

# Estimation of Annual Effective Dose due to Intake of Radionuclides from Total Diet of Lebanese Population

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**Abstract:** Natural and artificial radionuclides could enter the food chain and reach the human being via ingestion. Hence, several studies were conducted to assess public dose due to food ingestion. In this work, a total of 388 food samples were collected and analyzed during the period 2016-2020 in order to determine activity concentrations of present radionuclides and to estimate annual effective dose due to ingestion. The samples covered the majority of the constituents of food groups of the total Lebanese diet, fruits, vegetables, meat, chicken, fish, several milk powder brands, canned food, cereals and pulses. Cesium-137 was detected in milk samples in a range of  $0.30 \pm 0.04$  Bq  $\text{kg}^{-1}$  to  $2.00 \pm 0.08$  Bq  $\text{kg}^{-1}$ , and its content was lower than minimum detectable activity in other food species. The main natural radionuclides contributing in the public dose were  $^{40}\text{K}$  in various food species and  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in fish. Their activity concentrations were presented and discussed. The total annual doses due food ingestion in the years 2016-2020 were  $0.247$  mSv  $\text{year}^{-1}$ ,  $0.296$  mSv  $\text{year}^{-1}$ ,  $0.311$  mSv  $\text{year}^{-1}$ ,  $0.202$  mSv  $\text{year}^{-1}$ ,  $0.171$  mSv  $\text{year}^{-1}$  respectively with an average value of  $0.245$  mSv  $\text{year}^{-1}$ , a value comparable to the worldwide average value of internal dose due to ingestion  $0.3$  mSv  $\text{year}^{-1}$ . ANOVA test showed that there are no significant differences between estimated doses over years. Spearman correlation test proved that there is strong correlation between annual effective dose and consumption rate.

**Keywords:** Food ingestion, Radionuclides intake, Annual effective dose, ANOVA test, Spearman correlation test.

## 1 Introduction

Since many decades, studies had shown that different kinds of foodstuff contain certain level of radioactivity. This is attributed to the natural and artificial radionuclides that enter the food chain through many pathways, and then reach human beings via ingestion [1]. When ingested, radionuclides accumulate in specific organs it delivers radiation dose. For example,  $^{40}\text{K}$  accumulates in muscles and  $^{137}\text{Cs}$  in bones [2]. As reported by UNSCEAR 2000, the worldwide average value of internal dose due to food ingestion is  $0.3$  mSv  $\text{year}^{-1}$ , about eighth of the total annual worldwide average dose due to natural background radiation,  $2.4$  mSv  $\text{year}^{-1}$ . The public exposure through food ingestion is mainly attributed to natural radionuclides, uranium series, thorium series and  $^{40}\text{K}$  [3]. Radionuclides can reach the food chain mainly from soil to food crops, for this reason their activity concentration differs from species to another based on plants roots uptake and depending on their concentration in soil that differs upon various geographic and geological factors [3,4]. Fruits and vegetables could intake radionuclides by dry or wet

deposition, and from soil through soil-plant transfer. The use of fertilizers could increase the activity concentration of primordial radionuclides such as  $^{238}\text{U}$  and  $^{232}\text{Th}$  and their decay products, and  $^{40}\text{K}$  in soil and hence in agricultural products. The International Commission on Radiological Protection-ICRP stated that phosphate fertilizers had doubled the human's exposure to radionuclides from food ingestion [5]. In addition, artificial radionuclides could be transferred to the food chain, as they were released to the environment by past nuclear test weapons and nuclear accidents, and deposited by dry fallout or wet deposition over soil and plants. The main anthropogenic radionuclides are Cesium isotopes, strontium isotopes and Iodine-131. Potassium-40 and  $^{137}\text{Cs}$  are easily transferred to milk by grass-cow-milk route [6]. Meat and chicken can assimilate radionuclides through feeding of living animals by contaminated food. Radionuclides in sea are assimilated in various seafood [7]. Polonium-210 and  $^{210}\text{Pb}$  in fish are the main contributors to radiation dose received by humans [8]. Many studies were conducted to assess public dose due to food ingestion. In Lebanon, the first study to estimate the public dose from food ingestion was carried out in 2007

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[9]. Later, researches were conducted to screen some selected species that were representing the prime constituents of Lebanese diet [4]. In this work, a comprehensive study of all food groups was carried out during the last five years, through the determination of activity concentrations of natural and artificial gamma emitters in various kinds of fruits, vegetables, meat, chicken, fish, several milk powder brands, canned food, cereals and pulses, in addition to the determination of  $^{210}\text{Po}$  in fish, in order to assess the public dose from food ingestion. More species were taken in consideration in this study to cover the total diet. Values obtained were presented and discussed. The impact of COVID-19, prevailing in the year 2020, on the sampling procedures and analysis was highlighted. ANOVA statistical test was applied to study the variation of total dose over years, while spearman correlation test was carried out to discuss the relationship between activity concentration and consumption rate with the effective dose.

## 2 Materials and Methods

### 2.1 Sampling and Sample Preparation

A total of 388 samples of food groups of the total Lebanese diet were collected from local markets during the period 2016-2020. Eighty-five samples were gathered in 2016 and eighty-one samples in 2017. The number of collected samples was increased in 2018 to ninety-four, and it was eighty-six in 2019. During the year 2020, the period of the prevailing pandemic COVID-19, the number of gathered samples was reduced to forty-two, due to the lock down and the restrictions imposing the decrease of the number of competent staff to minimum. Samples were chosen regardless the country of origin, except for wheat and milk powder, where samples of different provenance were analyzed. Analyzed samples covered different kind of fruits and vegetables, meat, chicken, milk powder, canned food, grain and pulses. Benthic and pelagic fish samples were collected twice a year from the fresh catch at three coastal cities. Samples were prepared for measurement as collected without any processing or cooking procedures. For the measurement of gamma emitters by gamma spectroscopy technique, samples were crushed and homogenized. Then prepared in the adopted counting geometry, 500 ml polyethylene cylindrical container. For the determination of  $^{210}\text{Po}$  in fish sample, radiochemical separation was applied,  $^{208}\text{Po}$  tracer was used, followed by deposition on silver disk [4, 10].

### 2.2 Measurement

For the measurements of gamma emitting radionuclides, gamma spectrometers with high purity germanium detectors of relative efficiency 30%, 40% and 50% were

used. Measurements were conducted at the accredited gamma spectroscopy laboratory at the Lebanese Atomic Energy Commission. Detectors were housed in thick lead to reduce background radiation and copper layer to attenuate x-rays emitted by lead. Energy calibration and efficiency calibration were conducted. Background spectra were measured routinely to correct the activity concentrations of natural radionuclides. Genie 2000 V3.1a software from Canberra was used. A low-level extended range HPGe detector with Beryllium window, was utilized to measure  $^{210}\text{Pb}$  directly from its corresponding gamma line at 46.5 keV. The  $^{137}\text{Cs}$  and  $^{40}\text{K}$  were determined from their relevant gamma lines at 661.6 keV and 1460.8 keV respectively.

For the measurement of  $^{210}\text{Po}$  in fish samples, the prepared silver disks were counted using Canberra alpha spectrometer with passivated implanted planar silicon detectors of resolution 10.5 at 5486 keV, active area 450  $\text{mm}^2$ , mounted in a vacuum chamber, and connected to standard electronics to display spectra. Collected spectra showed the peak of  $^{210}\text{Po}$  at 5.15 MeV and that of  $^{208}\text{Po}$  tracer at 5.3 MeV. The activity concentrations were calculated and corrected for chemical recovery and for radioactive decay starting from sampling time.

### 2.3 Dose Estimation

The annual effective dose received by adults due to food ingestion was calculated as described in ICRP1996 [11], taking into account the annual consumption rate of a specific species, the activity concentration of the detected radionuclide and the dose conversion factor of ingestion intake [4].

### 2.4 Statistical Analysis

Analysis of variance test, ANOVA, was conducted to study the variation between estimated doses due to ingestion over the five years [4]. Spearman correlation test was applied to data set of each year in order to discuss the relationship or correlation between each of activity concentration and consumption rate with the calculated dose. The test referred to the calculation of spearman correlation coefficient designated by ( $\rho$ ) that will vary between -1 and +1. A zero value indicates that there is no correlation, the strength of correlation increases as getting closer to -1 and +1.

## 3 Results and Discussion

### 3.1 Activity Concentration

The anthropogenic radionuclide  $^{137}\text{Cs}$  was detected in milk samples, of different brands and origins, with activity concentration ranging from  $0.30 \pm 0.04 \text{ Bq kg}^{-1}$  to  $2.00 \pm 0.08 \text{ Bq kg}^{-1}$ . Values comparable to those reported in the

screening conducted previously [4], and to those stated by other countries [12,13,14,15]. While its activity concentration in the plurality of analyzed samples over 5 years was below the minimum detectable activity  $MDA=0.2 \text{ Bq kg}^{-1}$ . Among natural radionuclides,  $^7\text{Be}$  was detected in leafy vegetables with an activity concentration varying between  $6.0\pm 0.4 \text{ Bq kg}^{-1}$  in lettuce and  $20.2\pm 0.6 \text{ Bq kg}^{-1}$  in grape leaf. As declared by UNSCEAR 2000, the content of radionuclides in fruits and vegetables could be reduced by processing procedures such as washing, peeling and cooking [3]. The activity concentration of  $^{226}\text{Ra}$  and its daughters  $^{214}\text{Bi}$ ,  $^{214}\text{Pb}$  and  $^{210}\text{Pb}$  were below minimum detectable activity in all analyzed samples.

The main natural radionuclides contributing in the public dose due to food ingestion were  $^{40}\text{K}$  in various food species and  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in fish. The activity concentration of  $^{40}\text{K}$  was found comparable to values detected in the species selected and covered by the previous screening [4]. In fruits, its minimum content was  $36\pm 1 \text{ Bq kg}^{-1}$  determined in raspberry while its maximum content was  $117\pm 4 \text{ Bq kg}^{-1}$  in banana. Values were comparable to those stated in studies carried out in neighboring countries [16,17] and higher than values reported in east and southeast Asia [18, 19]. While in vegetables, the activity concentration of  $^{40}\text{K}$  varied between  $40\pm 2 \text{ Bq kg}^{-1}$  in onion and  $350\pm 10 \text{ Bq kg}^{-1}$  in Chard. Determined values were comparable to those detected in Syria and Japan [16,18], higher than values reported in Turkey [20] and lower than activity concentrations measured in Jordan and Tanzania [21, 22].

The content of  $^{40}\text{K}$  in milk powder samples of different country of origin ranged from  $220\pm 2 \text{ Bq kg}^{-1}$  to  $720\pm 20 \text{ Bq kg}^{-1}$ . Detected values are higher than those reported in other Arab countries [12, 16, 23, 24]. Its activity concentration in grain and pulses laid between  $93\pm 3 \text{ Bq kg}^{-1}$  in wheat imported from Canada and  $475\pm 14 \text{ Bq kg}^{-1}$  in beans, indicating content higher than that reported in other Arab countries [16, 25, 26]. The minimum value determined in white bread was  $73\pm 3 \text{ Bq kg}^{-1}$  while the maximum value was  $145\pm 5 \text{ Bq kg}^{-1}$  in brown bread.

Regarding canned food, the lowest activity concentration  $21.3\pm 0.3 \text{ Bq kg}^{-1}$  was detected in honey, a value lower than those reported in other Arab countries [27,28] and comparable to values determined in European countries [29, 30]. Highest value was determined in tomato paste with activity concentration  $412\pm 12 \text{ Bq kg}^{-1}$ . The activity concentration of  $^{40}\text{K}$  in beef meat and chicken ranged from  $60\pm 1 \text{ Bq kg}^{-1}$  to  $100\pm 2 \text{ Bq kg}^{-1}$ , and from  $75\pm 3 \text{ Bq kg}^{-1}$  to  $110\pm 3 \text{ Bq kg}^{-1}$  respectively. Values higher than those declared in other Arab countries [31, 32, 33]. The difference between activity concentrations of  $^{40}\text{K}$  determined in this work and those detected in other countries could be attributed to the diversity of prevailing background levels, climate and agricultural conditions and techniques [3].

Concerning fish samples, chosen species were the most commonly consumed by Lebanese population. Potassium-40 tenor varied between  $178\pm 5 \text{ Bq kg}^{-1}$  in Mugil sp. collected from Saida and  $530\pm 28 \text{ Bq kg}^{-1}$  in Mugil sp from the Capital Beirut. The determined activity concentrations were comparable to those declared in some countries [34] and higher than others [32, 35]. The activity concentration of  $^{210}\text{Po}$  ranged between  $20.0\pm 0.7 \text{ Bq kg}^{-1}$  in *Siganus rivulatus* and  $64\pm 2 \text{ Bq kg}^{-1}$  in *Pagellus erythrinus*. While that of  $^{210}\text{Pb}$  varied from  $10.0\pm 0.4 \text{ Bq kg}^{-1}$  in Mugil sp to  $26\pm 2 \text{ Bq kg}^{-1}$  in *Diplodus sargus*. These results were found higher than the values measured in other studies [32, 34, 36].

### 3.2 Annual Effective Dose

The annual effective doses due to  $^{40}\text{K}$  intake from food in the years 2016-2020 were  $0.209 \text{ mSv year}^{-1}$ ,  $0.258 \text{ mSv year}^{-1}$ ,  $0.282 \text{ mSv year}^{-1}$ ,  $0.168 \text{ mSv year}^{-1}$ , and  $0.143 \text{ mSv year}^{-1}$  respectively, with an average value of  $0.211 \text{ mSv year}^{-1}$ . A value comparable to that stated by Nassreddine et al. 2008, as both studies covered the total Lebanese diet. The slight difference between yearly values could be attributed to the selected species and their corresponding consumption rate [3]. However, ANOVA test showed that no significant variation between values over years. The lowest value was determined in 2020 as less consumed species were covered, and the number of analyzed samples was decreased due to Covid-19 pandemic.

The annual effective doses due to ingestion of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  from fish during 2016-2020 were  $0.038 \text{ mSv year}^{-1}$ ,  $0.038 \text{ mSv year}^{-1}$ ,  $0.029 \text{ mSv year}^{-1}$ ,  $0.034 \text{ mSv year}^{-1}$  and  $0.028 \text{ mSv year}^{-1}$  respectively, with an average value of  $0.033 \text{ mSv year}^{-1}$ . ANOVA test showed no significant variation over years.

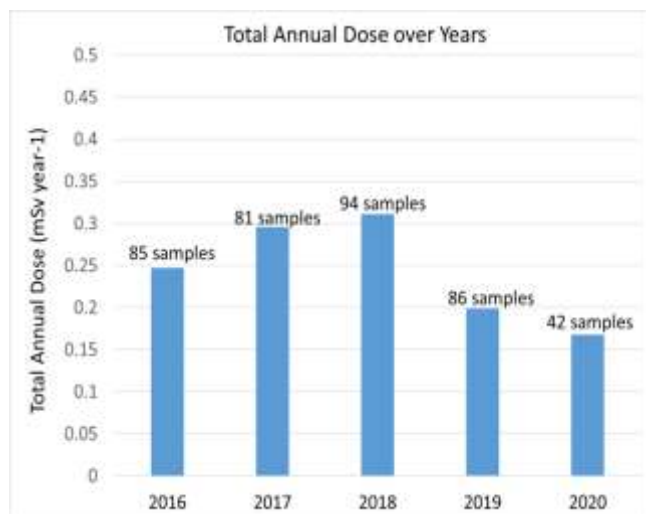
Spearman test showed that the calculated annual effective dose is highly dependent on the consumption rate of each of food species rather than the activity concentration of radionuclide. Calculated correlation coefficient ( $\rho$ ) proved that there is strong correlation between the annual effective dose and the consumption rate in all years. Moderate to low relationship between dose and activity concentration was recognized. Based on Spearman test, strong, moderate and low relationships are assigned to ( $\rho$ ) values of 0.7-1, 0.5-0.7, and 0.3-0.5 respectively. Table 1 represents the determined ( $\rho$ ) in each year.

**Table 1:** Spearman's correlation coefficients ( $\rho$ ) between each of activity concentration of radionuclides and consumption rate with the calculated effective dose.

Year	Activity concentration and calculated effective dose ( $\rho$ )	Consumption rate and calculated effective dose ( $\rho$ )
2016	0.61	0.86
2017	0.40	0.87
2018	0.64	0.83

2019	0.26	0.88
2020	0.66	0.79

The yearly total annual doses due to food ingestion during the years of 2016-2020 were 0.247 mSv year<sup>-1</sup>, 0.296 mSv year<sup>-1</sup>, 0.311 mSv year<sup>-1</sup>, 0.202 mSv year<sup>-1</sup> and 0.171 mSv year<sup>-1</sup> respectively. Figure 1 showed the total annual effective dose with the number of samples analyzed by year.



**Fig. 1:** Total annual effective dose with years and the corresponding number of samples.

The highest value was found in 2018 with larger number of samples analyzed, i.e. more species were taken in consideration. The value obtained in 2017 is slightly higher than that in 2016 even with lower number of samples. This could be attributed to the difference of chosen species and consumption rate [3]. Lowest value was determined in 2020 where minimum number of samples were collected and analyzed due to COVID-19 pandemic.

The average total dose due to food ingestion over 5 years was equal to 0.245 mSv year<sup>-1</sup>. A value comparable to the worldwide average value of internal dose due to ingestion 0.3 mSv year<sup>-1</sup> [3] and higher than that reported in 2018 [4]. This could be attributed to the fact that more species were added and analyzed in the current study to cover the food groups of the total diet. This is a complementary work for the previous one, which covered some selected species representing main constituents of Lebanese diet.

Table 2 represents the average annual effective dose determined in this work, the values reported by previous studies and by other countries. The difference between the values reported in this work and those stated by other countries could be attributed to the differences in the species of consumed local food and the differences in

consumption rates of each category between various population [3]

**Table 2:** Annual effective dose due to <sup>40</sup>K intake and annual effective dose

Country	Annual effective dose due to <sup>40</sup> K intake	Total Annual effective dose	References
Lebanon	0.211	0.245	Current study
Lebanon	0.186	-----	Nasreddine et al. (2008)
Lebanon	-----	0.077	El Samad et al. (2018)
Worldwide average	0.178	0.3	UNDCEAR 2000
Spain	-----	0.362	Hernández et al. (2004)
Sri Lanka	0.030–0.050	-----	Jayasinghe et al. (2019)
India	0.093 <sup>a</sup>	0.601 <sup>b</sup>	Basu et al. (2015) Giri et al. (2013)
Vietnam	-----	0.320	Van et al. (2019)
Japan	0.170	-----	Terada et al. (2020)
Arabian Gulf	-----	0.059	Ababneh et al. (2018)

<sup>a</sup> Basu et al. (2015)

<sup>b</sup> Giri et al. (2013)

## 4 Conclusions

In this study around 388 samples constituting the food groups of Lebanese Diet were analyzed over five years. It was found that <sup>40</sup>K is the main contributor in ingestion dose, in addition to <sup>210</sup>Po and <sup>210</sup>Pb in fish. The average total annual effective dose was equal to 0.245 mSv year<sup>-1</sup>, a value comparable to the worldwide average value of internal dose due to ingestion 0.3 mSv year<sup>-1</sup>. ANOVA test showed that there is no significant variation between determined values over years, while spearman test proved that the calculated effective dose is more dependent to the consumption rate of food species rather than the activity concentration of radionuclide. The COVID-19 pandemic had affected the work and inhibit to carry out the planned sampling and analysis due to lock down and restrictions that imposed minimum competent staff.

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