



Radioactive map of soil at Mount Lebanon province and external dose assessment

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Abstract

A total of 150 samples were collected from uncultivated areas at 87 locations distributed uniformly along the Mount Lebanon province. Measurements were carried out using gamma spectrometers with high purity germanium detectors. The activity concentrations of primordial radionuclides ranged from 14.0 ± 2.0 to 132.0 ± 8.0 Bq kg⁻¹ for ²³⁸U, from 5.4 ± 0.3 to 77.0 ± 3.0 Bq kg⁻¹ for ²³²Th, from 12.0 ± 1.0 to 227.0 ± 10.0 Bq kg⁻¹ for ²²⁶Ra, and from 10.7 ± 0.7 to 714.0 ± 16.0 Bq kg⁻¹ for ⁴⁰K, with average values of 37, 24, 46, and 206 Bq kg⁻¹, respectively. The average activity concentrations of ²³⁸U and ²³²Th were comparable to the worldwide average value, while that of ⁴⁰K was found to be half of the world average values stated in UNSCEAR (Sources, effects, and risks of ionizing radiation, United Nations, New York, 2000). The average value of ²²⁶Ra was found to be slightly higher than the worldwide value. The activity concentration of the ¹³⁷Cs at the measurement time in 2015 was found to lie between 0.30 ± 0.06 and 102.00 ± 2.00 Bq kg⁻¹ with an average of 23 Bq kg⁻¹. Surface activity concentrations were calculated, and radioactive maps were plotted. The average value of total absorbed dose rate and the annual effective dose was found to be lower than the world average values. Radium equivalent and external hazard index were within the international recommended values.

Keywords Primordial radionuclides · Artificial radionuclides · Absorbed gamma dose rate · Total effective dose · Radium equivalent · External hazard index

Introduction

The ionizing radiation exposure to humans arises from natural and artificial radionuclides. This exposure could be internal or external. External exposure arises mainly from natural radiation due to cosmic rays and terrestrial radioactivity. Terrestrial radionuclides are the primordial radionuclides, or radionuclides with half-lives comparable to the age of Earth, mainly ²³⁸U, ²³²Th, and their decay products as well as ⁴⁰K (Khan et al. 2012). They are present at trace levels in all ground formations (Awudu et al. 2012), and their occurrence is the main contributor,

about 85%, in the dose received by living organisms due to natural radiation. The concentration of primordial radionuclides in soil varies from region to another across the world depending on local geographical and geological factors (UNSCEAR 2000). Their distribution in soil depends on the radionuclides distribution in rocks from which they are originated and on the processes through which the soil is concentrated (Song et al. 2012). Man-made or anthropogenic radionuclides have been released to the environment by different nuclear activities. In the period 1950s–1960', 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. Artificial radionuclides may be transported with the radioactive plume for long distances across boundaries away from the accident zone. They could be deposited by fallout or wet deposition over soil. For this reason, the determination of natural and artificial radionuclides in soils has gained great interest during the last decades in different countries, and many studies were carried out (Al-Masri et al. 2006; Ababaneh et al. 2012; Ademola et al. 2014;

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Aközcan 2014; Bai et al. 2017; Dolhańczuk-Śródka 2012; Guiditto et al. 2015; Cho et al. 2014; Khan et al. 2012) in order to assess public exposure from terrestrial radiation, to establish radiation maps and baseline data necessary for the assessment of any radiation emergency, and for the detection and evaluation phases, and consequently, to take appropriate response.

In Lebanon, in the framework of a National Environmental Radiation Survey Program, many studies were carried out to determine the radioactivity levels in different compartment of the Lebanese environment (El Samad et al. 2007, 2013, 2017). These were the milestone for the public dose assessment of the Lebanese Population.

This study aims to determine the activity concentration of natural and artificial radionuclides in the Lebanese soil at Mount Lebanon province, deduce the surface activity concentration, and establish the radioactive maps of ^{238}U , ^{232}Th , ^{226}Ra , ^{137}Cs , and ^{40}K in the studied area, as well as to assess the external public exposure and radiation hazards due. This could be achieved through the estimation of total gamma dose rate, annual effective dose, radium equivalent, and external hazard index.

Materials and methods

Studied area

The Mount Lebanon province extends along Lebanon at a distance of 170 km between the Mediterranean coast and the western mountains chain till the highest mountain peak, 3088 m altitude. It surrounds the capital Beirut from three sides, south, east, and north. The studied area receives by substantial amount of precipitation and snow distributed over the winter season. It is characterized by oak and pine forests.

Sampling and sample preparation

A total of 150 samples were collected from different layers of soil down to 25 cm depth, at 87 uncultivated locations, distributed along the Mount Lebanon province. Sample gathering and measurements were carried out in the year 2015. Sampling locations are shown in Fig. 1. The coordinates of sampling sites were obtained based on the global positioning system. The collected samples were grinded, sieved, and homogenized. An aliquot from each sample was dried at 80 °C for 24 h and dry/wet ratio was calculated. Samples were prepared in the adopted counting geometry for gamma spectroscopy measurement (El Samad et al. 2013).

Gamma spectrometry analysis

Gamma spectrometry analysis was performed using three sets of gamma spectrometers with p-type coaxial high purity germanium detectors of relative efficiency 30, 40, and 50%. The laboratory is accredited by Hellenic Accreditation System ESYD, Certificate No. 788 dated January 20, 2012, according to ISO/IEC 17025 standards for calibration and testing laboratories. Energy calibration was carried out routinely by counting a standard multigamma sand sample (EG-ML-1195-11-1) from Eckert and Zeigler of 500 ml volume in polyethylene container. This is done with a tolerance 0.1 keV. The efficiency calibration was carried using mixed gamma—ray standard solution R8/31/32 of 500 ml volume from GE Healthcare Limited, and then efficiency curves were plotted for various compositions, densities, and geometries using EFFTRAN software taking in consideration attenuation and absorption factor (Vidmar et al. 2010). The estimated efficiency uncertainty is 3%. To test the performance of the detector in function of time, ^{152}Eu point source is counted periodically; the resolution (FWHM) and the full energy peak efficiency were checked at low and high energies. To subtract background radiation from samples spectra, an empty counting container was measured periodically. The sample counting time ranged from 130,000 to 173,000 s. The spectra were analyzed offline using the Genie 2000 software from Canberra Version V3.1b. Uranium-238 and Thorium-232 were determined from the gamma line of their daughters $^{234\text{m}}\text{Pa}$ and ^{228}Ac , respectively. Radium-226 was determined from its gamma line at 186.2 keV after correction for ^{235}U interference (Völgyesi et al. 2014). As well, ^{137}Cs and ^{40}K were determined from their corresponding gamma lines 661.7 and 1460.8 keV, respectively. The surface activity concentration per unit area (Bq m^{-2}) was then calculated. The inverse distance weighted (IDW) method was used to interpolate and predict unmeasured surfaces over investigated areas. Using this geometric method, the value of the variable at the unsampled locations is estimated based on the data from surrounding locations. The weights for IDW are proportional to the square distance between the prediction points and the observation points (Kravchenko and Bullock 1999; Yao et al. 2013).

Results and discussion

Radioactivity concentration in soil

The ranges of activity concentration and the average values for natural radionuclides and ^{137}Cs in the analyzed

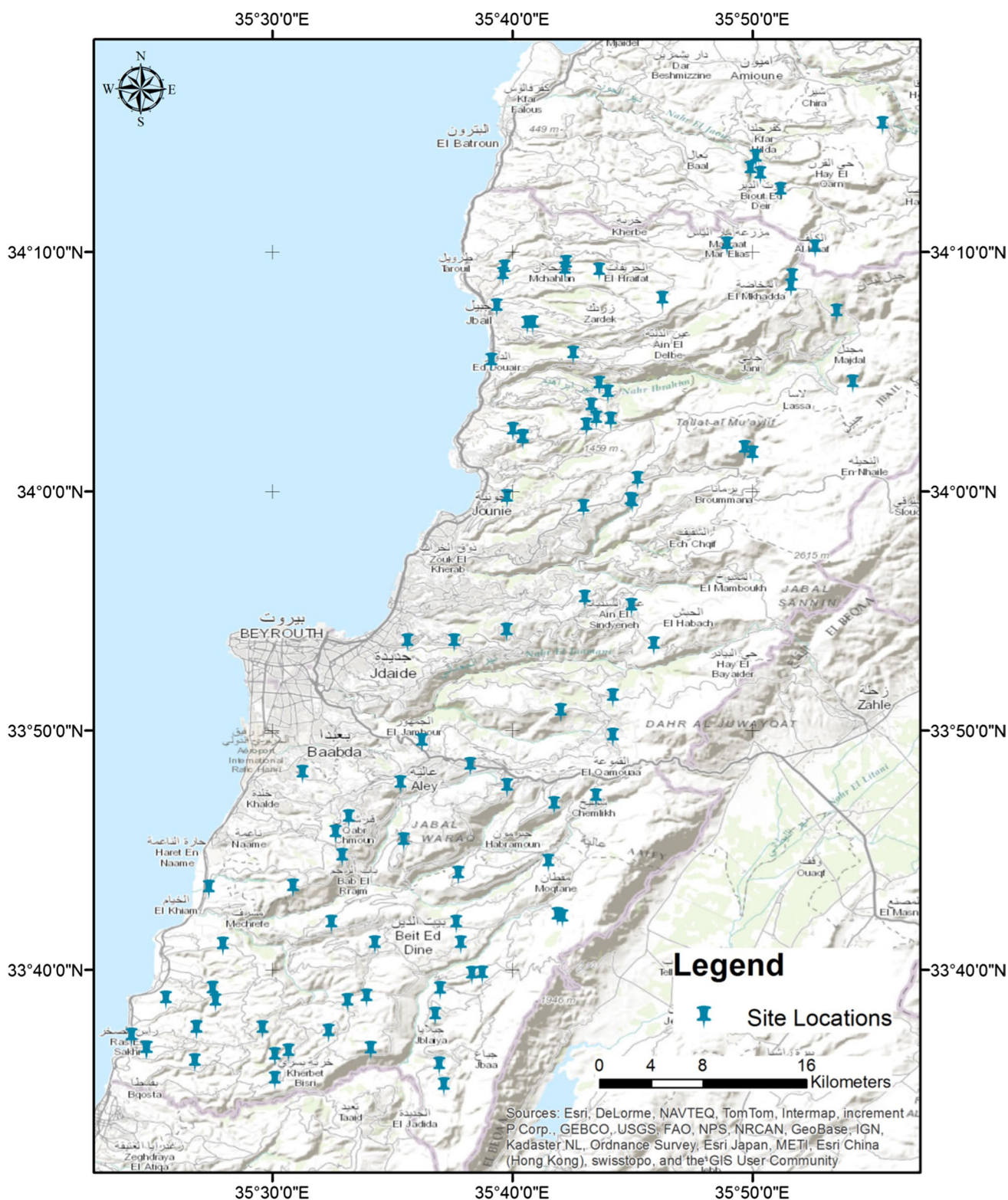


Fig. 1 Map of Mount Lebanon province with sampling locations

samples are presented in Table 1. The average values were found to be 37 Bq kg⁻¹ for ²³⁸U, 24 Bq kg⁻¹ for ²³²Th, 46 Bq kg⁻¹ for ²²⁶Ra, and 206 Bq kg⁻¹ for ⁴⁰K.

Values were comparable to those reported in other Lebanese province (El Samad et al. 2013). The average activity concentration of ²³⁸U and ²³²Th was comparable

Table 1 Ranges of determined activity concentrations (Bq kg⁻¹), average values and ranges of calculated surface activity concentrations (kBq m⁻²)

Radionuclide	Range activity concentration (Bq kg ⁻¹)	Average activity concentration (Bq kg ⁻¹)	Range surface activity concentration (kBq m ⁻²)
²³⁸ U	14.0 ± 2.0–132.0 ± 8.0	37	0.10–8.10
²³² Th	5.4 ± 0.3–77.0 ± 3.0	24	0.11–7.00
²²⁶ Ra	12.0 ± 1.0–227.0 ± 10.0	46	0.05–9.21
⁴⁰ K	10.7 ± 0.7–714.0 ± 16.0	206	0.60–38.00
¹³⁷ Cs	0.30 ± 0.06–102.00 ± 2.00	23	0.01–4.51

to the worldwide average values, 35 and 30 Bq kg⁻¹, respectively, while that of ²³²Th was lower than values reported in USA, 35 Bq kg⁻¹ (NCRP No. 45 1975) and in Europe, 34 Bq kg⁻¹ (Trevisi et al. 2012). The average value of ²²⁶Ra was found to be slightly higher than the worldwide value and European value 35 and 36 Bq kg⁻¹, respectively, while it was found comparable to US value, 40 Bq kg⁻¹ (NCRP No. 45 1975). The value obtained for ⁴⁰K was found to be half of the world average values stated in UNSCEAR (2000), 400 Bq kg⁻¹ and lower than that reported in USA and Europe, 370 and 483 Bq kg⁻¹, respectively. The variation in the activity concentrations of natural radionuclides in soils in different locations

across the world differs depending on soil type, geochemical composition, geological, and geographical conditions of the studied areas (Mir and Rather 2015). The average activity concentration of the ¹³⁷Cs was 23 Bq kg⁻¹. The main cause to the presence of ¹³⁷Cs in the Lebanese soil could be the deposition by fallout during earlier nuclear weapons testing in 1960s and Chernobyl accident in 1986. The ¹³⁴Cs was not detected in the analyzed samples, due to its short half-life. The content of ¹³⁷Cs was comparable to previous studies carried out in other Lebanese province (El Samad et al. 2013). However, it was lower than values reported in some European countries such as Poland and Greece (Dolhańczuk-Śródka 2012;

Table 2 Comparison of the average activity concentrations determined in the analyzed samples with values determined at North Lebanon and values reported by other countries

Country	²³⁸ U (Bq kg ⁻¹)	²³² Th (Bq kg ⁻¹)	²²⁶ Ra (Bq kg ⁻¹)	⁴⁰ K (Bq kg ⁻¹)	¹³⁷ Cs (Bq kg ⁻¹)	References
Kuwait	18	15	–	385	–	Alazemi et al. (2016)
Iraq	34.8	18.8	–	289.2	1.16	Taqi et al. (2016)
Turkey (Küçük)	–	20.5	48.4	744.8	3.3	Aközcan (2014)
Turkey (Mersin)	–	34.3	27.1	370.5	18.6	Karataş et al. (2016)
Turkey (Kilis)	–	15	16.1	206	9.5	Canbazoğlu et al. (2013)
Nigeria	55.3	26.4	–	505.1	–	Ademola et al. (2014)
Saudi Arabia	22.98	21.24	–	244.97	12.71	Al Mugren (2015)
Syria	15	24	–	116	–	Al-Masri et al. (2006)
Jordan	20	29.4	29.8	312.2	8.02	Ababaneh et al. (2012)
Ghana	23.19	31.10	–	143.78	2.88	Darko et al. (2015)
Taiwan	24.22	26.39	–	435.74	7.10	Tsai et al. (2008)
China	40	49	37	580	5.6	Bai et al. (2017)
Italy	73	47	–	623	16.2	Guiditto et al. (2015)
Greece	56.64	–	32.44	565.62	275.79	Kioupi et al. (2015)
Poland	6.94	7.86	–	147.6	178.9	Dolhańczuk-Śródka (2012)
USA	–	35	40	370	–	NCRP no. 45
Europe	–	34	35	483	–	Trevisi et al. (2012)
Worldwide	35	30	35	400	–	UNSCEAR (2000)
North Lebanon	27	24	–	246	21	El Samad et al. (2013)
Mount Lebanon	37	24	46	206	23	Present study

Kioupi et al. 2015), comparable to values determined in some areas in turkey (Karataş et al. 2016), and it was found higher than the content detected in Ghana and Taiwan, and in other Arab countries and Middle Eastern countries (Ababaneh et al. 2012; Aközcan 2014; Bai et al. 2017; Canbazoglu et al. 2013; Darko et al. 2015; Al Mugren 2015; Taqi et al. 2016; Tsai et al. 2008). These variations could be attributed to many factors, mainly the soil texture and the weather conditions such as the rainfall during and after the radioactive cloud passage (El Samad et al. 2007). Table 2 represents a comparison between activity concentration of natural and artificial radionuclides determined in this study and previous data determined at other Lebanese area and those reported by other countries.

Vertical distribution

For vertical distribution studies of natural radionuclides and ^{137}Cs , seven locations were selected, four samples at four depths (0–3, 3–8, 8–15, and 15–25 cm) were collected from each location. In order to study the variation between the data obtained for ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K at different depths, an analysis of variance test, one-way ANOVA was applied. The applied statistical test showed that there is no significant variation between activity concentrations at various soil layers as $p > 0.05$ and $F < F_{\text{crit}}$. This confirms with other studies carried out in other countries (Ramzaev et al. 2006; Belivermiş et al. 2010; Belivermiş 2012). Concerning the ^{137}Cs , theoretically data should show exponential pattern specially that the soil at the studied area is characterized by low carbonate content and high amounts clays minerals and organic matters that played an important role in maintaining the cesium at the top layer (El Samad et al. 2007; Belivermiş 2012). However, the vertical distribution of ^{137}Cs fits exponential function from layer 3–8 to 15–25 cm, with the exception of the value obtained at the first layer. This could be attributed to the environmental conditions, and to the soil erosion and degradation that have affected and disturbed the composition of the top layer over years, specially that sampling was carried out 3 decades after Chernobyl accident. This caused an underestimation of the activity concentration of ^{137}Cs by 40–50%. The vertical

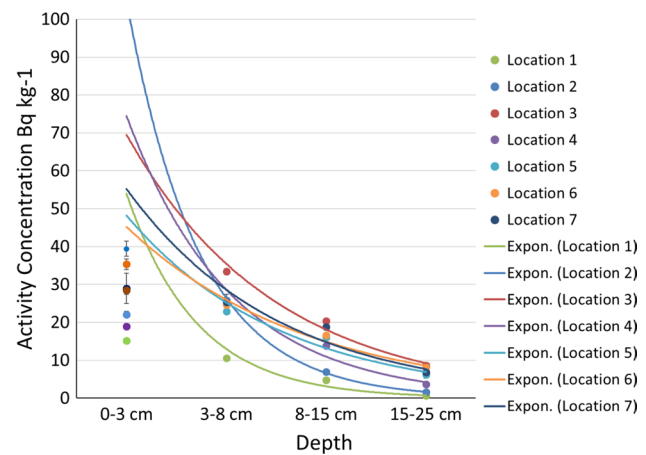


Fig. 2 Vertical distribution of ^{137}Cs for the studied locations

distribution of ^{137}Cs activity concentration at the studied locations was presented in Fig. 2.

Radiation maps

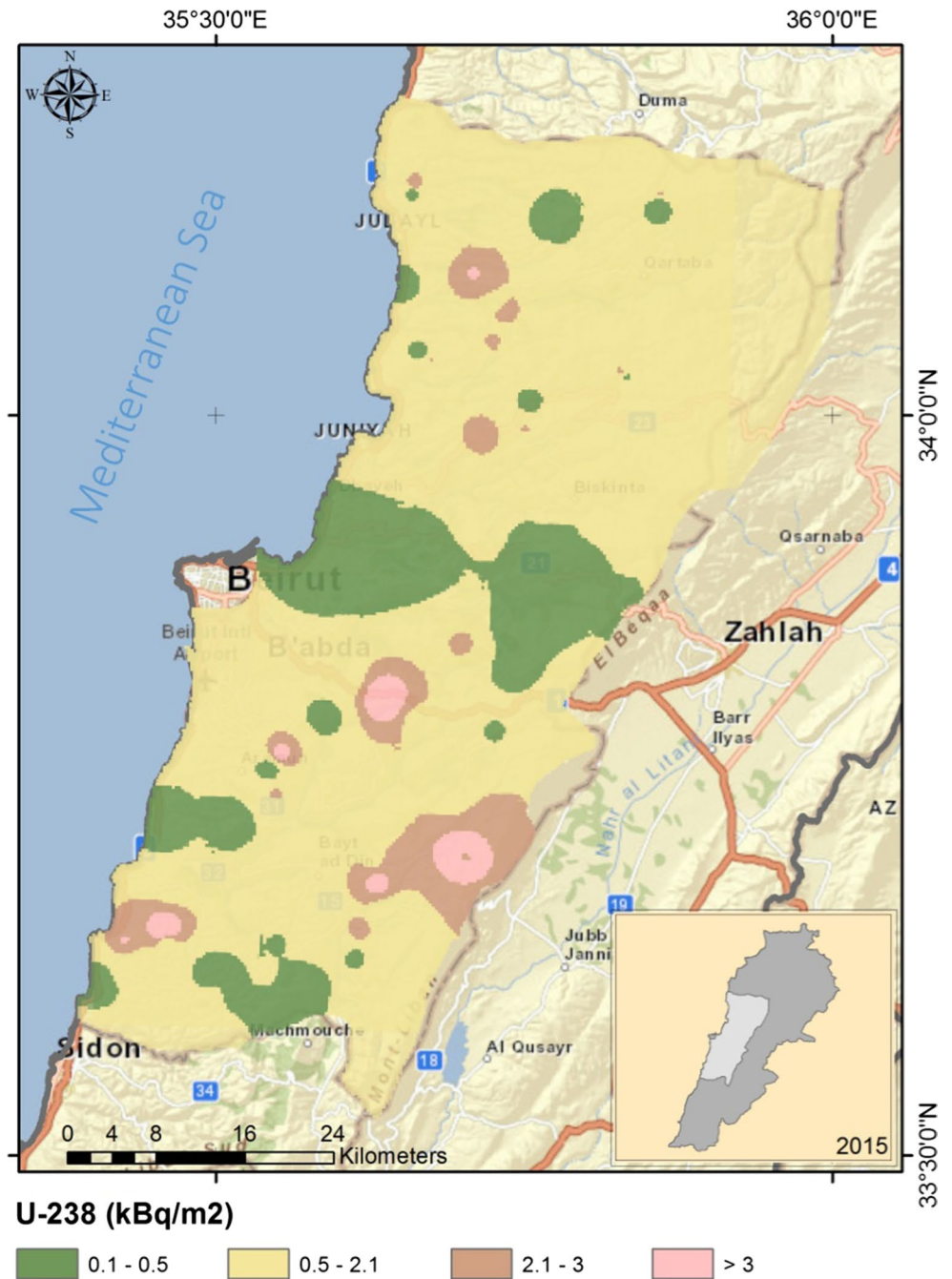
The ranges of surface activity concentrations for ^{238}U , ^{232}Th , ^{226}Ra , ^{40}K , and ^{137}Cs are presented in Table 1. Values were comparable to those calculated in other Lebanese province (El Samad et al. 2013). These surface activity concentrations were used to establish radioactive maps for Mount Lebanon province for natural and anthropogenic radionuclides. Maps for ^{238}U , ^{232}Th , ^{226}Ra , ^{40}K , and ^{137}Cs are represented in Figs. 3, 4, 5, 6, and 7, respectively.

Assessment of radiological hazards

Absorbed dose rate and annual effective dose

In order to assess the radiation risk, the total absorbed dose rates D (nGy h^{-1}) in air at 1 m above ground level due to the presence of natural and artificial radionuclides were estimated taking into account their activity concentrations and the corresponding dose rates per unit activity concentrations (UNSCEAR 2000; Kurnaz et al. 2011;

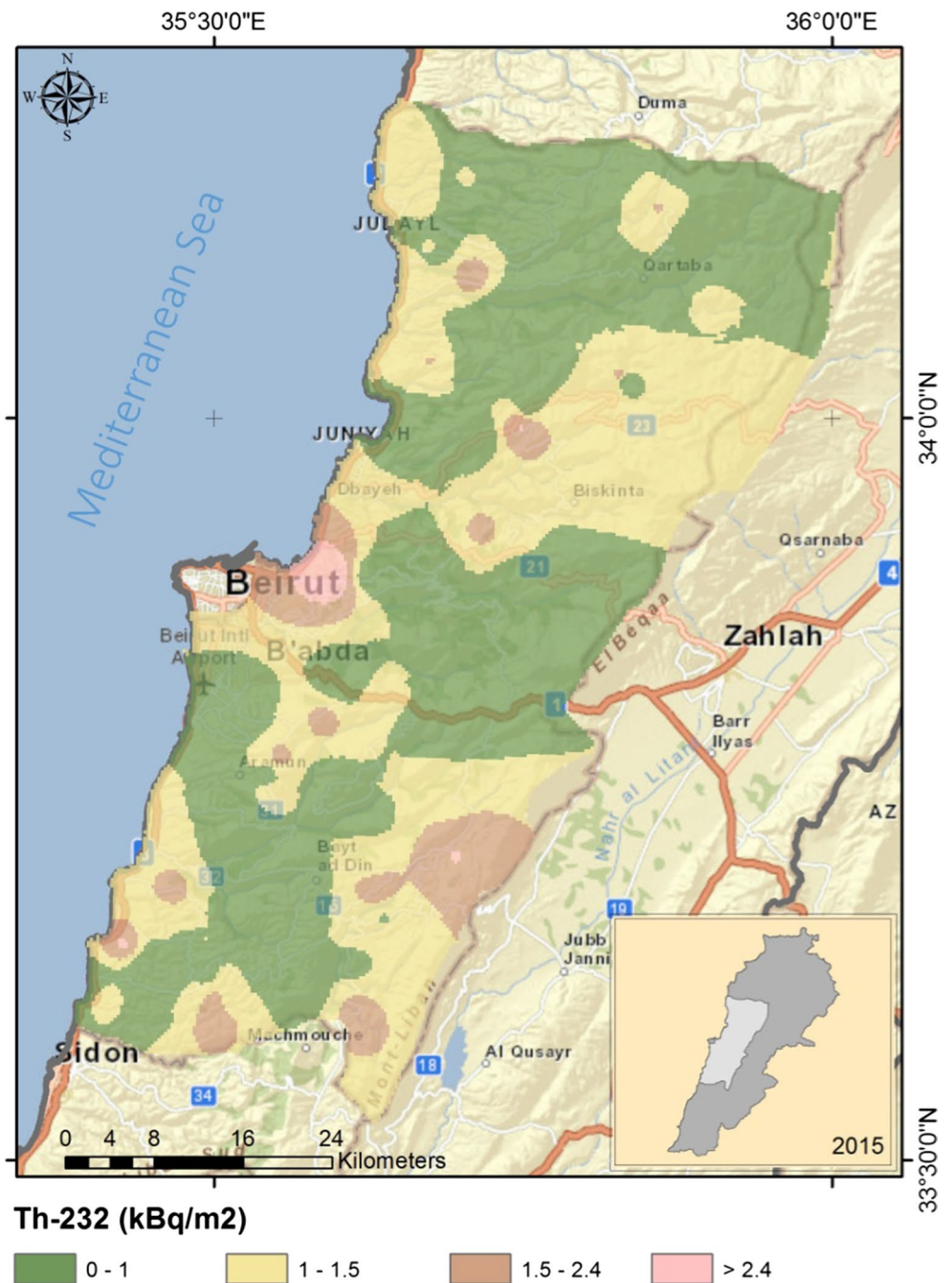
Fig. 3 Activity concentration per surface area for ^{238}U



Ademola et al. 2014). Values varied up to 91 nGy h^{-1} with an average of 40 nGy h^{-1} , where 99% of this dose is due to the natural radiation. The average value was found to be lower than the world average 57 nGy h^{-1} and the average value estimated in other Lebanese province 55 nGy h^{-1} (El Samad et al. 2013) and comparable to value reported in USA, 47 nGy h^{-1} .

The annual effective doses were calculated in order to assess the biological hazard to which public in the sampling locations are exposed. This was achieved using the conversion coefficient of 0.7 Sv Gy^{-1} from absorbed dose in air to effective dose received by an adult, and 0.2 for the outdoor occupancy factor. Values varied between 5 and $112 \text{ } \mu\text{Sv year}^{-1}$ with an average of $49 \text{ } \mu\text{Sv year}^{-1}$,

Fig. 4 Activity concentration per surface area for ^{232}Th

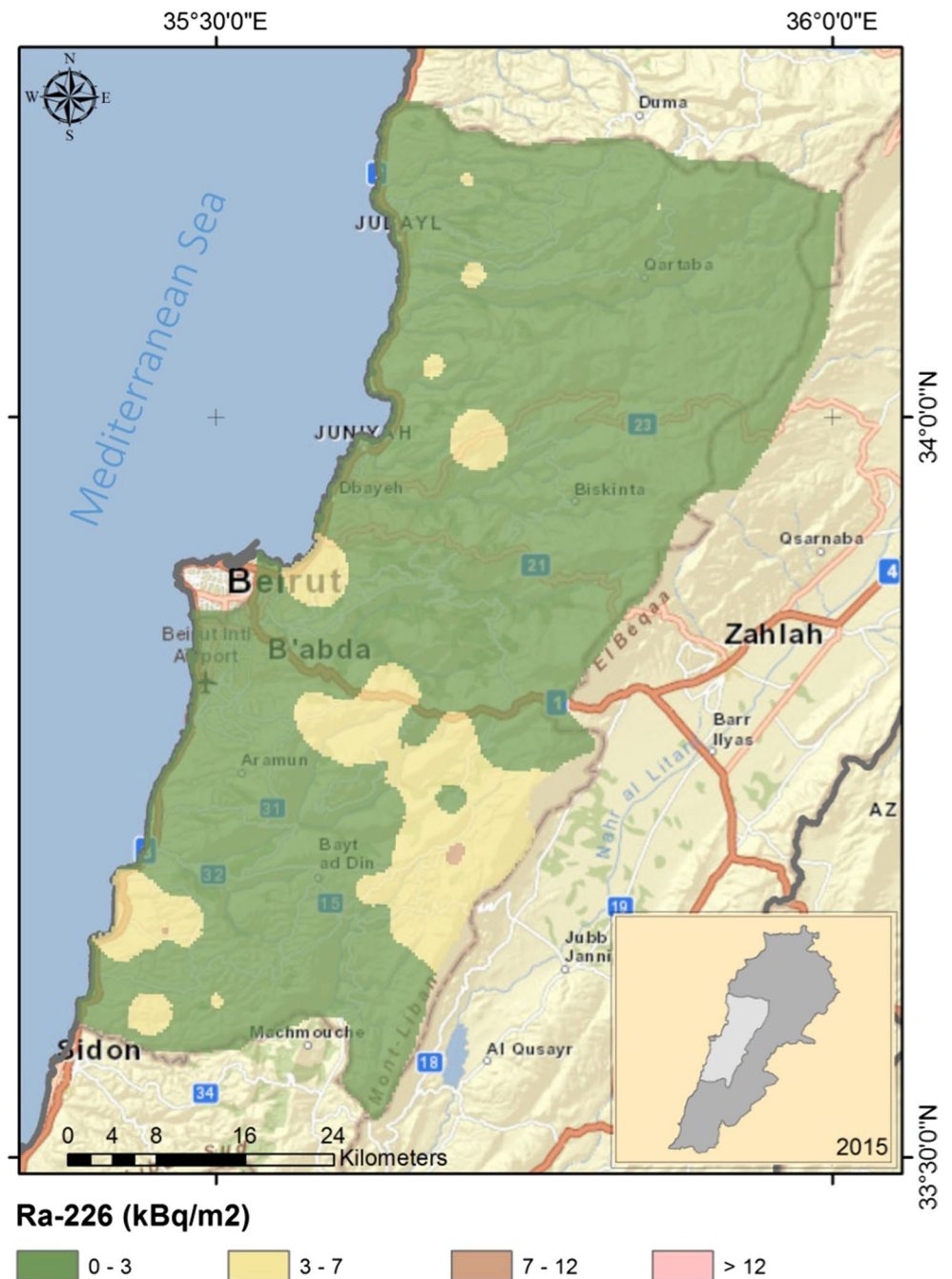


lower than the value calculated in North Lebanon province $69 \mu\text{Sv year}^{-1}$, as well as lower than the world average $70 \mu\text{Sv year}^{-1}$ and values reported in Japan and USA, 400 and $210 \mu\text{Sv year}^{-1}$ respectively.

The total absorbed dose rate and the annual effective dose were comparable to those reported in Jordan, Kuwait, Saudi Arabia, and Ghana (Ababaneh et al. 2012; Alazemi et al.

2016; Darko et al. 2015; Al Mugren 2015). Values were higher than those estimated in Poland and some area in Turkey (Canbazoğlu et al. 2013; Dolhańczuk-Śródka 2012) and lower than those in Küçük and Mersin in Turkey, Nigeria, China, and Taiwan (Ademola et al. 2014; Aközcan 2014; Bai et al. 2017; Karataşh et al. 2016; Tsai et al. 2008).

Fig. 5 Activity concentration per surface area for ^{226}Ra



Radium equivalent (Ra_{eq})

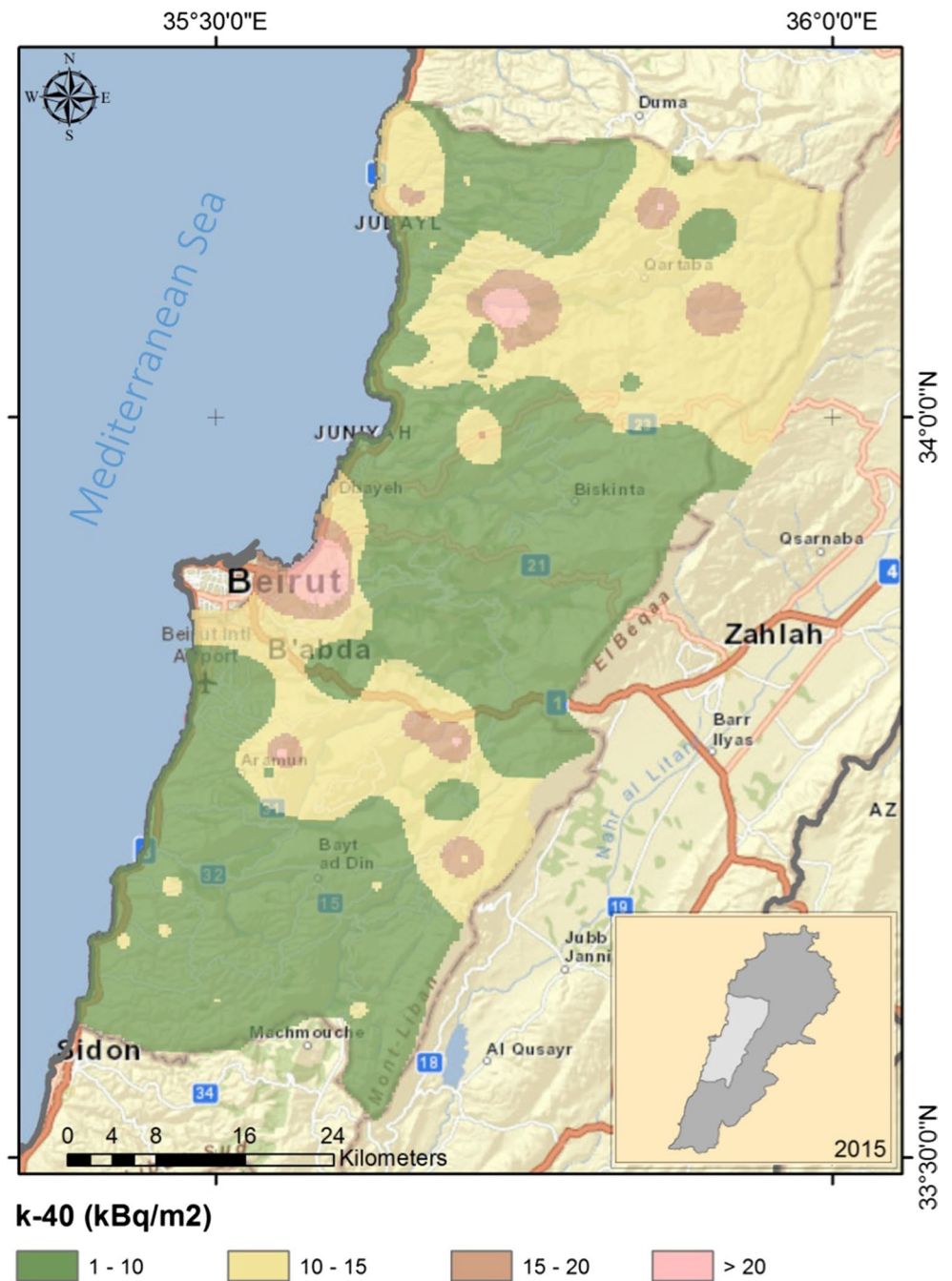
In order to assess the radiation hazards caused by gamma ray of specific radionuclides of ^{226}Ra , ^{232}Th , and ^{40}K , the most widely used hazard index, radium equivalent Ra_{eq} in Bq kg^{-1}

was calculated at all sampling locations. This was carried out using the following equation (Beretka and Mathew 1985)

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \tag{1}$$

where C_{Ra} , C_{Th} , C_K are the activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in Bq kg^{-1} , respectively. Values were found

Fig. 6 Activity concentration per surface area for ⁴⁰K

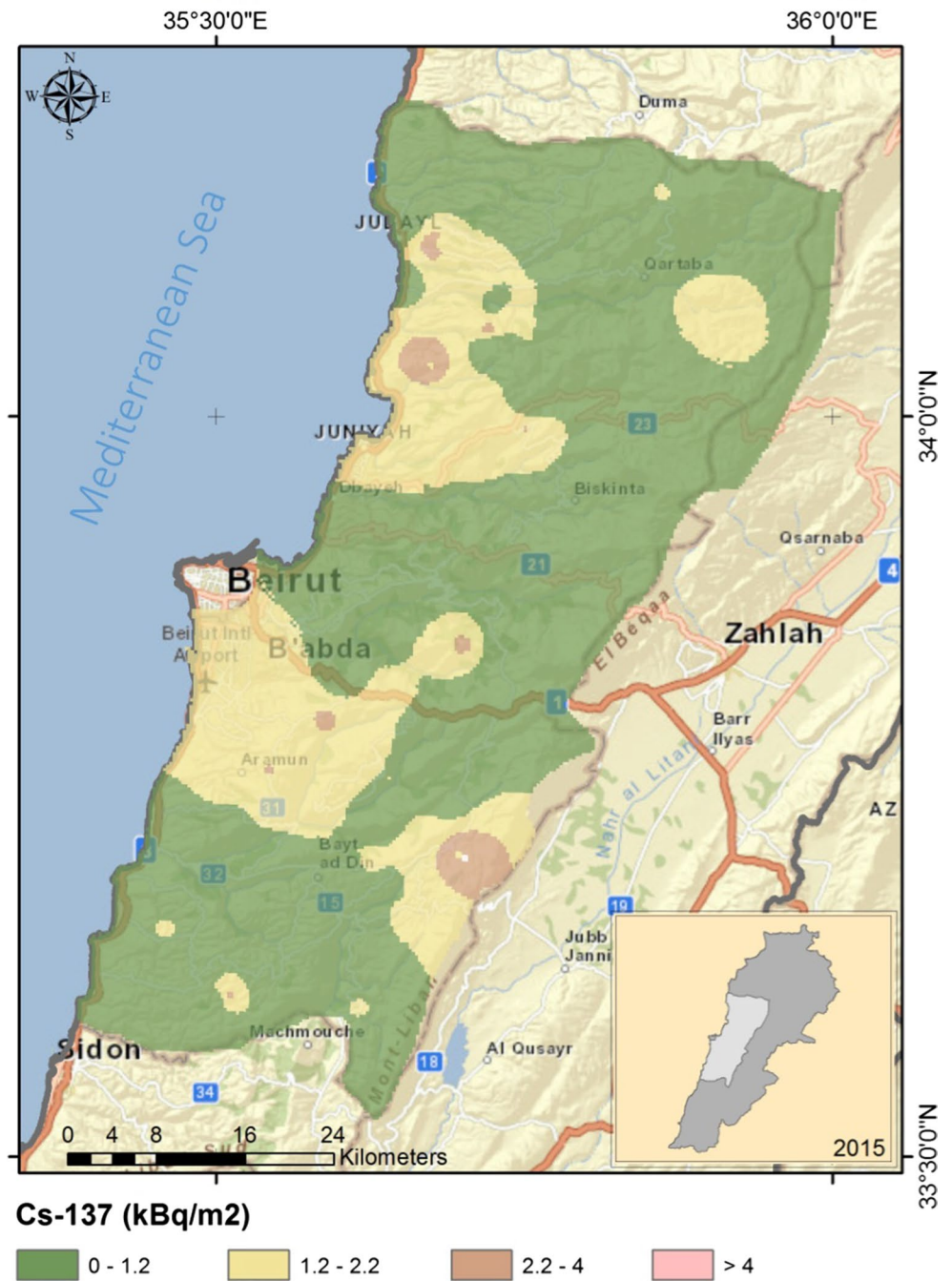


lower than the recommended value 370 Bq kg^{-1} (UNSCEAR 2000; Baykara et al. 2011; Aközcan 2014). They ranged from 13 to 304 Bq kg^{-1} with an average of 95 Bq kg^{-1} .

The average value of $R_{a_{eq}}$ was found comparable to that calculated in Taiwan (Tsai et al. 2008), higher than

that in Kuwait and Saudi Arabia (Alazemi et al. 2016; Al Mugren 2015), and lower than that in Küçük-Turkey, Nigeria, and China (Ademola et al. 2014; Aközcan 2014; Bai et al. 2017).

Fig. 7 Activity concentration per surface area for ^{137}Cs



External hazard index (H_{ex})

The external hazard index was calculated for all analyzed samples as follow (Beretka and Mathew 1985; Sroor et al. 2002).

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \quad (2)$$

All results were found less than the maximum value 1 mSv year⁻¹ which corresponds to the upper limit of Ra_{eq} (370 Bq kg⁻¹). Values ranged from 0.03 to 0.8 mSv year⁻¹ with an average value of 0.3 mSv year⁻¹.

Table 3 shows a comparison between radiation hazards parameters calculated in this study and those reported in other countries.

Conclusion

A total of 150 samples were collected from 87 locations distributed uniformly at the Mount Lebanon province. Gamma analysis was performed, and activity concentrations of natural and artificial radionuclides were

Table 3 Comparison of radiation hazards parameters with values determined at North Lebanon and values reported in other studies

Country	Dose rate (nGy h ⁻¹)	Annual effective dose (μSv year ⁻¹)	Ra _{eq} (Bq kg ⁻¹)	H _{ex} (mSv year ⁻¹)	References
Kuwait	33.16	40.7	68.5	0.19	Alazemi et al. (2016)
Turkey (Küçük)	66.77	80	129.8	0.37	Aközcan (2014)
Turkey (Mersin)	51	62	–	–	Karataş et al. (2016)
Turkey (Kilis)	25	31	–	–	Canbazoğlu et al. (2013)
Nigeria	66.3	81.3	132.14	0.36	Ademola et al. (2014)
Saudi Arabia	32.30	40	69.52	0.2	Al Mugren 2015
Jordan	44.2	54.3	–	–	Ababaneh et al. (2012)
Ghana	35.49	40	–	–	Darko et al. (2015)
Taiwan	46.63	57.19	98.18	0.27	Tsai et al. (2008)
China	76.7	83.1	138	0.37	Bai et al. (2017)
Poland	14	17.4	–	–	Dolhańczuk-Śródka (2012)
Japan	50	400	–	–	Furukawa and Shingaki (2012), Cho et al. (2017)
USA	47	210	–	–	NCRP Report No. 45 (1975), NCRP Report No. 160 (2009)
Worldwide	57	70	370	1	UNSCEAR (2000)
North Lebanon	55	69	–	–	El Samad et al. (2013)
Mount Lebanon	40	49	95	0.3	Present study

determined. Results were comparable to those determined in other Lebanese province. Radioactive maps for the studied area were established. The radiological hazards parameters, including absorbed gamma dose rate, total annual effective dose, radium equivalent, and external hazard index, were calculated. All results were within the limit of international recommended values. The data obtained are the basis of a national radioactivity baseline level and could be used as a reference for future studies on radionuclide distribution in the country.

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