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# Lead Dietary Intake in a Spanish Population (Canary Islands)

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For most people diet is the main route of exposure to trace metals, so information about dietary intake is also important to assess risks to human health for these elements. The purpose of this study was to determine the levels of Pb in the foods and drinks of highest consumption in the authors' our community to estimate daily intakes of Pb for each of the seven Canary Islands. Four hundred and twenty samples were analyzed using GFAAS. The total Pb intake of the Canarian population is 72.8  $\mu$ g/day, 29.12% (for a person of 70 kg body weight) of the provisional tolerable weekly intake limit of 25  $\mu$ g/kg fixed by the FAO/WHO. The island that presents the highest lead intake is La Gomera, followed by Lanzarote, Tenerife, and Gran Canaria islands. These four islands present a lead dietary intake over the mean Pb intake for the whole archipelago. The islands with lower Pb intakes are La Palma and Fuerteventura, with intakes under 70  $\mu$ g/day. These results have also been compared with the values found for other national and international communities.

KEYWORDS: Lead; AAS; dietary intake; Spanish population

## INTRODUCTION

Heavy metals are considered to be most harmful to living systems. The main source of these metals for humans is food and water. Some authors consider that alcohol and tobacco represent major sources of lead exposure (1). Increased dietary lead intake may cause functional disturbances, especially in children and adolescents (2). Nevertheless, many studies have concluded that risks due to dietary Pb intake seem to be low, for example, in Germany and Brazil (3, 4).

High concentrations of lead have been linked to human health problems including nervous system dysfunction of the fetus and infants and, in adults, hemotoxic effects, reproductive dysfunction, gastrointestinal tract alterations ("saturnine colic"), nephropathies, and Alzheimer's disease. It is also well-known that Pb poisoning may lead to anemia, because the activity of heme synthesis enzymes is inhibited by Pb exposure (5-8). Lead is also considered to be a risk factor for hypertension in women (9).

Pb poisoning and iron deficiency are often two associated problems. Iron deficiency increases susceptibility to Pb poisoning. Both conditions are known to cause anemia and appear to produce a more severe anemia when in combination (10). A low iron intake has also been related with high blood Pb concentration (11, 12). Dietary iron has been strongly recommended as a secondary preventive intervention against lead toxic effects in Korean lead workers (11).

Agricultural technology, industrial pollution, geological sources, and food processing are the most outstanding sources of food contamination by metals (13). During early childhood, lead-contaminated house dust can also be considered as an important source of Pb intake (14). It is well-known that vegetables absorb these metals from the atmosphere by dusts deposited on their surfaces (15). The application of sewage sludges on croplands may increase the accumulation of this metal in plants and their transfer to the human food chain (16). The use of a copper fungicide contaminated with lead has been considered to be the cause of the high Pb levels in Turkish raisins (0.93 mg/kg) sold in Canada (17).

The paint and varnish industries and the use of tetraethyl lead as antiknock preservative in petroleum products have been important lead sources for the environment (7, 18-20). Nevertheless, the introduction of unleaded petroleum products has significantly reduced levels of atmospherically deposited lead in soils and plants and lowered blood lead concentrations and dietary intakes in human populations (5, 21).

Moreover, some glazed pottery, especially of the craft and homemade kind, which has not been kilned at a sufficiently high temperature or has been made with poorly formulated frits, is capable of releasing toxic amounts of lead into food (22). Glass containers used in food products cannot contain >24%of lead oxide in order to avoid toxicity after lead migration from the glass to the food due to the acidity and heat (7).

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Table 1. Temperature Program of the Graphite Furnace in the Determination of  $\mathsf{Lead}^a$ 

step	temp (°C)	ramp time (s)	hold time (s)	argon flow (mL/min)
drying 1	110	1	20	250
drying 2	130	5	30	250
pyrolysis	850	10	20	250
atomization	1500 <sup>b</sup>	0	5	0
clean out	2400	1	2	250

<sup>a</sup> Injection temperature, 20 °C; matrix modifier, Mg (NO<sub>3</sub>)<sub>2</sub>/NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub>; wavelength, 217 nm; injection volume, 15 μL. <sup>b</sup> Reading temperature.

Developed countries present a Pb intake of substantially less than 250  $\mu$ g of Pb/day. Lead intake is generally higher in men than in women (23). In 1980–1988, average adult intakes of lead varied from 1 to 63  $\mu$ g/kg of body weight (bw)/week, approaching or exceeding the provisional tolerable weekly intake (PTWI) of 25  $\mu$ g/kg of bw in four countries providing data (24). Nevertheless, the lead intake in Denmark during 1993–1997 showed a decrease in comparison with similar estimates from the previous monitoring cycles (25). The same happens in the United Kingdom and The Netherlands. The Pb dietary intake for the general population of the United Kingdom has been reduced from 24  $\mu$ g/day in 1994 to 7.4  $\mu$ g/day in 2004, and since 1976–1978 the dietary intake of lead in The Netherlands has been reduced by a factor 3 (26, 27). This downward trend was observed consequent on reduction in the use of lead-

Table 2. Pb Recovery Study

soldered cans and of lead in petroleum products (24). Ikeda et al. (28) considered that Japan's decision in 1975 to withdraw alkyl lead from automobile gasoline had helped to lower the Pb intake levels and the blood lead levels.

The maximum limits for lead in certain food groups have been modified recently by Regulation (CE) 221/2002 of the European Commission (February 6, 2002) applicable from April 5, 2002 (29). The present WHO standard for Pb in drinking water is 10  $\mu$ g/L (30), and the maximum limit set for Pb content in wine by the International Office of Vine and Wine (OIV) is 200  $\mu$ g/L (31). Dietetic products have been recently associated with lead levels exceeding maximum allowed amounts (MAA) (32).

As food is usually the most important source of heavy metal intake, it is important to monitor the heavy metal dietary intakes (13, 33, 34). In this study we are determining the levels of Pb in the foods and drinks of highest consumption in our community in order to study the risk posed by the presence of this toxic heavy metal in food. For this aim we have used the intakes presented by the last Canarian Nutritional Survey (35).

#### MATERIALS AND METHODS

Pb was determined according to a standard method using graphite furnace atomic absorption spectrometry (GFAAS) (**Table 1**). This technique was, in fact, judged to be the most suitable for determining the element in different matrices given its good reproducibility and its high detection power. For the ashing of the samples we decided to select the dry-ashing procedure because the resulting ash can be

			concentration			
material	Na	measured <sup>b</sup>	certified <sup>c</sup>	accuracy (%)	procedure	
NBS SRH 1577a bovine liver	11	$0.13 \pm 0.02$	$0.14 \pm 0.02$	92.85	AAS graphite furnace	
BCR-278 R mussel tissue	11	$1.98 \pm 0.05$	$1.97 \pm 0.07$	100.51	AAS graphite furnace	
1515 apple leaves	11	$0.47 \pm 0.02$	$0.48 \pm 0.01$	97.91	AAS graphite furnace	
1573 tomato leaves	11	$4.70 \pm 0.14$	$4.71 \pm 0.12$	99.79	AAS graphite furnace	
1575 pine needles	11	$10.8 \pm 0.5$	$10.7 \pm 0.6$	100.93	AAS graphite furnace	
V-10 hay powder	11	$1.6 \pm 0.8$	$1.6 \pm 0.9$	100	AAS graphite furnace	

<sup>a</sup> Number of samples. <sup>b</sup> Mean  $\pm$  standard deviation. <sup>c</sup> Confidence interval = 95%.

#### Table 3. Food Consumption in the Canary Islands (ENCA, 2000)

	consumption (g/day)							
food	Canary Islands	Gran Canaria	Lanzarote	Fuerteventura	Tenerife	La Palma	La Gomera	El Hierro
milk	300.7	334.3	287.3	203.4	292.0	301.7	250.4	275.7
cheese	25.1	27.0	20.3	18.0	23.1	31.4	15.3	34.5
yogurt	45.7	52.2	34.5	60.8	49.1	30.4	33.8	46.3
other dairy products	19.2	19.1	17.2	9.5	20.7	20.9	13.6	19.5
fish	45.8	44.7	54.7	57.0	44.1	35.8	43.5	49.3
eggs	25.1	25.0	26.2	21.2	26.1	24.0	22.3	43.7
red meat	45.9	46.6	51.4	41.7	51.1	47.5	42.0	47.2
cold meats and sausages	25.9	26.9	23.6	27.5	29.2	28.2	24.8	30.3
viscera	1.2	0.5	0.0	3.3	1.5	1.9	0.2	0.4
poultry and rabbit	32.1	34.4	28.8	27.9	35.4	30.3	24.3	33.8
fats, oils	27.9	31.6	24.3	16.3	30.9	26.3	28.5	25.2
cereals	125.3	130.6	155.4	97.5	124.4	126.1	114.7	159.8
bakery products	33.1	31.0	37.8	29.7	37.8	39.4	42.6	45.2
legumes	27.2	21.2	32.9	38.5	26.2	27.4	36.9	29.7
fruits	218.4	235.8	196.0	161.8	215.9	215.6	152.3	239.7
nuts and dried fruits	1.9	2.2	1.2	0.1	1.8	1.9	1.8	1.7
vegetables	107.8	110.0	70.8	75.4	118.8	98.2	76.5	105.9
potatoes	143.2	137.0	115.7	81.7	163.7	144.1	238.8	119.2
cakes and sweet foods	48.8	55.3	60.5	27.3	47.5	43.1	40.8	50.3
alcoholic drinks	62.8	50.1	91.5	37.2	74.7	39.3	185.0	46.2
nonalcoholic drinks	590.5	801.8	456.5	110.0	606.0	389.5	395.7	396.9

 Table 4. Pb Mean Concentration in the Different Food Groups (Fresh Weight Basis)

food	no. of analyzed samples	Pb mean concn $(\mu g/kg) \pm SD$
milk	20	$12.0 \pm 56$
cheese	20	$2.3 \pm 4.9$
yogurt	20	$66.0 \pm 62.20$
other dairy products	20	$1.56 \pm 0.43$
fish	20	$367.00 \pm 241.64$
eggs	20	$10.0 \pm 18.55$
red meat	20	$37.30 \pm 58.61$
cold meats and sausages	20	$345.0 \pm 587.67$
viscera	20	$91.66 \pm 34.43$
poultry and rabbit	20	$22.43 \pm 21.87$
fats, oils	20	$0.8\pm0.083$
cereals	20	$1.66\pm0.928$
bakery products	20	$1.21 \pm 0.65$
legumes	20	$0.13\pm0.058$
fruits	20	$52.0\pm45.89$
nuts and dried fruits	20	$31.58 \pm 16.23$
vegetables	20	$0.14\pm0.045$
potatoes	20	$0.71\pm0.324$
cakes and sweet foods	20	$0.82\pm0.11$
alcoholic drinks	20	$120.0 \pm 305.51$
nonalcoholic drinks	20	$6.0\pm4.96$
water	20	7.3 ±1.56

dissolved in a small amount of diluent and this provides a much better detection limit than wet digestion (36-38).

**Biological Materials.** Food samples were randomly obtained from January to June 2000 from local markets, supermarkets, and grocery stores on each of the seven islands. The total diet was divided into groups. In every case, the number of samples analyzed by group was 20. Several brand names of each product, representing the most widely accepted and most frequently consumed in the Canary Islands, were selected for testing. These groups of selected foods are those that the Canarian Nutritional Survey (*35*) established as the groups of highest consumption in our community in 1997–1998. This survey was conducted using a method of food consumption at the individual level. Specifically, it used two 24-h recalls—administered on two nonconsecutive days—and a comprehensive food frequency questionnaire of 77 food items.

Table 5. Pb Intakes in the Canary Islands

The analytical quality recovery study was controlled with blanks, reference materials, and blind replicates to check the reliability of the method. The recovery percentages obtained were in all cases >92% (**Table 2**). The selected reference materials were those that covered the wider range of foods. Blind replicates were used to check the absence of lead contamination in the reagents used. All blind replicates showed a lead concentration below the detection limit of the GFAAS. The NIST SRH 1577a bovine liver from the National Institute of Standards and Technology (NIST) (Office of Standard Reference Materials) has been used before by many authors (21) in Pb, CD, and Cu analysis.

**Reagents.** Milli-Q deionized water (Millipore, Milford, MA), hydrochloric acid (10%) (Merck, Darmstadt, Germany), 1000 mg/kg Fisher lead standard solution (1000 mg/kg) certified for EAA, NBS SRH 1577a bovine liver, BCR-278 R mussel tissue, 1515 apple leaves, 1573 tomato leaves, 1575 pine leaves, and V-10 hay powder were used.

A 2% Acationox detergent solution, Sherwood, was used to wash all of the glass material.

Plastic and glass laboratory materials were kept in a 5% HNO<sub>3</sub> solution for one night and then washed with double-distilled water and dried in a laboratory heater with dust-free atmosphere.

**Apparatus.** A Perkin-Elmer 4100 ZL atomic absorption spectrophotometer (Perkin-Elmer, Norwalk, CT) fitted with a graphite furnace, an AS70 automatic injector, and a Zeeman effect background correction was used for determination. Background correction systems measure the background absorption signal and subtract it from the total absorbance signal to give a correct signal.

A Heraeus furnace oven with automatic adjustable temperature was used.

Two hundred and fifty watt infrared lamps were from Osram.

Analytical Procedures. Oils, drinking water, and alcoholic and nonalcoholic drink samples needed no preparation; therefore, they were directly injected in the GFAAS. For the rest of the food groups, 10 g of each homogenized sample was weighed in a china capsule and totally dried under an infrared lamp. Later, the samples were brought to the muffle oven furnace where the pyrolysis process took place at  $450 \pm 10$  °C during 24-48 h. The white ashes obtained were then dissolved in 10 mL of concentrated nitric acid. This volume was later adjusted at 100 mL with double-distilled water. The resulting solutions were then taken to the graphite furnace atomic absorption spectrometer. For each measurement  $15 \,\mu$ L of sample and  $15 \,\mu$ L of matrix modifier were injected. The resonance line for measurement was at 228.8 nm. Table 1 presents the temperature program of the graphite furnace (*39*, *40*).

	intake (µg/day)							
food	Canary Islands	Gran Canaria	Lanzarote	Fuerteventura	Tenerife	La Palma	La Gomera	El Hierro
milk	3.60	4.011	3.447	2.441	3.504	3.620	3.005	3.308
cheese	0.05	0.062	0.046	0.041	0.053	0.072	0.035	0.079
yogurt	3.01	3.445	2.277	4.013	3.240	2.006	2.231	3.056
other dairy products	0.03	0.029	0.027	0.015	0.032	0.032	0.021	0.03
fish	16.80	16.405	20.075	20.919	16.185	13.138	15.964	18.093
eggs	0.25	0.250	0.262	0.212	0.261	0.240	0.223	0.437
red meat	1.71	1.738	1.917	1.555	1.906	1.772	1.566	1.760
cold meats and sausages	8.93	9.280	8.142	9.487	10.074	9.729	8.556	10.453
viscera	0.11	0.046	0	0.302	0.137	0.174	0.018	0.036
poultry and rabbit	0.72	0.771	0.646	0.626	0.794	0.679	0.545	0.758
fats, oils	0.02	0.025	0.019	0.013	0.025	0.021	0.023	0.020
cereals	0.208	0.217	0.258	0.162	0.206	0.021	0.190	0.265
bakery products	0.04	0.037	0.046	0.036	0.046	0.047	0.051	0.054
legumes	0.003	0.002	0.004	0.005	0.003	0.003	0.005	0.004
fruits	11.35	12.261	10.192	8.413	11.227	11.211	7.919	12.464
nuts and dried fruits	0.06	0.069	0.038	0.003	0.057	0.06	0.057	0.053
vegetables	0.01	0.015	0.010	0.010	0.016	0.014	0.011	0.015
potatoes	0.10	0.097	0.082	0.058	0.116	0.102	0.169	0.084
cakes and sweet foods	0.04	0.045	0.05	0.022	0.039	0.035	0.033	0.041
alcoholic drinks	7.53	6.012	10.98	4.464	8.964	4.716	22.2	5.544
nonalcoholic drinks	3.54	4.811	2.739	0.660	3.636	2.337	2.374	0.238
water	14.6	14.6	14.6	14.6	14.6	14.6	14.6	14.6
total	72.8	74.25	75.857	68.057	75.12	66.63	79.796	71.40

 
 Table 6. Main Food Groups Contributing to the Pb Intake in the Canary Islands

Table 7. Comparison with the Results in Other Spanish Communities

community	ref	Pb intake (µg/day)		
Canary Islands	this study	72.8		
Madrid	44	574		
Madrid	53	574		
Galicia	44	106		
Galicia	53	106		
Valencia	44	40.3		
Valencia	53	40		
Valencia	56	120		
Andalucía	44	56.6		
Andalucía	53	57		
Tarragona (Cataluña)	44	114.77 <sup>a</sup>		
Tarragona (Cataluña)	13	49		
Cataluña	49	28.4 (male adults)		
País Vasco	54	43 `		

<sup>a</sup> Only fish.

The recovery study using six standard reference materials is presented in **Table 2**. Food consumption data from the last Nutritional Survey in the Canary Islands are present in **Table 3**.

#### **RESULTS AND DISCUSSION**

**Table 4** presents the Pb concentrations for each of the analyzed food groups. Alongside each item are given the number of samples analyzed and the mean Pb content  $\pm$  standard

deviation. Fish, cold meats and sausages, and alcoholic drinks are the food groups with higher Pb levels. As many studies before found, the fish group presents the highest Pb levels  $(367.00 \pm 241.64 \,\mu\text{g/kg} \text{ of fresh weight})$ , with tuna fish and sardines having the highest levels. The high variability in the fish Pb concentrations is due to the wide range of fishes analyzed and the differences of Pb levels in the seawaters and in the marine fishes' diets. Among the cold meat and sausage group, sausages followed by chorizo were the samples with higher Pb levels. Alcoholic drinks present a mean Pb concentration of 120.0  $\mu$ g/kg, a higher value than that obtained by Jorhem et al. (41) when studying the mean levels of Pb (73  $\mu$ g/L) in 67 different table wines on the Swedish market. The mean lead content obtained for the 20 water samples was 7.3  $\mu$ g/L. Therefore, we can affirm that the waters consumed by the Canarian population fulfill the maximum value of 10  $\mu$ g/L fixed by the WHO guidelines for drinking-water quality (30). The mineral content in legumes and nuts widely consumed in Spain was analyzed in 2003 by Cabrera et al. (42). These authors found that Pb levels ranged from 320 to 700  $\mu$ g/kg and from 140 to 390  $\mu$ g/kg in legumes and nuts, respectively. These results are surprisingly high and quite different from our results, 0.13 and 31.58  $\mu$ g/kg, for legumes and nuts and dried fruits, respectively. Nevertheless, the intake of nuts and dried fruits is so poor in the Canary Islands that the contribution of this food group to the total Pb intake is irrelevant. Certain differences were also observed when results were compared with levels in other countries. In Finland, mean lead content was 1.7  $\mu$ g/kg in milk, 17  $\mu$ g/kg in Finnish cheese, 17–60  $\mu$ g/kg in imported cheese, and 1  $\mu$ g/kg in eggs (43); in the Canary Islands the Pb concentrations were 12.0  $\mu$ g/kg in milk, 2.3  $\mu$ g/kg in cheese, and 10  $\mu$ g/kg in eggs. The highest Pb levels for dairy products were observed in the "other dairy products" food group (66.0  $\mu g/kg$ ).

In **Table 5** the total Pb intakes in the Canary Islands are shown. Even if fish consumption in the Canary Islands is

Table 8. Comparison of the Pb Dietary Intake in the Canary Islands and Other Countries

	date of nutritional		Pb intake	
country	survey	ref	(µg/day)	% of the PTWI
Canary Islands	1997–1998	this study	72.8	29.12 for a person of 70 kg
France		57	<150	
		58	34	
Croatia	1988–1993	59	100.14	19.9
Croatia	1991-2001	60		6–49
Poland		61	72–136	
Slovenia		62	60.32	24.2
Finland	1993–1994	55	12.2	
Sweden (diet excluding any contribution from wines and spirits)	1982	41	30	
Sweden (for an average wine consumption of 45 mL/day)	1982		3 (from wine)	
Sweden	1987	63	17	
Denmark (men)		64	7	
Denmark	1993–1997	25		11
The Netherlands		65	32	
		26	34	8
former German Democratic Republic (GDR)	1988	66	34 (men) 25 (women)	11–13
Germany	1994—1995	3	, ,	16—36
Greece	1993	67		23.6
United States		57	80-95	
Canada		68	53.8	
Greenland		69	2.42	
China		70	103.77	
Tainan, Taiwan	1994	52	22	
Bombay, India	1991–1994	71	27	

moderate, it is the food group that contributes most to the Pb intake because of its high lead concentration (367  $\mu$ g/kg). Fruits, water, and sausages are, behind the fish group, the main food groups contributing to the Pb intake.

These results coincide with those obtained by other authors in other Spanish communities (13, 44-46). In Valencia and Andalucía (Mediterranean areas) the main contributing food group is seafood, including crustaceans and molluscs (44), as in our community. The fish group it is also considered to be the food group that most contributes to the Hg and Cd dietary intakes in the Canarian population (47, 48). A recent study carried out in Cataluña also pointed out that fish and shellfish was the food group showing the highest contribution to the As, Cd, Hg, and Pb intakes (49).

Although Llobet et al. (13) found that in Tarragona (Cataluña, Spain) vegetables followed by tubers and fruits were the most important food sources contributing to the total daily intake of lead, vegetables are not an important source of dietary Pb in our community. Due to the low ingestion of cereal products by the Canarian population, cereal foodstuffs contribute very little to the Pb dietary intake in the Canary Islands. In Finland, cereals contributed ~15% of dietary lead (50). Nevertheless, cereals, especially rice, are the main source of dietary Pb for other populations. In adult women from Manila, rice accounted for 18% of dietary Pb (51), and, in Taiwan, rice accounted for 1.4% of daily Pb (52).

The island that presents the highest Pb intake in our study is La Gomera. The high consumption of alcoholic drinks by the La Gomera population (185 g/day) results in a great Pb contribution of these drinks to the total intake (22.2  $\mu$ g of Pb/day). La Gomera is followed by Lanzarote, Tenerife, and Gran Canaria islands. These four islands present a Pb dietary intake over the mean Pb intake (72.8  $\mu$ g of Pb/day) of the whole archipelago. The islands with lower Pb intakes are La Palma and Fuerteventura, with intakes of <70  $\mu$ g/day (**Table 5**).

On La Gomera and La Palma islands dietary lead from the fish food group has been overcome by lead coming from the water and alcoholic drinks food groups. It is surprising how lead from fruits is in fifth place of the lead-contributing food groups, whereas for the rest of the islands it is in second or third place (**Table 6**).

In **Table 7** we compare the Pb intake in the Canary Islands with the lead intake in other Spanish communities. The Pb intake in the Canary Islands is lower than that in Madrid and Galicia (44, 53) but it is higher than that in País Vasco, Andalucía, Tarragona, and Valencia (13, 44, 53, 54).

In **Table 8** we are compare the Canarian population's Pb intake with the dietary intakes of Pb in other countries. The date of the food consumption intake data is also included. Pb intake in the Canary Islands is lower than the Pb intake in France, Croatia, and China and quite close to the Pb intakes in Poland, Slovenia, and the United States. Sweden, Denmark, The Netherlands, and Finland present Pb dietary intakes of less than half the value obtained in the Canary Islands. The low Pb intakes for women, 24.6, 14.6, and  $11 \mu g/day$  in China, Japan, and The Philippines, respectively, are surprising (*51*). In Finland fish contributes 4% of the mean daily Pb intake (*55*).

In conclusion, Pb levels in foodstuffs consumed in the Canary Islands are low and within the range observed in other national and international communities. Fish, water, and fruits followed by sausages and cold meats are the main food groups contributing to the total Pb intake. The total intake of lead in the Autonomous Canarian Community in 1998 was 72.8  $\mu$ g/day less than the recommendation of the PTWI. Therefore, the risks

for the Canarian population's health are low, but there is a need to fix new limits of tolerance for lead in many food products.

### LITERATURE CITED

- Weyermann, M.; Brenner, H. Alcohol consumption and smoking habits as determinants of lead levels in a national population sample from Germany. *Arch. Environ. Health.* **1997**, *52* (3), 233–239.
- (2) Krejpcio, Z.; Olejnik, D.; Wojciak, R. W.; Gawecki, J. Lead and cadmium content in daily food rations of children and adolescents from Koper basin of Legnica region. *Rocz. Panstw. Zakl. Hig.* **1999**, *50* (4), 353–360.
- (3) Wilhelm, M.; Wittsiepe, J.; Schrey, P.; Feldmann, C.; Idel, H. Dietary intake of lead by children and adults from Germany measured by duplicate method. *Int. J. Hyg. Environ. Health.* 2003, 206 (6), 493–503.
- (4) Favaro, D. I.; Hui, M. L.; Cozzolino, S. M.; Maihara, V. A.; Armelin, M. J.; Vasconcellos, M. B.; Yuyama, L. K.; Boaventura, G. T.; Tramonte, V. L. Determination of various nutrients and toxic elements in different Brazilian regional diets neutron activation analysis. *J. Trace Elem. Med. Biol.* **1997**, *11* (3), 129– 136.
- (5) Halliwell, D.; Turoczy, N.; Stagnitti, F. Lead concentrations in *Eucalyptus* sp. in a small coastal town. *Bull. Environ. Contam. Toxicol.* 2000, 65, 583–590.
- (6) Kurasaki, M.; Hartoto, D. I.; Saito, T.; Suzuki-Kurasaki, M.; Iwakuma, T. Metals in water in the Central Kalimantan, Indonesia. *Bull. Environ. Contam. Toxicol.* 2000, 65, 591–597.
- (7) Rodríguez, M. A.; Navarro, M.; Cabrera, C.; López, M. C. Elementos tóxicos en alimentos, bebidas y envases. *Alimentaria* 2001, May 1, 23–31.
- (8) Sierra, A.; Hardisson, A. La contaminación química de los alimentos. Aditivos alimentarios. In *Medicina Preventiva y Salud Pública*, 9th ed.; Piédrola, G., Domínguez, M., Cortina, P., et al., Eds.; Salvat: Barcelona, Spain, 1991; pp 293–303.
- (9) Houston, D. K.; Johnson, M. A. Lead as a risk factor for hypertension in women. *Nutr. Rev.* **1999**, 57 (9 Part 1), 277– 279.
- (10) Kwong, W. T.; Friello, P.; Semba, R. D. Interactions between iron deficiency and lead poisoning: epidemiology and pathogenesis. *Sci. Total. Environ.* **2004**, *330* (1–3), 21–37.
- (11) Kim, H. S.; Lee, S. S.; Hwangbo, Y.; Lee, B. K. Cross-sectional study of blood lead effects on iron status in Korean lead workers. *Nutrition* **2003**, *19* (7–8), 571–576.
- (12) Schell, L. M.; Denma, M.; Stark, A. D.; Ravenscroft, J.; Parsons, P.; Schulte, J. Relationship between blood lead concentration and dietary intake in infants from 3 to 12 months of age. *Environ. Res.* **2004**, *96* (3), 264–273.
- (13) Llobet, J. M.; Granero, S.; Schuhmacher, M.; Corbella, J.; Domingo, J. L. Biological monitoring of environmental pollution and human exposure to metals in Tarragona, Spain IV. Estimation of the dietary intake. *Trace Elem. Elec.* **1998**, *15*, 136– 141.
- (14) Lanphear, B. P.; Hornung, R.; Ho, M.; Howard, C. R.; Eberly, S.; Knauf, K. Environmental lead exposure during early childhood. J. Pediatr. 2002, 140 (1), 40–47.
- (15) Rahlenbeck, S. I.; Burberg, A.; Zimmermann, R. D. Lead and cadmium in Ethiopian vegetables. *Bull. Environ. Contam. Toxicol.* **1999**, 62, 30–33.
- (16) Dudka, S.; Miller, W. P. Accumulation of potentially toxic elements in plants and their transfer to human food chain. J. Environ. Sci. Health B 1999, 34 (4), 681–708.
- (17) Dabeka, R. W.; McKenzie, A. D.; Pepper, K. Lead contamination of raisins sold in Canada. *Food Addit. Contam.* 2002, 19, 47– 54.
- (18) Baran, E. J. *Química Bioinorgánica*; McGraw-Hill: Madrid, Spain, 1994.
- (19) Gual García, J. Intoxicación por plomo. *LAB2000* **1994**, *53*, 5–17.

- (20) Gallicchio, L.; Scherer, R. W.; Sexton, M. Influence of nutrient intake on blood lead levels of young children risk for lead poisoning. *Environ. Health Perspect.* **2002**, *110* (12), A767– 772.
- (21) Schuhmacher, M.; Belles, M.; Rico, A.; Domingo, J. L.; Corbella, J. Impact of reduction of lead in gasoline on the blood and hair levels in the population of Tarragona Province, Spain, 1990– 1995. *Sci. Total Environ.* **1996**, *184* (3), 203–209.
- (22) González-Soto, E.; González-Rodríguez, V.; López-Suárez, C.; Castro-Romero, J. M.; Pérez-Iglesias, J.; Fernández-Solis, J. M. Migration of lead and cadmium from ceramic materials used in food preparation. *Bull. Environ. Contam. Toxicol.* **2000**, *65*, 598–603.
- (23) Alberti-Fidanza, A.; Burini, G.; Perriello, G.; Fidanza, F. Trace element intake and status of Italian subjects living in the Gubbio area. *Environ. Res.* 2003, *91* (2), 71–77.
- (24) Galal-Gorchev, H. Dietary intake, levels in food and estimated intake of lead, cadmium and mercury. *Food Addit. Contam.* 1993, 10, 115–128.
- (25) Larsen, E. H.; Andersen, N. L.; Moller, A.; Petersen, A.; Mortensen, G. K.; Petersen, J. Monitoring the content and intake of trace elements from food in Denmark. *Food Addit. Contam.* 2002, 19, 33–46.
- (26) Ellen, G.; Egmond, E.; Van Loon, J. W.; Sahertian, E. T.; Tolsma, K. Dietary intakes of some essential and nonessential trace elements, nitrate, nitrite and N-nitrosamines, by Dutch adults: estimated via 24-hour duplicate portion study. *Food Addit. Contam.* **1990**, 7 (2), 207–221.
- (27) Brussaard, J. H.; Van Dokkum, W.; Van der Paauw, C. G.; De Vos, R. H.; De, W. L.; Lowik, M. R. Dietary intake of food contaminants in The Netherlands (Dutch Nutrition Surveillance System). *Food Addit. Contam.* **1996**, *13* (5), 561–573.
- (28) Ikeda, M.; Watanabe, T.; Koizumi, A.; Fujita, H.; Nakatsuka, H.; Kasahara. Dietary intake of lead among Japanese farmers. *Arch. Environ. Health* **1989**, *44*, 23–29.
- (29) Reglamento (CE) 221/2002 de la Comisión de 6 de febrero de 2002 por el que se modifica el Reglamento (CE) 466/2001 por el que se fija el contenido máximo de determinados contaminantes en productos alimenticios.
- (30) World Health Organization. Guidelines for Drinking-Water Quality. Recommendations, 3rd ed.; Geneva, Switzerland, 2004; Vol. 1.
- (31) International Office of Vine and Wine (OIV). Recueil des Methodes Internationales d'Analyse des Vines et des Mouts; Résolution oenol/96; 2005.
- (32) Dragun, Z.; Puntaric, D.; Prpic-Majic, D.; Bosnir, J.; Gmajnic, R.; Klaric, M. Toxic metals and metalloids in dietetic products. *Croat. Med. J.* 2003, 44 (2), 214–218.
- (33) Storelli, M. M.; Ceci, E.; Marcotrigiano, G. O. Distribution of heavy metal residues in some tissues of Caretta caretta (Linnaeus) specimen beached along the Adriatic Sea (Italy). *Bull. Environ. Contam. Toxicol.* **1998**, *60*, 546–552.
- (34) Robberecht, H.; Van Cauwenbergh, R.; Bosscher, D.; Cornelis, R.; Deelstra, H. Daily dietary total arsenic intake in Belgium using duplicate portion sampling and elemental content of various foodstuffs. *Eur. Food Res. Technol.* **2002**, *214*, 27–32.
- (35) ENCA Encuesta Nutricional de Canarias 1997–1998. Servicio Canario de Salud; Consejería de Sanidad y Consumo: Gobierno de Canarias, 2000.
- (36) AOAC. Official Methods of Analysis of the AOAC, 15th ed.; Association of Official Analytical Chemists: Washington, DC, 1990; pp 237–273.
- (37) Jorhem, L. Determination of metals in foods by atomic absorption spectrometry after dry ashing: NMKL collaborative study. J. AOAC Int. 2000, 83 (5), 1204–1211.
- (38) Subramanian, K. S. Determination of metals in biofluids and tissues: sample preparation methods for atomic spectroscopic techniques. *Spectrochim. Acta B* **1996**, *51*, 291–319.
- (39) Horwitz, W. Official Methods of Analysis of the Association of Official Analytical Chemists, 13th ed.; AOAC: Washington, DC, 1980.

- (40) Hardisson, A.; Galindo, L.; García Montelongo, F. Niveles de concentración de Pb, Cd, Cu, Zn y Fe en túnidos en conserva. *Anal. Bromatol.* **1985**, *37* (2), 327–332.
- (41) Jorhem, L.; Mattsson, P.; Slorach, S. Lead in table wines on the Swedish market. *Food Addit. Contam.* **1988**, 5 (4), 645–649.
- (42) Cabrera, C.; Lloris, F.; Jiménez, R.; Olalla, M.; López, M. C. Mineral content in legumes and nuts: contribution to the Spanish dietary intake. *Sci. Total Environ.* **2003**, *308* (1–3), 1–14.
- (43) Tahvonen, R.; Kumpulainen, J. Lead and cadmium contents in milk, cheese and eggs on the Finnish market. *Food Addit. Contam.* **1995**, *12* (6), 789–798.
- (44) Cuadrado, C.; Kumpulainen, J.; Moreiras, O. Lead, cadmium and mercury contents in average Spanish market basket diets from Galicia, Valencia, Andalucía and Madrid. *Food Addit. Contam.* **1995**, *12*, 107–118.
- (45) Schuhmacher, M.; Bosque, M. A.; Domingo, J. L.; Corbella, J. Dietary intake of lead and cadmium from foods in Tarragona Province, Spain. *Bull. Environ. Contam. Toxicol.* **1991**, *56*, 106– 113.
- (46) López Artíguez, M.; Soria, M. L.; Camean, A.; Repetto, M. Cadmium in the diet of the total population of Seville (Spain). *Bull. Environ. Contam. Toxicol.* **1993**, *50*, 417–424.
- (47) Rubio, C.; Frías, I.; Hardisson, A. Toxicología del plomo y su presencia en los alimentos. *Alimentaria* 1999, Sept, 77–85.
- (48) Hardisson, A.; Lafuente, M. A.; Rubio, C.; Frías, I. Mercury concentration in foods. Contribution to the dietary intake of the Canarian Autonomous Community. Presented at the 8th International Congress of the European Association for Veterinary Pharmacology and Toxicology (EAVPT), Jerusalem, Israel, July 30–Aug 3, 2000.
- (49) Llobet, J. M.; Falcó, G.; Casas, C.; Teixido, A.; Domingo, J. L. Concentrations of arsenic, cadmium, mercury, and lead in common foods and estimated daily intake by children, adolescents, adults and seniors of Catalonia, Spain. J. Agric. Food Chem. 2003, 51, 838–842.
- (50) Tahvonen, R.; Kumpulainen, J. Lead and cadmium in some cereal products on the Finnish market 1990–91. *Food Addit. Contam.* **1993**, *10* (2), 245–255.
- (51) Zhang, Z. W.; Qu, J. B.; Watanabe, T.; Shimbo, S.; Moon, C. S.; Ikeda, M. Exposure of citizens in China and in Japan to lead and cadmium: a comparative study. *Toxicol. Lett.* **1999**, *108* (2–3), 167–172.
- (52) Ikeda, M.; Zhang, Z. W.; Moon, C. S.; Imai, Y.; Watanabe, T.; Shimbo, S.; Guo, Y. L. Background exposure of general population to cadmium and lead in Tainan city, Taiwan. *Arch. Environ. Contam. Toxicol.* **1996**, *30*, 121–126.
- (53) Moreiras, O.; Cuadrado, C.; Lamand, M.; Tressol, J. C. The adequacy of essential minerals intake in four areas of Spain as assessed by direct analysis and a database. *Nutr. Res.* 1995, 13, 851–861.
- (54) Urieta, I.; Jalón, M.; Eguileor, I. Food surveillance in the Basque Country (Spain). II Estimation of the dietary intake of organochlorine pesticides, heavy metals, arsenic, aflatoxin M1, iron, and zinc through the Total Diet Study, 1990/91. *Food Addit. Contam.* **1996**, *13*, 29–52.
- (55) Tahvonen, T.; Kumpulainen, J. Contents of lead and cadmium in selected fish species consumed in Finland in 1993–1994. *Food Addit. Contam.* **1996**, *13* (6), 647–654.
- (56) Barberá, R.; Farré, R.; Mesado, D. Oral intake of cadmium, cobalt, copper, iron, lead, nickel, manganese and zinc in the University student's diet. *Nahrung* **1993**, *3*, 241–245.
- (57) Boudene, C. Toxicidad de los metales. In *Toxicología y Seguridad de los Alimentos*; Derache, R., Ed.; Omega: Barcelona, Spain, 1990; pp 133–163.
- (58) Noel, L.; Leblanc, J. C.; Guerin, T. Determination of several elements in duplicate meals from catering establishments using closed vessel microwave digestion with inductively coupled plasma mass spectrometry detection: estimation of daily dietary intake. *Food Addit. Contam.* 2003, 20, 44–56.

- (59) Sapunar-Postrunik, J.; Bazulic, D.; Kubala, H. Estimation of dietary intake of arsenic in the general population of the republic of Croatia. *Sci. Total Environ.* **1996**, *191* (1–2), 119–123.
- (60) Blanusa, M.; Juresa, D. Lead, cadmium and mercury dietary intake in Croatia. Arh. Hig. Rada. Toksikol. 2001, 52 (2), 229– 237.
- (61) Marzec, Z.; Bulinski, R. Evaluation of cadmium, mercury and lead intake with daily food rations. *Rocz. Panstw. Zakl. Hig.* **1990**. *41*, 35–38.
- (62) Erzen, I.; Ursic, S.; Bosnjak, K. Assessment of dietary intake of cadmium, lead and mercury via foods of the plant and animal origin in Slovenia. *Med. Arh.* 2002, 56 (2), 105–109.
- (63) Becker, W.; Kumpulainen, J. Contents of essential and toxic mineral elements in Swedish market-basket diets in 1987. Br. J. Nutr. 1991, 66 (2), 151–160.
- (64) Bro, S.; Sandstrom, B.; Heydorn, K. Intake of essential and toxic trace elements in random sample of Danish men as determined by the duplicate portion sampling technique. *J. Trace Elem. Electrolytes Health Dis.* **1990**, *4* (3), 47–155.
- (65) Van Dokkum, W.; de Vos, R. H.; Muys, T.; Wesstra, J. A. Minerals and trace elements in total diets in The Netherlands. *Br. J. Nutr.* **1989**, *61*, 7–15.
- (66) Muller, M.; Anke, M. Investigations into the oral lead exposure of adults in the former German Democratic Republic. Z. Lebensm. Unters. Forsch. 1995, 200, 38–43.

- (67) Voutsa, D.; Samara, C. Dietary intake of trace elements and polycyclic aromatic hydrocarbons via vegetables grown in an industrial Greek area. *Sci. Total Environ.* **1998**, *218* (2–3), 203– 216.
- (68) Dabeka, R. W.; McKenzie, A. D.; Lacroix, G. M. Dietary intakes of lead, cadmium, arsenic and fluoride by Canadian adults: a 24-hour duplicate diet study. *Food Addit. Contam.* **1987**, *4* (1), 89–101.
- (69) Johansen, P.; Pars, T.; Bjerregaard, P. Lead, cadmium, mercury and selenium intake by Greenlanders from local marine food. *Sci. Total Environ.* 2000, 245 (1–3), 187–194.
- (70) Yang, H. F.; Luo, X. Y.; Shen, W.; Zhou, Z. F.; Jin, C. Y.; Yu Liang, C. S. National food contamination monitoring programmes-levels of mercury, lead and cadmium in Chinese foods. *Biomed. Environ. Sci.* **1994**, 7 (4), 362–368.
- (71) Tripathi, R. M.; Raghunath, R.; Krishnamoorthy, T. M. Dietary intake of heavy metals in Bombay city, India. *Sci. Total Environ.* **1997**, 208 (3), 149–159.

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