



## Dietary intake of arsenic, cadmium, mercury and lead by the population of Catalonia, Spain: Analysis of the temporal trend

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### ABSTRACT

In 2017, a monitoring study was conducted in Catalonia (Spain) to analyse, in widely consumed foodstuffs, the concentrations of arsenic (As), cadmium (Cd), mercury (Hg) and lead (Pb), as well as those of inorganic As (InAs) and methylmercury (MeHg). Health risks were estimated for various population groups, classified according to age. A continued reduction of dietary exposure to these elements was observed when comparing the results from the current and previous studies performed during the last 17 years. This reduction would be associated to a decrease of As, Cd, Hg and Pb concentrations in food, as well as by changes in dietary habits. None of the adult groups exceeded the safety threshold established by the EFSA, but toddlers, infants and children exceeded the PTWI for Cd and MeHg. The greatest intake of Cd and MeHg corresponded to infants, being due to the consumption of cuttlefish and hake. The consumption of these species should be reduced to two or three weekly portions, being combined with the intake of other non-predator species. Anyhow, the current results indicate that is necessary to conduct further periodical surveys, paying special attention to the trend in the intake of Cd and MeHg.

### 1. Introduction

Arsenic (As), cadmium (Cd), lead (Pb) and mercury (Hg) are naturally occurring elements present in the earth's crust. These elements are environmentally persistent and can bioaccumulate in living organisms, being also among the most toxic elements according to the US Agency for Toxic Substances and Disease Registry (ATSDR, 2017). Moreover, anthropogenic activities, such as traffic or industrial emissions, increase their presence in the nature (Nadal et al., 2004; De Lurdes Dinis and Fiúza, 2011; Beckers and Rinklebe, 2017). Exposure to these elements may have a wide range of adverse health effects, including Minamata (for Hg) or Itai-Itai (for Cd) diseases, as well as increased cancer risks (for As and Cd) (Zahir et al., 2005; Nordberg, 2009; Buha et al., 2017; Djordjevic et al., 2019).

It is well known that diet is the main exposure pathway to environmental contaminants (Bocio et al., 2005; Domingo, 2011; Domingo and Nadal, 2015; Linares et al., 2010; Perelló et al., 2015a). Actually, foodstuffs can be contaminated in any of the steps of the food chain (Domingo, 2011; Rather et al., 2017). Since children may be in contact with toxic elements during growth and in early stages of

development, they are one of the most susceptible groups of population to metal exposure. In fact, notable differences exist in the behaviour and development of children when compared to those of adults, thus requiring a special attention (Au, 2002).

Food safety agencies have the responsibility to assure that foods present in the market do not pose any risk for the health of the consumers. With this purpose, surveys such as total diet studies (TDSs) or duplicate diet studies (DDSs), are periodically conducted (Akhandaf et al., 2015; Martorell et al., 2012). In 2000, the Catalan Agency of Food Safety (ACSA) started a surveillance program consisting on a TDS aimed at estimating the exposure to a number of food contaminants and assessing the potential risk for the population of Catalonia (Spain). A wide range of chemical substances, including heavy metals (Llobet et al., 2003a), as well as several persistent organic pollutants (POPs) and emerging pollutants (Bocio et al., 2003; Falcó et al., 2003; Llobet et al., 2003b), were analysed, being the results considered as baseline for subsequent studies. In 2005, 2008 and 2012, follow-up surveys were conducted in order to update the information regarding not only the concentrations of heavy metals in food, but also their dietary intakes (Martí-Cid et al., 2008; Martorell et al., 2011; Perelló et al., 2014).

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In 2017, a new campaign was conducted to analyse again the levels of the same metals in foodstuffs of wide consumption by the population of Catalonia, as well as to assess the trends of the risks associated to their intakes, 17 years after the baseline study. The speciation of inorganic As (InAs) and methylmercury (MeHg) was also determined, in contrast with the information from previous surveys, in which the content of both species were merely estimated according to data from the scientific literature.

## 2. Materials and methods

### 2.1. Sampling

Sampling was similar to that designed and applied in previous TDSs (Martí-Cid et al., 2008; Martorell et al., 2011; Perelló et al., 2014). From January to July 2017, samples of different foodstuffs were acquired in different stores (local markets, small stores, supermarkets and big groceries stores) from 12 Catalan cities: Tarragona, Reus, Tortosa, Lleida, Manresa, Girona, Barcelona, Vilanova i la Geltrú, Hospitalet de Llobregat, Mataró, Sabadell, and Terrassa. Food items were selected among the most consumed foodstuffs by the general population of Catalonia. They were selected according to information from ENALIA (National Dietary Survey on the Child and Adolescent Population) and ENALIA-2 (National Food Survey on Adults, the Elderly and Pregnant Women), both developed by the Spanish Agency for Consumer Affairs, Food Safety and Nutrition (AECOSAN, 2016a,b), and co-financed by the European Food Safety Authority (EFSA).

Food samples included: meat and meat products (veal, pork, chicken, boiled ham, “frankfurt” sausages, cured ham and turkey ham), fish and seafood (cod, sea bream, tuna, canned tuna, European anchovy, swordfish, salmon, hake, monkfish, sole, cuttlefish, squid, sea bass, mussels and panga), vegetables (lettuce, tomato, zucchini, green bean, onion, leek and carrot), tubers (potato), fruit (pear, apple, orange, banana and tangerine), eggs (hen eggs), milk and dairy analogues (whole milk, semi-skimmed milk, soy milk and oat milk), dairy products (yogurt, flavoured yogurt, pudding and custard, mozzarella, cheese triangles (“quesitos”), and cheese II-fat cheese), bread and cereals (French bread, sliced bread, rice and pasta), pulses (lentils and chickpeas), oils (olive oil and sunflower oil), industrial bakery (milk bread, cookies, croissants, muffins, sponge cake and chocolate cookies), sauces (tomato sauce), chocolates (chocolate powder) and infant food (follow-on formulae, infant cereals and baby food: chicken, fruit, veal and fish). In order to get homogenized samples, three composite samples were prepared for each food item, being each composite a mixture of 20 individual units. Mixing and homogenization of food was conducted by means of a domestic shredder, and considering only the edible parts of food.

### 2.2. Analytical determination

The analysis of the concentrations of As, Cd and Pb was performed after a microwave digestion treatment with nitric acid (65% Suprapur, E. Merck, Darmstadt, Germany). One-half g of each food sample was digested with 9 mL of 22% nitric acid and 0.5 mL of 30% H<sub>2</sub>O<sub>2</sub> (30% Suprapur, E. Merck, Darmstadt, Germany). Samples were subjected for 30 min to microwave-assisted acid digestion at high pressure and increasing temperature, being kept at 200 °C for the last 15 min. Once it was performed, extracts were made up to 30 mL with purified water. Samples containing high levels of oils and fats were pre-digested with concentrated nitric acid (65% Suprapur, E. Merck, Darmstadt, Germany). On the other hand, the content of total Hg in food was performed by direct determination in an elemental mercury analyser (AMA), following a standard method of amalgamation with gold, using 0.1 g of sample for the direct analysis.

The chemical speciation of As and Hg was also conducted. MeHg was only analysed in samples with detectable levels of total Hg, using

0.8 g of sample. MeHg determination was done by extraction with toluene (E. Merck, Darmstadt, Germany) and retro-extraction with cysteine (E. Merck, Darmstadt, Germany), according to the standard operating procedure of the EC Joint Research Centre (JRC, 2013). On the other hand, the levels of InAs in all food items were determined by a microwave extraction method, using 0.25 g of sample and 10 mL of an acidic-oxidant solution (0,2% w/v nitric acid + 1% w/v hydrogen peroxide). The method of extraction last for 1 h up to a final temperature of 95 °C (Llorente-Mirandes et al., 2012).

The determination of the total content of each trace element, excepting Hg, was performed by using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7700x, with collision cell). For Hg and MeHg, an Automated Mercury Analyzer (AMA 254, Leco Instruments, Spain) was used. For InAs, analysis was done by liquid chromatography (HPLC, Agilent Series 1100) coupled to the ICP-MS above (Llorente-Mirandes et al., 2012). Quality control/quality assurance (QC/QA) of the analytical procedure was ensured by analysing the standards and blanks every batch of samples, plus spiked samples and reference materials (from several FAPAS interlaboratory proficiency tests). The limit of detection (LOD, fresh weight) was set at 0.002 µg/g for each of the analysed trace elements.

### 2.3. Calculations of the dietary exposure

Consumption data for the analysed foodstuffs were obtained from ENALIA and ENALIA-2 surveys (AECOSAN, 2016a,b). They are consumption data studies representative of different Spanish subpopulations, which are based on a random selection of 1862 children and adolescents, 623 adults, 310 seniors, and 157 pregnant women. The method included a 24-h recall in two non-consecutive days (at least 14 days in between) and a food frequency questionnaire. According to the available information, the population was divided into 8 groups: toddlers (6–11 months), infants (12–36 months), children (3–9 years), adolescents (10–17 years), young adults (18–39 years), adults (40–64 years), seniors (65–74 years), and a specific group of pregnant women. The dietary intake of each food item was calculated by multiplying the concentration of each chemical in food and the consumption of that food. The total dietary intake was obtained by summing the respective intakes of all food groups. Finally, the intake was also estimated according to the respective average body weight. Literature data corresponding to the mean body weight of each population group were obtained (Carrascosa et al., 2010; López-Sobaler et al., 2016; Martínez et al., 2017; WHO, 2003). These data were the following: 8.4 kg for toddlers, 12.3 kg for infants, 24 kg for children, 51 kg for adolescents, 72 kg for young adults, 77 kg for adults, 70.5 kg for seniors and 65 kg for pregnant women.

For calculations, when the concentration of a heavy metal was under the respective limit of detection (LOD), that value was assumed to be one-half of that limit (ND = 1/2LOD). It was done according to WHO guidelines, as well as our previous studies. Exceptionally, to avoid an overestimation of exposure to MeHg, the intake of this species was considered as zero for those foods containing levels below the LOD.

## 3. Results and discussion

The mean concentrations of the 4 toxic elements (As, Cd, Hg, and Pb) here analysed, as well as those of the analysed species (InAs and MeHg), in the 15 evaluated food groups, are depicted in Fig. 1. Fish and seafood showed the maximum concentrations of As (3.592 µg/g of fresh weight), Cd (0.117 µg/g of fresh weight), Hg (0.152 µg/g of fresh weight), and MeHg (0.135 µg/g of fresh weight). In turn, the maximum levels of InAs were found in bread and cereals (0.034 µg/g of fresh weight), while the highest concentration of Pb corresponded to chocolate (0.045 µg/g of fresh weight). Since no traces of InAs were detected in fish and seafood, it can be concluded that most of the As contained in fish and seafood is organic As, which is much less toxic

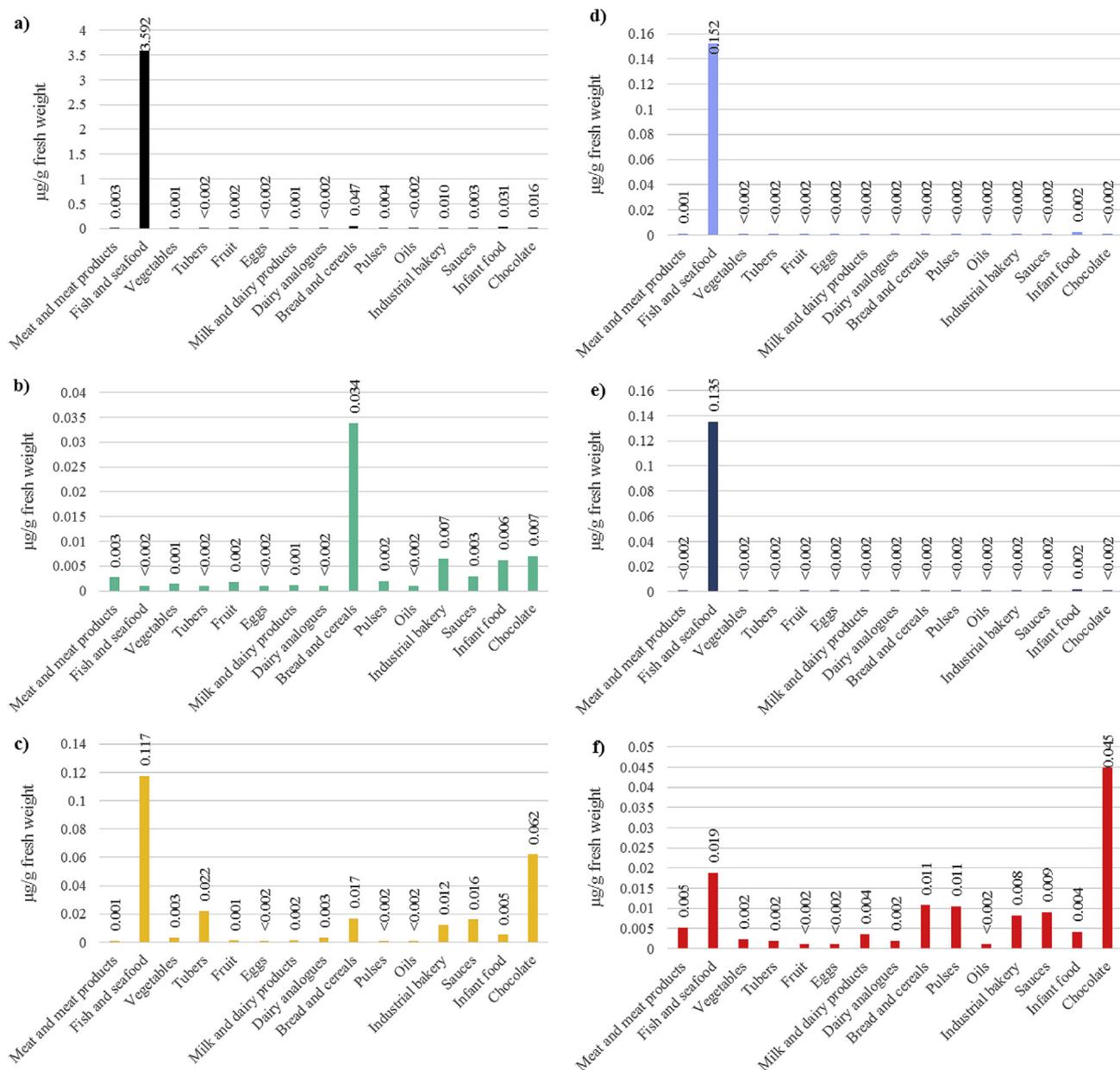


Fig. 1. Concentrations of As (a), InAs (b), Cd (c), Hg (d), MeHg (e) and Pb (f) in different foodstuffs.

than InAs. In turn, the mean percentage of InAs vs. total As in bread and cereals was up to 71%, ranging from 68% to 100% for rice and pasta, respectively. The presence of Pb in chocolate has been largely stated in the scientific literature (Guérin et al., 2017), being concluded that this element might mean a significant source of Pb ingestion, particularly in children (Villa et al., 2014).

Because of the occurrence of heavy metals is remarkable in fish and seafood, a detailed study of this food group was specifically performed. The levels of heavy metals in each one of the 14 species of fish and seafood here analyzed are summarized in Table 1. Sole, cuttlefish and European anchovy showed the highest levels of As (9.28, 8.73 and 7.85 µg/g of fresh weight, respectively), while the lowest values corresponded to pangas and sea bass (0.010 and 0.461 µg/g of fresh weight, respectively). In previous studies, sole had been already highlighted as a species that easily accumulates As (Martorell et al., 2011). It also occurred with red mullet (Martí-Cid et al., 2008; Perelló et al., 2014), a species that was not now included given its low incidence in the current consumption pattern of the Catalan population. Regarding Cd, squid and mussel presented the maximum levels (1.27 and 0.220 µg/g of fresh

Table 1

Concentrations (µg/g of fresh weight) of toxic elements in fish and seafood species purchased from Catalonia, in 2017.

Food item	As	InAs	Cd	Hg	MeHg	Pb
Cod	2.92	< 0.002	< 0.002	0.054	0.046	0.005
Sea bream	1.35	< 0.002	< 0.002	0.105	0.103	0.004
Tuna	1.21	< 0.002	0.013	0.430	0.348	< 0.002
Canned tuna	0.777	< 0.002	0.015	0.214	0.175	< 0.002
European anchovy	7.85	< 0.002	0.013	0.060	0.044	0.002
Swordfish	1.57	< 0.002	0.062	0.856	0.781	0.003
Salmon	0.866	< 0.002	< 0.002	0.019	0.008	< 0.002
Hake	6.14	< 0.002	< 0.002	0.126	0.126	0.002
Monkfish	5.97	< 0.002	< 0.002	0.155	0.145	0.004
Sole	9.28	< 0.002	< 0.002	0.072	0.062	0.010
Cuttlefish	8.73	< 0.002	0.157	0.048	0.046	0.043
Squid	4.32	< 0.002	1.274	0.036	0.033	0.028
Sea bass	0.461	< 0.002	< 0.002	0.095	0.095	0.003
Mussel	2.42	< 0.002	0.220	0.011	0.005	0.171
Pangas	0.010	< 0.002	< 0.002	0.003	0.003	0.003
<b>Fish and seafood</b>	<b>3.59</b>	<b>&lt; 0.002</b>	<b>0.117</b>	<b>0.152</b>	<b>0.135</b>	<b>0.019</b>

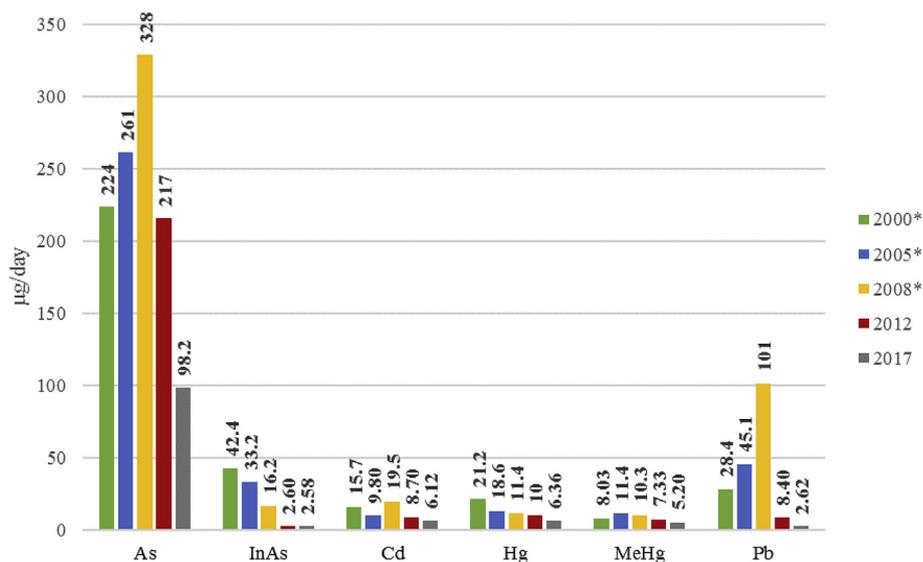
**Table 2**  
Food consumption (g/day) and dietary intake of total As, InAs, Cd, Hg, MeHg and Pb ( $\mu\text{g/day}$ ) by the adult population from Catalonia, Spain.

Food item	Consumption (g/day)	Intake ( $\mu\text{g/day}$ )					
		As	InAs	Cd	Hg	MeHg	Pb
Veal	23	0.046	0.037	0.023	0.023	–	0.120
Pork	11.6	0.010	0.011	0.011	0.010	–	0.056
Chicken	32.4	0.033	0.034	0.032	0.076	–	0.064
Boiled ham	2.07	0.011	0.013	0.002	0.002	–	0.018
Frankfurt sausages	1.48	0.005	0.003	0.002	0.002	–	0.008
Cured ham	8.48	0.009	0.010	0.008	0.009	–	0.034
Turkey ham	4.35	0.026	0.019	0.004	0.006	–	0.022
<b>Meat and meat products</b>	<b>83.4</b>	<b>0.140</b>	<b>0.127</b>	<b>0.082</b>	<b>0.128</b>	–	<b>0.322</b>
Cod	2.75	6.99	0.004	0.004	0.135	0.125	0.014
Sea bream	1.51	1.82	0.001	0.001	0.127	0.162	0.007
Tuna	3.14	3.31	0.003	0.052	1.551	0.918	0.003
Canned tuna	5.77	4.06	0.006	0.079	1.327	1.019	0.006
European anchovy	1.03	6.98	0.0008	0.012	0.055	0.042	0.002
Swordfish	1.10	1.81	0.001	0.057	1.022	0.853	0.004
Salmon	3.90	3.31	0.004	0.004	0.084	0.036	0.003
Hake	9.94	51.8	0.012	0.010	1.13	1.16	0.025
Monkfish	0.6	0.094	0.0004	0.0006	0.009	0.031	0.0007
Sole	0.55	0.0003	0.0005	0.0009	0.0005	0.002	0.001
Cuttlefish	0.79	5.78	0.0008	0.115	0.046	0.029	0.024
Squid	1.94	7.55	0.002	1.83	0.069	0.058	0.048
Sea bass	0.65	0.268	0.0008	0.0006	0.050	0.054	0.003
Mussel	0.33	0.902	0.0003	0.090	0.004	0.002	0.068
Panga	0.47	0.006	0.0006	0.0003	0.002	0.001	0.001
<b>Fish and seafood</b>	<b>34.5</b>	<b>94.7</b>	<b>0.037</b>	<b>2.26</b>	<b>5.61</b>	<b>4.49</b>	<b>0.210</b>
Lettuce	28	0.027	0.027	0.118	0.026	–	0.110
Tomato	39.7	0.039	0.041	0.091	0.043	–	0.039
Zucchini	2.17	0.002	0.002	0.002	0.002	–	0.009
Green bean	6.21	0.006	0.006	0.005	0.005	–	0.006
Onion	11.9	0.031	0.026	0.057	0.012	–	0.022
Leek	1.15	0.003	0.002	0.006	0.001	–	0.001
Carrot	6.79	0.017	0.015	0.028	0.007	–	0.023
<b>Vegetables</b>	<b>95.9</b>	<b>0.125</b>	<b>0.119</b>	<b>0.307</b>	<b>0.096</b>	–	<b>0.210</b>
Potato	41.3	0.039	0.040	0.842	0.038	–	0.086
<b>Tubers</b>	<b>41.3</b>	<b>0.039</b>	<b>0.040</b>	<b>0.842</b>	<b>0.038</b>	–	<b>0.086</b>
Apple	42.8	0.041	0.046	0.041	0.046	–	0.043
Orange	32.6	0.035	0.036	0.032	0.030	–	0.033
Pear	12.6	0.069	0.066	0.023	0.014	–	0.013
Banana	15.6	0.017	0.016	0.019	0.015	–	0.017
Tangerine	11.1	0.011	0.010	0.015	0.009	–	0.011
<b>Fruit</b>	<b>115</b>	<b>0.173</b>	<b>0.174</b>	<b>0.130</b>	<b>0.114</b>	–	<b>0.117</b>
Hen eggs	17.2	0.017	0.015	0.017	0.017	–	0.018
<b>Eggs</b>	<b>17.2</b>	<b>0.017</b>	<b>0.015</b>	<b>0.017</b>	<b>0.017</b>	–	<b>0.018</b>
Whole milk	59.5	0.057	0.053	0.059	0.062	–	0.055
Semi-skimmed milk	81.1	0.078	0.076	0.078	0.080	–	0.083
Soy milk	12.4	0.014	0.020	0.078	0.012	–	0.012
Oat milk	3.03	0.003	0.003	0.003	0.003	–	0.003
<b>Milk and dairy analogues</b>	<b>156</b>	<b>0.152</b>	<b>0.152</b>	<b>0.218</b>	<b>0.157</b>	–	<b>0.153</b>
Yogurt	22.2	0.020	0.021	0.024	0.023	–	0.020
Flavoured yogurt	8.56	0.009	0.011	0.009	0.008	–	0.012
Pudding and custard	8.01	0.008	0.008	0.009	0.009	–	0.008
Mozzarella	4.13	0.004	0.003	0.005	0.003	–	0.027
“Quesitos”	2.32	0.004	0.003	0.011	0.002	–	0.017
Cheese II-fat cheese	1.54	0.001	0.002	0.002	0.001	–	0.018
<b>Dairy products</b>	<b>46.8</b>	<b>0.046</b>	<b>0.048</b>	<b>0.060</b>	<b>0.046</b>	–	<b>0.102</b>
French bread	62.3	0.474	0.346	1.24	0.065	–	0.699
Sliced bread	11.4	0.044	0.037	0.234	0.011	–	0.123
Rice	11	1.78	1.09	0.068	0.011	–	0.029
Pasta	14.3	0.243	0.239	0.322	0.015	–	0.270
<b>Bread and cereals</b>	<b>99</b>	<b>2.54</b>	<b>1.71</b>	<b>1.86</b>	<b>0.102</b>	–	<b>1.12</b>
Lentils	3.71	0.005	0.004	0.004	0.004	–	0.042
Chickpeas	2.33	0.016	0.007	0.002	0.003	–	0.021
<b>Pulses</b>	<b>6.04</b>	<b>0.021</b>	<b>0.011</b>	<b>0.006</b>	<b>0.007</b>	–	<b>0.063</b>

(continued on next page)

Table 2 (continued)

Food item	Consumption (g/day)	Intake ( $\mu\text{g}/\text{day}$ )					
		As	InAs	Cd	Hg	MeHg	Pb
Olive oil	15.7	0.016	0.016	0.016	0.015	–	0.016
Sunflower oil	1.82	0.002	0.002	0.002	0.002	–	0.002
<b>Oils</b>	17.5	<b>0.018</b>	<b>0.018</b>	<b>0.018</b>	<b>0.017</b>	–	<b>0.018</b>
Milk bread	1.88	0.007	0.006	0.020	0.002	–	0.015
Cookies	6.05	0.039	0.038	0.047	0.006	–	0.019
Muffins	3.01	0.008	0.009	0.014	0.003	–	0.005
Sponge cake	3.13	0.023	0.014	0.015	0.004	–	0.013
Chocolate cookies	1.12	0.014	0.005	0.021	0.001	–	0.018
Croissant	2.70	0.016	0.014	0.040	0.003	–	0.041
Breakfast cereals	0.62	0.023	0.013	0.010	0.0006	–	0.003
<b>Industrial bakery</b>	18.5	<b>0.130</b>	<b>0.099</b>	<b>0.167</b>	<b>0.020</b>	–	<b>0.114</b>
Tomato sauce	5.73	0.018	0.018	0.082	0.006	–	0.050
<b>Sauces</b>	5.73	<b>0.018</b>	<b>0.018</b>	<b>0.082</b>	<b>0.006</b>	–	<b>0.050</b>
Chocolate powder	0.92	0.017	0.007	0.067	0.001	–	0.041
<b>Chocolate</b>	0.92	<b>0.017</b>	<b>0.007</b>	<b>0.067</b>	<b>0.001</b>	–	<b>0.041</b>
<b>TOTAL</b>	737	<b>98.2</b>	<b>2.58</b>	<b>6.12</b>	<b>6.36</b>	4.49	2.62



\*InAs and MeHg were not analysed, but their intake was estimated according to bibliographic values.

Fig. 2. Temporal trend of the dietary intake of toxic elements by the adult population of Catalonia (Spain). \*InAs and MeHg were not analysed, but their intake was estimated according to bibliographic values.

weight, respectively). In contrast, this metal could not be detected in several species, namely cod, sea bream, salmon, hake, monkfish, sole, sea bass and panga. In agreement to the results of previous studies (Cano-Sancho et al., 2015a), the highest levels of Hg were found in swordfish and tuna (0.856 and 0.430  $\mu\text{g}/\text{g}$  of fresh weight, respectively), which were also the species with the highest content of MeHg (0.781 and 0.348  $\mu\text{g}/\text{g}$  of fresh weight, respectively). The mean percentage of MeHg vs. total Hg was 89%, ranging from 42% for salmon, to 100% for hake, sea bass and panga. This percentage is slightly higher than that reported in previous surveys performed in Catalonia, in which the MeHg vs. total Hg mean percentage was estimated in 77% (Perelló et al., 2014). However, it falls well within usual values according to data from the scientific literature (Alves et al., 2018; Cano-Sancho et al., 2015a; Zhang et al., 2018). In addition to chocolate, relatively high levels of Pb were also observed in fish and seafood (mean: 0.019  $\mu\text{g}/\text{g}$  of fresh weight), being molluscs the species with the highest content of this element (0.171, 0.043 and 0.028  $\mu\text{g}/\text{g}$  of fresh weight in mussel, cuttlefish and squid, respectively).

The estimated dietary intake of the trace elements here analysed by the adult population living in Catalonia (Spain) is presented in Table 2. Because of the relatively high concentrations of found in fish and seafood, this food group was identified as the main contributor to the intake of As (94.7  $\mu\text{g}/\text{day}$ ), Cd (2.25  $\mu\text{g}/\text{day}$ ), as well as both total Hg (5.61  $\mu\text{g}/\text{day}$ ) and MeHg (4.49  $\mu\text{g}/\text{day}$ ). Up to 95.5% of the ingestion of total As would be due to the consumption of fish and seafood, with hake in a key position (52.8% of the total). Hake is actually the fish species with the highest consumption by the adult population, with a mean intake of 9.94 g/day. Furthermore, hake was the main contributor to the intake of MeHg (22%), while the species with the highest contribution to the total ingestion of total Hg was tuna, fresh and canned (24% and 21% of the total, respectively). With respect to Cd, despite the relatively low consumption of squid, this species showed the highest percentage of Cd intake, reaching up to 30% of the total. The consumption of bivalves and cephalopods has been linked to a substantial relative risk, particularly with one or more weekly meals (Ventura et al., 2018).

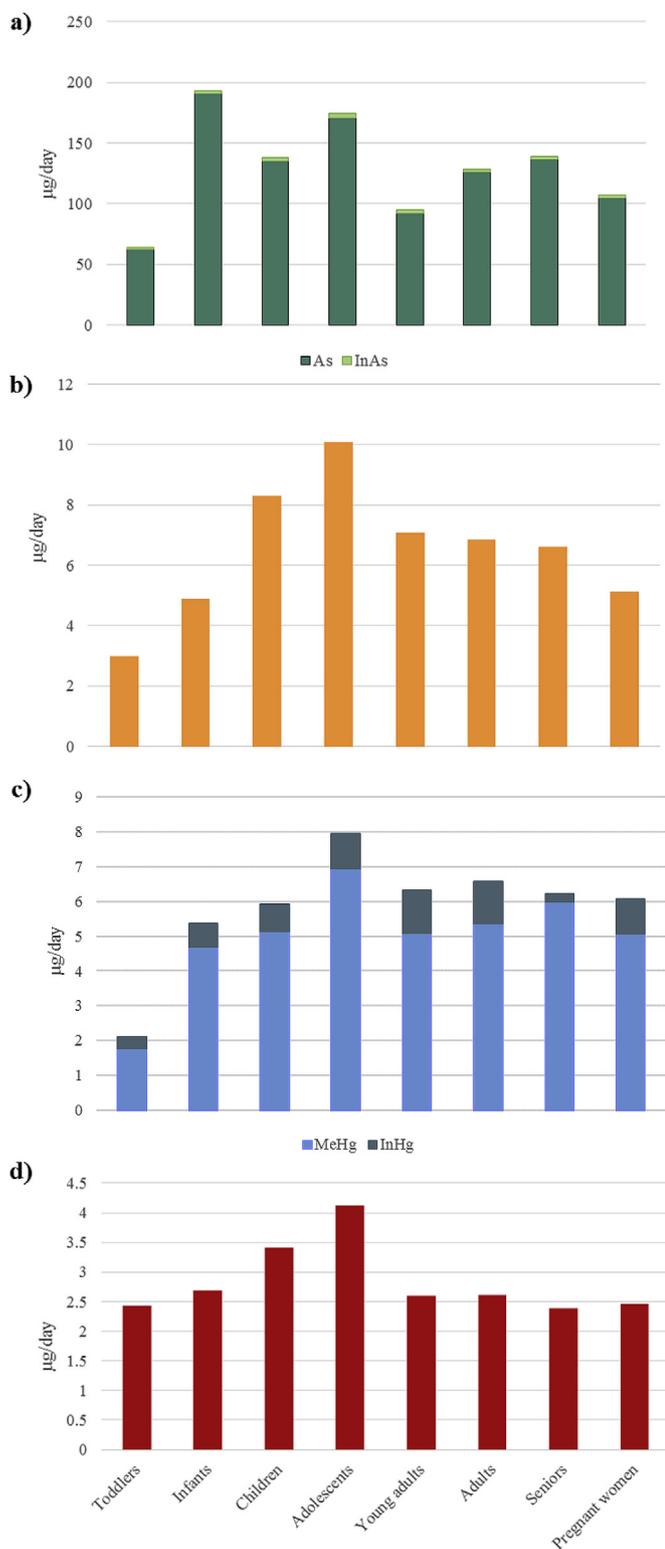


Fig. 3. Dietary intake of As and InAs (a), Cd (b), Hg and MeHg (c), and Pb (d) by various population groups in Catalonia (Spain).

The group of bread and cereals was the major contributor to the intake of InAs and Pb by Catalan adults, with mean percentages of 67% and 42% of the total, respectively. This food group was also identified as an important contributor of Cd ingestion (31%), holding a second position just after the groups of fish and seafood. When evaluating the role of each individual food item, rice was the individual foodstuff with the highest predominance in the intake of InAs, being French bread the

most important item for the Pb intake. Finally, although chocolate was the food item with the highest content of Pb, the importance of this particular foodstuff was only residual, because of the intake of this element dropped to only 1.5% of the total.

Data of the current study were compared with those obtained in previous surveys of the Catalan TDS, including the baseline survey, which was performed in 2000 (Llobet et al., 2003a). The temporal trends in the intake of As, Cd, Hg and Pb, as well as those of InAs and MeHg, are depicted in Fig. 2. It must be noticed that in the first 3 surveys, conducted in 2000, 2005 and 2008 (Llobet et al., 2003a; Martí-Cid et al., 2008; Martorell et al., 2011), the intake of InAs and MeHg species was only estimated, but not specifically analysed. Contrastingly, the levels of both InAs and MeHg were chemically determined in the last (2012) and present (2017) studies (Perelló et al., 2014). In general terms, a remarkable reduction of the intake of all elements was found when comparing the current levels with those of the baseline study. The reductions in the intake of As, Cd, Hg and Pb were: 56%, 61%, 70% and 91%, respectively. It must be remarked that the values corresponding to As and Pb followed a similar trend between 2000 and 2008, with both elements showing a clear increasing tendency (from 224 to 328 µg/day for As, and from 28.4 to 101 µg/day for Pb). However, their intakes dramatically dropped since then, with reductions of 70% and 97%, respectively. In contrast to As and Pb, the reduction in the intake of Cd and Hg was more gradual. Finally, the intake of InAs was identical in both monitoring campaigns (2.60 and 2.58 µg/day in 2012 and 2017), while that of MeHg has slightly reduced (from 7.33 to 4.49 µg/day). Unfortunately, since experimental values are available only for the last two surveys (2012 and 2017), long-term temporal trends cannot be yet evaluated.

The dietary intake of As, Cd, Hg, and Pb for different population groups living in Catalonia is depicted in Fig. 3. Details on the percentages of InAs vs. total As, and MeHg vs. total Hg, are also shown. Adolescents aged between 10 and 17 years was the age group showing the highest intake of Cd (10.1 µg/day), Hg (7.95 µg/day), and Pb (4.12 µg/day). Although this group also presented a relatively high intake of As (175 µg/day), the highest value corresponded to the group of infants aged 1–3 years (191 µg/day). In turn, toddlers (6–11 months old) showed the lowest intake of most analyzed elements, being especially relevant those of As, Cd and Hg (only 61.7, 2.67 and 1.80 µg/day, respectively).

The intake of InAs, Cd, MeHg and Pb for each population group was recalculated in terms of body weight, being then compared with the safety levels established by the European Food Safety Authority (EFSA) (Fig. 4). In 2004, the International Agency for Research on Cancer (IARC, 2004) included As and related compounds in the Group 1, being considered as carcinogenic to humans. Data demonstrated that this element could cause severe effects on the urinary tract, lungs and skin. In 2009, the EFSA considered that the Provisional Tolerable Weekly Intake (PTWI) of 15 µg/kg/week was no longer suitable, setting a new benchmark dose lower confidence limit (BMDL<sub>01</sub>) of 0.3–8 µg/kg/day for InAs (EFSA, 2009a). In the present study, none of the studied population groups exceeded that threshold, being the infants the group showing the highest exposure (0.18 µg/kg/day) (Fig. 4a).

The IARC also classified Cd as a carcinogen to humans (Group 1), with sufficient evidence for carcinogenicity to humans (IARC, 2004). In 2005, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a PTWI of 7 µg/kg/week. Subsequently, in 2009, the EFSA re-evaluated data about Cd and fixed a new TWI of 2.5 µg/kg/week (EFSA, 2009b). In the current study, we found that Catalan infants from 1 to 3 years old would exceed this threshold (2.69 µg/kg/week), while both toddlers and children would be also close to that limit (2.22 and 2.41 µg/kg/week, respectively) (Fig. 4b). It contrasts with the results found in our previous survey (Perelló et al., 2014), when children did not surpass the safety value. Therefore, special attention must be paid to this element.

With respect to Hg and MeHg, the EFSA (2012) recommended a TWI

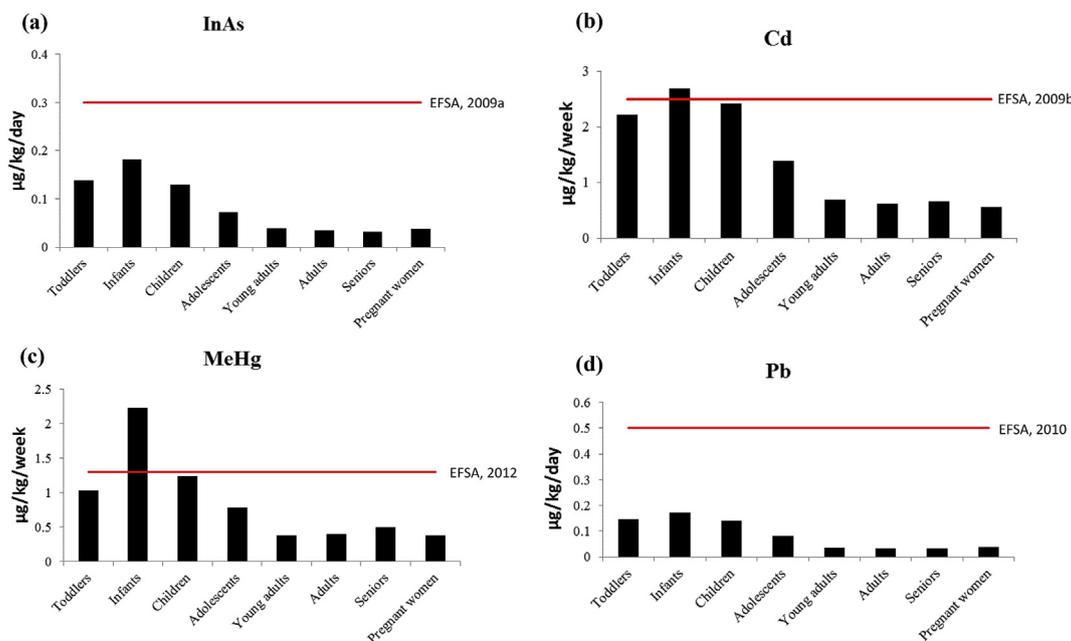


Fig. 4. Dietary intake of InAs (a), Cd (b), MeHg (c) and Pb (d) by various population groups, and comparison to the safety levels established by the EFSA.

Table 3

Dietary intake of toxic elements: A summary of recent (2012–2018) data in a number of countries.

Trace element	Country	Intake (µg/day)	Reference
As (InAs)	Spain	98.2 (2.58)	Present study
	Chile	73 (19.8)	Muñoz et al. (2017)
	Korea	145 (10.4)	Seo et al. (2016)
	Spain	265	Perelló et al. (2015b)
Cd	Spain	6.12	Present study
	Chile	181	Muñoz et al. (2017)
	USA	10.4	Quraishi et al. (2016)
	Spain	33.9	Perelló et al. (2015b)
	Japan	26.4	Itoh et al. (2014)
	Denmark	14	Eriksen et al. (2014)
	France	11.2	Arnich et al. (2012)
Hg (MeHg)	Spain	6.36 (4.49)	Present study
	Chile	5.70	Muñoz et al. (2017)
	Spain	11.6	Puerto-Parejo et al. (2017)
	Korea	4.74 (3.72)	Kim et al. (2016)
	Poland	4.47	Koch et al. (2016)
	Spain	8.48	Perelló et al. (2015b)
Pb	Spain	2.62	Present study
	Spain	46.2	Puerto-Parejo et al. (2017)
	China	28.7 <sup>a</sup>	Luo et al. (2018)
	Spain	41.7	Perelló et al. (2015b)
	Germany	37.1 <sup>a</sup>	Schneider et al. (2014)

<sup>a</sup> Original data given in µg/kg/day, but recalculated for a 70-kg adult.

of 4 µg/kg/week for inorganic mercury and 1.3 µg/kg/week for MeHg. The profile of MeHg intake -according to the age-is similar to that of Cd, with infants exceeding the threshold, and both toddlers and children close to the TWI. Values of MeHg weekly intake were estimated in 1.03, 2.23 and 1.24 µg/kg/week for individuals aged 6–11 months old, 1–3 years old, and 4–9 years old, respectively (Fig. 4c). By contrast, the intake of MeHg by adolescents and adults, including pregnant women, would be well below the maximum intake recommended by the EFSA (2012).

Since the ingestion of Cd and MeHg by infants was particularly high, and in some cases, the EFSA TWI was even exceeded, a particular analysis of the main foodstuffs contributing to that intake was

performed. Regarding Cd, potato was identified as the food item with the highest contribution for toddlers (60%), being followed by baby formula (12%). In turn, the dietary profile of infants and children was very similar, being squid and potato the two food items mostly contributing to Cd intake (28% and 24%, respectively, for infants, and 38% and 13%, respectively, for children). Therefore, a first recommendation to reduce the intake of Cd in young populations is the replacement of squid by other species of cephalopods containing less Cd, such as cuttlefish, whose Cd concentration was notably lower (1.274 vs. 0.157 µg/g of fresh weight).

Interestingly, the intake of MeHg by infants aged 1–3 years exceeded the TWI established by the EFSA. Hake was identified as the individual food item with the highest contribution of MeHg (56%) for this particular age group. This percentage would be directly linked to the fact that hake is the species of fish and seafood with the greatest consumption by infants aged 1–3 (20.3 g/day). Given these estimations, it is clear that the consumption of hake should be -individually-maintained in a moderate level, which can be combined with the intake of other non-predator species. However, it must be noticed that these recommendations are given exclusively considering the health risks associated to the ingestion of MeHg. It is well known that consumption of fish and seafood is very beneficial from many perspectives, being especially relevant the intake of ω-6 and ω-3 fatty acids (Cano-Sancho et al., 2015b; Domingo et al., 2007; Vilavert et al., 2017).

With respect to Pb, the IARC classified this element as possibly carcinogen to humans (Group 2B), and the JECFA established a PTWI of 25 µg/kg/week. However, in 2010, the EFSA concluded that this PTWI was no longer appropriate, since there is not sufficient evidence on the critic Pb effects. Instead of using the PTWI, the EFSA proposed to assess the Pb risk with BMDLs derived from Pb levels in blood (in µg/L), according to data on neurotoxicological development, BMDL<sub>01</sub> 12 (0.50), effects on systolic blood pressure, BMDL<sub>01</sub> 36 (1.50), and the effects on the prevalence of chronic kidney disease, BMDL<sub>10</sub> 15 (0.63) (EFSA, 2010). When compared to the lowest BMDL, none of the groups exceeded the safety levels for Pb (Fig. 4d).

A detailed comparison of metal dietary intake values for adults, reported recently for a number of countries, is summarized in Table 3. In general, data relative to As, Cd and Pb found in the current study would be in the lowest part of the range, while the ingestion of Hg would be similar to that reported in other countries. However, it is

important to remark that data cannot be entirely comparable, as it is affected by a number of miscellaneous parameters, such as consumption habits and selected species (Renieri et al., 2019).

#### 4. Conclusions

Seventeen years after the first TDS on exposure to chemical pollutants, including As, Cd, Hg and Pb, performed in Catalonia (Spain), a clear and continued reduction of the exposure to these elements through food consumption, has been observed. This decrease would be associated to a reduction in food concentrations of these trace elements, as well as to some changes in the dietary patterns of the population. Specific calculations for different population groups according to age were also performed. According to our estimations, none of the adult groups exceeded the safety threshold established by the EFSA. Adolescents aged 10–17 were identified as the population group with the highest intake of Cd, Hg and Pb. This group was also the second in importance for As exposure, just after infants aged between 1 and 3 years. Notwithstanding, when the intake is considered in terms of body weight, the youngest people (namely toddlers, infants and children) showed the highest intake of most elements, exceeding in some cases the PTWI set by the EFSA. Following a standard diet, the dietary intake of MeHg by infants aged 1–3 years was higher than the maximum recommended value (2.23 vs. 1.3 µg/kg/week). A similar pattern was also found for Cd, with infants slightly exceeding the EFSA PTWI (2.69 vs. 2.5 µg/kg/week). According to the data of the current study, to decrease the ingestion of Cd and MeHg by infants aged 1–3, the consumption of fish and seafood should be reduced to two or three weekly portions of varied species of fish and seafood, especially considering that it is an age of high importance for the development.

Although there is a clear decreasing tendency of the dietary intake of toxic elements by the adult population living in Catalonia (Spain), food monitoring studies should continue to assure this decrease is also followed by younger population groups. Therefore, further periodical surveys, paying special attention to the trends in the intake of Cd and MeHg, are clearly necessary.

#### Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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