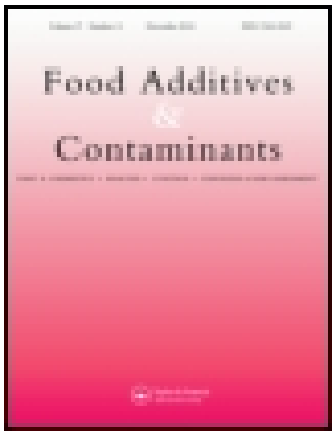

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Dietary intake of trace elements by the population of Catalonia (Spain): results from a total diet study

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Dietary intake of trace elements by the population of Catalonia (Spain): Results from a total diet study

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Abstract

This study aimed at analysing the concentrations of Al, Ba, Bi, Cu, Cr, Ge, Mn, Mo, Ni, Sb, Se, Sr and Zn in food samples collected in 2008 in Catalonia (Spain). The dietary intake of these 13 trace elements was subsequently estimated by different age–gender groups of the population: children, adolescents, adults, and seniors. In general terms, fish and shellfish, cereals, and pulses were the food groups showing the highest levels for most elements. Higher dietary intakes were associated to male groups (adolescents, adults and seniors). However, none of them exceeded the tolerable levels. When exposure was estimated based on body weight, children were the group with the highest dietary intake. Notwithstanding, only the weekly intake of Al by children exceeded the recommendations of the European Food Safety Authority. It is a consequence of the higher intake of cereals in relation to their respective body weights. In addition to the

periodical food surveillance of toxic metals (As, Cd, Hg and Pb), it is also important to determine the levels of other trace elements in order to ensure that the dietary exposure by the Catalan population is under control.

Keywords: trace elements; food; dietary intake; human exposure; Total Diet Study; Catalonia (Spain)

Introduction

Metals are natural components of the Earth's crust, being present in different environmental compartments such as air, soil, and water (Nadal et al. 2008). Some of these elements can be also important environmental contaminants, particularly in areas with high anthropogenic pressure (Mari et al. 2014; Rovira et al. 2004a,b,; Vilavert et al. 2015). A number of elements, such as arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) have no beneficial effects, being potentially toxic to humans at very low quantities (Martí-Cid et al. 2008; Martorell et al. 2011; Perelló et al. 2014a).

In recent years, exposure assessment studies have focused on toxic metals, whilst limited research on other elements has been published. It is well established that dietary intake is the major source of exposure to metals for the general population (Llobet et al. 2003; Arnich et al. 2012; Perelló et al. 2014a). Similarly, food may be the key entrance route of other trace elements into the organism (Gimou et al. 2013, 2014). Although some of these elements may be essential for humans, their dietary intake should be controlled in order to ensure that threshold values are not exceeded.

In 2000, our laboratory initiated a wide surveillance programme aimed at determining the content of toxic metals (As, Cd, Hg and Pb) in a number of foodstuffs purchased in Catalonia, Spain, by using a Total Dietary Study (TDS) approach (Llobet et al. 2003). Since then, metal levels have been periodically assessed (Martí-Cid et al. 2008; Martorell et al. 2011; Perelló et al. 2014a), and the dietary intakes of toxic metals, according to age/gender subpopulation groups, have been also estimated. These surveys have confirmed a decreasing tendency. The only exception was Pb, whose levels in the 2008 survey significantly increased with respect to previous sampling campaigns (Martorell et al. 2011). Based on this result, we hypothesised that other trace elements, not previously analysed, could have followed a similar trend. Therefore, we increased the list of target elements in subsequent studies and focused on determining the total dietary intake of a number of elements, in addition to As, Cd, Hg and Pb.

In Catalonia, other investigations have also allowed the acquisition of data on the concentrations of trace elements in foodstuffs, as well as their dietary intake by non-occupationally exposed populations (Martí-Cid et al. 2009; Perelló et al. 2015). Chromium and Mn were the elements showing the highest concentrations in foodstuffs acquired in an area surrounding a hazardous waste incinerator (HWI), located in Tarragona County (Perelló et al. 2015). Compared with data from a pre-operational survey in the vicinity of the HWI, which was conducted in 1996 (Llobet et al. 1998), a decreasing trend in the dietary exposure of most elements has been noted in the last 15 years, with the only exceptions of Cr and Hg. The intake of these two elements by the adult population living near the plant significantly increased with respect to the baseline study, being up to 5-fold higher for Cr. Furthermore, the dietary exposure to Cd exceeded the provisional tolerable weekly intake (PTWI) established by the European Food Safety Authority (EFSA) (3.36 vs. 2.5 µg/kg b.w../week).

The aim of the present study was to analyse the concentrations of 13 trace elements in foods of high consumption in Catalonia, Spain, and to estimate their dietary intake for different population groups. The temporal trends were also assessed. In addition, data were compared with guidelines or threshold values set by international organisations.

Materials and methods

Sampling

In November and December of 2008, food samples were purchased in local markets, small stores, supermarkets, and big grocery stores of 12 cities of Catalonia (Barcelona, Hospitalet de Llobregat, Vilanova i la Geltrú, Mataró, Sabadell, Terrassa, Girona, Tarragona, Reus, Tortosa, Lleida, and Manresa). The foodstuffs selected for the study were among those most consumed in Catalonia according to Serra-Majem et al. (2003). Samples included meat, meat products, fish and seafood, vegetables, tubers, fruits, eggs of hen, milk, dairy products, cereals, pulses, oils and fats, and industrial bakery. A detailed list of food items included in each category has been published elsewhere (Martí-Cid et al. 2009). For elemental analyses, two composite samples were prepared for each food item. Each composite sample consisted of 24 individual units. Only edible parts of each food were included in the composites.

Analytical determination

Before metal analyses, samples were homogenised with a domestic mixer. Subsequently, 0.5 g of each food sample were treated with 5 mL of 65% HNO_3 (Suprapur, E. Merck, Darmstadt, Germany) in hermetic Teflon vessels for 8 h at room temperature. Solutions were heated at 80°C for 8 h. On completion of the digestion and after adequate cooling, solutions were filtered and made up to 25 mL with ultrapure water. The concentrations of aluminium (Al), barium (Ba), bismuth (Bi), copper (Cu), chromium total (Cr), germanium (Ge), manganese (Mn), molybdenum (Mo), nickel (Ni), antimony (Sb), selenium (Se), strontium (Sr) and zinc (Zn) were determined by inductively coupled plasma-mass spectrometry (Perkin-Elmer Elan 6000, Woodbridge, ON, Canada) (Martí-Cid et al., 2009). The accuracy of the instrumental methods and analytical procedures was checked by duplication of the samples, as well as by using 2 reference materials for trace elements: TORT-2 Lobster hepatopancreas (National Research Council, Canada) and NIST-1570a Spinach leaves (National Institute of Standards and Technology Trace elements, US). Rhodium was used as internal standard. Replicate measurements were performed. The ISO 17025:2005 standard was followed to identify and quantify uncertainty sources. Mean recovery rates of the analysed elements were 101% and 112% for TORT-2 and NIST-1570a, respectively. Selenium showed the lowest percentage (83% for TORT-2), while Zn had the highest recovery values (129 and 135% for TORT-2 and NIST-1570A, respectively).

Dietary exposure estimates

Consumption data of the analysed foodstuffs by the general population of Catalonia were obtained from Serra-Majem et al. (2003). The population was divided into four age groups: children (4–9 years), adolescents (10–19 years), adults (20–65 years), and seniors (>65 years). In turn, each group was sub-divided according to gender. Total dietary intake of trace elements for each food group was calculated by summing the results of multiplying the element concentration in each specific food item by the amount (proportionally estimated) consumed of that item. Total dietary intake of each element was finally obtained by summing the respective intakes from each food group. For calculations, when the concentration of an element was under the respective LOD, that value was assumed to be equal to one half of the LOD (ND=1/2 LOD), according to WHO guidelines and previous studies (WHO 1985; Martí-Cid et al. 2009; Perelló et al. 2014a).

Results and discussion

Table 1 shows the concentrations of all analysed elements in foods, classified according to the following groups: meat and meat products, fish and shellfish, vegetables, tubers, fruits, eggs, milk, dairy products, cereals, pulses, oils and fats, and industrial bakery. The highest concentrations differed for the respective elements. For Al, fish and shellfish, as well as industrial bakery, showed the highest concentrations, while they were under the LOD in all samples of eggs and milk. Barium and Zn showed similar patterns, with eggs showing the highest levels (1.32 and 48.0 μg/g for Ba and Zn, respectively). In contrast, oils and fats was the food group with the minimum values. In turn, total Cr showed concentrations ranging from 0.23 to 1.50 μg/g, being sunflower oil the individual food item with the highest level (2.19 μg/g). Cereals and pulses showed the highest levels of Cu, Mn and Mo. More specifically, chickpeas (Cu: 3.17 μg/g), pasta (Mn: 8.58 μg/g) and haricot beans (Mo: 0.97 μg/g) were the individual food items with the highest concentrations. On the other hand, pulses and fruits were the food groups with the highest concentrations of Ni (0.53 and 0.46 μg/g, respectively), while this element was not detected in eggs and milk. With respect to Sr, fish and shellfish (especially, clam) and dairy products (especially, cheese-II) were the foodstuffs with the maximum concentrations of this element. Finally, Bi, Ge, Sb and Se were only detected in some food samples, being the results close to their respectively detection limits. It is interesting to remark the notable concentrations of Se found in the group of fish and shellfish. This group may also contain high concentrations of methylmercury and total Hg (Llobet et al. 1998, 2003; Martí-Cid et al. 2008, 2009; Martorell et al. 2011; Perelló et al. 2014a,b), constituting also an important source of Se (Olmedo et al. 2013). In order to identify the fish species whose Hg contents may be associated with increased health risks for humans and wildlife, Se:Hg molar ratios efforts should be assessed (Ralston 2010). In general, Se:Hg molar ratios in most saltwater fish species, excepting Mako shark, are above 1 (Kaneko and Ralston 2007; Burger and Gochfeld 2011). Considering the data on the Hg concentrations in fish and shellfish from Catalonia (Martí-Cid et al. 2008), a mean Se:Hg of 5.8 is estimated.

The daily dietary intakes of the 13 analysed elements by different population groups living in Catalonia are summarised in Figures 1a and 1b. Male adults were the group with the highest intake of Al, Ni and Se (5.5, 404 and 140 μg/day, respectively), while male seniors showed the highest intakes of Ge, Sb and Sr (9.5 μg/day, 10.7 μg/day and 2.15 mg/day, respectively). The highest intake of the remaining elements

was associated to male adolescents, with the only exception of Bi, for which children showed the highest intake (3.93 µg/day). For all populations, chicken breast, potatoes, whole milk and white bread were the major contributors to Bi intake. However, these foods have a lower consumption by females, so that their overall Bi intake was less.

Table 2 summarises the dietary intake of the elements here analysed by different population groups, according to the body weight (b.w.). In addition, data are compared with minimum requirements and maximum recommended values established by a number of international regulatory organizations. EFSA (2011) indicated a tolerable weekly intake (TWI) of 1 mg/kg b.w. for Al. In the present study, male adults showed a lower Al intake (0.55 mg/kg b.w./week). In contrast, the weekly intake of Al through food consumption by children (1.14 mg/kg b.w./week) slightly exceeded the TWI. This would be linked to the high consumption of bread and cereals, as well as to the low body weight of this population group in relation to their consumption rates.

For Ba, the US EPA (2005) established an oral reference dose (RfD_o) of 0.20 mg/kg b.w./week, which is higher than the intakes associated to all population groups in Catalonia (Table 2). With respect to total Cr, WHO (1996) established a dietary requirement of 50 µg/day, while the Expert Group on Vitamins and Minerals stated in its guidelines a maximum consumption of 160 µg/kg b.w./day (FSA 2003). It must be highlighted that Cr may be present in food in different chemical forms. Most notably, Cr(III) is considered to be a trace element essential for the proper functioning of living organisms, whereas Cr(VI) may exert toxic effects on biological systems (Uluozlu et al. 2009; Rakhunde et al. 2012). Unfortunately, there is a lack of data on the presence of Cr(VI) in food, with only a few studies. For instance, Soares et al. (2012) analysed the contents of Cr(VI) and total Cr in samples of white and whole bread samples, estimating that hexavalent Cr was about 12% of the total. In its recent Scientific Opinion, the EFSA CONTAM Panel decided to consider all the reported analytical results in food as Cr(III), when assessing the risks to public health related to the presence of chromium in food and drinking water (EFSA 2014b). This assumption was based on the outcome of recent speciation work, the fact that food is by-and-large a reducing medium, and that oxidation of Cr(III) to Cr(VI) would not be favoured in such medium. In all cases, the current dietary intake of Cr was found to be above daily requirements and below maximum recommended values (Table 2), even in a conservative scenario of 12% of Cr(V) vs. total Cr. The same occurs for Mn. WHO (1996) proposed a daily requirement of 2-5 mg of Mn. Although all population groups showed a higher intake, the levels of

children and female seniors were in the lowest part of the range (2.0 mg/day). Consequently, the current exposure to Mn was also found to be lower than the maximum recommended value established at 200 µg/kg b.w./day by FSA (2003). In agreement to the conclusions of the Committee on Toxicity of Chemicals in Foods (COT 2008), the dietary exposure to total Cr and Mn is not likely to cause toxicological problems.

The dietary requirements for Cu were established by WHO (1996) in 0.33-1.15, 1.15 and 1.35 mg/day for children, female and male groups, respectively. Contrastingly, EFSA (2006) set the threshold of Cu at 1-4 mg/day for children, and 10 mg/day for adults. All population groups showed daily intakes within the range, with values of 1.26, 1.04-1.40 and 1.04-1.35 mg/day for children, female and male groups. Similarly, the current intake of Mo by the Catalan population is higher than the reference daily intake set by WHO (1996) (1.5-2.5 µg/kg b.w./day), but lower than the threshold established by EFSA (2006) (0.1-0.5 mg/day for children, and 0.6 mg/day for adults).

The WHO (2005) established a TDI of 22 µg/kg b.w./day for Ni, which is near 4-fold higher than the intake estimated for the adult population of Catalonia (5.8 µg/kg b.w./day). The same organization had established a dietary requirement of 100 µg/day for the same element (WHO 1996). With respect to Se, the EFSA established a UL of 300 µg/day (EFSA 2006), while WHO recommended a minimum intake of 6-30, 30 or 40 µg/day by children, females and males, respectively. The dietary intake of both Ni and Se was well within the range of recommendations. Regarding Sb, FSA (2003) considers a maximum tolerable value of 450 µg/day (FSA 2003), being significantly higher than the present results (Table 2). With respect to Zn, EFSA (2006) indicated a NOAEL of 50 mg/day in adults, and 7-22 mg/day for children. Very recently, EFSA (2014) also suggested a minimum range of 9.4-16.3 mg/day for Zn in adults. Again, Zn intakes in Catalonia were well below the NOAEL and above the daily requirement, being estimated in 17.4 and 15.0-23.8 mg/day for children and adults, respectively. Finally, neither maximum values nor recommended levels have been established yet for Bi, Ge and Sr.

The results of the present survey are similar to those published elsewhere. For example, a number of studies have been published under the Food Standards Australia New Zealand (FSANZ) Acts of 2003, 2008 and 2011. Relatively lower intakes of Al, Ni and Sb were found in comparison to those currently detected (36 vs. 79 µg/kg b.w./day for Al, 1.8-2.7 vs. 5.8 µg/kg b.w./day for Ni, and <0.01-0.08 vs. 0.15 µg/kg b.w./day for

Sb). In the UK, Rose et al. (2010) evaluated the dietary intake of some elements also applying a TDS approach. They found similar values to those estimated for the Catalan population. The following dietary exposure levels by British adults were calculated: 4.97 mg/day for Al; 658 µg/day for Ba; 1.05-1.54 µg/day for Bi; 1.21 mg/day for Cu; 19.6-656 µg/day for Cr; 0.07-1.29 µg/day for Ge; 104-114 µg/day for Ni; 113-115 µg/day for Mo; 2.24-2.31 µg/day for Sb; 58-67 µg/day for Se; and 9.9 mg/day for Zn.

On the other hand, Zhang et al. (2011) estimated the dietary exposure of 14 trace elements in two areas of China with high lung cancer incidence. The reported intakes of Cu, Cr, Mn, Ni, Mo and Zn were 1210, 80, 2460, 100, 100 and 25.9 µg/day, respectively. In another recent study performed in Japan, Yamada et al. (2014) estimated the Cu, Zn and Mg intake in Japanese in male and female adult populations, finding ranges of 1.2-1.4, 8.2-10.1 and 4.9-5.1 mg/day, respectively.

In 2012, Guerin et al. (2011) investigated the concentrations of various elements in fish and shellfish from the French market, finding similar values to those observed in Catalonia. Arnich et al. (2012) also reported similar findings when investigating the intake of several elements in a TDS national survey. Aluminium, Ni and Sb intakes by the French population were estimated in 62.2, 3.83 and 0.037-0.055 µg/kg b.w./day, respectively. Similarly, Gimou et al. (2013, 2014) studied the dietary intake of some of these elements in Cameroon, evaluating also the health risks for the population. For Cr, Se and Zn, dietary intake values were up to 230, 114 and 6540 µg/day, respectively (Gimou et al. 2013), lower than those obtained in the current survey. In turn, the intake of Al, Ba, Mo, Ni and Sb (67, 20.8, 195, 4.37 and 0.014-0.034 µg/kg b.w./day, respectively) through food was similar than that found in Catalonia (Gimou et al. 2014).

In conclusion, the dietary intake of a number of trace elements (Al, Ba, Bi, Cu, Cr, Ge, Mn, Mo, Ni, Sb, Se, Sr, and Zn), other than the well known toxic elements As, Cd, Hg, and Pb, by the population of Catalonia is similar to that reported in other countries. The only exception would be Al, whose current intake by children slightly exceeded the dietary recommendations. The concentrations in food items acquired in the market are also in the same range, according to data from the scientific literature. We must highlight the high Se:Hg molar ratio in species of fish and shellfish, confirming the importance of this food group as an entrance pathway of Se.

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Table 1. Concentrations of 13 trace elements (µg/g of fresh weight) in samples of food acquired in Catalonia (Spain)

	Al	**Ba**	**Bi**	**Cr**	**Cu**	**Ge**	**Mn**	**Mo**	**Ni**	**Sb**	**Se**	**Sr**	**Zn**
Limit of detection	**2.76**	**0.073**	**0.005**	**0.054**	**0.134**	**0.007**	**0.037**	**0.008**	**0.096**	**0.012**	**0.16**	**0.107**	**2.54**
Meat and meat products	4.64	0.137	0.003	0.870	0.949	0.007	0.450	0.025	0.445	0.008	0.101	0.742	43.3
Fish and shellfish	9.85	0.179	0.004	0.784	1.28	0.010	0.915	0.035	0.367	<0.012	0.626	5.27	23.8
Vegetables	6.02	0.486	<0.005	0.162	0.943	0.008	2.24	0.158	0.358	<0.012	<0.16	3.13	7.51
Tubers	2.27	0.192	0.006	0.356	1.47	0.008	1.48	0.063	0.081	<0.012	<0.16	0.346	4.45
Fruits	1.54	0.200	<0.005	0.226	0.787	0.007	0.901	0.046	0.455	0.008	<0.16	1.16	6.11
Eggs	<2.76	1.32	<0.005	1.15	1.95	<0.007	1.093	0.165	<0.096	<0.012	0.258	1.17	48.0
Milk	<2.76	0.104	<0.005	0.272	<0.134	0.006	0.034	0.038	<0.096	<0.012	<0.16	0.884	9.45
Dairy products	2.89	0.510	0.007	0.748	0.489	0.007	0.230	0.074	0.113	0.008	<0.16	3.90	27.5
Cereals	4.96	0.760	<0.005	1.02	2.16	0.007	7.01	0.285	0.391	0.008	<0.16	1.71	19.2
Pulses	3.03	0.817	0.004	0.417	2.20	0.008	5.45	0.745	0.526	0.008	<0.16	2.96	12.9
Oils and fats	2.15	0.061	<0.005	1.50	0.123	0.006	0.069	0.012	0.175	<0.012	<0.16	0.162	4.17
Industrial bakery	7.89	0.544	<0.005	1.28	0.897	0.009	3.49	0.133	0.361	0.012	<0.16	1.50	7.00

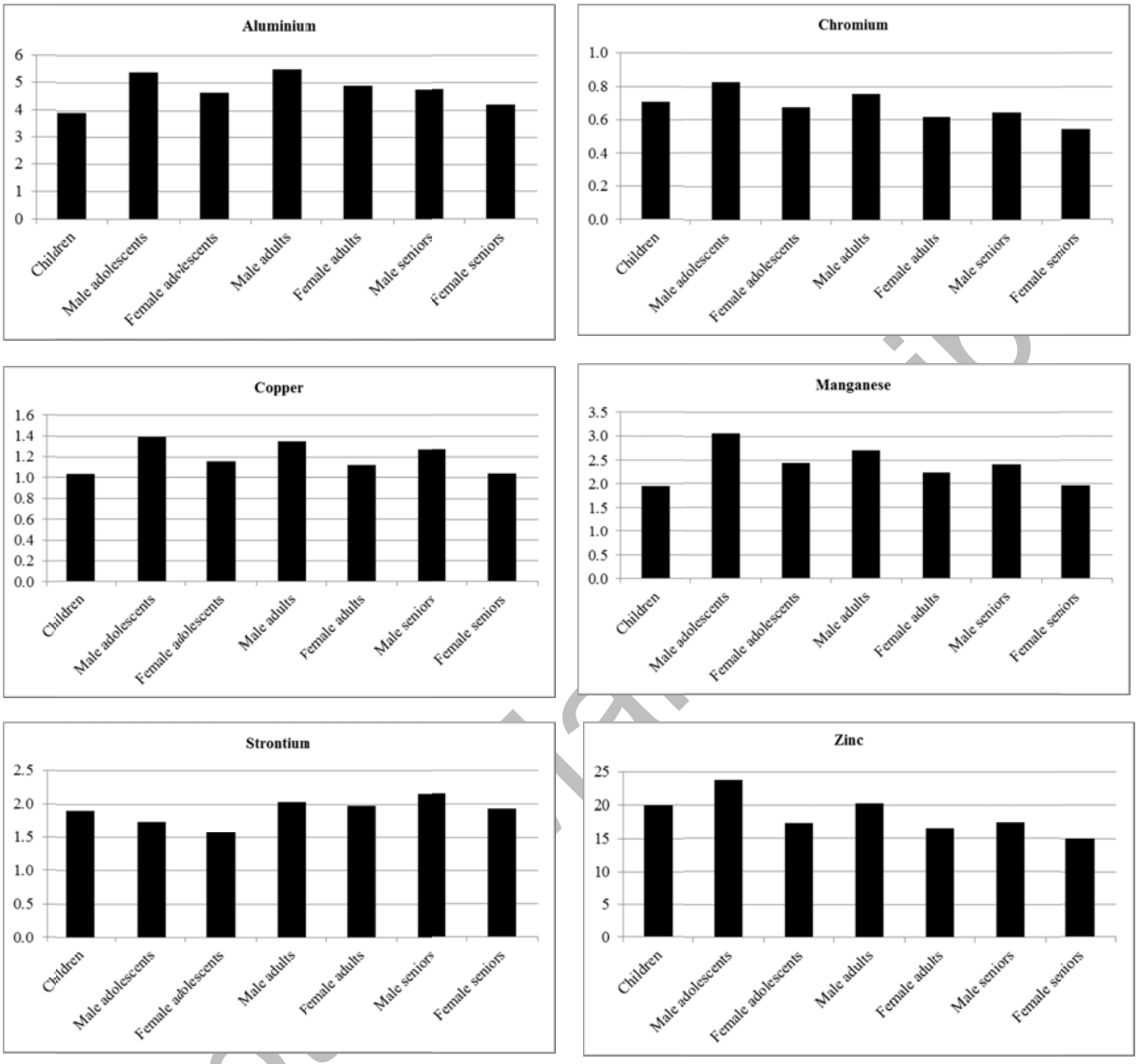

Figure 1a. Daily intake to elements studied according to different population groups in Catalonia (Spain) (mg/day).

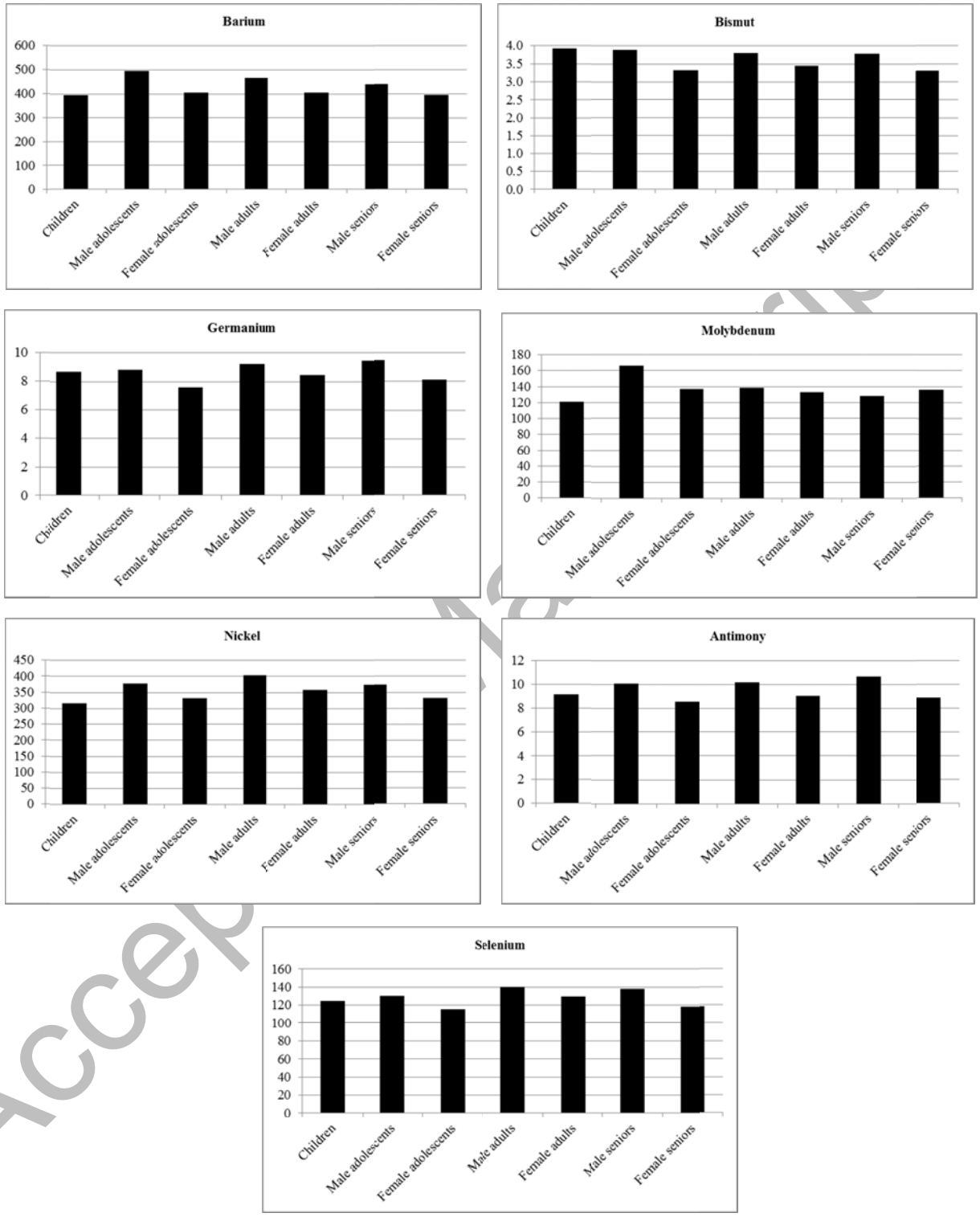

Figure 1b. Daily intake to elements studied according to different population groups in Catalonia (Spain) (µg/day).

Table 2. Estimated dietary intake of trace elements by different population groups in Catalonia (Spain). Comparison with guidance values

Element	Reference daily intake		Recommended maximum		Children	Male adolescents	Female adolescents	Male adults	Female adults	Male seniors	Female seniors
	Value	Reference	Value	Reference							
Al			1 mg/kg b.w./week	EFSA (2011)	1.14	0.67	0.62	0.55	0.62	0.51	0.49
Ba			0.20 mg/kg b.w./week	US EPA (2005)	0.12	0.06	0.05	0.05	0.05	0.05	0.05
Cr	50 µg/day	WHO (1996)			706	828	675	755	616	646	545
			150 µg/kg b.w./day*	FSA (2003)	29.4	14.8	12.7	10.8	11.2	9.9	9.1
Cu	0.33-1.15 (children) 1.15 (females), and 1.35 (males) mg/day	WHO (1996)	1-4 (children) and 10 (adults) mg/day	EFSA (2006)	1.26	1.35	1.13	1.04	1.40	1.27	1.04
Mn	2-5 mg/day	WHO (1996)			2.0	3.1	2.4	2.7	2.2	2.4	2.0
			200 µg/kg bw/day	FSA (2003)	81.5	54.7	46.1	38.7	40.8	37.2	32.8
Mo	1.5-2.5 µg/kg b.w../day	WHO (1996)			5.1	3.0	2.6	2.0	2.4	2.0	2.3
			0.1-0.5 (children) and 0.6 (adults) mg/day	EFSA (2006)	0.14	0.14	0.13	0.12	0.17	0.13	0.14
Ni	100 µg/day	WHO (1996)			316	379	333	404	359	373	332
			22 µg/kg b.w./day	WHO (2005)	13.2	6.8	6.3	5.8	6.5	5.7	5.5
Sb			450 µg/day	FSA (2003)	8.58	10.19	9.05	9.15	10.11	10.66	8.93
Se	6-30 (children), 30 (females) and 40 (males) µg/day	WHO (1996)	60-250 (children) and 300 (adults) µg/day	EFSA (2006)	115	140	129	125	130	138	118
Zn	9.4-16.3 mg/day	EFSA (2014a)	7-22 (children) and 50 (adults) mg/day	EFSA (2006)	17.4	20.3	16.6	19.9	23.8	17.4	15.0

*This value corresponded to Cr^{3+}. For Ba, Ge and Sr, neither maximum nor minimum recommended values have been established yet.