Chapter 33
Dietary Exposure to Heavy Metals and Radionuclides in Lebanon: A Total Diet Study Approach

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Introduction

The continuous surveillance of food safety has lagged behind in Lebanon, a Mediterranean country with a territory of 10,452 km^2 and a population estimated to be around four million. Limited reports are available suggesting the presence of particular contaminants in specific food items [1]. However, the magnitude and the severity of the dietary exposure of the population to food chemicals have not been appropriately addressed.

The estimation of dietary exposure to food contaminants combines data on the levels of the contaminant in particular foods with data on the quantities of those foods consumed by the population in question. The World Health Organization (WHO) supports the use of the total diet study (TDS) as one of the most cost-effective means of achieving accurate dietary exposure estimates, and the WHO's GEMS/Food Programme has encouraged all countries, and particularly developing ones, to conduct a TDS as a matter of public health significance. A TDS provides a first step exposure assessment based on food consumption surveys and point towards priority nutrients or contaminants that need to be further investigated. In this context, a TDS has been initiated in 2004 in Lebanon and has provided the first estimates of the dietary exposure of the population to heavy metals (lead, cadmium, and mercury) and to gamma-emitting radionuclides, the results of which are related below. In addition a TDS was performed to estimate the dietary exposure of children to mycotoxins [2] and to certain food additives [3].

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Source of Food Consumption Data

The first step in a dietary exposure assessment study is the generation of data on the types and amounts of foods typically consumed in the population under study. In theory, food consumption data can be generated by three different means: (1) Food Balance Sheets (FBS), (2) household budget or consumption surveys, and (3) individual food consumption surveys. Because the data provided by both FBS and household-based surveys represent food availability rather than actual food consumption data, individual food consumption surveys more closely reflect actual dietary habits and practices within a country or a region.

In Lebanon, the last comprehensive individual food consumption survey was conducted in 1961 by the Interdepartmental Committee on Nutrition for National Defense [4]. Afterwards studies on the dietary habits of the Lebanese population were only sparsely described in the scientific literature. In order to generate individual-based food consumption data that more closely reflect consumption patterns in the country, an individual dietary survey was conducted in 2001 on a representative sample of the population of Beirut, which has been called the "melting pot" of the country. The survey provided data on 210 men and 234 women aged 25–54 years. The age and sex distribution of this sample was proportionate to the baseline population according to the Lebanese Central Administration for Statistics [5]. Food consumption data were collected by means of a quantitative food frequency questionnaire (QFFQ) specifically designed for the study. It consisted of a list of 112 culture-specific food items/beverages and included a number of composite dishes that may contain multiple ingredients. The questionnaire permitted not only the estimation of the frequency of consumption of each food item, but also the identification of the portion size that the individual usually consumes [6].

Total Diet Study Design

Selection of Food Items

The selection of food items to be included in the TDS was based on two criteria. First, foods were selected based on their ranking in consumption by adults: the food items with a mean consumption > 1 g/day per person were included. Second, foods identified by the WHO GEMS/Food Programme in its comprehensive list [7] as potential sources of lead (Pb), cadmium (Cd), and mercury (Hg) were selected. The second criterion has thus led to the exclusion of certain food items from the total diet list. The excluded food items comprised soft drinks, coffee, tea and infusions, nuts, added fats and oils (except for those incorporated in the cooked recipes), olives, alcoholic beverages, chocolate, added white sugar, jam, honey and candies. By combining the two selection criteria, 77 food items, including drinking water, were selected for the analytical determination of Pb, Cd, Hg, and radioisotopes (see Table 33.1). These foods

Table 33.1 Aggregation of the 77 food items into 21 food groups, weight of each item as consumed (g/day) and percentage weight of each food item in its group

Food group	Daily intake (g/day)	% weight	Food group	Daily intake (g/day)	% weight
1. Bread and Toast			*7. Fruit juices*		
Traditional bread	136.8	93.6	Juice, canned	65.2	50.1
Traditional crackers (Ka'ak)	6.2	4.2	Juice, fresh	65.0	49.9
Toast	3.2	2.2	*Total*	*130.2*	*100.0*
Total	*146.2*	*100.0*			
			8. Fruits		
2. Biscuits and croissants			Oranges	75.3	32.7
Biscuits	13.6	68.7	Apples	61.0	26.5
Croissant	4.9	24.7	Bananas	20.7	8.9
Doughnuts	1.3	6.6	Watermelon	15.0	6.5
Total	*19.8*	*100.0*	Fruit-based deserts	10.9	4.8
			Grapes	10.0	4.4
3. Cakes and pastries			Cherries	5.6	2.4
Cakes	11.8	46.3	Peaches	5.3	2.3
Traditional pastry (Knefah)	8.2	32.2	Pears	5.2	2.3
Other traditional pastry	5.5	21.6	Fruit salad	4.7	2.1
Total	*25.5*	*100.0*	Melon	4.2	1.8
			Strawberry	3.5	1.5
4. Pasta and other cereals			Exotic fruits	2.7	1.2
Pasta, cooked	23.9	74.0	Apricots	2.1	0.9
Burghol, cooked	5.5	17.0	Canned fruits	2.2	1.0
Burghol, raw	2.8	9.0	Prunes	1.4	0.6
Total	*32.2*	*100.0*	*Total*	*230.0*	*100.0*
5. Pizzas and pies			*9. Cheese*		
Pies, type Manaeesh	32.1	64.0	Cheese Akkawi	9.8	28.0
Pizza	11.3	23.0	Cheese Halloum	9.8	28.0
Other traditional pies	6.6	13.0	Cheese Kashkawal	8.1	23.0
Total	*50.0*	*100.0*	Packaged cheese	7.4	21.0
			Total	*35.1*	*100.0*
6. Rice and rice-based products	50.1	100.0	*10. Milk*		
Rice, cooked			Milk, reconstituted from powder	69.8	70.0
			Milk liquid	29.9	30.0
			Total	*99.7*	*100.0*

(continued)

Table 33.1 (continued)

Food group	Daily intake (g/day)	% weight	Food group	Daily intake (g/day)	% weight
11. Milk-based ice cream and pudding			*15. Vegetables, raw and salads*		
Pudding	6.1	50.2	Other salads	56.6	42.1
Milk-based ice cream	6.0	49.8	Vegetables, raw	49.4	36.7
Total	*12.1*	*100.0*	Traditional salad, type Fattouch	15.5	11.6
			Traditional salad, type Tabbouli	12.9	9.6
12. Yogurt and yogurt-based products			*Total*	*134.5*	*100.0*
Yogurt	68.3	71.1	*16. Potatoes*		
Strained yogurt, type Lebneh	27.8	28.9	Potatoes, boiled	57.8	91.0
Total	*96.2*	*100.0*	Potato chips	5.7	9.0
			Total	*63.5*	*100.0*
13. Vegetables, canned					
Corn	4.6	39.8			
Mixed vegetables	3.2	27.8	*17. Pulses*		
Artichoke	1.5	12.6	Chickpeas	12.7	32.1
Mushrooms	1.2	10.3	Lentils	9.8	24.7
Asparagus	1.1	9.4	Fava beans	9.1	22.9
Total	*11.6*	*100.0*	Beans	5.3	13.4
			Fava bean-based falafel	2.7	6.8
14. Vegetables, cooked					
Green beans, stew	13.7	13.8	*Total*	*39.5*	*100.0*
Mixed vegetables, stew	13.7	13.8			
Zucchini, stuffed	12.9	13.1	*18. Fish*		
Eggplant	8.7	8.8	Fish, fresh or frozen	11.2	62.4
Jews mallow, stew	7.0	7.1	Tuna, canned	6.7	37.6
Peas, stew	6.9	6.9	*Total*	*17.9*	*100.0*
Cauliflower	6.7	6.8			
Grape leaves, stuffed	6.3	6.3	*19. Meat, cooked and cured*		
Cabbage, stuffed	4.5	4.6			
Spinach, stew	4.5	4.5	Meat, cooked	47.6	91.5
Okra, stew	4.4	4.4	Cured meat	4.4	8.5
Eggplant, stuffed	3.9	3.9	*Total*	*52.0*	*100.0*
Chicory	3.3	3.4			
Artichoke	2.3	2.3	*20. Poultry*		
Total	*98.9*	*100.0*	Chicken, grilled	36.1	96.1
			Chicken liver, fried	1.5	3.9
			Total	*37.6*	*100.0*
			21. Drinking water[a]	*985.9*	*100.0*

[a]Drinking water is a composite of water collected from the nine districts that were included in the dietary survey

represented approximately 80 %, on a weight basis, of the daily ration of the average individual. For the evaluation of the dietary exposure to Hg, only fish and fish products were included.

Food Collection, Preparation and Aggregation

The composite approach, as recommended by the WHO, has been applied. Accordingly, for each of the 77 foods listed, five subsamples, i.e. five different brands or varieties, were purchased and combined into one composite sample. Since market shares of the different brands are not determined in Lebanon, the contribution of each subsample to total weight was equal to 20 %. Individual samples were collected from different retail outlets in the city of Beirut. Food items were transported to one central laboratory where they were prepared and cooked in a manner similar to local cooking practices. To take into account the occurrence of changeable contamination, five complete sets of foods (market baskets) were collected during 2004 [8].

For each market basket, the 77 food items were aggregated into 21 groups of similar foods (see Table 33.1). For practical reasons, these 21 food groups were further combined into 12 aggregates for the analytical determination of radionuclides. Food items of each group were combined and blended and homogenized using an ordinary domestic mixer. Samples were then stored at −18 °C prior to analysis.

Source of Food Contamination Data

The analytical quantification of Pb, Cd and Hg was performed using inductively coupled plasma mass spectrometry (ICPMS). For quality control, each test run included spiked test samples and certified reference materials for the comparison of measured and certified concentrations of the elements of interest. All samples were analyzed in duplicate, which were digested and measured in separate batches to eliminate any batch specific error.

The quantitative determination of radionuclides was performed using two gamma ray spectrometry systems (Canberra) equipped with two high purity coaxial germanium detectors with relative efficiency of 30 % and 40 %, respectively, and of high resolution [1.85 and 2.0 keV (FWHM) at 1,332 keV, respectively] [9].

Dietary Exposure Assessment

In the Lebanese TDS, the calculation of dietary exposure was performed using a deterministic model in which the average value for food consumption was multiplied by the average concentration of the contaminant and the exposures from all food sources were then summed to provide the average dietary exposure value. For foods containing levels of elements below the limit of quantification (LOQ), a value equal to half the LOQ was assigned and used for calculation purposes [10]. Consumer exposure estimates to Pb, Cd and Hg were then expressed in mg/kg body weight/week to allow comparison with the respective Provisional Tolerable Weekly Intake (PTWI) values. For this purpose, a body weight of 72.8 kg was used, which is the average body weight of the participants in the dietary survey.

Findings from the Lebanese Total Diet Study

Dietary Exposure to Lead and Cadmium

The highest levels of Pb and Cd were found in cereal-based products with bread presenting the highest concentrations of both elements (35.4 μg/kg for Pb and 17.5 μg/kg for Cd). However, when compared with the Maximum Levels (ML) proposed by the Codex Alimentarius Commission [11], none of the food samples analyzed within the Lebanese TDS were found to exceed these limits. The Lebanese TDS showed that the average dietary exposures to Pb and Cd represented 7 % and 17 %, respectively, of their corresponding PTWIs [12], [13] and thus did not, at least for Cd, represent a risk for the average consumer. Because the PTWI for Pb has been withdrawn, a further evaluation of Pb exposure is necessary. The TDS showed that the food groups that contributed most to the dietary exposure to Pb and Cd were cereals and cereal-based products (45.3 % and 36 % respectively), vegetables and potatoes (17.6 % and 28.5 % respectively) and drinking water (16.2 % and 24 % respectively) (see Table 33.2). The contribution of a food group to the dietary exposure depends not only on the contamination level of that particular food but also on the consumption level of this food by the population under study. In Lebanon, the diet relies heavily on cereals and vegetables. In fact, breads and cereals alone were found to provide 35.0 % of the total energy intake of the average Lebanese urban adult [6]. In addition, the mean consumption of fresh fruits and vegetables by the average Lebanese urban adult was reported at 367 g person/day, a value approaching the WHO/FAO minimum recommended value of 400 g daily. This plant-centered diet, which is a characteristic shared by other Mediterranean countries, partly explains the fact that cereals and other plant-based food products were the main dietary sources of Pb and Cd in Lebanon.

The TDS findings suggest that, in Lebanon, the dietary exposure to the potentially toxic elements Pb and Cd is low, and that for the average consumer, there is no risk of exceeding their respective PTWIs. However, it is important to keep in mind that the exposure assessment conducted in the present study was based on the analysis of the Lebanese total diet food list that represented 80 % of the average daily energy intake of the average individual. It might thus be argued that some food items that typically contain high levels of heavy metals may have been excluded

Table 33.2 Lead and cadmium concentrations (μg/kg) in food samples and mean daily exposures (μg/person/day) from "total diet" food groups

	Lead			Cadmium		
Food group	Range (μg/kg)	Mean[a] (μg/kg)	Mean exposure (μg/day)	Range (μg/kg)	Mean[a] (μg/kg)	Mean exposure (μg/day)
1. Bread and toast	23–47.1	35.4	5.2	14.2–19.8	17.5	2.6
2. Biscuits and croissant	6.9–23.3	14.0	0.3	8.7–16.7	10.7	0.2
3. Cakes and traditional pastry	27.9–40.5	35.2	0.9	3.6–5.5	4.4	0.1
4. Pasta and other cereal products	6.7–12.3	9.4	0.3	13.8–19.2	15.8	0.5
5. Pizzas and pies	21.6–41.8	27.3	1.4	10.2–14.8	12.5	0.6
6. Rice and rice-based products	4.1–5.5	4.6	0.2	3.2–8.8	6.6	0.3
7. Fruits and fruit-based products	2.6–14.6	6.1	1.4	0.4–0.9	0.6	0.1
8. Fruit juices	<6–<6	3.0	0.4	<6–<6	3.0	0.4
9. Pulses	3.9–11.3	5.9	0.2	0.9–2.1	1.6	0.1
10. Vegetables, canned	13.4–16.0	14.8	0.2	5.7–8.4	7.1	0.1
11. Vegetables, cooked	12.7–22.5	16.0	1.6	8.5–21.2	13.1	1.3
12. Vegetables, raw and salads	5.7–15.5	7.9	1.1	6.8–22.0	10.4	1.4
13. Potatoes	1.6–4.5	2.8	0.2	5.0–15.7	10.0	0.6
14. Cheese	14.8–22.3	17.6	0.6	1.4–1.5	1.4	0.1
15. Milk and milk-based beverages	<6–<6	3.0	0.3	<6–<6	3	0.3
16. Milk-based ice cream and pudding	4.1–6.3	4.8	0.1	1.1–3.3	2.0	0.02
17. Yogurt and yogurt-based products	1.7–3.0	2.1	0.2	0.4–0.5	0.5	0.1
18. Meat and meat-based products	6.2–11.8	9.0	0.5	1.2–3.4	2.0	0.1
19. Poultry	5.2–9.9	6.8	0.3	3.8–16.6	6.6	0.3
20. Fish and fish-based products	4.4–8.8	5.9	0.1	3.8–6.5	5.0	0.1
21. Drinking water	<6–<6	3.0	3	<6–<6	3.0	3.0
Total			*18.5*			*12.3*

[a]Mean concentrations are calculated based on the concentrations determined in the five "total diet" sets

from the TDS. Examples of such foods would include mollusks and crustaceans, which accumulate high levels of Pb, oysters, which accumulate high levels of Cd, and kidneys, which may contain high amounts of both Pb and Cd. According to the individual food consumption survey data [4], the average dietary intake of these food items was very low and did not exceed 0.5 g/day for the typical consumer. This suggests that these types of foods, such as crustaceans, mollusks and kidneys, are very rarely consumed by the Lebanese population and that their contribution to dietary exposure would be rather limited. It would be of interest, however, to evaluate the dietary exposure of the excessive consumers of these types of foods since this group of the population may be exposed to higher levels of Pb and Cd than the average consumer.

Dietary Exposure to Mercury

Being the primary source of dietary Hg and particularly methylmercury (MeHg), the most toxic form of the element, fish was the only food group included in the dietary exposure of the population to Hg. Accordingly, the average concentration of total Hg in fish samples was 165 mg/kg, ranging between 140 mg/kg and 203 mg/kg, and the mean daily exposure to Hg was estimated as 3 mg/day, representing 5.6 % of the PTWI (5 mg/kg bw/week) [14]. Based on the assumption that 100 % of Hg in fish is in the MeHg form, the levels of MeHg in the analyzed fish samples were not found to exceed the Guideline Levels specified in the Codex Alimentarius of 0.5 mg/kg for non-predatory fish and 1.0 mg/kg for predatory fish [9]. Based on the same assumption, the mean dietary exposure to MeHg was found to represent 17.5 % of the PTWI of MeHg (1.6 mg/kg bw/week) [15], and thus did not represent a risk for the average consumer.

Dietary Exposure to Gamma-emitting Radionuclides

Since the primary factor contributing to the internal effective dose in the human organism is contaminated food, the control of radionuclides in food represents the most important means of protecting public health. In this context, the Lebanese TDS was also used to assess the dietary exposure of the consumer to gamma-emitting radionuclides and for determining baseline levels of these radionuclides in foods, especially given that in Lebanon, there were no previous evaluations of radioisotopes in foods. The selection of the TDS design for this purpose was linked to many reasons: the TDS allows the derivation of ingestion doses by combining measurements of radionuclides in different food groups with food consumption data; it differs from other food surveillance programs because it focuses on the diet as a whole and not on individual foods [16]. Most importantly, the TDS deals with

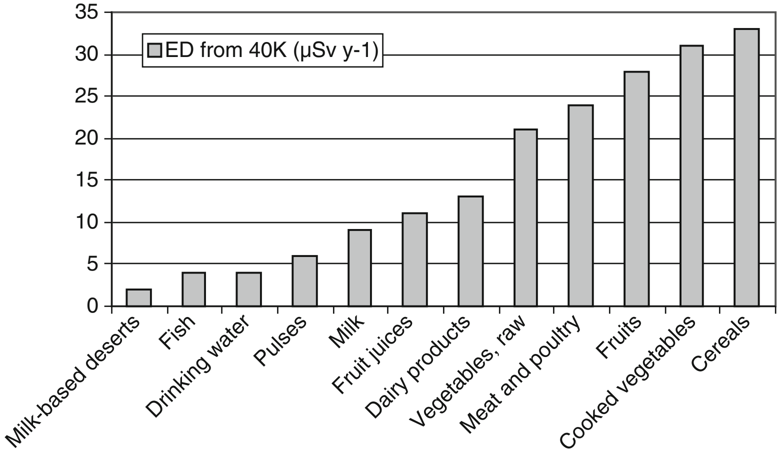

Fig. 33.1 Contribution of the different food groups to the effective dose (*ED*) from 40 K (μSv y^{-1}) in the reference typical diet of an adult urban population in Lebanon

foods that are processed as for normal consumption, thus taking into consideration the impact of home cooking and food processing on the levels of radionuclides.

Accordingly, the radioisotopes cesium-134 (Cs-134) and iodine-131 (I-131) were not detected in any of the food samples. The artificial radionuclide Cs-137 was measured above detection limits in only fish, meat and milk-based deserts, with the highest activity concentration being equal to 0.1 Bq/kg. In comparison, the maximum permitted levels of Cs-137 in foodstuffs (with the exception of foods for infants) are of 1,000 Bq/kg as specified by the Codex Alimentarius Commission [11] and the International Atomic Energy Agency [17].

The activity concentrations of the naturally occurring radionuclide potassium (K-40) varied between 31.1 and 120.9 Bq/kg in the analyzed food samples, which is in accordance with the range reported in different parts of the world (35–380 Bq/kg) [18]. By associating the activity concentrations of K-40 (in Bq/kg) in the different food groups with the consumption levels of these food groups (in g/person/day), the total daily exposure to K-40 was found to be equal to 81.86 Bq/person/day, a value that is comparable to estimates provided by other TDSs conducted in several other countries (42.79–94.8 Bq/person/day) [19]. The mean annual effective dose resulting from the dietary exposure to K-40 was estimated at 186 μSv/year for the average adult consumer (see Fig. 33.1), a value that is comparable to the world average value (178 μSv/year) and lies well within the range reported by other countries.

The applied TDS design has thus shown that the activity concentration of the gamma-emitting radionuclide K-40 in foodstuffs available on the Lebanese market is consistent with values reported in the literature and that the levels of Cs-137 in foods do not present any public health hazard.

Conclusion

The TDS that has been applied in Lebanon is a small-scale TDS, the findings of which may be limited by the fact that it was not a nationally representative study and by the use of the deterministic approach in the assessment of the dietary exposure. In fact, inherent to the deterministic model is the assumption that the average consumption of specified food(s) represents the average diet [20]. This approach therefore does not provide an insight into the range of possible exposures that may occur within a population or the main factors influencing the results of the assessment. However, while not excluding the possibility that the daily exposures determined in the present study may not be representative of the population as a whole, this TDS has provided a first estimate of consumers' exposure to heavy metals and radionuclides through the diet in Lebanon. The main output of this study, other than characterizing the risk for the consumer, is the fact that it has provided the framework for other total diet studies, which are currently ongoing and which target the dietary exposure of the Lebanese consumer to pesticide residues, mycotoxins and essential minerals

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