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Soil Radioactive Maps for Natural and Artificial Radionuclides and Estimation of Public Dose at East and North Bekaa – Lebanon

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Abstract: The long-lived radionuclides, ²³⁸U, ²³²Th and their decay products and ⁴⁰k are the major sources of radioactivity in soil. Cesium isotopes are the main artificial radionuclides deposited over soil by fallout or wet deposition. In addition, cosmic rays at ground levels are major contributor to external dose rate. In this work public dose arising from terrestrial and cosmic radiation was assessed. Sixty-five samples were collected from uncultivated areas, situated at East and North Bekaa province, cleaned, dried and prepared in the adopted counting geometry and measured by gamma spectrometer with high purity germanium detectors. The average activity concentrations of primordial radionuclides ²³⁸U, ²³²Th and ⁴⁰K were comparable to worldwide values; 35 Bq kg⁻¹, 30 Bq kg⁻¹ and 400 Bq kg⁻¹ respectively, while that of ²²⁶Ra was higher than worldwide average 35 Bq kg⁻¹. Surface activity concentrations were deduced and radioactive maps were established using inverse distance weighted (IDW) method. The average activity concentration of the artificial radionuclide, ¹³⁷Cs was found to be lower than that reported in soil samples studied at other Lebanese provinces. The average absorbed dose rate from terrestrial radiation was 65 nGy h⁻¹ while the average annual effective dose was 79 µSv y⁻¹, values lower than worldwide average value stated in UNSCEAR- 0.5 mSv y⁻¹ and higher than those reported in studies carried out at other Lebanese provinces. Four stations belonging to the radiation early warning network system fixed in the studied area were used for continuous monitoring of ambient air dose rate including cosmic rays. These are composed of dose rate meters Gamma Tracer XL2 from Saphimo-Bertin, with Geiger Muller tubes. The average annual ambient gamma dose rate was 0.61 mSv y ¹, a value double worldwide value 0.3 mSv y⁻¹. The average total annual public dose due to external exposure arising from both terrestrial radiation and cosmic rays was 690 μ Sv y⁻¹, a value lower than the sum of worldwide values stated by UNSCEAR.

Keywords: Radioactive maps, terrestrial radiation, ambient air dose rate, total annual effective dose.

1 Introduction

Humans are continuously exposed to natural background radiation at different levels due to the presence of natural radionuclides in the various environmental components. The worldwide average public dose due to natural radiation is 2.4 mSv y⁻¹. This value arises from external exposure and internal exposure attributed to ingestion and inhalation. Terrestrial radiation and cosmic radiation are main contributors of external exposure and they take a large part of the population dose [1]. The long-lived radionuclides, ²³⁸U, ²³²Th and their decay products and ⁴⁰k constitute the essential source of natural radioactivity in soil, and their concentrations differs from one place to another across the world depending on the geological conditions and types of

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rocks. Higher radiation levels are associated with igneous rocks such as granite, and lower levels with sedimentary rocks [2,3]. The testing of nuclear weapons and the nuclear accidents leaded to the release of anthropogenic radionuclides to the environment [4]. Cesium isotopes are the main artificial radionuclides deposited over soil by fallout or wet deposition. Thus many studies were carried out in different countries to determine the natural and artificial radioactivity in soil, and to assess public dose attributed to terrestrial radiation [5,6,7,8,9].

Cosmic rays are attributed to high-energy particles originating in outer space. These cosmic radiations interact with nuclei present in the atmosphere producing a cascade of interactions and secondary reactions that contribute to cosmic ray exposure. These cosmic particle flux varies with



latitude and altitude. The cosmic rays contribute to about 13% of the total annual effective dose received by the population [10]. Hence, the assessment of cosmic ray exposure at ground level is of great interest to estimate the total public dose. For this reason, several data for ambient air dose rate values recorded in different countries were published [11,12,13].

In Lebanon several studies were carried out to assess public dose from internal and external exposure [14], and some researches focused on establishing the radioactive maps for different provinces, and on assessment of exposure from terrestrial radiation [15, 16]. This work is a complementarity of previous studies, it aims to establish the radioactive maps of soil for the analyzed radionuclides (²³⁸U, ²³²Th, ²²⁶Ra, ¹³⁷Cs, ⁴⁰K) at East and North Bekaa province, as well as to assess the total annual dose to which public is exposed due to terrestrial radiation and ambient air dose rate including cosmic rays.

2 Materials and methods

2.1 Study area

The Bekaa province is located about 30 km east of Beirut and it descends from the highest peak in Lebanon (Qornet el Saouda) at an altitude 3080 meters to reach the Syrian borders and the mountains of Baalbek in the north and the east. It extends between the west and east Lebanese Mountain chains. This area is characterized by harsh and semi-arid climate. The studied region, where samples were collected, occupies the east and north of Bekaa valley, the most important agricultural area in the country.

2.2 Sampling

Sixty-five soil samples were collected during the year 2015 from uncultivated area distributed along East and North of Bekaa province. Sampling was carried out from the first layer of the terrestrial depth (0-3cm) using a stainless steel template of 25x20 cm. The coordinates of the sampling sites were defined based on the global positioning system. For the determination of the ambient air dose rate, including cosmic rays at ground level, four dose rate meters with Geiger Muller tubes were fixed in the populated areas of the studied region to monitor air continuously. The sampling locations and the XL2 stations, for dose rate records are illustrated in figure 1.

2.2 Sample preparation

The soil samples were cleaned physically from roots and stones. Then they were subjected to grinding, sieving and homogenization. After that, samples were prepared in 500 ml polyethylene containers, in the adopted counting geometry for the gamma spectroscopy measurement. An aliquot was taken from each sample and dried for 24 hours at 80° C. Then the dry/wet ratio was calculated to express the result in Bq kg⁻¹ dry weight.

2.3 Measurement

The measurement of soil samples was carried out at the Gamma Spectroscopy Laboratory at the Lebanese Atomic Energy Commission (LAEC). The laboratory is accredited since 2012 according to ISO/IEC 17025 standards for testing and calibration laboratories. Two sets of gamma spectrometers with high purity germanium detectors of relative efficiency 40% and 50% were used in this study. The detectors were calibrated for energy and efficiency [15,16]. Efficiency curves were plotted using EFFTRAN software taking in consideration attenuation and absorption factors (Vidmar et al. 2010). Accuracy of results was tested through the periodic application of quality control procedure. Analysis of spectra was carried out using Genie 2000 software version V3.1b from Canberra. Results were expressed in Bq kg⁻¹. These activity concentrations were then used to calculate the surface activity concentration per unit area expressed in Bq m⁻². Inverse distance weighted (IDW) method was applied to interpolate and predict unmeasured surfaces in order to plot radioactive maps [17]. For the measurement of ambient dose rate, four stations belonging to the radiation early warning network system, consisted of twenty-six stations distributed along the Lebanese territory, were fixed in the studied area. These are composed of dose rate meters GammaTracer XL2 from Saphymo- Bertin with two Geiger Muller tubes each, for low and high dose rate measurements. Dose rates are recorded continuously and data are transmitted to the central station at LAEC via binary SMS messages and via FTP 4G transmission, where they are converted to digital quantities.



Figure 1. Sampling sites and locations of the radiation early warning network system stations.

2.4 Dose estimation

The total public dose due to external exposure was estimated taking into account the dose attributed to terrestrial radiation and the ambient dose rate in air including cosmic rays. The external absorbed dose rate (nGy h^{-1}) at 1 m above ground due to radioactivity in soil was estimated according to UNSCEAR 2000 [18] as shown in Equation (1).

$$D = aC_U + bC_{Th} + cC_K + dC_{Cs}$$
(1)

Where a, b, c and d are the dose rates per unit activity concentrations of U, Th, K and Cs, whose values are 0.462, 0.604, 0.0417 and 0.0.3 nGy h^{-1} per Bq kg⁻¹, and C_U, C_{Th}, C_K, C_{Cs} are the activity concentrations in Bq kg⁻¹ of ²³⁸U, ²³²Th, ⁴⁰K and ¹³⁷Cs respectively.

The annual effective dose in μ Sv y⁻¹ was then deduced taking in consideration the conversion factor 0.7 Sv Gy⁻¹ and the occupancy factor that specifies the fraction of total time spent outdoors 0.2 (UNSCEAR, 2000)

3 Results and Discussion

3.1 Activity concentration and radioactive maps

The average activity concentration of ²³⁸U, ²³²Th and ⁴⁰K were comparable to worldwide values stated in UNSCEAR 2000, 35 Bq kg⁻¹, 30 Bq kg⁻¹ and 400 Bq kg⁻¹ respectively. Their average values were 35 Bq kg⁻¹, 34 Bq kg⁻¹ and 385 Bq kg⁻¹, with ranges between 14 ± 1 Bq kg⁻¹ and 82 ± 2 Bq kg⁻¹ for 238 U, 4.5 ± 0.3 Bq kg⁻¹ and 61 ± 3 Bq kg⁻¹ for 232 Th and between 42 \pm 2 Bq kg^-1 and 868 \pm 25 Bq kg^-1 for $^{40}K.$ The average activity concentration of ²²⁶Ra was 66 Bq kg⁻¹, a value higher than worldwide value, 35 Bq kg⁻¹ and that reported in other Lebanese provinces [15, 17]. The values varied between 22 ± 3 Bq kg⁻¹ to 140 ± 14 Bq kg⁻¹. The average values of ²³²Th and ⁴⁰K were higher than those calculated in North Lebanon and Mount Lebanon provinces, while that of ²³⁸U was comparable to that in other studied Lebanese provinces. The variation in activity concentrations of primordial radionuclides between different regions and their distribution in soils are dominated by the type of parent bed rocks from which they are originated [19]. The majority of bed rocks in Lebanon are sedimentary rocks mainly limestone, rich in mineral content and carbonate. However, the mineral and organic composition of Lebanese soil varies from site to site within the Lebanese territory [20]. This could explain the high activity concentration of ²²⁶Ra that was found double the worldwide average value, as highest radium content could be dependent on mineral composition of parent bed rock [3]. Cesium -137 was determined in the analyzed soil samples and its activity concentration ranged between 0.3 ± 0.04 Bq kg⁻¹ and 53 ± 1 Bq kg⁻¹. This range was comparable to other values reported in studies carried out in other Middle-East countries such as Syria, Jordan, Turkey and Greece [21, 22, 23, 24]. The average activity

concentration of ¹³⁷Cs was 12 Bq kg⁻¹, a value comparable to that determined in North Lebanon province, when corrected for radioactive decay to the year 2015, and lower than the value determined in Mount Lebanon province for samples collected in same year 2015. This could be attributed to the difference in rainfall depositions during and immediately after the radioactive cloud passage, as this cesium could be attributed to Chernobyl accident. The topographical characteristics of the country imposes a less rainfall in Bekaa region through the year [20]. The surface activity concentrations were expressed in kBq m⁻², ranges are shown in the plotted radioactive maps for ²³⁸U, ²³²Th, ²²⁶Ra, ⁴⁰K and ¹³⁷Cs presented in figures 2, 3, 4, 5 and 6 respectively.

3.2 Dose assessment

The calculated absorbed dose rate due to terrestrial radiation, ranged from 19 nGy h⁻¹ to 122 nGy h⁻¹ with an average of 65 nGy h⁻¹. A value double that reported in Saudi Arabia and Kuwait [8, 25], and higher than the worldwide value and those calculated in Mount and North Lebanon provinces. While the annual effective dose varied between 23 μ Sv y⁻¹ and 149 μ Sv y⁻¹. The calculated average value was found to be 79 μ Sv y⁻¹, a value lower than worldwide average value stated in UNSCEAR- 0.5 mSv y⁻¹ and higher than those reported in other Lebanese provinces and Jordan, while it is double that calculated in Saudi Arabia and Kuwait [3, 23, 25]. Absorbed dose rate and annual effective dose due to terrestrial radiation are presented in table 1.



Figure 2. Surface activity concentration of ²³⁸U





Figure 3. Surface activity concentration of ²³²Th



Figure 5. Surface activity concentration of ²²⁶Ra



Figure 4. Surface activity concentration of 40 K



Figure 6. Surface activity concentration of ¹³⁷Cs

 Table 1. Absorbed dose rate, annual effective dose due terrestrial radiation at East and North of Bekaa province and comparison with other Lebanese provinces and some Arab countries

| Absorbed | Annual | References |
|----------------|--|--|
| dose rate | effective dose | |
| $(nGy h^{-1})$ | (µSv y ⁻¹) | |
| 19-122 | 23-149 | Present |
| | | study |
| 65 | 79 | Present |
| | | study |
| 55 | 69 | [15] |
| 40 | 49 | [16] |
| | | |
| 57 | 500 | [18] |
| 32.30 | 40 | [25] |
| 33.16 | 40.7 | [3] |
| 44.2 | 54.3 | [23] |
| | dose rate (nGy h^{-1}) 19-122 65 55 40 57 32.30 33.16 | dose rate (nGy h ⁻¹)effective dose (μ Sv y ⁻¹)19-12223-1496579556940495750032.304033.1640.7 |

The gamma dose rate recorded at four stations over the whole year 2015, varied between 63 nSv h⁻¹ and 115 nSv h⁻¹ with an average value of 70 nSv h⁻¹. The variation between recorded values could be attributed to the different meteorological conditions prevailing in the studied area over the year [18]. The rainfall and snow cover could cause fluctuation in the readings due to the possible washout of radon progeny from air that induce short increase of gamma dose rate in air [18]. However, the applied statistical test, analysis of variance test, ANOVA showed that this variation is not significant. The monthly average value recorded dose rate at the four stations are shown in figure 7. The average value was higher than the worldwide value 30.9 nSv h⁻¹ stated by UNSCEAR 2008. The difference in the recorded ambient radiation levels at various places could be attributed to the variation in the geographical location (latitude and altitude). The annual effective dose ranged between 567 µSv y^{-1} and 1156 μ Sv y^{-1} with an average 690 μ Sv y^{-1} , a value higher than worldwide value, 270 μ Sv y⁻¹ [18].





The total public annual effective dose due to external exposure arising from both terrestrial radiation and ambient dose in air including cosmic rays was found to vary between 549 μ Sv y⁻¹ and 1156 μ Sv y⁻¹ with an average 742 μ Sv y⁻¹. A value slightly lower than the sum of worldwide average value for terrestrial radiation, 0.5 mSv y⁻¹, and that for cosmic rays 0.3 mSv y⁻¹ stated in UNSCEAR.

4 Conclusion

Total Sixty-five soil samples were collected from uncultivated areas at east and north of the Bekaa province. Activity concentration of natural and artificial radionuclides were determined. Radioactive maps for ²³⁸U, ²³²Th, ²²⁶Ra, ¹³⁷Cs and ⁴⁰K were established. Absorbed dose rate and annual effective dose were calculated. Ambient dose rate including cosmic rays were recorded. Public dose due to external exposure arising from terrestrial and cosmic rays was assessed. Average value was found slightly lower than the sum of worldwide average values 0.5 mSv y⁻¹ and 0.3 mSv y⁻¹ respectively as stated in UNSCEAR.

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