Hegde AG (2010) Evaluation and assessment of 25 years of environmental radioactivity monitoring data at Tarapur (India) nuclear site. *J. Environ. Radioactivity* 101(8):630–642
- Roselli C, Desideri D, Rongoni A, Saetta D, Feduzi L (2013) Radioactivity in coffee. *J Radioanal Nucl Chem* 259(3):1813–1818
- Suriyanarayanan S, Brahmanandhan GM, Malathi J, Ravi Kumar S, Masilamani V, Shahul Hameed P, Selvasekarapandian S (2008) Studies on the distribution of ^{210}Po and ^{210}Pb in the ecosystem of Point Calimere Coast (Palk Strait), India. *J Environ Radioactivity* 99:766–771
- Tchokossa P, Olomo JB, Balogun FA, Adesanmi CA (2013) Assessment of radioactivity contents of food in the oil and gas producing areas in Delta State. *Nigeria Int J Sc Technol* 3(6):315–321
- Technical reports series No. 295 (1989) Measurement of radionuclides in food and the environment. International Atomic Energy Agency. IAEA, Vienna
- United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR. UNSCEAR 2000. Sources, effects and risks of ionizing radiation, (New York : United Nations) pp.4–5.
- Völgyesi P, Kis Z, Szabó Z, Szabó C (2014) Using the 186-keV peak for ^{226}Ra activity concentration determination in Hungarian coal-slag samples by gamma-ray spectroscopy. *J Radioanal Nucl Chem* 302: 375–383
- WHO, Guidelines for drinking-water quality (2011), fourth edition, Switzerland.