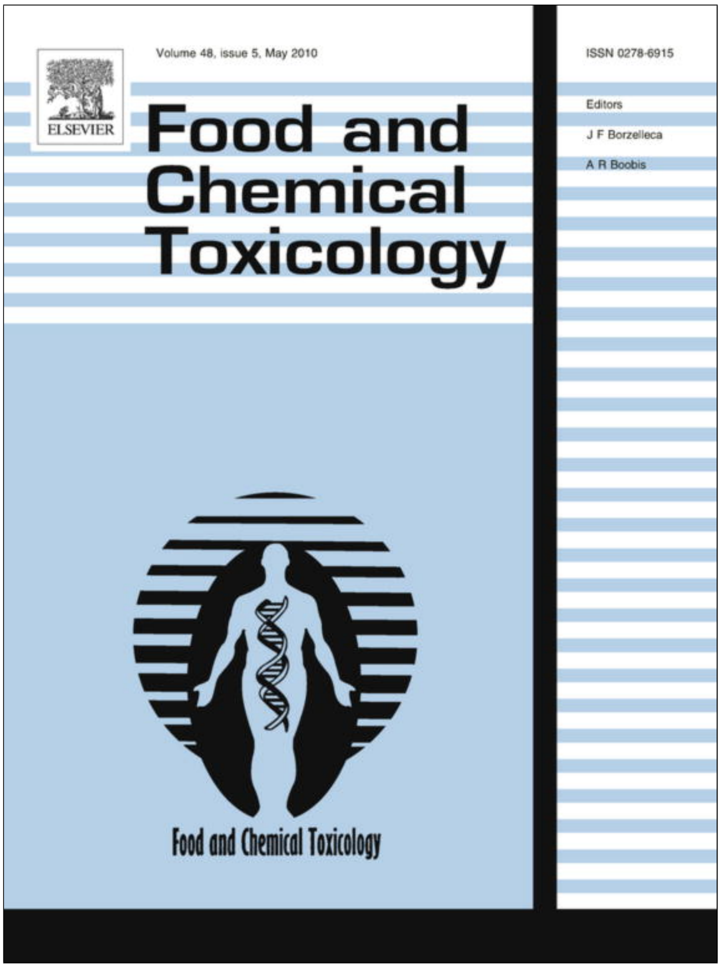
Volume 48, issue 5, May 2010
ISSN 0278-6915
ELSEVIER
Food and Chemical Toxicology
Editors
J F Borzelleca
A R Boobis
Food and Chemical Toxicology

Food and Chemical Toxicology 48 (2010) 1262–1269

Contents lists available at ScienceDirect

Food and Chemical Toxicology

journal homepage: www.elsevier.com/locate/foodchemtox

Dietary exposure to essential and toxic trace elements from a Total diet study in an adult Lebanese urban population

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ARTICLE INFO

Article history:
Received 19 August 2009
Accepted 16 February 2010

Keywords:
Adult
Dietary exposure
Trace elements
Total diet study
Urban
Lebanon

ABSTRACT

This study assesses, by the Total diet study approach, the adequacy of micronutrient intake (Co, Cu, Fe, Mn, Ni, Zn) and the dietary exposure of a Lebanese adult urban population to two toxic elements (Cd, Pb). The foods that made up the average 'total diet' were derived from a previous individual consumption survey. A total of 1215 individual foods were collected, prepared and cooked prior to analysis. Analytical quantification was performed using inductively coupled plasma mass spectrometry. Average daily intakes of Co (11.4 μg/day), Cu (1104.19 μg/day), Fe (13.00 mg/day), Mn (2.04 mg/day), Ni (126.27 μg/day) and Zn (10.97 mg/day) were below toxicological reference values and were found to satisfy nutritional recommendations, except for manganese in men and iron in women. Average dietary exposure to Pb and Cd represented 3.2% and 21.7% of the respective provisional tolerable weekly intakes. Estimates of dietary intakes of iron appeared to be inadequate for 63% of adult women. These findings should constitute a current measure of assessing the adequacy and safety of foods consumed in Lebanon and may be a basis for future monitoring studies.

1. Introduction

Some trace elements such as Fe, Co, Cu, Mn, Ni and Zn are essential micronutrients that need to be consumed in adequate amounts to maintain normal physiological functions (Goldhaber, 2003). In contrast, dietary exposure to other elements such as Cd and Pb has been associated with toxic and adverse health effects (FAO/WHO, 2007). All trace elements, including the essential ones, may be toxic when taken in excessive amounts and to paraphrase Paracelsus (1493–1541) "*All things are toxic and there is nothing without toxic properties. It is only the dose which makes something a poison*". Thus, the estimation of dietary exposure is crucial for risk evaluation, and possibly for the determination of the relationships between adverse effects observed in humans and dietary exposure to particular substances. The evaluation of the dietary exposure consists of associating food consumption data with concentration/contamination data and typically includes the application of statistical adjustment factors that allow conclusions about the amount of a substance being consumed on a 'usual' basis or over a lifetime (FAO/WHO, 2006).

For toxic trace elements present in foods, the risk assessment process is similar to that applied for any non-nutrient chemical and is undertaken by identifying the adverse effect(s) produced at high intakes, defining the dose–response relationship for the effect(s), and then selecting an appropriate safety margin to establish levels of intake that can be consumed daily over a lifetime without significant adverse health effects (Renwick, 2006). In contrast, in the case of nutrients, such as essential trace elements, the process of risk assessment examines two ends of the dose–response relationships: (1) that associated with intakes that are too high and the resulting toxicity and (2) that associated with intakes that are too low and the resulting nutritional deficiencies (Goldhaber, 2003; Renwick and Walker, 2008; Renwick et al., 2008). Accordingly, the FAO/WHO has defined micronutrient requirements, including trace elements as "an intake level, which will meet specified criteria of adequacy, preventing risk of deficit or excess" (WHO, 1996). The World Health Organization (WHO) has carried out risk assessments dealing with the toxicity end of the spectrum, thus establishing acceptable daily intakes (ADIs) and provisional maximum tolerable daily intakes (PMTDIs) for a large number of chemicals, including some essential trace

Abbreviations: AI, adequate intake; GEMS/Food, Global Environment Monitoring System/Food Contamination Monitoring and Assessment Programme; PMTDI, Provisional maximum tolerable daily intake; PTWI, provisional tolerable weekly intake; RDA, recommended dietary allowance; TDI, tolerable daily intake; TDS, Total diet study; UL, tolerable upper intake level; WHO, World Health Organization.

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doi:10.1016/j.fct.2010.02.020

elements. The US Food and Nutrition Board of the Institute of Medicine has dealt with nutritional deficiency problems as well as toxicity by setting Dietary Reference Intakes (DRIs), which include the recommended dietary allowance (RDA), the estimated average requirement (EAR), the adequate intake (AI), and the tolerable upper intake level (UL) for essential trace elements (Institute of Medicine, 2001; Goldhaber, 2003).

These nutritional and toxicological reference values can be used as benchmarks in the risk assessment process. Numerous studies investigating the dietary exposure to trace elements have been carried out by several countries using the Total diet study (TDS) approach (WHO, 1985; Van Dokkum et al., 1989; Ysart et al., 2000; Egan et al., 2002; Kroes et al., 2002; Leblanc et al., 2005b), which consists of purchasing, at retail level, foods commonly consumed by the population (market basket), processing them as for usual consumption, often combining them into food composites or aggregates, homogenizing them and analyzing them for toxic chemicals and certain nutrients (WHO, 1985); it thus takes into account the effects of kitchen preparation on the levels of contaminants and nutrients in foods and provides dietary intake data for use by regulatory agencies and the public. It also identifies which food groups are the main dietary sources of the different contaminants/nutrients, and when repeated regularly, it constitutes a reliable monitoring tool (WHO, 1985).

In Lebanon, an Eastern Mediterranean country with a population estimated to be around 4 million, there are currently no reports on the dietary intake of essential trace elements by the population. Data on the dietary exposure to trace elements are also scarce in other parts of the Middle East. The objective of the present study is to assess, by the TDS approach, the dietary exposure of the population to six essential trace elements (Co, Cu, Fe, Mn, Ni and Zn) and two toxic ones (Cd and Pb). The types and quantities of foods that make up the average 'total diet' are based on the results of a previous study of an adult urban population living in Beirut, the capital of Lebanon (Nasreddine et al., 2006a). Consumer exposure estimates are compared with the appropriate nutritional and toxicological reference values and with data provided from other countries. The findings may shed light on the link between shortages or excesses of trace elements in the diet and disease risk in the country.

2. Material and methods

2.1. Food selection and collection of food samples

The first step in the TDS consisted of establishing the list of foods to be analyzed. This list was based on food consumption data provided by an individual food consumption survey that was conducted on an adult urban population in Lebanon, the details of which have been published elsewhere (Nasreddine et al., 2006a). Accordingly, 81 food items, including drinking water and representing 80% of the average daily energy intake were selected for the Total diet study (Table 1). The selected food items represented, on a weight basis, 79% of the daily ration of the average individual. Based on general guidelines provided by the WHO (WHO, 1985) and on sampling schemes described in the literature (Leblanc et al., 2005a), a composite sampling approach has been applied in this study and consisted of purchasing the same item from five different sites or in five different brands/varieties and of combining the five items to represent a composite sample of the food product in question. Since market shares of the different brands are not established in Lebanon, the contribution of each sub-sample to total weight was equal to 20%. Even though the composite sampling scheme may dilute high contamination levels that may be found in one of the collected sub-samples, this scheme has the advantage of increasing the representativeness of food sampling (WHO, 1985).

Three complete sets of foods (market baskets) were collected at 3 months intervals and the sampling of foods was performed at the most popular retail markets in Beirut. Thus, 81 food items × five sub-samples × three market baskets = 1215 food items were collected for this study. The quantities of purchased foods reflected that necessary for analysis of trace elements, as well as for the storage of reserve portions after allowing for amounts lost during preparation and cooking.

2.2. Preparation of the food samples

A popular local cookbook was consulted for the preparation of composite dishes (Kamal and Osman, 1995) and food items were prepared and cooked in a manner similar to local cooking practices. In compliance with international WHO/GEMS/Food recommendations, foods were prepared using drinking water from the city where the foods were bought, i.e. Beirut for this study. Salt and spices were also added as in regular cooking practices. Suitable stainless steel cooking equipment was used to prepare the food and the use of storage containers and cooking utensils made of aluminum, ceramic or enamel was excluded. For each of the three market baskets and 81 foods specified on the food list, the five sub-samples were combined (20% w/w) and blended to give a homogeneous sample representative of the food item in question. In accordance with good laboratory practices and with the internal standard operating procedures of the Environment Core Laboratory (EVL), in compliance with the ISO/IEC 17025 norm (ISO/IEC 17025, 2005), the equipment used for preparing and homogenizing the composite samples was thoroughly washed between each preparation (e.g. cleaning with a laboratory-grade detergent, rinsing thoroughly with hot tap water, rinsing or soaking with acid solution, rinsing thoroughly with deionized water) to avoid the risk of cross-contamination.

For each of the three market baskets, the 81 food items were aggregated into 11 groups of similar foods for the analysis of essential trace elements. The appropriate amount of each raw, prepared or cooked food item to be included in its composite food group was determined from the food consumption data (Table 1). Food items of each group were combined and blended using an ordinary domestic mixer to give a homogeneous sample. Thus, in total, 11 composite food groups per market basket × three market baskets = 33 composite food group samples were prepared for analysis in this study. Samples were stored at −18 °C prior to analysis. The study was conducted in 2008.

2.3. Analytical determination of trace elements

Food samples, except for liquid ones (fruit juice, milk and water), were dried in an oven at 105 °C until a fixed weight was obtained. Weights of food samples were recorded before and after the drying process to allow the determination of the moisture content in each sample. Dried samples were then ground into fine powder and digested in Teflon vessels with 3 ml nitric acid (70% v/v, trace metal grade), using a closed microwave digestion system (Ethos plus, Milestone Microwave Milestone, Italy). A volume of 2 ml hydrogen peroxide (50% v/v, Certified Stabilized), was added to all samples containing fat. For quality control, a number of Certified Reference Material (CRM) were used and included oyster tissue (NIST 1566b), Tomato leaves (NIST 1573a), Fucus seaweed (IAEA-140-TM), Wheat flour (NIST 1567a), whole milk powder (NIST 8435), and rye flour (IAEA-V-8). All CRMs were used as provided without further grinding. CRMs were stored under the same conditions and digested with the same protocol as the analyzed food samples.

The analytical quantification of trace elements was performed using inductively coupled plasma mass spectrometry (Agilent ICP-MS 7500-Ce, Japan). For the comparison of measured and certified concentrations of the elements of interest, each test run included spiked test samples and reference materials. All samples were analyzed in duplicates, which were digested and measured in separate batches to eliminate any batch specific error. The limits of detection (LOD) were equal to 0.0005 mg/kg for Mn, Co, Ni, Cu, Zn, Pb and Cd while the LOD for Fe was equal to 0.1 mg/kg. The limits of quantification (LOQ) for Mn, Co, Ni, Cu, Zn, Pb and Cd were equal to 0.002 mg/kg while the LOQ for Fe was equal to 0.5 mg/kg.

Recovery percentages based on CRMs ranged between 86.2% and 112.5% for Mn, 82.8% and 120.0% for Co, 80.1% and 114.4% for Ni, 84.9% and 111.5% for Cu, 82.5% and 113.4% for Zn, 87.9% and 116.6% for Fe, 87.3% and 119.2% for Cd and 82.1% and 110.55 for Pb. Corrections based on recovery percentages were not performed. The average spiking recovery percentages were 104.1% (±4.3%) for Mn, 114.0% (±10.9%) for Co, 107.4% (±13.3%) for Ni, 102.8% (±4.9%) for Cu, 102.1% (±7.1%) for Zn, 103.8% (±15.9%) for Fe, 101.8%(±14.4%) for Cd and 98.0% (±12.9%) for Pb.

2.4. Calculation of dietary intake

The concentrations of trace elements are expressed in mg or µg element/kg fresh matter and intakes in mg or µg/day/person. For foods containing levels of elements below the LOQ, a value equal to half the LOQ was assigned and used for calculation purposes (GEMS/Food-Euro, 1995). For foods containing levels of elements below the LOD, a value equal to half the LOD was assigned and used for calculation purposes (GEMS/Food-Euro, 1995).

To take into account the variability that exists in food-consumption patterns and to provide data on the distribution of exposure levels within the studied population, the distribution of food intake as provided by the individual dietary survey was combined with the average concentration for each trace element under study. Accordingly, mean, median, average and percentiles 2.5th and 97.5th intakes were identified, the contributions of each food group to the total average dietary intake of minerals and trace elements were provided and risk characterization was performed for average and high intake levels and for percentage of individuals exceeding toxicological reference values (PMTDI, PTWI pr TDI) or not meeting 2/3 RDAs or AIs.

Table 1
Aggregation of the 81 food items into 11 composite food groups: weight of each item as consumed (g day^{-1}) and percentage weight of each food item in its group.

Composite food group	Daily intake (g/day)	% Weight	Food group	Daily intake (g/day)	% Weight
1. Breads and cereals			*4. Vegetables*		
Traditional bread	136.8	42.2	Other salads	56.6	23.1
Rice, cooked	50.1	15.5	Vegetables, raw	49.4	20.2
Pies, type Manaeesh	32.1	9.9	Traditional salad, type Fattouch	15.5	6.3
Pasta, cooked	23.9	7.4	Green beans, stew	13.7	5.6
Biscuits	13.6	4.2	Mixed vegetables, stew	13.7	5.6
Cakes	11.8	3.6	Traditional salad, type Tabbouli	12.9	5.3
Pizza	11.3	3.5	Zucchini, stuffed	12.9	5.3
Traditional pastry (Knefah)	8.2	2.5	Eggplant, cooked	8.7	3.6
Other traditional pies	6.6	2.0	Jews mallow, stew	7	2.9
Traditional crackers (Káak)	6.2	1.9	Peas, stew	6.9	2.8
Other traditional pastry	5.5	1.7	Cauliflower, cooked	6.7	2.7
Burghol, cooked	5.5	1.7	Grape leaves, stuffed	6.3	2.6
Croissant	4.9	1.5	Corn, canned	4.6	1.9
Toast	3.2	1.0	Cabbage, stuffed	4.5	1.8
Burghol, raw	2.8	0.9	Spinach, stew	4.5	1.8
Doughnuts	1.3	0.4	Okra, stew	4.4	1.8
Total	**323.8**	100.0	Eggplant, stuffed	3.9	1.6
2. Potatoes			Chicory, cooked	3.3	1.3
Potatoes, boiled	57.8	91	Mixed vegetables, canned	3.2	1.3
Potato chips	5.7	9	Artichoke, cooked	2.3	0.9
Total	**63.5**	100	Artichoke, canned	1.5	0.6
3. Pulses			Mushrooms, canned	1.2	0.5
Chickpeas	12.7	32.1	Asparagus, canned	1.1	0.4
Lentils	9.8	24.7	Total	**244.8**	100.0
Fava beans	9.1	22.9	*5. Fruit juices*		
Beans	5.3	13.4	Juice, canned	65.2	50.1
Fava bean-based falafel	2.7	6.8	Juice, fresh	65	49.9
Total	**39.5**	100	Total	**130.2**	100
6. Fruits			*8. Fish*		
Oranges	75.3	32.7	Fish, fresh or frozen	11.2	62.4
Apples	61	26.5	Tuna, canned	6.7	37.6
Bananas	20.7	8.9	Total	17.9	100
Watermelon	15	6.5	*9. Milk*		
Fruit-based deserts	10.9	4.8	Milk, reconstituted from powder	69.8	70
Grapes	10	4.4	Milk liquid	29.9	30
Cherries	5.6	2.4	Total	**99.7**	100
Peaches	5.3	2.3	*10. Dairy products*		
Pears	5.2	2.3	Yogurt	68.3	47.7
Fruit salad	4.7	2.1	Strained yogurt, type Lebneh	27.8	19.4
Melon	4.2	1.8	Cheese Akkawi	9.8	6.8
Strawberry	3.5	1.5	Cheese Halloum	9.8	6.8
Exotic fruits	2.7	1.2	Cheese Kashkawal	8.1	5.7
Apricots	2.1	0.9	Packaged cheese	7.4	5.2
Canned fruits	2.2	1	Pudding	6.1	4.3
Prunes	1.4	0.6	Milk-based ice cream	6	4.2
Total	**230**	100	Total	**143.3**	100.0
7. Meat and poultry			*11. Drinking water* *	985.9	100
Meat, cooked	47.6	53.1			
Chicken, grilled	36.1	40.3			
Cured meat	4.4	4.9			
Chicken liver, fried	1.5	1.7			
Total	**89.6**	100.0			

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

It is important to mention that, average dietary exposure estimates are based on the chemical analysis of a diet for which the consumption data were not differentiated by sex. Thus, for the comparison of the daily intakes of trace elements with the RDAs, which may be sex specific, we assumed that the average adult woman and the average adult man consumed the same diet i.e. the average typical diet of this population group (the market basket).

To compare dietary exposure levels with toxicological reference values which are expressed per kg body weight (TDI, PTWI or PMTDI), an average body weight of 72.8 kg was used since it was the average weight of the participants in the food consumption survey.

3. Results

3.1. Concentrations of trace elements in the composite food groups

The mean concentration of each element in the different food categories is shown in Table 2. The analytical results showed that all elements were undetectable (<LD) in water samples except for Zn, the levels of which were quantifiable (i.e. >LQ). For Pb, Cd and Co, 2 out the 11 samples had levels below the LQ, while for Ni one sample only had a concentration below the LQ.

Per kilogram of food item (fresh weight), meat and poultry were among the richest dietary sources of iron (38.57 mg/kg) and zinc (44.55 mg/kg). Pulses contained high levels of manganese (4.42 mg/kg), copper (2708.06 μg/kg), nickel (387.27 μg/kg) and cobalt (36.55 μg/kg). Potatoes were among the richest sources of cobalt (37.53 μg/kg), while bread and cereal products contained the highest levels of manganese (3.0 mg/kg), nickel (169.01 μg/kg), and iron (15.04 mg/kg). The average concentrations of lead and cadmium in the analyzed composite food groups ranged between 0.54 and 16.43 μg/kg for lead and between 3.07 and 30.23 μg/kg for cadmium, with the highest concentrations being observed in vegetable-based products.

Table 2
Range and mean concentrations (µg/kg or mg/kg fresh weight) of Cd, Co, Cu, Fe, Pb, Mn, Ni and Zn in the 11 composite food groups of the Total diet study of an adult urban population in Lebanon.

Composite food group (n = 3)*	Cd (µg/kg)	Co (µg/kg)	Cu (µg/kg)	Fe (mg/kg)	Mn (mg/kg)	Ni (µg/kg)	Pb (µg/kg)	Zn (mg/kg)
Breads and cereals	15.09 (14.18–15.9)	6.36 (4.65–6.72)	1503.85 (1492.94–1536.82)	15.04 (13.85–17.62)	3 (2.83–3.17)	169.01 (162.82–174.11)	8.02 (7.91–8.12)	9.48 (9.15–9.62)
Dairy	3.07 (2.98–3.14)	1.64 (1.58–1.70)	191.64 (183.66–195.62)	0.32 (0.28–0.32)	0.10 (0.09–0.11)	74.28 (68.12–81.90)	0.54 (0.52–0.57)	15.22 (14.84–16.49)
Fish	6.95 (4.26–9.0)	<2.00	254.67 (246.41–259.16)	7.65 (6.79–8.26)	0.19 (0.18–0.21)	17.46 (15.42–20.20)	6.13 (5.8–6.5)	5.00 (4.89–5.12)
Fruits	6.27 (5.7–7.2)	4.85 (4.51–5.34)	443.87 (402.77–522.98)	4.60 (4.21–4.94)	1.35 (1.29–1.53)	29.00 (24.70–35.34)	0.98 (0.89–1.1)	0.89 (0.72–1.2)
Juices	<2.00	<2.00	69.65 (66.12–73.55)	0.16 (0.15–0.16)	0.84 (0.82–0.86)	<2.00	<2.00	0.10 (0.09–0.11)
Meat and Poultry	5.77 (4.89–6.69)	0.44 (0.31–0.51)	1644.41 (1463.97–1891.05)	38.57 (36.87–40.12)	0.49 (0.43–0.52)	25.24 (23.75–27.91)	3.03 (2.77–3.30)	44.55 (40.65–47.69)
Milk	<2.00	1.56 (1.51–1.62)	178.00 (165.98–185.67)	1.50 (1.47–1.51)	0.02 (0.019-.02)	18.83 (17.71–20.05)	<2.00	1.40 (1.38–1.43)
Potato	3.57 (3.3–3.8)	37.53 (34.50–44.10)	1295.86(1163.04–436.81)	3.57 (3.42–3.74)	0.97 (0.91–1.04)	139.15 (113.24–148.3)	6.18 (5.99–6.31)	2.76 (2.69–2.83)
Pulses	7.89 (7.33–8.28)	36.55 (34.24–38.26)	2708.06 (2585.81–2979.31)	2.70 (2.24–2.85)	4.42 (4.34–4.57)	387.27 (357.91–411.21)	2.22 (1.89–2.61)	11.26 (10.91–11.64)
Vegetables	30.23 (24.43–36.14)	14.60 (13.38–16.90)	488.08 (465.10–497.30)	11.52 (9.09–12.46)	1.43 (1.35–1.48)	103.22 (93.81–116.62)	16.43 (14.07–18.49)	2.30 (2.18–2.39)
Water	<0.5	<0.5	<0.5	<0.1	<0.0005	<0.5	<0.5	0.09 (0.08–0.12)

* Reported range is based on three analyzed food samples per food group. In total, the number of analyzed samples was 33 (11 food groups per market basket) and three per food group (n = 3).

3.2. Estimation of the dietary intake of trace elements

The estimated average daily intake of manganese was 2.04 mg/day (Table 3), representing 113.3% and 88.6% of the AI values of 1.8 and 2.3 mg/day for adult women and adult men, respectively (Table 4). The composite food groups that contributed the most to the dietary intake of Mn were breads and cereals (0.97 mg/day), accounting for 47.6% of the total dietary intake of manganese, followed by vegetables (0.35 mg/day) and fruits (0.31 mg/day), which accounted for 17.1% and 15.2%, respectively (Table 3).

Copper mean intake was estimated at 1104.19 µg/day, accounting for 122.7% of the RDA value for adults (900 µg/day) and 3% of the PMTDI (0.5 mg/kg BW/day or 36,400 µg/day) of the element (Table 4). Breads and cereals were the biggest contributor to the intake of Copper providing 486.95 µg/day (44.1%), followed by meat and poultry with 147.34 µg/day (13.3%) and vegetables with 119.48 µg/day (10.8%) (Table 3).

The estimated mean daily intake of Zn was found to be equal to 10.97 mg/day, thus contributing 137.1% and 99.7% of the RDA values (8 mg/day; 11 mg/day) for adult women and men respectively. Average daily intake of Zn represented 15% of the PMTDI value (1 mg/kg BW/day or 72.8 mg/day). The composite food groups that contributed the most to the intake of Zn were meat and poultry (3.99 mg/day), which represented 36.4% of total dietary intake of zinc followed by breads and cereals (3.07 mg/day) and dairy products (2.18 mg/day) which accounted for 28% and 19.9% of the total dietary intake of zinc respectively (Table 3).

Table 3
Contribution (%) of each composite food group to the mean daily intake of Cd, Co, Cu, Fe, Pb, Mn, Ni and Zn for an adult urban population in Lebanon.

Composite food group	Cd		Co		Cu		Fe		Mn		Ni		Pb		Zn	
	µg/d	%	µg/d	%	µg/d	%	mg/d	%	mg/d	%	µg/d	%	µg/d	%	mg/d	%
Breads and cereals	4.89	30.9	2.06	18.1	486.95	44.1	4.87	37.5	0.97	47.6	54.73	43.3	2.60	31.4	3.07	28.0
Dairy	0.44	2.8	0.24	2.1	27.46	2.5	0.05	0.3	0.01	0.7	10.64	8.4	0.08	0.9	2.18	19.9
Fish	0.12	0.8	0.02	0.2	4.56	0.4	0.14	1.0	0.003	0.2	0.31	0.2	0.11	1.3	0.09	0.8
Fruits	1.44	9.1	1.12	9.8	102.09	9.2	1.06	8.1	0.31	15.2	6.67	5.3	0.23	2.7	0.21	1.9
Juices	0.13	0.8	0.13	1.1	9.07	0.8	0.02	0.2	0.11	5.4	0.13	0.1	0.13	1.6	0.01	0.1
Meat and poultry	0.52	3.3	0.04	0.3	147.34	13.3	3.46	26.6	0.04	2.2	2.26	1.8	0.27	3.3	3.99	36.4
Milk	0.10	0.6	0.16	1.4	17.75	1.6	0.15	1.1	0.002	0.1	1.88	1.5	0.10	1.2	0.14	1.3
Potato	0.23	1.4	2.38	20.9	82.29	7.4	0.23	1.7	0.06	3.0	8.84	7.0	0.39	4.7	0.18	1.6
Pulses	0.31	2.0	1.44	12.7	106.97	9.7	0.11	0.8	0.17	8.5	15.30	12.1	0.09	1.1	0.45	4.0
Vegetables	7.40	46.8	3.57	31.3	119.48	10.8	2.82	21.7	0.35	17.1	25.27	20.0	4.02	48.7	0.56	5.1
Water	0.25	1.6	0.25	2.2	0.25	0.02	0.09	0.8	0.0002	0.01	0.25	0.2	0.25	3.0	0.09	0.8
Average intake	15.82		11.4		1104.19		13.0		2.04		126.27		8.26		10.97	

Table 4
Estimates of the distribution of the dietary exposure of an adult Lebanese urban population (n = 444) to selected trace elements and comparison of the mean, 2.5th and 97.5th percentile exposure to toxicological and nutritional reference values.

Trace element	Dietary exposure				Contribution of mean to		Contribution of 97.5th percentile intake to	Percentage of consumers consuming <2nd/3rd RDA or AI	
	2.5th percentile	50th percentile	97.5th percentile	Mean ± SD	RDA or AI[b] (%)	PMTDI, PTWI or TDI[c] (%)	PMTDI, PTWI or TDI c (%)	Males (n = 210)	Females (n = 234)
Cd (μg/d)	10.03	15.22	23.57	15.82 ± 3.65	NA	21.7%	32.4%	NA	NA
Co (μg/d)	6.88	11.05	17.39	11.40 ± 2.73	NA	NA	NA	NA	NA
Cu(μg/d)	654.52	1075.3	1812.16	1104.19 ± 295	122.7	3.0	4.9	0.0	1.7
Fe (mg/d)[a]	7.77	12.50	21.65	13.00 ± 3.26	72.2 (F) 162.5 (M)	22.3	37.2	0.0	62.5
Mn (mg/d)	1.23	1.98	3.24	2.04 ± 0.52	113.3 (F) 88.6 (M)	NA	NA	1.9	3.8
Ni (μg/d)	76.17	122.95	204.00	126.27 ± 3.29	NA	34.7	56.0	NA	NA
Pb (μg/d)	5.19	7.83	12.48	8.26 ± 1.93	NA	3.2	4.8	NA	NA
Zn(mg/d)	5.74	10.51	18.55	10.97 ± 3.26	137.1 (F) 99.7 (M)	15.1	25.5	0.0	0.4

[a] Females between 25and 45 years were included to be comparable to the19–50 years RDA.
[b] F = females and M = males; AI for Mn = 1.8 mg/day (F) and 2.3 mg/day (M); RDA for Cu = 900 μg/day; RDA for Fe = 18 mg/day (F) and 8 mg/day (M); RDA for Zn = 8 mg/day (F), 11 mg/day (M).
[c] PTWI for Pb = 25 μg/kg BW/week (JECFA, 1987); PTWI for Cd = 7 μg/kg BW/week (JECFA, 1993); TDI for Ni = 5 μg/kg BW/day (WHO, 1993); PMTDI of Cu = 0.5 mg/kg BW/day (FAO/WHO, 2007); PMTDI Zn = 1 mg/kg BW/day (FAO/WHO, 2007); PMTDI Fe = 0.8 mg/kg BW/day (FAO/WHO, 2007); NA is used when nutritional or toxicological reference value have not been proposed for the element.

The estimated average daily intake of Fe was found to be 13 mg/day, representing 72.2% and 162.5% of the RDA values of iron (18 mg/day and 8 mg/day) for adult women and men respectively. The mean intake of iron was found to represent 22.3% of the PMTDI value (0.8 mg/kg BW/day or 58.2 mg/day). The most important sources of Fe in the diet were breads and cereals (4.87 mg/day), which contributed 37.5% of the total intake of this element, followed by meat and poultry (3.46 mg/day), and vegetables (2.82 mg/day) which accounted for 26.6% and 21.7%, respectively.

The average daily intake of Ni was estimated at 126.27 μg/day, thus contributing 34.7% to the TDI of Ni (5 μg/kg BW/day or 364 μg/day). Breads and cereals contributed the most to the intake of Ni (54.73 μg/day), accounting for 43.3% of the daily intake of this element, and were followed by vegetables (25.27 μg/day) and pulses (15.3 μg/day), which accounted for 20% and 12.1% of the dietary intake of Ni, respectively.

The average daily intake of Co was estimated at 11.4 μg/day. Vegetables (3.57 μg/day) represented the basic source of Co in the diet, providing 31.3% of the total intake of this element. Potatoes, breads and cereals were also significant sources, contributing 20.9% and 18.1% respectively.

Based on lead concentrations in food samples, the average daily intake of lead was estimated at 8.26 μg/day thus accounting for 3.2% of the PTWI of lead (25 μg/kg BW/week or 260 μg/day) (JECFA, 1987). The mean intake of cadmium was calculated at 15.82 μg/day and represented 21.7% of the PTWI value (7 μg/kg BW/week or 72.8 μg/day) (JECFA, 1993). The main contributors to the dietary intake of Pb and Cd were vegetables (48.7% and 46.8% respectively) followed by breads and cereal-based products (31.4% and 30.9% respectively).

Table 4 also shows, for each element, the range of dietary intake between the 2.5th and 97.5th percentile levels. It indicates that, for all the studied elements, the 97.5th percentile level does not exceed the PMTDI, TDI or PTWI values. In addition, none of the study subjects had an intake that exceeded the toxicological reference values for any of the studied trace elements (data not shown). When stratifying the data by gender, 62.5% of adult females appeared to consume less than 2/3 RDA of Fe (18 mg/day). Furthermore, 3.8% of adult females had an inadequate intake of Mn, 1.7% an inadequate intake of Cu and 0.4% an inadequate intake of Zn. Among adult males, 1.9% appeared to have an inadequate intake of Mn, while the intake of all the other trace elements appeared to be adequate (>2/3 RDA).

4. Discussion

Total diet studies are a reliable tool for investigating the population's diet, both in terms of food-consumption patterns and intake levels of nutrients, bioactive compounds and contaminants, providing important information about the nutritional quality of a population's diet and about potential nutritional deficiencies or exposure to food contaminants (WHO, 1985; Lombardi-Boccia et al., 2003). The WHO supports Total diet studies as one of the most cost-effective ways of achieving accurate dietary exposure estimates and the WHO's Global Environment Monitoring System/Food Contamination Monitoring and Assessment Programme (GEMS/Food) has encouraged all countries, and particularly developing ones, to conduct Total diet studies as a matter of public health significance (WHO, 1999). The TDS has the advantage of yielding more realistic exposure data as compared to other dietary exposure assessment methods such as the Duplicate Portion Study and the Selective Study of Individual Foods (WHO, 1985; Kroes et al., 2002), since it consists of analyzing a representative 'market basket' of foods prepared as for usual consumption (WHO, 1985). The TDS differs from other chemical surveillance programmes since it addresses contaminants/nutrients in the diet as a whole rather than in individual foods and the exposure assessment focuses on the long-term average intake by the population. In fact, Total diet studies are designed to measure the average amount of each chemical/element ingested by the population of interest, thus addressing the chronic situation, whereby the average concentration over a long time is at stake and not the variety at a particular moment. In this context, the TDS provides a first step exposure assessment based on food consumption surveys and points out towards priority nutrients or contaminants that need to be further investigated. The intake data can then be used to examine a specific element of interest (Lombardi-Boccia et al., 2003).

This study provides, for the first time in Lebanon, an estimate of the dietary intake of six essential trace elements and examines the dietary exposure to two other toxic elements; data are then compared to those provided by studies conducted worldwide. In fact, the amount of trace elements and other metals ingested by man may be country-specific since it is directly related to dietary habits of the population as well as to the content of trace elements in foodstuff, the latter factor being largely determined by local soil characteristics such as organic matter content, pH and clay mineralogy as well as the addition of chemical products used as fertilizers, pesticides or insecticides which can increase the metal amounts in soil and water (Fernícola, 1983; Barnard et al., 1997). In the present study, mean exposure estimates of nutritional elements were found to be below the toxicological reference values and were found to satisfy nutritional recommendations for most trace elements except for manganese in males and iron in females. This study is not the first to identify iron as a potential nutrient of concern. For instance, a TDS conducted on Italian adults showed that mean exposure estimates of essential trace elements were all found to satisfy nutritional recommendations, except for iron, the intake of which was found to be inadequate among adult females (Turconi et al., 2008).

In this study, average dietary exposure to lead and cadmium was found to represent respectively 3% and 22% of the PTWIs. When compared to a previous TDS conducted 5 years earlier (Nasreddine et al., 2006b), the dietary exposure to cadmium is of the same order of magnitude (15.82 μg/day as compared to 12.3 μg/day) while the exposure to lead is two times lower (8.26 μg/day as compared to 18.5 μg/day) (Nasreddine et al., 2006b). This downward trend in lead intake has been reported from several other countries and coincides with a considerable reduction in the use of lead-soldered cans and in the lead content of petrol (Galal-Gorchev, 1993). Alternatively, the discrepancy in exposure estimates between the present study and those reported in 2006 may be due to differences in food sampling schemes whereby the number of market baskets (three in the present study vs. five in 2006) and the level of aggregation and compositing of food samples differed between the present study and that of 2006. The discrepancy in exposure estimates may also be an artifact of the difference in analytical limits for both lead and cadmium, given that the limits of detection and of quantification were much lower in the present study as compared to those used in 2006, a fact that may have direct repercussions on exposure estimates and the contribution of non detected or non quantifiable results to dietary exposure.

The results of this study suggest that the dietary exposure to the toxic elements Pb and Cd is low, and that for the average consumer, there is no risk of exceeding the 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 a "total diet" 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 form the TDS. Examples of such foods would include mollusks and crustaceans which accumulate high levels of lead, oysters which accumulate high levels of cadmium, and kidneys which may contain high amounts of both lead and cadmium. According to the individual food consumption survey data (Nasreddine et al., 2006a), 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 (crustaceans, mollusks, 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 lead and cadmium than the average consumer.

As shown in Table 5, mean intake estimates of the studied trace elements fall within the range of values reported by Total diet studies conducted in different parts of the world, except for Pb, the intake of which appeared to be lower than that reported by most other countries. These comparisons provide valuable insight as to inter-country variability in dietary exposure to trace elements. Nevertheless, such comparisons should be interpreted with caution since in many instances the studies may differ in the limit of quantification and detection of the analytical technique, the assumptions made in intake calculations if concentrations are below the limit of quantification or detection, the weight and energy content of the 'total diet', the degree of preparation of the 'total diet' as well as the model used for estimating dietary exposure (Kroes et al., 2002).

This study helped in identifying the specific food groups most responsible for major or minor contributions to the intakes of the studied elements. This information is warranted since contribution of any food group to daily intakes depends on the concentration of the element in this particular food but also on the amount consumed. Thus a food item that is rich in a certain element but infrequently consumed by the population would not have an important contribution to the daily intake of that element. Consequently, food groups that represent the major source of an element in one country may not be the same in another where the population has a different dietary pattern. This can be illustrated by data provided by Total diet studies conducted in different parts of the world. For instance, the main dietary source of Cu was black beans (60%) in Brazil (Santos et al., 2004), meat and poultry in the USA (Egan et al., 2002) while cereals were the main contributor to the intake of this element in Italy (35%) (Lombardi-Boccia et al., 2003) as well as in the present study (44.1%). It is important to note that, in this study, plant-based products including breads, cereals and vegetables were found to be the main contributors to most of the studied nutritional elements. More particularly, breads and cereals were

Table 5
Comparison of the average dietary exposure to Cd, Cu, Co, Fe, Mn, Ni, Pb and Zn as determined by different Total diet studies in several countries and in the present study.

Country	Cd (μg/day)	Co (μg/day)	Cu (mg/day)	Fe (mg/day)	Mn (mg/day)	Ni (μg/day)	Zn (mg/day)	Pb (μg/day)	References
China	21.2[b]	–	4.8[a]	22.7[a]	5.9[a]	–	9.8[a]	81.2[b]	Chen and Gao (1993)[a], Gao (2004)[b]
France	2.7	7.53	0.98	–	2.31	93.7	8.66	18.0	Leblanc et al. (2005a,b)
Italy	13.6	29.0	1.14	11.0	1.38	361.1	12.0	55.2	Turconi et al. (2008)
Nine Asian countries	–	9.6–17.9	0.87–2.17	6.6–31.4	2.08–10.54	–	4.3–13.5	–	Iyengar et al. (2002)
Spain	16–29	–	1.6–8.7	11–14.3	–		10.1–15.2	37–521	Cuadrado et al. (1995)
Sweden	12.0	–	1.2	16	3.7 mg/day	82	12	17.0	Becker and Kumpulainen (1991)
The Netherlands	21.0	–	1.5	14	–		14	32.0	Van Dokkum et al. (1989)
UK	14.0	–	1.2		–	130	8.4	24.0	Ysart et al. (1999)
USA	11.5–14.2[a]	14.0[b]	0.73–1.36[a]	9.0–13.9[a]	2.07–2.81[a]		7.6–12.7[a]	4.2–18.8[a]	Egan et al. (2002)[a], Iyengar et al. (2000)[b]
Lebanon	15.82	11.4	1.10	13.00	2.04	126.27	10.97	8.26	Present study

[a,b] These superscripts are used to indicate, within each row, the references from which dietary exposure estimates were derived.

the main dietary source of Mn, Cu, Fe, Ni while vegetables represented the primary source of cobalt. This can be explained by the fact that the Lebanese diet relies heavily on cereals and vegetables. In fact, breads and cereals were found to provide alone 35% of the total energy intake of the average Lebanese urban adult (Nasreddine et al., 2006a). 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 is a characteristic shared by other Mediterranean countries such as Greece, Italy and Spain (Cuadrado et al., 1995; Lombardi-Boccia et al., 2003), where cereals and other plant-based food products were also found to be the main dietary source of several trace elements including Mn, Cu, Fe, Ni and Co. In the present study, the main contributors to the dietary intake of Pb and Cd were vegetables (48.7% and 46.8% respectively) followed by breads and cereal-based products (31.4% and 30.9% respectively). It is noteworthy that most of the foods identified as being the major contributors to lead and cadmium intake were also reported by several other countries as being the main dietary source of these heavy metals (Van Dokkum et al., 1989; Dabeka and McKenzie, 1995; Leblanc et al., 2005a; Sommerfeld, 2004).

The distribution of exposure levels, which reflects the variability in food-consumption patterns, showed that the extreme level of intake (97.5th percentile) was below the toxicological reference values for all trace elements and none of the individuals' dietary intake exceeded these reference values (TDI, PTWI or PMTDI). However, the dietary intake of Fe appeared to be inadequate for the majority of adult females. This indicates that this group of the population is at a potential risk of iron deficiency anemia, a risk that may be exacerbated by the fact that the major source of dietary iron was the group of breads and cereals which represent a source of non-heme iron. In fact, the results of this study indicate that in the average diet of the average individual, about 71% of the Fe is consumed as non-heme Fe, which raises a concern about the bioavailability of the element. A previous study conducted in Lebanon showed that 13.4% of women of child bearing age have iron deficiency anemia and 20.2% are iron deficient, thus indicating that iron status may be problematic in this population group in Lebanon (Hwalla et al., 2004). Iron nutrition has also been documented as a serious public health problem in most countries of the Eastern Mediterranean (WHO, 2004). It is estimated that more than one third of the population is anemic in the region: anemia ranged from 11% in Egypt to over 40% in the Syrian Arab Republic and Oman among women of childbearing age (WHO, 2004).

In conclusion, this study examined by the TDS approach the dietary intake of a Lebanese adult population to several trace elements of nutritional or toxicological significance. Accordingly, mean exposure estimates were found to be below the toxicological reference values for all the studied elements and were found to satisfy nutritional recommendations for most trace elements except for manganese in males and iron in females. Based on the distribution of exposure levels, none of the individuals' dietary intake exceeded any of the toxicological reference values, but the dietary intake of iron appeared to be inadequate for the majority of adult females, thus suggesting that the nutritional status of Fe should be further investigated, particularly in vulnerable population groups such as women of child bearing age. Even though the daily intake data provided by a TDS are insufficient to ascertain the adequacy of the diet, they offer important information about the nutritional quality of the national diet and about potential nutritional problems. The intake data can then be used to examine a specific element of interest. The study's findings should thus constitute a current measure of assessing the adequacy and safety of the foods consumed by the adult urban Lebanese population with respect to the studied trace elements and may be a basis for future monitoring studies.

The study has some limitations as the population sample was an urban one, selected from the capital Beirut. The choice of Beirut can be explained by the fact that it comprises 40% of the Lebanese population and is usually considered a melting pot of the country. However, the extrapolation of the results to the country as a whole remains controversial. Furthermore, this study was confined, like many exposure assessments conducted around the world, to the adult fraction of the population (Buchet et al., 1983; WHO, 1985; Chen and Gao, 1993; Tsuda et al., 1995; Jalon et al., 1997). The average dietary intake estimates were based on the chemical analysis of a diet for which the consumption data were not differentiated by sex. Thus, while not excluding the possibility that the daily intakes determined in the present study may not be representative of the population as a whole, this study has provided a first estimate of the consumer exposure to trace elements through the diet in Lebanon. However, a more complete assessment of the dietary exposure of the Lebanese population to trace elements as well as to other food contaminants such as pesticide residues and mycotoxins is needed in order to ensure the safety of the food supply (WHO, 1999).

Conflict of interest statement

The authors declare that there are no conflicts of interest.

Acknowledgements

The study was supported by the French–Lebanese Cooperative Research Programme (CEDRE) and by the Lebanese National Council for Scientific Research through its support to the Associated Research Unit on Undernutrition and Obesity in Lebanon.

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