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Concentrations of Arsenic, Cadmium, Mercury, and Lead in Common Foods and Estimated Daily Intake by Children, Adolescents, Adults, and Seniors of Catalonia, Spain

J. M. LLOBET,[†,‡] G. FALCÓ,[†] C. CASAS,[§] A. TEIXIDÓ,[§] AND J. L. DOMINGO*[,‡]

Toxicology Unit, School of Pharmacy, University of Barcelona, 08034 Barcelona, Spain; Laboratory of Toxicology and Environmental Health, School of Medicine, "Rovira i Virgili" University, 43201 Reus, Spain; and Department of Health and Social Security, Generalitat de Catalunya, 08028 Barcelona, Spain

This study was designed to estimate the dietary intake of arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) by the general population of Catalonia, Spain. The concentrations of these elements were determined in food samples randomly acquired in seven cities of Catalonia between June and August 2000. A total of 11 food groups were included in the study. As, Cd, Hg, and Pb levels were measured by ICP-MS and AAS. The dietary intake of these elements was determined by a total diet study. Calculations were carried out on the basis of recent data on the consumption of the selected food items. Trace element intake was estimated for five population groups: children, adolescents, male and female adults, and seniors. The highest dietary intakes of As (223.6 μg/day), Cd (15.7 μg/day), Hg (21.2 μg/day), and Pb (28.4 μg/day) corresponded to male adults. For all analyzed elements, fish and shellfish was the group showing the highest contribution to the respective intakes. In comparison with previous results, a general decrease in As, Cd, Hg, and Pb intake has occurred. The dietary intake of these elements was also compared with the provisional tolerable weekly intake (PTWI). Dietary intakes of As, Cd, Hg, and Pb by the population of Catalonia are currently well below the respective PTWIs.

KEYWORDS: Arsenic; cadmium; mercury; lead; food; dietary intake; Catalonia, Spain

INTRODUCTION

Arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) are widely dispersed in the environment. These elements have no beneficial effects in humans, and there is no known homeostasis mechanism for them (*1*). Although toxicity and the resulting threat to human health of any contaminant are, of course, a function of concentration, it is well-known that chronic exposure to As, Cd, Hg, and Pb at relatively low levels can cause adverse effects. Although some individuals are primarily exposed to these contaminants in the workplace, for most people the main route of exposure to these toxic elements is through the diet. Consequently, information about dietary intake is very important to assess risks to human health. To evaluate the health risks to consumers, it is necessary to determine the specific dietary intake of each pollutant for comparison with toxicologically acceptable levels (*2*). In relation to this, it is well established that there are notable differences in both food consumption and food contamination by metals or other contaminants among different regions and countries (*3*−*5*).

During the past decade, we estimated the dietary intake of a number of trace elements by the population of Tarragona, a province of Catalonia (northeastern Spain) (*6*−*9*). As, Cd, Hf, and Pb were included in the monitoring program, which includes repeated surveys over time. Taking into account the important efforts that in recent years have been carried out in most industrialized countries to reduce the environmental levels of toxic elements, the main purpose of the present survey was to update and extend the information about dietary intake of As, Cd, Hg, and Pb by the general population of Catalonia. Intakes were estimated for subjects from five groups: children (aged 4−9 years), adolescents (aged 10−19 years), male adults (aged 20−65 years), female adults (aged 20−65 years), and seniors (aged >65 years).

According to the FAO/WHO (*10*), three basic approaches can be used to determine the intake of a food contaminant: (1) total diet study, (2) duplicate diet method, and (3) diary study, which combines data for specific contaminants in food with individual (or household) consumption data. In the present study, the total diet study was used to determine As, Cd, Hg, and Pb dietary intakes. Samples of a number of food items were individually analyzed to obtain detailed information on the variation of these elements in foods eaten by the population of

* Author to whom correspondence should be addressed (telephone +34 977 759380; fax +34 977 759322; e-mail jlldr@fmcs.urv.es).
[†] University of Barcelona.
[‡] "Rovira i Virgili" University.
[§] Generalitat de Catalunya.

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Catalonia. To estimate the dietary intake of As, Cd, Hg, and Pb, these results were combined with food consumption data.

MATERIALS AND METHODS

Sampling. Between June and August 2000, food samples were randomly obtained in local markets, large supermarkets, and grocery stores from seven cities (Barcelona, Tarragona, Lleida, Girona, L'Hospitalet de Llobregat, Badalona, and Terrassa) of Catalonia, Spain, which have populations between 150,000 and 1,800,000 inhabitants. For collection of samples two groups were made up. The first group included meat of beef (steak, hamburger), pork (loin, sausage), chicken (breast), and lamb (steak); fish (hake, sardine) and shellfish (mussel); vegetables (lettuce, tomato, potato, green beans, cauliflower); fresh fruits (apple, orange, pear); and eggs. The second group included cow's milk (whole, semiskimmed) and dairy products (yogurt, cheese); cereals (bread, pasta, rice); pulses (lentils, beans); fats (margarine) and oils (olive, sunflower); tinned fish (tuna, sardine); and meat products (ham, hot dogs, salami). Because in the first group most products are usually retailed, their origins could be very diversified in the different cities. Therefore, in that group four composite samples were analyzed for each food item. Each composite was made up by 10 individual samples. In contrast, most food items included in the second group corresponded to brands/trademarks that could be obtained in many different places. Consequently, in this group only two composite samples were analyzed for each food item. Each composite was made up by eight individual samples. A total of 108 samples were analyzed for As, Cd, Hg, and Pb concentrations.

Analytical Methods and Instrumentation. As, Cd, Hg, and Pb in food samples were determined according to previously described methods (*5*, *7*). A microwave (Milestone 1200) assisted digestion procedure was used. Between 0.5 and 3 g (depending on each food item) of homogenized samples was digested under pressure in Teflon vessels with 4 mL of nitric acid (Suprapur, E. Merck, Darmstadt, Germany) and 1.5 mL of hydrogen peroxide (E. Merck). Samples with a low aqueous content were ashed at 450 °C in a Selecta furnace. On completion of the digestion and after adequate cooling, solutions were filtered and made up to 50 mL with 1% nitric acid. For the ashes, 1% nitric acid was used as quantitative solution.

As, Cd, and Pb contents in samples of meat, eggs, milk and dairy products, cereals, and fats and oils were determined by inductively coupled plasma-mass spectrometry (ICP-MS, Varian-Vista with an ultrasonic nebulizer U5000AT), whereas Hg concentrations were determined using a Perkin-Elmer 2380 spectrophotometer coupled to a hydride generation Perkin-Elmer MHS-10 model. As, Cd, Hg, and Pb contents in the remaining food samples were determined using a Varian spectrophotometer (Spectra 600 ABQ) coupled to a hydride generation/cold vapor VGA-77 Varian.

The accuracy of the instrumental methods and analytical procedures were checked by duplication of the samples, as well as by using the following reference solutions: BCR(CMR-186) porcine kidney for Cd, Hg, and Pb; BCR(CMR-422) cod muscle for As; and MR-119(FAPAS) powdered milk for Cd and Pb. The coefficients of variation for the different samples were between 15 and 30%, whereas the recovery rates for the elements analyzed under the experimental conditions were found to be between 80 and 120%.

Dietary Exposure Estimates. The daily intake of As, Cd, Hg, and Pb from each food item was calculated by multiplying the respective concentration in each food by the weight of that food group consumed by an *average* individual from Catalonia (*11*, *12*). Finally, total dietary intake was obtained by summing these products for all food groups. For calculations, when an element concentration was under the limit of detection (LOD), the value was assumed to be half of the respective detection limits (ND = $^1/_2$ LOD), which were in the following ranges depending on the analyzed food item: As, 0.03−0.20 μg/g; Cd, 0.01−0.07 μg/g; Hg, 0.02−0.15 μg/g; and Pb, 0.02−0.30 μg/g.

RESULTS AND DISCUSSION

Figures 1−4 show the concentrations of As, Cd, Hg, and Pb in a number of foods classified into the following 11 groups:

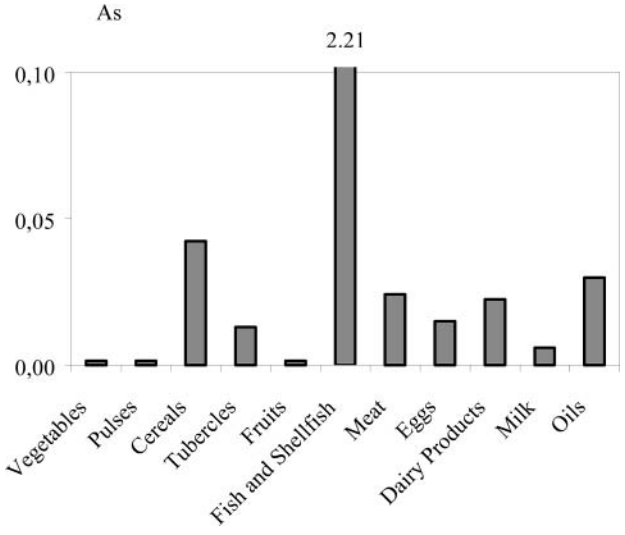

Figure 1. Arsenic concentrations (μg/g of wet weight) in foods from Catalonia.

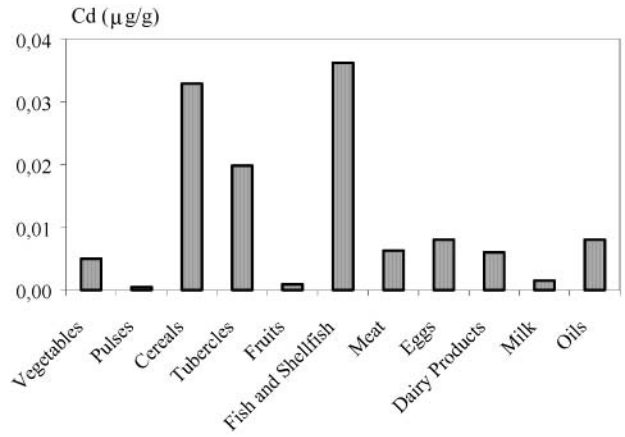

Figure 2. Cadmium concentrations (μg/g of wet weight) in foods from Catalonia.

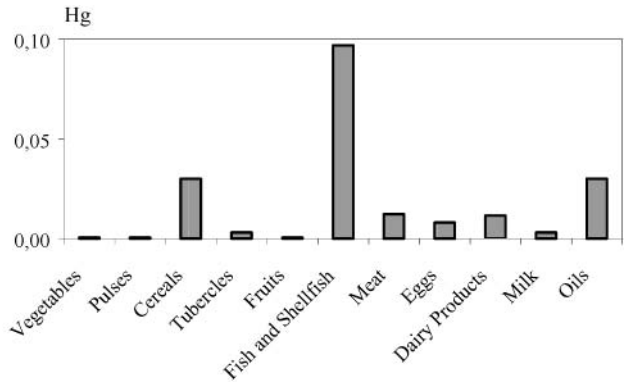

Figure 3. Mercury concentrations (μg/g of wet weight) in foods from Catalonia.

vegetables, pulses, cereals, tubercles, fruits, fish and shellfish, meat, eggs, milk, dairy products, and fats and oils. The highest concentrations of the four elements were found in the group of fish and shellfish, whereas the levels of these elements in cereals, especially for Cd, were also comparatively high. In contrast, the lowest As, Cd, and Hg concentrations were found in pulses, vegetables, and fruits, whereas the lowest Pb levels were detected in pulses and milk, followed by fruits.

Tables 1−5 summarize data on daily consumption of the 11 food groups for children, adolescents, male adults, female adults,

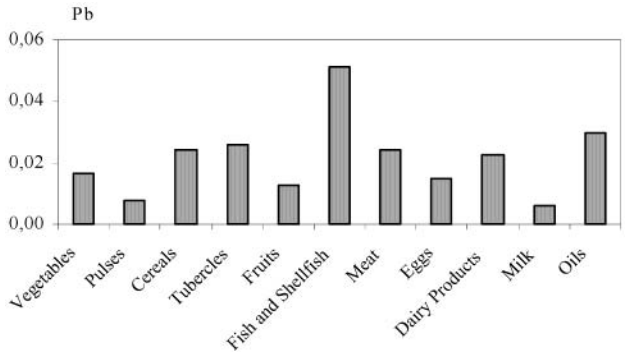

Figure 4. Lead concentrations (μg/g of wet weight) in foods from Catalonia.

Table 1. Food Intake and Intake of Arsenic, Cadmium, Mercury, and Lead through the Diet of Children in Catalonia, Spain

food group	food intake, g/day	As intake, μg/day	Cd intake, μg/day	Hg intake, μg/day	Pb intake, μg/day
vegetables	125	0.19	0.63	0.06	2.04
pulses	25.5	0.04	0.01	0.01	0.20
cereals	200.5	8.42	6.62	6.02	4.81
tubercles	63.5	0.83	1.26	0.19	1.64
fruits	16	0.29	0.18	0.10	2.47
fish and shellfish	51.5	113.82	1.86	5.00	2.64
meat	140	3.36	0.84	1.68	3.36
eggs	26.5	0.40	0.21	0.21	0.40
dairy products	114	2.62	0.68	1.37	2.62
milk	309	1.85	0.62	0.93	1.85
fats and oils	33.5	1.01	0.27	1.01	1.01
total		132.82	13.17	16.57	23.04

Table 2. Food Intake and Intake of Arsenic, Cadmium, Mercury, and Lead through the Diet of Adolescents in Catalonia, Spain

food group	food intake, g/day	As intake, μg/day	Cd intake, μg/day	Hg intake, μg/day	Pb intake, μg/day
vegetables	162.5	0.24	0.81	0.08	2.65
pulses	24	0.04	0.01	0.01	0.18
cereals	221	9.28	7.29	6.63	5.30
tubercles	76.5	0.99	1.51	0.23	1.98
fruits	202	0.30	0.18	0.10	2.55
fish and shellfish	62	137.02	2.24	6.01	3.17
meat	167	4.01	1.00	2.00	4.01
eggs	25.5	0.38	0.20	0.20	0.38
dairy products	122.5	2.82	0.74	1.47	2.82
milk	266.5	1.60	0.53	0.80	1.60
fats and oils	36	1.08	0.29	1.08	1.08
total		157.77	14.82	18.63	25.73

and seniors, respectively, living in Catalonia, as well as the dietary intake of As, Cd, Hg, and Pb for each of these five age and/or sex groups. In turn, the daily intakes for the same groups are depicted in **Figures 5−8** for As, Cd, Hg, and Pb, respectively.

The highest As intake corresponded to the group of male adults, 223.6 μg/day, whereas the lowest intake of this element corresponded to children and adolescents, 132.8 and 157.8 μg/day, respectively. For all five groups, the consumption of fish and shellfish was the main food group responsible of these

Table 3. Food Intake and Intake of Arsenic, Cadmium, Mercury, and Lead through the Diet of Male Adults in Catalonia, Spain

food group	food intake, g/day	As intake, μg/day	Cd intake, μg/day	Hg intake, μg/day	Pb intake, μg/day
vegetables	226	0.34	1.13	0.11	3.68
pulses	24	0.04	0.01	0.01	0.18
cereals	206	8.65	6.80	6.18	4.94
tubercles	74	0.96	1.47	0.22	1.92
fruits	239	0.36	0.22	0.12	3.01
fish and shellfish	92	203.32	3.33	8.92	4.71
meat	185	4.44	1.11	2.22	4.44
eggs	34	0.51	0.27	0.27	0.51
dairy products	106	2.44	0.64	1.27	2.44
milk	217	1.30	0.43	0.65	1.30
fats and oils	41	1.23	0.33	1.23	1.23
total		223.59	15.73	21.22	28.37

Table 4. Food Intake and Intake of Arsenic, Cadmium, Mercury, and Lead through the Diet of Female Adults in Catalonia, Spain

food group	food intake, g/day	As intake, μg/day	Cd intake, μg/day	Hg intake, μg/day	Pb intake, μg/day
vegetables	202.3	0.30	1.01	0.10	3.30
pulses	22.6	0.03	0.01	0.01	0.17
cereals	138.3	5.81	4.56	4.15	3.32
tubercles	57	0.74	1.13	0.17	1.48
fruits	226.6	0.34	0.20	0.11	2.86
fish and shellfish	79.3	175.25	2.87	7.69	4.06
meat	125	3.00	0.75	1.50	3.00
eggs	23.3	0.35	0.19	0.19	0.35
dairy products	91.3	2.10	0.55	1.10	2.10
milk	253.3	1.52	0.51	0.76	1.52
fats and oils	31	0.93	0.25	0.93	0.93
total		190.38	12.03	16.71	23.08

Table 5. Food Intake and Intake of Arsenic, Cadmium, Mercury, and Lead through the Diet of Seniors in Catalonia, Spain

food group	food intake, g/day	As intake, μg/day	Cd intake, μg/day	Hg intake, μg/day	Pb intake, μg/day
vegetables	189.5	0.28	0.95	0.09	3.09
pulses	22	0.03	0.01	0.01	0.17
cereals	156.5	6.57	5.16	4.70	3.76
tubercles	69.5	0.90	1.38	0.21	1.80
fruits	268	0.40	0.24	0.13	3.38
fish and shellfish	80	176.80	2.90	7.76	4.10
meat	114	2.74	0.68	1.37	2.74
eggs	22.5	0.34	0.18	0.18	0.34
dairy products	72	1.66	0.43	0.86	1.66
milk	253.5	1.52	0.51	0.76	1.52
fats and oils	29	0.87	0.23	0.87	0.87
total		192.12	12.67	16.95	23.41

intakes. This contribution was found to be between 85.7% of the total intake for children and 92.0% for adult females and seniors. Remarkably lower were the contributions of cereals and meat, as well as those of the remaining food groups.

For Cd, cereals was the group showing the highest contribution to dietary intake, which ranged from 37.9% for female

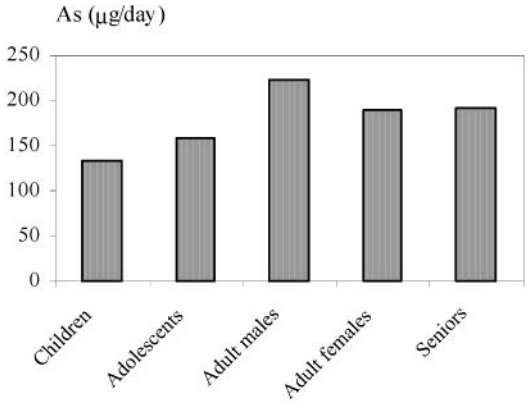

Figure 5. Dietary intake of As by the population of Catalonia.

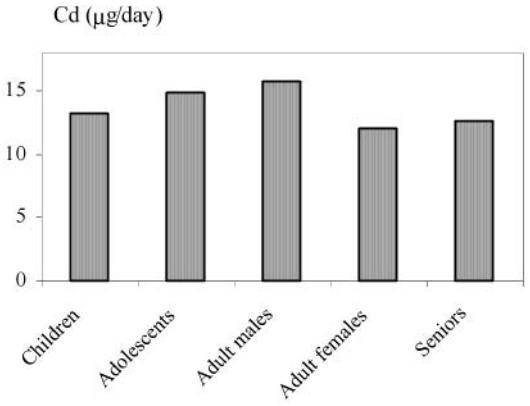

Figure 6. Dietary intake of Cd by the population of Catalonia.

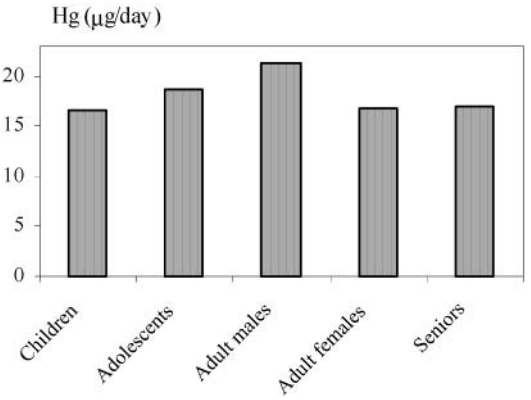

Figure 7. Dietary intake of Hg by the population of Catalonia.

adults to 50.3% for children. Fish and shellfish was the second group in quantitative importance to daily Cd intake. Its contribution varied between 23.9% for female adults and 14.1% for children. The highest Cd intake corresponded to male adults, with 15.7 μg/day, followed by adolescents, with 14.8 μg/day.

As for Cd, the highest Hg intake was observed in male adults (21.2 μg/day) and adolescents (18.6 μg/day). However, although in male adults as well as in the groups of female adults and seniors, fish and shellfish was again the main food group responsible of Hg intake (from 46.0% for female adults to 42.1% for male adults), cereals was the group showing the highest contribution to Hg intake in children and adolescents. In these age groups, fish and shellfish was the second contributor in percentage to Hg intake, whereas for all groups meat was the third contributor with an average percentage of ~10%.

With respect to Pb, the greatest intake again corresponded to the groups of male adults (28.4 μg/day) and adolescents (25.7 μg/day). In both groups, as well as in children, the food group

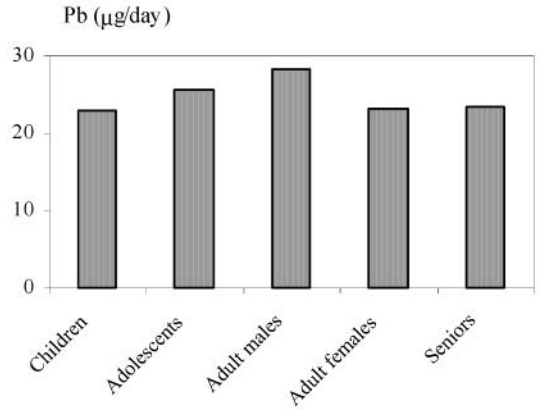

Figure 8. Dietary intake of Pb by the population of Catalonia.

showing the highest contribution to Pb intake was cereals. Although cereals were also an important contributor to Pb intake in the groups of female adults (14.3%) and seniors (16.1%), the main contributor in these age groups was fish and shellfish, with ~18.0%. Lead intake from vegetables (female adults) and fruits (seniors) was also quantitatively important, with percentages of 14.3 and 14.4%, respectively.

For an assessment of health risks of the above intakes, these have been compared with the current provisional tolerable weekly intakes (PTWI) for As, Cd, Hg, and Pb (*13*). For inorganic As, the PTWI is 15 μg/kg of body weight/week or 128 μg/day for a subject of 60 kg. In the present study, all analyses were carried out for total (organic and inorganic) arsenic. However, it is well-known that most As found in fish and shellfish is organic As, which is the less toxic form. In the literature, the percentage of inorganic As in fish and shellfish has been reported to be between 0.02 and 11% (*14*), whereas the maximum acceptable daily load for As, set by the WHO in 1967 and unrevised in 1989, is 3000 μg for a subject of 60 kg (*15*). Taking this into account, in the current study the intake of As estimated would not be of concern for any age group. The total As intake was lower than that found in traditional fish-consuming countries such as the Basque country (Spain) and Japan, with values of 297 μg/day (*16*) and 280 μg/day (*17*), respectively. In comparison with our previous study performed in Tarragona province there was also a remarkable reduction from an average value of 272.7 μg/day (*7*) to the current 223.6 μg/day (male adults).

For Cd, the intake represented between 26.2% of the PTWI (60 μg/day for a subject of 60 kg) for male adults and 20.1% of the PTWI for females. The average current Cd intake is similar to or lower than that recently reported for other European countries: United Kingdom, 12 μg/day (*18*); Basque country, 11−29 μg/day (*19*); France, 17 μg/day (*2*); Denmark, 15 μg/day (*20*); and Germany, 22.2 μg/day (for an adult of 60 kg) (*21*). It was also lower than the intake found in our recent survey in Tarragona province, 18 μg/day (*7*), and notably lower than that found in 1991 in the same province, 56 μg/day (*6*).

For Hg, the PTWI has been established at 5 μg/kg of body weight as total Hg, or 43 μg/day for an individual weighing 60 kg. In the present survey and as a percentage of the PTWI, Hg contribution varied between 49.3% for adult males and 38.5% for children. Although the current dietary Hg intakes were lower than the PTWI, they were higher than those previously reported for subjects living in Tarragona province (Catalonia), 4.8 μg/day (9), or for the British, 3 μg/day (*18*), and Danish, 3.3 μg/day (*20*), populations.

Finally, a PTWI of 25 μg/kg of body weight was established for Pb by the FAO/WHO (*13*), which is equivalent to 214 μg/

day for an individual of 60 kg. In the current study, the highest and lowest Pb intakes corresponded to adult males (28.37 μg/day) and children (23.04 μg/day), respectively. These values are equivalent to 13.3 and 10.8% of the PTWI. In agreement with the general tendency reported for most developed countries, Pb intake was remarkably reduced in relation with previous studies performed in Catalonia during the 1990s: 115 μg/day (*6*) and 49 μg/day (*9*). The current intakes are also lower or very similar to those recently reported for other industrialized countries: Basque country, 34 μg/day (*22*); United Kingdom, 26 μg/day (*18*); and France, 52 μg/day (*2*). They are, however, still higher than Pb intake in Denmark, with a median of 18 μg/day (*20*).

The results of this study show that in relation with previous surveys (*6*−*9*), the dietary intakes of As, Cd, Hg, and Pb by the population of Catalonia have been remarkably reduced. Taking into account that in comparison with our 1998 survey (*9*), changes in dietary habits of the population of Catalonia have not been especially relevant, the main reason for the notable decrease in the dietary intakes of As, Cd, Hg, and Pb should be the important reduction of these elements in foods.

The intakes estimated for As, Cd, and Pb were notably lower than the respective PTWIs, which indicates that these intakes do not cause any health concern for any age group of the population assessed. Although the current intakes of As, Cd, and Pb from the diet of the general population of Catalonia pose little risk, there may be specific groups of individuals whose dietary pattern might result in an increased intake of these elements. For example, because fish and shellfish has been the group showing the highest levels of As, Cd, and Pb, those subjects (i.e., vegetarians) following diets that imply a notable consumption of foods of this group could significantly increase the intake of these elements and, hypothetically, the potential health risks. With respect to Hg, although its dietary intake was also lower than the PTWI, it is still comparatively high. Future surveys should corroborate the decreasing trend observed in the present study.

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