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Assessment of metal levels in foodstuffs from the Region of Valencia (Spain)

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ABSTRACT

Concentrations of lead, mercury, cadmium, arsenic, tin, copper and chromium were measured in a study carried out in 2010–2011. A total of 8100 food samples were collected and composite samples for 12 food groups were analysed for metal concentration levels. Metal levels were, in general, below the maximum levels set by the current European legislation. The fish group presented the highest Cd, Hg and As levels, whereas sweeteners and condiments group was the most contaminated food group by Pb, Cr and Sn and the meat group had the highest concentrations of Cu. The results of this study are generally similar to or lower than those observed in other studies conducted in other countries, except in the case of Hg, for which high values were obtained, mainly in swordfish. In addition, this survey confirms a decreasing tendency when compared with other studies carried out in Spain.

1. Introduction

Environmental contamination through heavy metals is recognised as a public health hazard worlwide [1]. The general population is exposed to a large number of relevant contaminants such as metals through food consumption, water and other environmental matrices. Diet (food and water) is the main route of exposure to metals [2]. Some metals are relevant toxic elements such as Pb, Cd, As, Cr (VI) and Hg or minor toxic metals (Sn), whereas others are considered essential or probably essential trace elements with likely potential toxicity at excess intakes such as Cu and Cr (III). Besides, mercury can occur as inorganic mercury, mercuric cations and organic mercury. Methylmercury (meHg) is by far the most common form of organic mercury in the food chain [3]. Regarding arsenic, the organic form is less harmful than the inorganic form of arsenic (iAs) which can cause cancer [4]. Nevertheless, the last EFSA Scientific Opinion on arsenic in food [5] shows that occurrence data on arsenic are usually reported as total arsenic (approximately 98%).

Although the European Commission adopted the Regulation 1881/2006 [6] setting maximum levels for Cd, Hg, Sn, iAs and Pb in foodstuffs, Member States should monitor and report levels of these elements to allow the Commission to assess the need to modify existing measures or to adopt additional ones. In addition, it is of great importance to determine the concentrations of metals in foodstuffs in order to calculate the dietary exposure, required to evaluate the possible risk associated through food consumption.

The dietary exposure of a population to food contaminants can be assessed by different approaches [7]. The World Health Organization (WHO) recommended the so-called total diet studies (TDSs) [8] and nowadays the standardised methodology recommended by the WHO [8] or more recently by EFSA [9] is the most widely used in many countries.

In 2008, the results of a monitoring programme on cadmium, lead and mercury in fish and seafood was carried out by the Department of Public Health of the Valencian government, Spain [10]. The estimated dietary exposure of these pollutants was also reported. However, a representative dataset on food consumption is more appropriate to derive the dietary exposure. Consequently, a new study was carried out in the Region of Valencia in which a representative dataset on food consumption was combined with data on the concentration of the compounds of interest in foods to derive the exposure.Over the last years, some studies have reported metal occurrence data in several countries such as France [11], UK [12] or Chile [13]. In Spain, other studies have also allowed the acquisition of data on the concentrations of trace elements in foodstuffs from Catalonia [14,15] or Canary Islands [16]. In 2008, a study was carried out in Valencia [17] to determine the levels

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of mercury, cadmium and lead in fish and seafood marketed in the Region of Valencia using data from monitoring. To complement this study, in 2010–2011, the Public Health Directorate of the region of Valencia started the Valencia Total Diet Study, to estimate the dietary exposure to toxic and essential elements in order to assess the derived health risk. The data in the context of a health risk assessment was reported previously [18]. The present study contains more detail on analytical methods and more complete reporting of the results. The data presented are of great interest as it can be used for regulatory purposes.

The aim of this work was to present metal occurrence data in foodstuffs collected in the region of Valencia for Pb, Cd, As, Hg, Cu, Cr and Sn and to compare these results with those obtained in other countries or in different regions in Spain, and, when available, to compare these results with the maximum levels established by law [6].

2. Material and methods

2.1. Samples

Foodstuffs were selected to be representative of the diet of the population of the Region of Valencia. Two main criteria were considered for selecting the food in the study: (1) the most consumed foods in terms of quantity (> 2 g/person and day) according to the food consumption data of the region of Valencia and (2) foods that are known to contribute the most to exposure to the metals of interest (swordfish or tuna in the case of meHg or offal for Cd [22,30]). A total of 81 different individual foods were selected and aggregated into twelve food groups. To minimise the variability, each food was composed of a hundred samples, collected in different areas (covering rural and urban areas in different geographic locations) and seasons, so the total number of samples purchased was 8100. In order to reduce the number of analysis, a composite sample was formed by 10 individual samples of the same food, so the total number of analysis was 810 for each metal, except for mercury that was only analysed in fish and seafood products (120 analysis) (Table 1).

Two fundamental criteria were considered for designing the sampling plan: the type of establishment and its geographical location. The sampling was carried out in two stages: (1) Selection of a random cluster sample corresponding to different geographical areas or core areas of the Valencian Region; being the sample size assigned to each cluster proportional to the population that it represented, and (2) A new selection using stratified random sampling based on the type of establishment. Four types of establishments were considered: 3 food chains supplying an important part of the Valencian region population (30% each) and local markets (10%). Finally, samples were collected in 11 cities of the Region of Valencia, with over 25.000 inhabitants each, at their respective markets and supply chains (see Fig. S.I.1 in the supplementary information online).

Only edible parts of each food were included in the composites. Kitchen utensils were used for food handling. Food was homogenised with a Thermomix TM-21 food processor and the obtained mixture was divided into 100 g or mL aliquots. These composite samples were stored in high-density polyethylene bags. For maximum stability and homogeneity of samples, fresh samples (high water content) were previously lyophilised with a Telstar LyoAlfa 15 lyophiliser and sent to the laboratory for analysis.

2.2. Reagent and standard solutions

All reagents used in this study were Suprapur-type (Merck, Darmstadt, Germany), or of high analytical grade. Reagents and samples were prepared using analytical reagent grade chemicals and ultrapure water type I (ASTM) generated by purifying distilled water with a Milli-Q Gradient A10 system (Merck Millipore S.A., Merck KGaA, Darmstadt, Germany).

2.3. Analysis

The samples were analysed in two different laboratories: the Public Health Laboratory (Alicante) and the Institute of Agrochemical and Food Technology (Valencia), accredited following the ISO/IEC 17,025 standard [19]. The analytical techniques used fulfilled the criteria set in Regulation (EC) Nº333/2007 [20]. All analyses were performed according to protocols of quality assurance, including duplicate samples, reagent blanks, fortified samples and certified reference materials. Detailed methodologies are described in the following sections:

2.3.1. Analysis of Pb, Cd, total As (tAs), Cu, Cr and Sn

The digestion of lyophilised samples was carried out using a microwave digestion system, Ethos one (Milestone Inc., Shelton, USA), equipped with the Q-20 Quartz Rotor Ultratrace Analysis (20 mL quartz

Table 1
Foodstuffs included in the total diet study, data sampling design.

Food group	Foodstuffs	Nº Total samples	Nº total of composites (or analysis)
Vegetable oils (Vo)	Olive oil and sunflower oil	200	20
Mineral water (Mw)	Mineral water	100	10
Alcoholic beverages (Ab)	Wine and beer	200	20
Non-alcoholic beverages (nAb)	Soda and soft drinks, orange juice, multi-fruits juice	300	30
Meat and meat products (Meat)	Chicken, pork, beef, lamb, rabbit, hamburgers, sausages, cured ham, cooked ham, cured sausages, foie-grass and offal.	1200	120
Cereals, pulses, tuber, nuts and dried fruits (Cereal)	Rice, industrial bakery, cornflakes, cookies, beans, white bread, sliced bread, wholemeal bread, pasta, potatoes, dried fruits.	1100	110
Prepared dishes (Pd)	Pizzas, snacks, frozen prepared dishes and canned meals	400	40
Sweeteners and condiments (Sc)	Chocolate and cacao, sugar, salt, sweets and sauces and mayonnaise	500	50
Vegetables and fruits (Vf)	Spinaches and chards; lettuces; green beans; onions; garlic; peppers; aubergine, zucchini and cucumber; carrots and pumpkin; tomatoes; olives and pickles; cauliflower, cabbage and broccoli; artichokes, celery and leek; mushrooms; coffee and soluble coffee; oranges; strawberries; apples and pears; sherry and plum; melon and watermelon; banana; peach and apricot; grapes.	2200	220
Eggs (Egg)	Chicken eggs	100	10
Milk and dairy products (Milk)	Milk, cheese, yogurt, custards and smoothie, butter and soybean products	600	60
Fish and seafood (Fish)	Canned fish, tuna, squid and cuttlefish, sea bream and sea bass, swordfish, shellfish, mussels, whitefish, salmon and trout, sardine and anchovy, salting fish and smoked fish.	1200	120

Note: Number of samples/food item = 100.
Number of samples/composite = 10.

tubes, 250 °C and 40 bars operating parameters). A unique sample digestion procedure was applied to all samples, but immediately after digestion, two different approaches were used depending on the stability of the analytes.

Approximately 0.25 g of lyophilised or dried sample were weighed in quartz digestion vessels and 5 mL of suprapure HNO3 (65%) were added in a fume hood. The mixture was left to react for over an hour until the gas generation process finished. Samples were placed in the microwave digestion system and the digestion programme shown in Table S.I.1 in the supplementary information online was applied.

In each digestion sequence, at least one randomly-selected vessel was filled with reagents only and taken through the entire procedure as a reagent blank. After cooling at room temperature, sample solutions were quantitatively transferred into 25 mL glass volumetric flasks (Class A) and completed with ultra-pure water to the final volume. Solutions were transferred to 50 mL polypropylene tubes and two aliquots were immediately prepared:

Aliquot for tAs, Cd, Cr, Cu and Pb analysis by ICP–MS: 9.9 mL of the digestion solution was placed in a 10 mL polypropylene tube and 0.100 mL of 10 mg L^{-1} Internal Standard solution (containing scandium (Sc), germanium (Ge), rhodium (Rh), antimony (Sb) and bismuth (Bi)) was added to obtain a final concentration of 10 μg L^{-1}.

Aliquot for Sn analysis by ICP–MS: 9.4 mL of the digestion solution was placed in a 10 mL polypropylene tube, 0.5 mL of suprapur HCl 37% was added to stabilise Sn in solution and 0.1 mL of mg L^{-1} Internal Standard solution was added to obtain a final concentration of 10 μg L^{-1}.

Residual moisture was determined in all lyophilised samples, in order to correct the final results expressed in dry mass. The following procedure was applied: approximately 0.5 g of sample was weighed on a previously dried and stabilised ceramic container and introduced in an oven at 105 °C during 12 h. After this step, containers were introduced in a desiccator and weighed until a constant weight was obtained.

Analysis were performed on an ELAN DRC II ICP-MS (PerkinElmer, Inc., Shelton, USA) equipped with a standard concentric-glass nebulizer and a baffled-glass cyclonic spray chamber (both from Meinhard® Glass Products, Golden, Colorado, USA). The instrumental operating conditions are shown in Table S.I.1 in the supplementary information online. As a routine basis, several performance parameters (i.e., sensitivity in the whole mass range, reading precision, double-charges and oxide formation and background signal) were checked daily with the 1 μg L^{-1} tuning solution. In high-chloride sample matrices (i.e., mineral salt, salted fish and salted meet) tAs was analysed by means of dynamic reaction cell (DRC) technology using ultra-pure oxygen (O_2) as a reaction gas. In the rest of matrices, tAs was analysed in Standard mode. In all matrices, Cr was analysed in DRC mode using methane as a reaction gas in order to eliminate ArC-based interferences.

Multi-element standard solutions were used for external calibration. Six standards in 2% (w/w) HNO3 matrix for As, Cd, Cr and Pb, and in 2% (w/w) HCl matrix for Sn were prepared at levels ranging from 0 to 50 μg L^{-1}. For Cu, the calibration range was enlarged up to 100 μg L^{-1}. A standard linear regression approach was applied with internal standardisation.

Five different quality control samples (QCS) were chosen to monitor the analytical sequence: Initial Calibration Verification (ICV), Initial Calibration Blank (ICB), Reagent Blanks, Certified Reference Materials (CRM) and Continuous Calibration Verification (CCV) as well as internal standard signal monitoring. The CRM used to assess method performance criteria were BCR 150 (skimmed milk), BCR 191 (brown bread) and BCR 185R (bovine liver) from the Community Bureau of References, IRMM 804 (rice flour) from the Institute for Reference Materials and Measurement, European Commission, Joint Research Centre, DORM 3 (fish protein) from the National Research Council, Canada and LGC7162 (strawberry leaves) from the Laboratory of Government Chemist (LGC, UK) (see Table S.I.1 in the supplementary information online).

2.3.2. Analisys of total Hg (tHg)

Samples were digested in a microwave oven and mercury was measured by cold vapor generation coupled with atomic fluorescence spectrometry (CV-AFS), using a PSA team 10,023 model, Orpington, UK, in the samples of fish and seafood. Lyophilized samples (0.2 g) were placed in a Teflon PFA vessel and treated with 4 ml of HNO_3 concentrate (14 N) and 1 ml of H_2O_2. The Teflon PFA vessel was irradiated at 800 W (180 °C, 15 min). At the end of the digestion programme, the digest was placed in a 250 ml beaker and allowed to rest all night to eliminate nitrous vapour. It was then filtered through 0.45 μm and made up to volume with 5% HCl (v/v).

2.3.3. Analisys of meHg

For the extraction of Hg species, an ultrasonic acid extraction was employed. A volume of 10 mL of extractant solution (0.10% v / v HCl + 0.05% m / v L-cysteine + 0.10% v / v 2-mercaptoethanol) was added to the lyophilized samples (0.2–2 g). The mixture was sonicated for 5 min and centrifuged (2000 rpm/15 min). The resulting extract was filtered through a 0.45 μm Whatman Nylon before the quantification by HPLC-thermooxidation-CV-AFS, using polytetrafluoroethylene (PTFE) tubing and T-joints.

The instrumental conditions for tHg and meHg determination and the method performance criteria are shown in Table S.I.2 in the supplementary information online. As CRM, DORM-2, TORT-2 and DORM-3, from the Institute for Reference Materials and Measurements, European Commission, Joint Research Centre, were used.

2.3.4. Analisys of inorganic iAs

Analysis was performed by acid digestion, solvent extraction and hydride generation by flow injection (FI-HG-AAS) determination [21]. Deionised water (4.1 mL) and 12 mol L^{-1} HCl (18.4 mL) were added to lyophilised or dry food samples (0.5–1 g) and the mixture was left overnight (12-15 h). After reduction by HBr (2 mL) and hydrazine sulfate (1.5% w/v, 1 mL), iAs was extracted into chloroform (3 x 10 mL) and back-extracted into 1 mol L^{-1} HCl (2 x 10 mL). The determination of inorganic arsenic in the back-extraction phase was performed by means of the following procedure: 2.5 mL of ashing aid suspension (20% w/v $MgNO_3$ + 2% w/v MgO) and 10 mL of 14 mol L^{-1} HNO_3 were added to the combined back-extraction phases. The mixture was evaporated on a sand bath until total dryness and placed in a muffle furnace (425 ± 25 °C; 12 h). The white ash obtained was dissolved in 6 M HCl and reducing solution (5% m/v KI + 5% m/v ascorbic acid). After 30 min, the resulting solution was filtered through Whatman No. 1 filter-paper and diluted to final volume with 6 mol L^{-1} HCl.

The instrumental conditions and the analytical characteristics of the method are shown in Table S.I.3 in the supplementary information online. As CRM, rice flour SRM 1568a from the National Institute of Standards and Technology (NIST) was analyzed with each series of samples.

2.4. Consumption data

Intake estimates were based on consumption data obtained from a questionnaire-based dietary survey conducted and validated in 2010–11 by the Valencian Public Health Directorate (Fullana et al. 2010). Dietary data were collected through a 24-h recall in which 1478 subjects (195 young children between 6 and 15 years of age and 43.5 kg mean body weight; and 1281 adults between 16 and 95 years of age and 71.2 kg mean body weight) were asked in a face-to-face interview to recall and describe the kinds and amounts of all foods and beverages ingested during the previous 24-h period. It was conducted from June 2010 to February 2011 in three consecutive periods or waves in order to take into account of variations in consumption patterns according to season. The food consumption data and more detailed information can be found in a previous paper published by the authors [18] inwhich dietary exposure was assessed.

2.5. Statistical analysis

Ordinary statistical methods were used to calculate the arithmetical mean, minimum and maximum levels on numbers (n) samples of general food groups. Results below the LOQ, were set to LOQ/2, as in a middle-bound (MB) scenario assessment and were set to LOQ in the upper-bound (UB) scenario assessment.The article describes the metal concentration data by food but the study was not designed to allow statistical comparisons between foods (only ten data per food). However, food group data were enough for carrying out a statistical comparison and assessment of significant differences. This was made using Student's t-test. All statistics were performed using data analysis function in Microsoft Office Excel.

3. Results

Tables 2a and 2b (Pb, Cd, As, Cu, Cr and Sn) and Table 2c (Hg) show the concentration found in the different foodstuffs analysed. The distribution of element concentrations in food groups was represented graphically. All food group results were expressed as the mean on the corresponding figures.

3.1. Lead

Of the 810 samples analysed, 84% contained Pb at levels higher than the LoQ (Tables 2a and 2b and Fig. 1). All samples fell below the limits established by normative [6]. By food groups, the average levels of Pb found followed the sequence: "Sweeteners and condiments" (Sc) (0.0958 mg kg^{-1}) > Cereal (0.0438 mg kg^{-1}) > Fish (0.0349 mg kg^{-1}) > Meat (0.0273 mg kg^{-1}) > "Prepared dishes" (Pd) (0.0225 mg kg^{-1}) > "Vegetable oils" (Vo) (0.0192 mg kg^{-1}) (see Fig. 1).

In the Sc group, salt was by far the product with the highest Pb mean level (0.331 mg kg^{-1}). In the "cereal" group, bakery presented the highest average levels (0.0893 mg kg^{-1}) followed by pasta (0.0540 mg kg^{-1}) and pulses (0.0522 mg kg^{-1}) (see Table 2a). In the "Fish "group, mussels presented the highest average levels (0.2203 mg kg^{-1}). However, these values are below the maximum limit established by law [6]. In the "Meat" group, the food cured sausage presented the highest Pb levels (0.0607 mg kg^{-1}). Snacks were the food with the highest Pb average level in the "Prepared dishes" group (0.0376 mg kg^{-1}). The rest of food groups contained, in general, low average levels of Pb.

3.2. Cadmium

Of the 810 samples analysed, 54% contained Cd at levels higher than the LoQ (Tables 2a and 2b and Fig. 1). All of the samples fell below the limits established by normative [6].

The "fish" group presented the highest average levels of Cd, at 0.0816 mg kg^{-1} (ranging between 0.0018 and 0.5686 mg kg^{-1}); followed by Sc (0.0512 mg kg^{-1}); "Meat" (0.0281 mg kg^{-1}), "Cereal" (0.0271 mg kg^{-1}) and "Prepared dishes" (0.0246 mg kg^{-1}) (see Table 2a and Fig. 1).

In the "fish" group, mussels presented, once again, the highest average Cd levels (0.1967 mg kg^{-1}) followed by squid (0.1853 mg kg^{-1}). The second group with high Cd levels was "Sweeteners and condiments", mainly due to the contribution of chocolate and cocoa, with an average of 0.0938 mg kg^{-1}(see Table 2a). In the "meat" group, offal presented the statistically highest Cd average levels (0.1583 mg kg^{-1}) (see Table 2a). whereas dried fruits were the products with the highest level of Cd in the "Cereal" group, with a range between 0.040 and 0.232 mg kg^{-1} (Table 2a).

3.3. Total arsenic

Of the 810 samples analysed, 87% contained tAs at levels higher than the LoQ (Tables 2a and 2b and Fig. 1). All samples were below the normatively established limits [6].

The "fish" group presented the highest tAs levels. Results ranged between 0.3292 and 18.3130 mg kg^{-1} and the average level was 2.1669 mg kg^{-1} (see Table 2a). The other food groups presented low values in relation to fishery products. In the present study, the concentration range was between 0.0304 mg kg^{-1} for the "Cereal" and 0.0035 mg kg^{-1} for "Alcoholic beverages" group. In the "Cereal" group, rice presented the highest level, with an average level of 0.1468 mg kg^{-1} and a range between 0.1160 and 0.2330 mg kg^{-1} (see Table 2a).

3.4. Inorganic arsenic

Of the 810 samples analysed, 91% contained iAs at levels higher than the LoQ (Tables 2a and 2b and Fig. 1). All samples were below the normatively established limits [6].

The iAs average concentration was 0.007 mg kg^{-1} with a minimum of 0.0001 mg kg^{-1} for tomato and a maximum of 0.0502 mg kg^{-1} for shellfish (Tables 2a and 2b). By food groups, the highest levels of iAs were found in the "fish and fishery products" group, with an average of 0.0174 mg kg^{-1}. Shellfish showed the highest levels with an average of 0.0502 mg kg^{-1}, followed by the homogeneous mixture of "sardine and anchovy" with an average concentration of 0.03992 mg kg^{-1}. Mussel iAs average level was of 0.0270 mg kg^{-1}.

In the group of "cereals, pulses, tubers and nuts" of the present study, an iAs average value of 0.0133 mg kg^{-1} fresh weight was obtained. Again, the highest level was found in rice with an average of 0.0740 mg kg^{-1} (see Table 2a).

3.5. Mercury

Of the 120 fish and seafood samples analysed, 100% of tHg and meHg values were quantified (> LOQ) (Table 2c and Fig. 2). In the present study the average values were 0.2515 mg kg^{-1} for tHg and 0.1604 mg kg^{-1} for meHg (see Table 2c). The highest values of tHg and meHg were observed in swordfish (average of 1.4448 mg kg^{-1} for tHg and values from 1.0854 to 2.2875 mg kg^{-1}), in which all samples exceed the limit established by law of 1.0 mg kg^{-1} fresh weight [6]. Tuna average value was below the limit established by law [6], but 3 samples exceed it, with a maximum value of 1.6155 mg kg^{-1} (see Fig. 2). The rest of the samples were below the maximum levels established by legislation. The lowest levels were detected in mussels (see Table 2c), with an average value of 0.007 mg kg^{-1}.

3.6. Copper

Of the 810 samples analysed, 97% contained Cu at levels higher than the LoQ (Tables 2a and 2b and Fig. 1). Average values obtained varied from 0.0485 mg kg^{-1} in Alcoholic beverages group (Ab) to 5.1891 mg kg^{-1} in the "meat" group.

In the "meat" group the levels ranged between 0.2663 mg kg^{-1} (chicken) and 100.8016 mg kg^{-1} (offal). The product with the highest average level was the offal with 50.4074 mg kg^{-1} and the product with the lowest level was chicken (0.3589 mg kg^{-1}) (see Table 2a).

The "Cereals" group presented also high Cu levels, but below the "Meat" group, with an average of 3.0958 mg kg^{-1} mainly due to the contribution of dried fruits in which an average of 11.483 mg kg^{-1} was obtained (Table 2a).

In the"Sweeteners and condiments" group, the main contributors were chocolate and cocoa, with an average of 13.3355 mg kg^{-1}.

3.7. Chromium

Of the 810 samples analysed, 95% contained Cr at levels higher than the LoQ (Tables 2a and 2b and Fig. 1).

The highest mean levels were found in the food group "sweeteners

Table 2a
Levels of Pb, Cd, As, Cu, Cr and Sn in foods in mg kg^{-1} fresh mass.

FOODSTUFFS	Pb				Cd				tAs				iAs	
	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean
Vo (n = 20)	80	0,0192	0,0104	0,0480	0				0				100	0,0022
Olive oil	60	0,0204	0,0119	0,0270	0				0				100	0,0015
Sunflower oil	100	0,0185	0,0104	0,0480	0				0				100	0,0028
Mw (n = 10)	0				0				0				100	0,0002
Ab (n = 20)	70	0,0048	0,0007	0,0162	0				55	0,0035	0,0024	0,0101	100	0,0016
Wine	40	0,0121	0,0103	0,0162	0				10	0,0101	0,0101	0,0101	100	0,0007
Beer	100	0,0018	0,0007	0,0057	0				100	0,0029	0,0024	0,0033	100	0,0016
nAb (n = 30)	100	0,0068	0,0009	0,0250	0				30	0,0023	0,0014	0,0030	63	0,0014
Soda and soft drinks	100	0,0013	0,0009	0,0024	0				0				50	0,0004
Orange juice	100	0,0058	0,0014	0,0147	0				0				40	0,0004
Multi-fruits juice	100	0,0132	0,0060	0,0250	0				90	0,0023	0,0014	0,0030	100	0,0023
Meat (n = 120)	68	0,0273	0,0025	0,0684	23	0,0281	0,0043	0,1583	100	0,0233	0,0039	0,0582	84	0,0019
Chicken	40	0,0031	0,0025	0,0044	0				100	0,0086	0,0039	0,0255	100	0,0018
Pork	30	0,0052	0,0037	0,0062	0				100	0,0148	0,0106	0,0192	100	0,0003
Beef	10	0,0035	0,0035	0,0035	0				100	0,0185	0,0111	0,0239	100	0,0032
Lamb	20	0,0038	0,0035	0,0040	0				100	0,0140	0,0096	0,0243	100	0,0003
Rabit	10	0,0033	0,0030	0,0035	0				100	0,0118	0,0073	0,0150	100	0,0003
Hamburgers	100	0,0328	0,0129	0,0617	20	0,0044	0,0043	0,0045	100	0,0186	0,0124	0,0258	100	0,0007
Sausages	100	0,0273	0,0230	0,0369	40	0,0085	0,0046	0,0212	100	0,0261	0,0223	0,0300	100	0,0033
Cured ham	90	0,0119	0,0069	0,0253	0				100	0,0530	0,0423	0,0582	70	0,0042
Cured sausage	100	0,0607	0,0472	0,0684	20	0,0082	0,0082	0,0082	100	0,0435	0,0348	0,0532	20	0,0296
Cooked ham	100	0,0202	0,0154	0,0246	0				100	0,0331	0,0285	0,0386	100	0,0059
Foie-gras	100	0,0287	0,0200	0,0416	100	0,0176	0,0157	0,0230	100	0,0279	0,0235	0,0568	50	0,0045
Offal	100	0,0333	0,0200	0,0665	100	0,0553	0,0168	0,1583	100	0,0073	0,0059	0,0099	80	0,0008
Cereal (n = 110)	95	0,0438	0,0025	0,3925	87	0,0271	0,0087	0,2320	100	0,0304	0,0022	0,2330	92	0,0133
Rice	40	0,0123	0,0097	0,0153	10	0,0091	0,0091	0,0091	100	0,1468	0,1160	0,2330	100	0,0740
Industrial bakery	100	0,0893	0,0295	0,3925	80	0,0106	0,0084	0,0163	100	0,0153	0,0113	0,0374	100	0,0060
Cornflakes	100	0,0471	0,0360	0,0630	100	0,0238	0,0170	0,0340	100	0,0306	0,0151	0,0580	100	0,0144
Cookies	100	0,0508	0,0290	0,0960	80	0,0128	0,0102	0,0147	100	0,0180	0,0158	0,0238	100	0,0056
Beans	100	0,0522	0,0380	0,0690	100	0,0150	0,0127	0,0178	100	0,0188	0,0157	0,0221	80	0,0049
White bread	100	0,0253	0,0133	0,0362	100	0,0164	0,0143	0,0186	100	0,0161	0,0126	0,0198	90	0,0067
Sliced bread	100	0,0407	0,0252	0,0935	100	0,0159	0,0150	0,0175	100	0,0217	0,0196	0,0250	100	0,0062
Wholemeal bread	100	0,0306	0,0251	0,0371	100	0,0208	0,0173	0,0251	100	0,0191	0,0140	0,0470	80	0,0048
Pasta	100	0,0540	0,0290	0,1110	100	0,0182	0,0142	0,0240	100	0,0189	0,0112	0,0390	100	0,0107
Potatoes	100	0,0147	0,0025	0,0471	100	0,0233	0,0125	0,0334	100	0,0031	0,0022	0,0045	80	0,0013
Dried fruits	100	0,0462	0,0208	0,1260	100	0,1106	0,0400	0,2320	100	0,0254	0,0131	0,0600	80	0,0031
Pd (n = 40)	100	0,0225	0,0057	0,1740	100	0,0246	0,0038	0,1020	100	0,0300	0,0148	0,0523	42,5	0,0041
Pizzas	100	0,0214	0,0165	0,0286	100	0,0107	0,0093	0,0152	100	0,0424	0,0299	0,0523	80	0,0059
Snacks	100	0,0376	0,0121	0,1740	100	0,0769	0,0550	0,1020	100	0,0321	0,0249	0,0430	0	
Frozen prepared dishes	100	0,0128	0,0057	0,0200	100	0,0057	0,0038	0,0072	100	0,0286	0,0173	0,0512	0	
Canned meals	100	0,0180	0,0138	0,0205	100	0,0051	0,0044	0,0058	100	0,0170	0,0148	0,0195	90	0,0003

FOODSTUFFS	iAs		Cu				Cr				Sn			
	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max
Vo (n = 20)	0,0010	0,0040	100	0,1243	0,0480	0,5250	100	0,1499	0,1070	0,2300	0			
Olive oil	0,0010	0,0033	100	0,1496	0,0580	0,5250	100	0,1480	0,1070	0,2300	0			
Sunflower oil	0,0010	0,0040	100	0,0989	0,0480	0,1980	100	0,1517	0,1160	0,1670	0			
Mw (n = 10)	0,0001	0,0005	0				0				0			
Ab (n = 20)	0,0003	0,0027	100	0,0485	0,0271	0,0957	55	0,0107	0,0059	0,0173	50	0,0178	0,0026	0,0816

(continued on next page)

Table 2a (*continued*)

FOODSTUFFS	iAs		Cu				Cr				Sn			
	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max
Wine	0,0003	0,0009	100	0,0590	0,0271	0,0957	10	0,0101	0,0101	0,0101	0			
Beer	0,0009	0,0027	100	0,0381	0,0304	0,0573	100	0,0107	0,0059	0,0173	100	0,0178	0,0026	0,0816
nAb (n = 30)	0,0003	0,0035	100	0,1404	0,0026	0,3238	100	0,0239	0,0009	0,0803	83	0,0184	0,0024	0,0785
Soda and soft drinks	0,0003	0,0005	100	0,0108	0,0026	0,0356	100	0,0217	0,0061	0,0563	80	0,0048	0,0024	0,0136
Orange juice	0,0004	0,0005	100	0,2004	0,1679	0,2297	100	0,0086	0,0009	0,0313	70	0,0141	0,0038	0,0414
Multi-fruits juice	0,0014	0,0035	100	0,2100	0,1508	0,3238	100	0,0415	0,0090	0,0803	100	0,0323	0,0102	0,0785
Meat (n = 120)	0,0003	0,0124	100	5,1891	0,2663	100,8016	100	0,2384	0,0188	2,2237	81	0,1151	0,0006	0,6879
Chicken	0,0013	0,0037	100	0,3589	0,2663	0,5380	100	0,0804	0,0525	0,1163	30	0,0152	0,0026	0,0397
Pork	0,0003	0,0003	100	0,5044	0,3951	0,6498	100	0,7234	0,0260	1,5890	100	0,4490	0,2924	0,6879
Beef	0,0003	0,0057	100	0,7173	0,6025	1,0018	100	0,7932	0,0614	2,2237	100	0,1855	0,1268	0,2370
Lamb	0,0003	0,0004	100	0,9343	0,6230	1,4148	100	0,5463	0,0455	2,1383	100	0,1712	0,1411	0,2453
Rabit	0,0003	0,0003	100	0,4573	0,3895	0,6100	100	0,1404	0,0474	0,3471	100	0,0867	0,0285	0,1540
Hamburgers	0,0004	0,0023	100	0,9196	0,7508	1,2530	100	0,0851	0,0350	0,1770	70	0,0146	0,0051	0,0165
Sausages	0,0004	0,0062	100	1,0440	0,7506	1,5413	100	0,0827	0,0415	0,1217	40	0,0172	0,0048	0,0080
Cured ham	0,0005	0,0006	100	0,9106	0,7866	1,0154	100	0,1025	0,0936	0,1179	80	0,0600	0,0079	0,3341
Cured sausage	0,0042	0,0045	100	1,1343	1,0042	1,3063	100	0,1293	0,0756	0,3608	100	0,0055	0,0006	0,0100
Cooked ham	0,0016	0,0124	100	0,6003	0,5291	0,7551	100	0,1015	0,0790	0,1349	100	0,0258	0,0032	0,1496
Foie-gras	0,0024	0,0102	100	3,8912	3,7026	4,9292	100	0,0366	0,0329	0,0598	100	0,0727	0,0439	0,2441
Offal	0,0003	0,0029	100	50,4074	20,7820	100,8016	100	0,0356	0,0188	0,0786	50	0,0052	0,0038	0,0082
Cereal (n = 110)	0,0002	0,1048	100	3,0958	0,6168	13,6450	100	0,0992	0,0118	0,6720	47	0,0551	0,0070	1,7242
Rice	0,0602	0,1048	100	1,3931	1,2284	1,6330	100	0,0789	0,0118	0,2877	70	0,0317	0,0090	0,1175
Industrial bakery	0,0035	0,0107	100	0,9389	0,6168	1,4365	100	0,0985	0,0607	0,1472	60	0,0216	0,0086	0,0815
Cornflakes	0,0056	0,0294	100	2,2613	1,9900	2,7330	100	0,0978	0,0620	0,1300	70	0,0206	0,0103	0,0620
Cookies	0,0010	0,0096	100	1,0727	0,9720	1,1450	100	0,0705	0,0560	0,0980	10	0,0232	0,0232	0,0232
Beans	0,0028	0,0075	100	8,2161	7,5900	9,6170	100	0,3400	0,2530	0,6720	50	0,0545	0,0119	0,1620
White bread	0,0024	0,0102	100	1,3396	1,2227	1,4450	100	0,0951	0,0637	0,1349	40	0,3662	0,0100	1,7242
Sliced bread	0,0006	0,0191	100	1,3958	1,1602	2,5781	100	0,0842	0,0606	0,1213	40	0,0127	0,0070	0,0270
Wholemeal bread	0,0036	0,0072	100	1,9938	1,7565	2,3422	100	0,0918	0,0665	0,1471	80	0,0111	0,0080	0,0170
Pasta	0,0071	0,0143	100	3,1046	2,9660	3,2160	100	0,0503	0,0410	0,0590	30	0,0881	0,0105	0,0620
Potatoes	0,0002	0,0028	100	0,8546	0,6554	1,0599	100	0,0192	0,0144	0,0274	100	0,0340	0,0097	0,1404
Dried fruits	0,0010	0,0061	100	11,4830	9,5020	13,6450	100	0,0651	0,0400	0,0850	20	0,0174	0,0124	0,0224
Pd (n = 40)	0,0003	0,0159	75	1,5627	0,5799	3,7000	75	0,1419	0,0340	0,3940	60	0,0776	0,0059	0,3298
Pizzas	0,0030	0,0159	0				0				30	0,0458	0,0360	0,0652
Snacks			100	3,1897	2,7380	3,7000	100	0,2827	0,2110	0,3940	10	0,1480	0,1480	0,1480
Frozen prepared dishes			100	0,7507	0,5799	1,8447	100	0,0634	0,0463	0,0887	100	0,0268	0,0059	0,1461
Canned meals	0,0003	0,0004	100	1,2098	1,0268	1,4610	100	0,0797	0,0430	0,2480	100	0,1308	0,0472	0,3298

n = number of composite samples.
Number of samples/food = 10.
Vo: Vegetable oils; Mw: Mineral water; Ab: Alcoholic beverages; nAb: Non-alcoholic beverages; Meat: Meat and meat products; Cereal: Cereals, pulses, tuber, nuts and dried fruits; Pd: Prepared dishes; Sc: Sweeteners and condiments; Vf: Vegetables and fruits; Egg: Eggs; Milk: Milk and dairy products; Fish: Fish and seafood.

Table 2b
Levels of Pb, Cd, As, Cu, Cr and Sn in foods in mg kg^{-1} fresh mass.

FOODSTUFFS	Pb				Cd				tAs				iAs	
	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean
Sc (n = 50)	94	0,0958	0,0079	0,6320	40	0,0512	0,0069	0,1310	84	0,0216	0,0102	0,0370	98	0,0034
Chocolate and cacao	100	0,0597	0,0340	0,0910	100	0,0938	0,0630	0,1310	100	0,0224	0,0133	0,0320	90	0,0034
Sugar	80	0,0260	0,0211	0,0330	0				100	0,0184	0,0143	0,0225	100	0,0021
Salt	100	0,3310	0,1270	0,6320	0				100	0,0281	0,0220	0,0370	100	0,0040
Sweets	90	0,0271	0,0167	0,0440	0				20	0,0129	0,0102	0,0156	100	0,0030
Sauces and mayonnaise	100	0,0143	0,0079	0,0404	100	0,0085	0,0069	0,0100	100	0,0194	0,0170	0,0209	100	0,0045
Vf (n = 220)	77	0,0091	0,0010	0,0551	71	0,0094	0,0003	0,0706	89	0,0162	0,0011	0,1117	97	0,0042
Oranges	50	0,0016	0,0012	0,0020	0				50	0,0018	0,0012	0,0024	100	0,0008
Strawberries	90	0,0031	0,0020	0,0080	100	0,0025	0,0014	0,0042	100	0,0044	0,0027	0,0085	90	0,0028
Spinaches and swiss chard	100	0,0123	0,0022	0,0266	100	0,0389	0,0045	0,0603	100	0,0384	0,0101	0,0848	100	0,0076
Lettuces	60	0,0038	0,0010	0,0141	100	0,0098	0,0034	0,0562	100	0,0107	0,0038	0,0329	100	0,0023
Green beans	80	0,0028	0,0012	0,0079	100	0,0051	0,0029	0,0113	100	0,0058	0,0023	0,0088	100	0,0014
Onions	90	0,0025	0,0010	0,0060	100	0,0040	0,0022	0,0071	100	0,0077	0,0020	0,0108	100	0,0021
Garlic	60	0,0129	0,0053	0,0229	90	0,0088	0,0038	0,0181	100	0,0099	0,0029	0,0185	100	0,0031
Peppers	80	0,0056	0,0020	0,0196	100	0,0058	0,0042	0,0087	90	0,0179	0,0112	0,0711	100	0,0023
Aubergine, courgette and cucumber	90	0,0050	0,0020	0,0088	100	0,0065	0,0042	0,0117	100	0,0261	0,0112	0,0471	100	0,0200
Carrots and pumpkins	100	0,0067	0,0034	0,0118	100	0,0046	0,0023	0,0112	100	0,0179	0,0090	0,0272	100	0,0100
Tomatoes	0				100	0,0055	0,0017	0,0084	80	0,0056	0,0011	0,0105	90	0,0001
Olives and pickles	100	0,0427	0,0331	0,0551	50	0,0038	0,0024	0,0080	100	0,0360	0,0154	0,0493	100	0,0044
Apples and pears	80	0,0039	0,0025	0,0066	0				80	0,0032	0,0021	0,0071	90	0,0026
Sherry and plum	100	0,0029	0,0021	0,0043	0				30	0,0022	0,0020	0,0025	100	0,0016
Melon and watermelon	40	0,0119	0,0018	0,0402	70	0,0028	0,0017	0,0063	100	0,0096	0,0042	0,0173	100	0,0082
Bananas	100	0,0034	0,0022	0,0043	100	0,0005	0,0003	0,0006	100	0,0035	0,0028	0,0046	100	0,0006
Peach and apricot	100	0,0028	0,0015	0,0038	0				90	0,0026	0,0021	0,0041	100	0,0022
Grapes	100	0,0037	0,0025	0,0066	0				80	0,0026	0,0019	0,0034	80	0,0019
Cauliflower, cabbage and broccoli	20	0,0022	0,0017	0,0028	100	0,0049	0,0014	0,0136	90	0,0101	0,0015	0,0230	100	0,0039
Artichoke, leek, celery and chard	50	0,0039	0,0024	0,0065	100	0,0293	0,0092	0,0706	100	0,0362	0,0170	0,0560	100	0,0022
Mushrooms	100	0,0102	0,0013	0,0315	100	0,0123	0,0052	0,0219	100	0,0653	0,0304	0,1117	100	0,0083
Coffee and soluble coffee	100	0,0311	0,0167	0,0530	50	0,0099	0,0078	0,0106	80	0,0130	0,0102	0,0154	100	0,0028
Egg (n = 10)	90	0,0042	0,0023	0,0065	0				100	0,0054	0,0049	0,0058	100	0,0003
Milk (n = 60)	90	0,0109	0,0010	0,0474	20	0,0085	0,0025	0,0134	70	0,0158	0,0010	0,0691	92	0,0021
Milk	100	0,0022	0,0014	0,0035	10	0,0030	0,0030	0,0030	70	0,0048	0,0010	0,0015	100	0,0006
Cheese	100	0,0219	0,0057	0,0474	0				100	0,0379	0,0244	0,0691	100	0,0030
Yogurt	80	0,0029	0,0014	0,0076	0				80	0,0146	0,0128	0,0167	50	0,0008
Custards and smoothies	70	0,0064	0,0036	0,0112	10	0,0024	0,0024	0,0024	70	0,0168	0,0134	0,0197	70	0,0020
Butter	90	0,0201	0,0116	0,0350	0				0				100	0,0023
Soybean products	100	0,0098	0,0083	0,0131	100	0,0097	0,0077	0,0134	100	0,0041	0,0034	0,0048	100	0,0033
Fish (n = 120)	95	0,0349	0,0026	0,3494	68	0,0816	0,0018	0,5686	100	2,1669	0,3292	18,3130	99	0,0174
Canned fish	100	0,0090	0,0046	0,0157	100	0,0115	0,0077	0,0158	100	0,4622	0,3292	0,6356	100	0,0050
Tuna	90	0,0126	0,0054	0,0227	100	0,0117	0,0074	0,0195	100	1,3647	0,6461	3,1268	100	0,0131
Squid and cuttlefish	100	0,0144	0,0044	0,0287	100	0,1853	0,0018	0,5686	100	2,5796	0,5660	6,0025	100	0,0032
Seabream and seabass	90	0,0090	0,0041	0,0139	0				100	1,1293	0,7083	1,6824	100	0,0142
Swordfish	80	0,0107	0,0030	0,0315	100	0,0925	0,0385	0,1681	100	1,2501	0,9442	1,9732	100	0,0097
Shellfish	100	0,0243	0,0143	0,0365	100	0,0661	0,0182	0,1313	100	6,9377	3,1717	18,3130	100	0,0502
Mussels	100	0,2203	0,1017	0,3494	100	0,1967	0,1410	0,3880	100	2,1092	1,4601	3,0288	100	0,0270

(*continued on next page*)

Table 2b (*continued*)

FOODSTUFFS	Pb				Cd				tAs				iAs	
	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean
Whitefish	80	0,0055	0,0026	0,0107	0				100	2,8398	1,6578	4,6628	90	0,0066
Salmon and trout	100	0,0090	0,0037	0,0237	0				100	1,0682	0,5834	3,4174	100	0,0132
Sardine and anchovy	100	0,0380	0,0236	0,0667	100	0,0092	0,0062	0,0143	100	2,7810	1,8932	3,5512	100	0,0399
Salting fish	100	0,0343	0,0094	0,0911	100	0,0870	0,0183	0,1500	100	2,0989	1,1676	3,5119	100	0,0173
Smoked fish	100	0,0160	0,0121	0,0225	0				100	1,3822	0,6578	2,7079	100	0,0077

FOODSTUFFS	iAs		Cu				Cr				Sn			
	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max
Sc (n = 50)	0,0010	0,0091	100	2,9694	0,0310	17,5170	100	0,7925	0,0250	3,9340	34	1,8724	0,0119	25,6219
Chocolate and cacao	0,0021	0,0060	100	13,3355	10,6470	17,5170	100	1,6506	0,6670	3,9340	100	0,0215	0,0121	0,0370
Sugar	0,0010	0,0091	100	0,1192	0,0370	0,5340	100	0,0503	0,0250	0,1240	30	0,0172	0,0119	0,0229
Salt	0,0017	0,0062	100	0,4480	0,2200	0,5650	100	1,9861	1,8720	2,1280	0			
Sweets	0,0010	0,0061	100	0,3143	0,0310	0,4610	90	0,1258	0,0560	0,2300	100	0,0317	0,0122	0,1230
Sauces and mayonnaise	0,0034	0,0065	100	0,6302	0,5396	0,7107	100	0,0830	0,0603	0,1389	100	6,1207	0,0506	25,6219
Vf (n = 220)	0,0001	0,0457	100	1,1911	0,0847	12,5160	99	0,0598	0,0029	0,5477	34	0,0091	0,0003	0,1263
Oranges	0,0006	0,0020	100	0,3485	0,2976	0,3933	100	0,0470	0,0036	0,1179	90	0,0055	0,0012	0,0179
Strawberries	0,0002	0,0058	100	0,2380	0,1754	0,3027	100	0,0129	0,0062	0,0366	100	0,0155	0,0005	0,1157
Spinaches and swiss chard	0,0020	0,0245	100	0,9650	0,4558	1,6184	100	0,0677	0,0182	0,1335	0			
Lettuces	0,0004	0,0048	100	0,4353	0,1938	0,5747	100	0,0285	0,0157	0,0527	0			
Green beans	0,0006	0,0025	100	0,6377	0,3653	1,0027	100	0,0180	0,0072	0,0598	0			
Onions	0,0008	0,0031	100	0,4433	0,3597	0,5789	100	0,0121	0,0058	0,0203	0			
Garlic	0,0009	0,0059	100	1,4096	0,6423	2,3523	80	0,0310	0,0029	0,0674	0			
Peppers	0,0060	0,0037	100	0,6411	0,4286	0,8336	100	0,0119	0,0080	0,0204	0			
Aubergine, courgette and cucumber	0,0060	0,0457	100	0,6973	0,5637	1,0163	100	0,0178	0,0051	0,0475	0			
Carrots and pumpkins	0,0048	0,0149	100	0,5035	0,1982	0,8560	100	0,0136	0,0032	0,0373	0			
Tomatoes	0,0001	0,0003	100	0,4674	0,0847	0,8488	100	0,0189	0,0118	0,0306	0			
Olives and pickles	0,0014	0,0077	100	1,2533	0,9826	2,5611	100	0,3809	0,2791	0,5477	0			
Apples and pears	0,0011	0,0070	100	0,5808	0,3687	0,8363	100	0,0392	0,0257	0,0675	30	0,0057	0,0028	0,0111
Sherry and plum	0,0011	0,0026	100	0,6211	0,5373	0,6993	100	0,0325	0,0292	0,0358	10	0,0053	0,0053	0,0053
Melon and watermelon	0,0031	0,0168	100	0,3658	0,2793	0,4569	100	0,0219	0,0191	0,0288	40	0,0285	0,0021	0,1263
Bananas	0,0003	0,0021	100	0,6308	0,4537	0,7876	100	0,0549	0,0411	0,0811	100	0,0019	0,0003	0,0096
Peach and apricot	0,0015	0,0031	100	0,9204	0,5615	1,1520	100	0,0265	0,0217	0,0312	40	0,0055	0,0019	0,0122
Grapes	0,0003	0,0036	100	0,8825	0,7062	1,0108	100	0,0367	0,0271	0,0571	20	0,0044	0,0049	0,0356
Cauliflower, cabbage and broccoli	0,0022	0,0061	100	0,4658	0,1811	1,0885	100	0,0434	0,0081	0,0915	90	0,0072	0,0013	0,0212
Artichoke, leek, celery and chard	0,0010	0,0035	100	0,8906	0,6198	1,3820	100	0,0674	0,0456	0,1516	100	0,0084	0,0052	0,0099
Mushrooms	0,0028	0,0219	100	2,1921	1,2736	3,7229	100	0,0336	0,0259	0,0525	80	0,0021	0,0011	0,0051
Coffee and soluble coffee	0,0010	0,0071	100	10,6132	8,6050	12,5160	100	0,2928	0,2170	0,4260	50	0,0102	0,0112	0,0174
Egg (n = 10)	0,0003	0,0003	100	0,6583	0,6136	0,7360	100	0,0498	0,0407	0,0724	30	0,0389	0,0141	0,0556
Milk (n = 60)	0,0001	0,0083	95	0,4490	0,0126	2,0309	95	0,0575	0,0021	0,5213	48	0,0667	0,0012	0,5561
Milk	0,0004	0,0009	100	0,0467	0,0399	0,0612	100	0,0294	0,0133	0,0689	80	0,0031	0,0012	0,0143
Cheese	0,0005	0,0060	100	0,5764	0,5083	0,6875	100	0,0364	0,0247	0,0515	30	0,0325	0,0058	0,0664
Yogurt	0,0001	0,0013	100	0,0629	0,0455	0,1100	90	0,0858	0,0021	0,5213	50	0,0052	0,0020	0,0102
Custards and smoothies	0,0012	0,0044	100	0,1622	0,0147	0,3764	100	0,1435	0,0396	0,3057	30	0,0048	0,0038	0,0053
Butter	0,0010	0,0083	70	0,0649	0,0126	0,1890	80	0,0300	0,0172	0,0470	0			
Soybean products	0,0012	0,0082	100	1,6658	1,3139	2,0309	100	0,0172	0,0092	0,0289	100	0,1771	0,0021	0,5561
Fish (n = 120)	0,0002	0,1133	100	1,1390	0,1351	9,2838	98	0,1016	0,0003	0,6737	38	0,0642	0,0044	0,4089
Canned fish	0,0020	0,0108	100	0,3384	0,2545	0,4142	100	0,1387	0,1010	0,2144	100	0,0535	0,0285	0,1050

(*continued on next page*)

Table 2b (*continued*)

FOODSTUFFS	iAs		Cu				Cr				Sn			
	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max	> LOQ (%)	Mean	Min	Max
Tuna	0,0048	0,0325	100	0,5222	0,3854	0,9429	100	0,0607	0,0196	0,1371	70	0,0195	0,0147	0,0218
Squid and cuttlefish	0,0002	0,0086	100	1,6793	0,3029	3,0941	100	0,0457	0,0216	0,0747	20	0,0152	0,0126	0,0177
Seabream and seabass	0,0068	0,0307	100	0,4134	0,3448	0,4873	80	0,0341	0,0038	0,1259	60	0,0113	0,0055	0,0170
Swordfish	0,0041	0,0140	100	0,3749	0,3017	0,5059	100	0,0342	0,0108	0,0848	40	0,0106	0,0048	0,0184
Shellfish	0,0087	0,1133	100	4,9004	2,8326	9,2838	100	0,0462	0,0110	0,1071	40	0,0155	0,0061	0,0240
Mussels	0,0165	0,0426	100	1,0733	0,8318	1,4518	100	0,1077	0,0243	0,2999	90	0,0360	0,0070	0,0739
Whitefish	0,0019	0,0089	100	0,1954	0,1351	0,3047	100	0,0427	0,0207	0,0639	0			
Salmon and trout	0,0065	0,0403	100	0,3715	0,2372	0,5223	100	0,0213	0,0057	0,0419	30	0,0180	0,0082	0,0257
Sardine and anchovy	0,0287	0,0609	100	1,2284	1,0126	1,5555	100	0,0156	0,0003	0,0475	100	0,0240	0,0044	0,1081
Salting fish	0,0112	0,0216	100	1,8517	1,2890	2,5773	100	0,3803	0,1509	0,6737	100	0,0418	0,0301	0,0493
Smoked fish	0,0046	0,0106	100	0,7197	0,4697	0,8352	100	0,2784	0,1802	0,3953	100	0,2915	0,1828	0,4089

n = number of composite samples.
Number of samples/food = 10.
Vo: Vegetable oils; Mw: Mineral water; Ab: Alcoholic beverages; nAb: Non-alcoholic beverages; Meat: Meat and meat products; Cereal: Cereals, pulses, tuber, nuts and dried fruits; Pd: Prepared dishes; Sc: Sweeteners and condiments; Vf: Vegetables and fruits; Egg: Eggs; Milk: Milk and dairy products; Fish: Fish and seafood.
Number of samples/food = 10.

and condiments” (0.7925 mg kg^{-1}), due to the contribution of salt and cacao with average values of 1.986 mg kg^{-1} and 1.650 mg kg^{-1}, respectively (Table 2a). The “meat and meat product” had a Cr average level of 0.2384 mg kg^{-1}, ranging from 0.0188 mg kg^{-1} in viscera to 222.70 μg kg^{-1} in beef (Table 2a).

The lowest levels were detected in alcoholic and non-alcoholic drinks with average values of 10.66 μg kg^{-1} and 23.90 μg kg^{-1}, respectively (Table 2a).

3.8. Tin

Of the 810 samples analysed, 53% contained Sn at levels higher than the LoQ (Tables 2a and 2b and Fig. 1). The “sweeteners and condiments” group presented the highest Sn levels, with an average of 1.8725 mg kg^{-1}, mainly due to the contribution of sauces and mayonnaise in which an average of 6.1207 mg kg^{-1} and a maximum value of 25.6219 mg kg^{-1} were obtained (Table 2a).

The “Meat and meat products” group had Sn values from 0.0006 to 0.687 mg kg^{-1} for the cured sausage and pork products, respectively (Table 2a).

4. Discussion

Heavy metals are ubiquitous and chemically stable, so they can be expected to be present in all parts of the biotic and abiotic matter. Therefore, metals included in this study were analysed in all the food groups. Nevertheless, tHg and meHg were analysed only in fish because it is currently considered that consumption of fish is the main path for human exposure to mercury (Hg) [22].

In the following sections the concentration levels found for each metal are discussed.

It should be noticed that every dietary exposure assessment is affected by scientific uncertainties or scientific knowledge limitations. These are important for the correct interpretation of the results. First of all, the effect of cooking or processing was not taken into account for the calculation of the metal levels in the different products studied. Secondly, when samples were analyzed as composites, it is usual to find concentrations levels below the regulated levels because they correspond to mean levels. There are many uncertainties associated with the analytical methods, including sample representativeness or the use of different analytical limits if data are censored, and also with the methodology such as the different composition of samples or food groups and the different origin of the products. Tables 3a–3c show comparative data of levels of metals between the present study and different TDS carried out in various countries.

4.1. Lead

Regarding the Pb levels reported in this study, in the “Fish “group, the high average level presented for bivalve molluscs and crustaceans can be explained because they are filter feeders that accumulate metals from aquatic environment regardless of environmental pollution although contaminated water can also increase their metal content [23]. The average lead level in mussels obtained in the present study, was similar to that found in previous studies carried out in Valencia in 2005 and 2006 (0.220 mg kg^{-1}) [17], or in the 2nd French TDS (0.268 mg kg^{-1}) [11]. Although Rose et al. [12] reported high levels of Pb in offal (0.065 mg kg^{-1}) (see Table 3a), in the present study the values obtained for offal were not particularly high (0.0333 mg kg^{-1}). This difference could be explained by the different species included in both studies. In some foods such as the cured sausage, snacks, olives and pickels, the high average Pb levels found can be related with their high salt content, the food product with the highest Pb level.

The Pb average levels obtained in this study have been compared with other studies. As can be seen in Table 3a, the Pb levels found in this study are, in general, higher than those found in other countries as

Table 2c
Levels (mean) of tHg and meHg in Fish and seafood in mg kg^{-1} fresh mass.

Fish and seafood (n = 120)	tHg				meHg			
	> LOQ (%)	Mean	Min	Max	> LOQ%	Mean	Min	Max
	100	0,2515	0,0032	2,2874	100	0,1604	0,0015	1,7285
Canned fish	100	0,2165	0,1275	0,3691	100	0,1689	0,0973	0,2500
Tuna	100	0,9395	0,4409	1,6155	100	0,7212	0,2476	1,7285
Squid and cuttlefish	100	0,0240	0,0103	0,0550	100	0,0083	0,0015	0,0256
Sea bream and sea bass	100	0,0700	0,0433	0,0974	100	0,0119	0,0035	0,0219
Swordfish	100	1,4448	1,0851	2,2874	100	0,8186	0,6266	1,0791
Shellfish	100	0,0441	0,0090	0,0978	100	0,0129	0,0025	0,0251
Mussels	100	0,0070	0,0032	0,0132	100	0,0055	0,0033	0,0109
Whitefish,	100	0,0802	0,0207	0,1711	100	0,0487	0,0203	0,1147
Salmon and trout	100	0,0203	0,0117	0,0354	100	0,0176	0,0041	0,0386
Sardine and anchovy	100	0,0339	0,0107	0,0595	100	0,0275	0,0034	0,0558
Salting fish	100	0,1206	0,0466	0,2002	100	0,0787	0,0405	0,1162
Smoked fish	100	0,0165	0,0124	0,0225	100	0,0052	0,0044	0,0091

n = number of composite samples.
Number of samples/food = 10.

well as those reported previously in other regions of Spain [24]. Nevertheless, the levels observed in this study were similar to or lower than those observed in the data provided by EFSA [25], with mean lead levels between 0.0003 mg kg^{-1} for infant follow-on formulae to 4.3 mg kg^{-1} for dietetic products with an overall median across all categories of 0.021 mg kg^{-1}. In the present study infant follow-on formulae were not included because the study included subjects between 16 and 95 years of age. On the other hand, as none of the subjects interviewed reported the consumption of dietetic products, they were not selected in this study, because it was assumed that this product was not consumed in this region.

4.2. Cadmium

The high Cd levels obtained in the "fish" group mainly for mussels and squid are statistically similar (p-value 0.21) to those reported in France where crustaceans and molluscs had an average Cd of 0.1666 mg kg^{-1} [26] but statistically lower (p-value 0.02) than those reported by Korea, with Cd levels in molluscs of 0.677 mg kg^{-1} [27]. This difference could be explained by the different metal distribution in the fishery areas. In samples of fishery products collected in markets of Valencia in 2005–2006 average values obtained in molluscs (mussels) were 0.170 mg kg^{-1}; in cephalopods 0.230 mg kg^{-1} in squid and 0.140 mg kg^{-1} in cuttlefish [17]. The lowest value (0.0070 mg $kg^{-1)}$ found in fish were obtained in Lebanon [28] and in a market-basket study conducted in Sweden with values of 0.006 mg kg^{-1} [29] (see Table 3a). The high Cd values found in chocolate and cocoa could be explained by the naturally high Cd content in the soils of some regions in cocoa-producing countries. Millour, in France, obtained values for dark chocolate of 0.076 mg kg^{-1} [11] (see Table 3a). On the other hand, the Cd levels in offal from the present study were higher than those found in viscera in some studies such as those conducted in the UK (0.084 mg kg^{-1}; [12]), and in Santiago de Chile (0.079 mg kg^{-1}; [13]). In the 2nd TDS carried out in France, the levels reported in offal were indeed lower (0.020 mg kg^{-1}; [11]) (see Table 3a).

In general, similar results than those found in this study were reported by EFSA [30], with arround half of the food samples with levels below the limit of quantification and an overall median across all categories of 0. 1 mg kg^{-1}. Furthermore, similar results to those reported in this study were obtained for the food products molluscs (0.132 mg kg^{-1}) or chocolate (0.081 mg kg^{-1}). Although in the EFSA report algal supplements and seaweeds used as a vegetable had the highest average cadmium levels, this products were not included in this study because it was assumed that these products were not consumed in this region.

4.3. Total arsenic

The percentage of samples with tAs levels over the LoQ (87%) in the present study is higher than those reported by EFSA [31] among the EU members reported results (44%) or in the 2nd French total diet study (65%) [11], fact that reflects the effort made for decreasing the LoQ values (0.0004–0.010 mg kg^{-1}).

The iAs levels found in the "fish" group (average 2.1669 mg kg^{-1}) are in the range of average tAs values in fish reported in different studies such as 1.351 mg kg^{-1} in the Santiago de Chile TDS [13] or 3.990 mg kg^{-1} in the total diet study conducted in France (see Table 3a). In particular, shellfish tAs average values (6.9377 mg kg^{-1}) were consistent with the results from other studies carried out in Belgium [32] and Spain [14]. Data collected by EFSA from 19 EU countries, showed statistically similar average values (p-value 0.35) for fishery products (2.3837 mg kg^{-1}, UB) than those reported in the present study and the highest values were found also in crustaceans 5.691 mg kg^{-1}, cephalopods 3.923 mg kg^{-1} and molluscs 3.4078 mg kg^{-1} [4].

It is well known that rice and rice-based products could present high arsenic levels. This fact has been also confirmed in the present study, in which rice presented the highest level in the "cereal" group. Nevertheless, these values of tAs in rice, were lower than those detected in Catalonia, Spain, where the average value of tAs in rice was higher 0.18 mg kg^{-1} [24] and much lower than those found in Canada, with an average value of 1.240 mg kg^{-1} [32] (see Table 3a) but higher than those detected in the first TDS of France (0.016 mg kg^{-1} for white rice) [11]. This fact demonstrating the effectiveness of the global measures to reduce the environmental pollution.

4.4. Inorganic arsenic

Again, the percentage of samples with iAs levels over the LoQ (91%) in the present study is higher than those reported by EFSA [31] among the EU members reported results (68%), reflecting the effort made for decreasing the LoQ values (0.0001–0.0049 mg kg^{-1}).

The levels of iAs found in the "fish and fishery products" group were statistically similar (p-value 0.23) to those reported by EFSA of 1012 samples of fish and fishery products (0.0256 mg kg^{-1}) from 21 EU countries [31]. The iAs average level in mussel are agree with the study carried out with samples of foods purchased in Belgian markets, in which iAs was only detected in mussels and prawns with average values in a range of 0.005 to 0.022 mg kg^{-1} fresh weight (Ruttens, et al., 2012).

The high iAs level found in rice support some studies that suggest that rice and rice-based products could also contribute significantly to

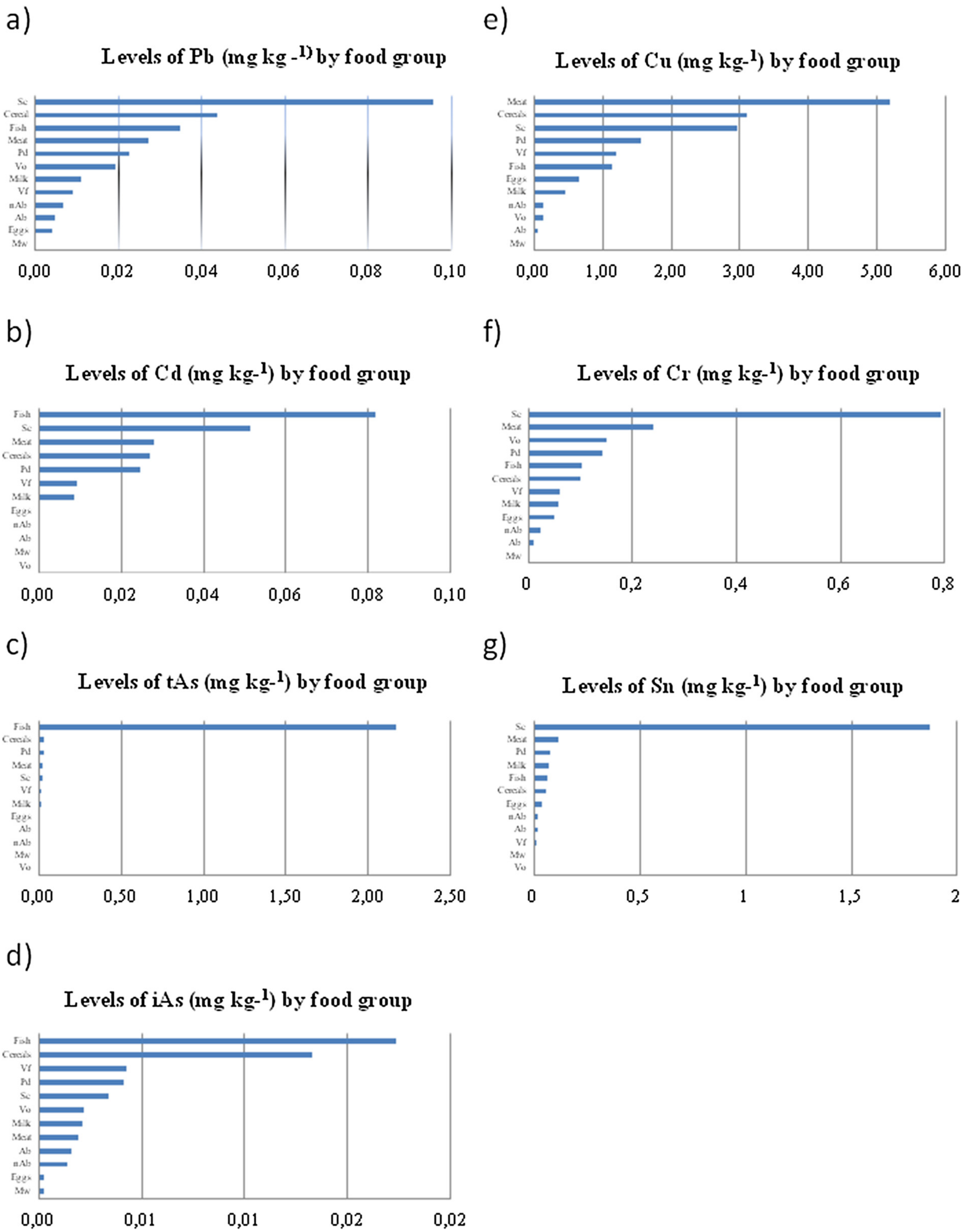

Fig. 1. Mean levels by food groups of a) Pb, b) Cd, c) tAs, d) iAs, e) Cu, f) Cr and e) Sn (mg kg^{-1}).

inorganic arsenic. In addition, similar data were reported by EFSA, mean value of 0.101 mg kg^{-1} for rice [31].

Although the speciation of As in the context of risk assessment is of great relevance, most of the studies determine the iAs content inferred from tAs by the use of conversion factors. Nevertheless, few studies such as a TDS carried out in the UK reported levels. In the aforementioned study, iAs levels were below the LOQ for most of the food groups and was only detected in cereals and fish [12]. On the other hand, in a TDS carried out in Hong Kong, the iAs detection frequency was 51%. The levels found by food group are presented in Table 3a.

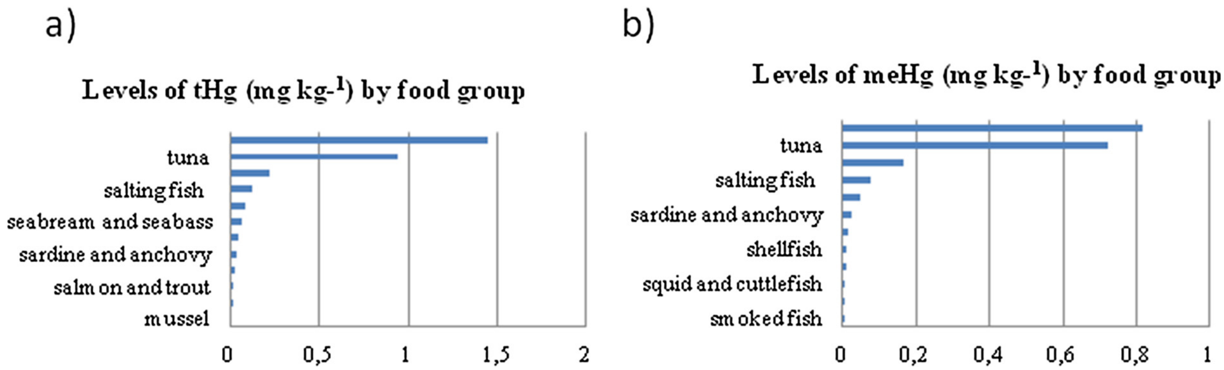

Fig. 2. Mean levels of a) tHg and b) meHg (mg kg^{-1}) in fish and seafood products. Number of composite samples per food = 1.

Spain lacks information on iAs content in foods. The data presented here are of great interest and contribute to the recommendation issued by the European Commission on the need to provide iAs content in food for regulatory purposes.

4.5. Mercury

Table 3b shows the comparison with other studies. As can be seen, the concentrations reported in most studies were lower than those found in the present study but, the same as in the present study, the highest levels were found in fish, specifically in tuna or swordfish and the lowest contents were reported in shellfish. In the European context, in France and UK values of 0.045 mg kg^{-1} [11] and 0.056 mg kg^{-1} [12], respectively, were obtained. In the 2nd French TDS fish had the highest Hg concentrations with an average value of 0.065 mg kg^{-1} [11]. Nevertheless, the highest average values (0.476 mg kg^{-1}) were found in tuna, whith a maximum value of 0.702 mg kg^{-1}. The lowest tHg values in food were obtained in the TDS carried out in Santiago (Chile) (0.048 mg kg^{-1}) [13]. In Asia, tHg values reported were relatively low, ranging from 0.0119 mg kg^{-1} in fish in Cambodia [35] to 0.770 mg kg^{-1} in swordfish in Taiwan [36]. In the New Zealand TDS, the highest values were detected in fish paste (0.195 mg kg^{-1} and 0.2655 mg kg^{-1}, lower-bound (LB) and upper-bound (UB) respectively), followed by fresh fish (0.1376 mg kg^{-1} and 0.0893 mg kg^{-1}, LB and UB respectively) [37]. Finally, in the Canadian TDS the highest value of tHg was observed in swordfish, with an average value of 1.820 mg kg^{-1}.

Hg levels found in the study were in good agreement with values reported by other authors in studies conducted in countries from the Mediterranean coast. In Italy detected Hg levels were in the range of 0.430–1.140 mg kg^{-1} for the five most consumed fish species [38]. In Catalonia (Spain), Perelló obtained the highest concentrations of Hg in fish, with an average of 0.22 mg kg^{-1} [24], which dropped in relation with values from a previous study, in which an average of 0.247 mg kg^{-1} was reported [14]. In Madrid (Spain), average values of 0.990 mg kg^{-1} for luvar and 0.930 mg kg^{-1} for sworthfish were obtained [39]. In Andalucia (Spain) an average value of 0.540 mg kg^{-1} for swordfish and 0.470 mg kg^{-1} for tuna [40] were found and in Valencia (Spain) values of 0.7666 mg kg^{-1} for swordfish and 0.666 mg kg^{-1} for tuna were reported [41]. And finally, in a study carried out in Canarias, tHg average levels in fish of 0.1189 mg kg^{-1} [16] were obtained.

In the present study meHg represents 60.2 ± 30.6% of tHg, varying by species between 18.1 ± 11.1% for sea bream and bass to 82.8 ± 30.2% for mussels. The contribution of meHg to tHg in swordfish was 58 ± 9.6% and in tuna was 73.4 ± 14.4%. Similar relations were reported in other studies, obtaining values ranging from 50% to 100% depending on the species in Hong Kong (Wang et al., 2013); 68% in China [42] and 38,16%, 74,6% y 91,2% in three different Cambodia regions [35]. According to WHO, the proportion of meHg contributing to tHg is between 30–100%, depending on the species, size, age and diet of the fish [43].

4.6. Copper

The average values of Cu found in the present study in meat (5.1891 mg kg^{-1}) were in the range of those in different studies conducted in Sweden [29] and Canada [33], respectively (see Table 3c), in which values from 0.740 to 10.723 mg kg^{-1} were reported. The average level of Cu found in offal (50.4074 mg kg^{-1}) was similar to the maximum reported levels in the UK (52.5000 mg kg^{-1}) [44]. On the other hand, the maximum level found in offal in the present study (100.8016 mg kg^{-1}) was also similar to those found in a TDS carried out in France (113.0000 mg kg^{-1}) [11] or Canada (127.687 mg kg^{-1}) [33].

Regarding "Cereals" group, the average Cu level (3.0958 mg kg^{-1}) in the present study was also statistically similar (p-value 0.18) to those in UK [44] in which values of 2.210 mg kg^{-1} in cereals and 9.150 mg kg^{-1} in dried fruits were obtained.

Finally, the Cu levels found in chocolate and cocoa (average of 13.3355 mg kg^{-1}) were higher than those reported in the 2nd French TDS in chocolate (average of 6.430 mg kg^{-1}) [11]. This fact could be explained by the different origin of the cocoa. In addition, in the 2nd French TDS, the main contributor to the Cu intake was group "Sweeteners, honey and confectionery".

4.7. Chromium

The Cr levels found in the food group "sweeteners and condiments" (0.7925 mg kg^{-1}) in the present study were statistically lower (p-value 0.03) than those obtained also for sweeteners in a total diet study from Brazil [43] but statistically similar (p-value 0.13) than those reported in France [11], with average values of 0.799 mg kg^{-1} and 0.574 mg kg^{-1}, respectively (see Table 3c). On the other hand, the Cr average level found in "meat and meat product" food group (0.2384 mg kg^{-1}) was also statistically similar (p-value 0.15) to the values obtained in France (0.299 mg kg^{-1}) [11].

The lowest levels were detected in alcoholic and non-alcoholic drinks with average values of 10.66 μg kg^{-1} and 23.90 μg kg^{-1}, respectively (Table 2a).

The highest values of Cr in food were reported in the total diet study of Catalonia, with average values ranging from 0.272 to 1.500 mg kg^{-1}

Table 3a
Comparative data of levels (mean) (mg kg^{-1}) to Pb, Cd and As, from TDS in different countries.

Food Groups	Australia[a] 2008 [47]	UK [b]2006 [12]	France[b] 2007-2009 [11]	Lebanon[c] 2008 [28]	Sweden 1999 [29]	Canada[b] 2007 [33]	Catalonia[c] 2012* [15]	Hong-Kong 1ºTDS[c] [34]	Present study Valencia 2010
Pb									
Vo	(0.0017-0.0046)	< 0.006 (LOD) ***	0.004		< 0.006 ***	0.0020	0.004 ***		0.0192
Mw	(0.0030-0.0031)		0.004	< 0.0005		0.0003			ND
Ab	(0.0062-0.0063)		0.010		0.018	0.0050			0.0048
nAb	(0.0013-0.0017)	< 0.001 (LOQ)	0.007		< 0.001 **	0.00034			0.0068
Meat	0.0091	< 0.005 (LOQ)(m. p.);0.065 offal	0.011	0.0030	0.004	0.0041	0.011		0.0273
Cereal	(0.0086-0.0088)	< 0.007(LOQ)(ce)	0.009 (c. p.)	0.0080 (c.p.)	0.009(c. p.)	0.0047	0.010(ce); 0.011 (pu); 0.014(pu);0.012 (ib).		0.0438
Pd	0.0321		0.008			0.0049			0.0225
Sc	(0.0203-0.0212)	< 0.006(LOD) (swee)	0.017 (swee);0.014 (cond)		0.007(swee)	0.0418			0.0958
Fv	(0.0064-0.0067)	0.004 (veg)-< 0.002(LOQ)(fr)	0.009	0.01643(veg);0.0010(fr)	< 0.003(veg);0.007(fr)	0.0053	0.006(veg); 0.004(fr)		0.0091
Eggs	(0.0019-0.0022)	0.003	0.006		< 0.004	< 0.0009	0.002		0.0042
Milk	(0.0023-0.0030)	< 0.003(LOD)(d. p.)	0.007	< 0.002(milk);0.0005(d. p.)	< 0.002	0.0025	< 0.002(milk); 0.01º(dp)		0.0109
Fish	0.0057	< 0.004(LOQ)	0.050	0.0061	0.006	0.0036	0.028		0.0349
Cd									
Vo	(0.009-0.0123)	< 0.005(LOD) (***)	0.001		< 0.003***	0.0131	< 0.002 ***		ND
Mw	(0.0001-0.0002)		0.001	< 0.0005		0.00002			ND
Ab	(0.0014-0.0015)		0.001		< 0.001	0.0002			ND
nAb	(0.0009-0.001)	< 0.001(LOD)	0.002		< 0.0004**	< 0.00004			ND
Meat	(0.0020-0.0025)	< 0.007(LOQ)(m. p.);0.084 offal	0.007	0.0058	0.002	0.0040	0.001		0.0281
Cereal	(0.00152-0.0156)	0.021(c. p.);0.065 (d.f.)	0.024 (c. p.)	0.0151(c. p.)	0.024 (c. p.)	0.0302	0.015(ce, pu); 0.001(pu); 0.010(ib);		0.0271
Pd	0.0079		0.012			0.0049			0.0246
Sc	(0.0122-0.0137)	< 0.006(LOQ) (swee)	0.021 (swee);0.017 (cond)		0.007(swee)	0.0079			0.0512
Fv	(0.0049-0.0052)	0.006 (veg)-< 0.001(LOD)(fr)	0.012	0.0302(veg);0.0063(fr)	0.007(veg); < 0.001(fr)	0.0155	0.006(veg);0.002 (fr)		0.0094
Eggs	(00002-0.0011)		0.001		< 0.002	0.0003	< 0.002		ND
Milk	(0.0007-0.0018)	< 0.003(LOD)(d. p.)	0.002	< 0.002(milk);0.0031(d.p.)	< 0.001	0.0010	< 0.002		0.0085
Fish	0.0088	0.015	0.055	0.0070	0.006	0.0032	0.050		0.0816
tAs									
Vo	(0.0005-0.0233)	< 0.005(LOD) ***	0.015			0.0675	< 0.002***		ND
Mw	(0.0002-0.0006)		0.010			0.0004			ND
Ab	(0.008-0.009)		0.009			0.0052			0.0035
nAb	(0-0.0025)	< 0.001(LOD)	0.012			0.0004			0.0023

(continued on next page)

Table 3a (*continued*)

Food Groups	Australia[a] 2008 [47]	UK [b]2006 [12]	France[b] 2007-2009 [11]	Lebanon[c] 2008 [28]	Sweden 1999 [29]	Canada[b] 2007 [33]	Catalonia[c] 2012* [15]	Hong-Kong 1ºTDS[c] [34]	Present study Valencia 2010
Meat	(0.019-0.020)	0.022 chicken	0.026			0.0065	0.001		0.0233
Cereal	(0.0253-0.0354)	< 0.018(c. p.)	0.021(c. p.)			0.0101	0.045(ce); 0.002(pu); < 0.002(tu); 0.013(i.b)		0.0304
Pd	0.0245		0.030			0.0130			0.0300
Sc	(0.0115-0.0216)	< 0.009(LOQ) (swee)	0.035 (swee);0.068(cond)			0.0255			0.0216
Fv	(0.0065-0.011)	0.004 (veg); < 0.001(LOQ)(fr)	0.013			0.0045	0.001		0.0162
Eggs	(0.011 -0.012)	< 0.003(LOQ)	0.015			0.0036	< 0.002		0.0054
Milk	(0.0022-0.0108)	< 0.003(LOD)(d. p.)	0.016			0.0060	< 0.002		0.0158
Fish	1.800	3.990	1.920			2.285	3.2		2.167
iAs									
Vo		< 0.01					< 0.002	0.0015	0.0040
Mw								0.0008	0.0005
Ab								0.0035	0.0027
nAb		< 0.01						0.0017	0.0035
Meat		< 0.01					< 0.002	0.0042	0.0124
Cereal		< 0.01					0.007(ce); 0.001(pu); < 0.002(tu); 0.011(ib)	0.0072	0.1048
Pd								0.0073	0.0159
Sc		< 0.01						0.0032 (swee);0.009 (cond)	0.0091
Fv		< 0.01					0.001	0.009(veg);0.0078(fr)	0.0457
Eggs		< 0.01					< 0.002	0.034	0.0003
Milk		< 0.01					< 0.002	0.0015	0.0003
Fish	(0-0.05)	0.015					0.017	0.0015	0.1133

ND: not detected; f: fish; s: shellfish; c:crustaceans; m: molluscs; veg: vegetables; fr: fruits; m. p.: meat products; c. p.: cereal products; ce: cereals; pu: pulses; tu:tubers; ib:industrial bakery; d. p.: dairy products; d.f.: dried fruits; swee: sweeteners; cond: condiments; [a]in braquets,own value calculated from the data of the author (LB-UB;); [b](UB); [c](MB); *median** include soft drinks,light beer and mineral water; *** include animal fats.

Table 3b
Comparative data of levels (mean) (mg kg^{-1}) to Hg in TDSs in different countries.

Countries	Fish and seafood (n = 120)	
	meHg	tHg
Australia[*] 2008[a] [47]		0.8725
N. Zealand[*] 2009[b] [37]		0.09053
Canada[*] 2002[c] [48]		0.26909
Santiago (Chile)[c] 2001–2002 [13]		0.048
UK 2006[c] [12]		0.056
France 2007–2009[c] [11]		0.045
Korea 2009 [27]		0.234(f);0.0285 (s. and c.);0.051(m)
China 2007 [42]	0.01254	0.01848
Kampong chan [35]	0.0227	0.0249
Kratie [35]	0.0603	0.158
Kandal [35]	0.0089	0.0119
Catalonia 2012[b] [15]	0.17	0.22
Present study Valencia 2010	0.16042	0.25145

f:fish; s:sellfish; c: crustaceans: m: molluscs.
* own value calculated from the data of the author.
[a] (LB-UB).
[b] (MB).
[c] (UB).

for the oils and fats and the fruits groups, respectively [15]. Conversely, in the UK study [12] most values were below the LOQ/LOD (0.003–0.020 mg kg^{-1}) and the detected values were in a range from 0.020 to 0.080 mg kg^{-1} for the oils and fats and the sugar and preserves groups, respectively (Table 3c).

4.8. Tin

The percentage of samples with Sn levels over the LoQ (53%) in the present study is lower than those reported by in the 2nd French total diet study (74%) [46], maybe because the different food products included in both studies.

Although high concentrations of tin in foods were found in tinned fruit and vegetables, in some multi-vitamin and mineral food supplements (levels up to 10 µg tin/tablet) (EGVM, 2002) or in “compotes and stewed fruits” [46], in this study these kind of products were not included in this study because it was assumed that these products were not consumed in this region.

Although in the 2nd French total diet study [46] high contents of tin were also found in the “sweeteners, honey and confectionery groups” (0.238 mg kg^{-1}), those are statistically lower (p-value 0.01) than the tin contents found in the present study for the Sc group. Nevertheless, it should be taken into account that in the French study high tin levels were also observed in some sauces such as tomato sauce (5.99 mg kg^{-1}), included in other different group called “condiments and sauces”, but in the Sc in the present study. Therefore, in the TDS, the conclusions should be interpreted with caution, because the food groups could include different food items.

Most of the total diet studies have not studied the levels of tin in food. Only the 2nd French TDS, a study carried out in UK (Rose, et al., 2010) and the 20th TDS in Australia reported Sn values in food with values in all food groups close to the LoQ value except for canned foods such as canned vegetables, canned fruits, canned tuna and baked (see Table 3c).

5. Conclusions

The results of this study indicate that the estimated levels of Pb and Cd in foodstuffs were, on the whole, satisfactory compared with the maximum levels set by European regulations. However, in the case of Hg, all swordfish samples (100%) and three samples of tuna (30%) out of the 10 composite samples analyzed of each foodstuffs, exceeded the limits established by law.

The fish group presented the highest Cd, Hg and As levels, whereas Sc was the most contaminated food group by Pb, Cr and Sn, mainly due to salt and the meat group had the highest levels of Cu. In the mineral water group only As was quantified and in the vegetable oils group, both Cu and Cr were detected.

The results of this study are generally similar to or lower than those observed in other TDSs conducted in other countries, except in the case of Hg, for which high values were obtained, mainly in swordfish. This survey confirms a decreasing tendency when compared with other studies carried out in Spain.

As has been mentioned in the discussion part, some scientific uncertainties should be taken into account for comparisons. First of all, the effect of cooking was not taken into account for the calculation of the metal levels in the different products studied and secondly, the samples were analyzed as composites, therefore concentrations found correspond to mean levels.

Heavy metals are related with some toxic effects, such as fish deformities [51]. For this reason, the contamination data has been compared with own-food consumption data, to estimate the exposure of the population of Valencia [18]. The results show that a percentage of population could be at risk, especially young children. This highlights the difficulties inherent to establishing maximum levels of metals in Europe, taking into account the different dietary patterns in the various countries, and the technological and market aspects involved.

For certain metals (e.g., Hg, As, Cr and Sn), speciation has become an essential tool that provides information on the chemical form present in the samples, which is crucial for accurately assessing toxicity. Therefore, it is important in future studies to obtain speciation data for Cr and Sn, not included in the present study.

Disclosure statement

No potential conflict of interest was reported by the authors.

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Transparency document

The Transparency document associated with this article can be found in the online version.

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Table 3c
Comparative data of levels (mg kg^{-1}) to Cu, Cr and Sn, from TDS in different countries.

Food Groups	Australia[a] 2008 [47]	UK[b] 2006 [12]	France[b] 2007-2009 [49] ****	Lebanon[c] 2008 [28]	Sweden 1999 [29]	Canada[b] 2007 [33]	Brazil[c] [45]	Catalonia[c] *2008 [50]	Present study Valencia 2010
Cu									
Vo	(2.1161-2.1213)	< 0.080 (LOQ) ***	0.095		< 0.150 ***	1.0051		0.123 ***	0.1243
Mw	(0.1757-0.182)	0.074	0.095	< 0.0005		0.0561			ND
Ab	(0.1587-0.163)		0.121		0.046	0.0581			0.0485
nAb	(0.0059-0.018)		0.207		0.018 **	0.0118			0.1404
Meat	0.8511	1.160 (m. p.); 52.500 (offal)	9.450	164.441	0.740	10.7237		0.949	5.1891
Cereal	1.5444	2.210 (c. p.)	1.460 (c. p.)	1.50385 (c. p.)	1.900 (c. p.)	1.7203		2.160 (ce)	3.0958
Pd	1.130		0.689			0.8993			1.5627
Sc	(0.9917-0.9948)	1.800 (swee)	3.690 (swee); 0.614 (cond)		1.820 (swee)	0.9514			2.9694
Fv	1.3163	0.580 (veg); 0.786 (fr)	0.819	0.48808 (veg); 0.44387 (fr)	0.670 (veg); 0.090 (fr)	0.5913		0.943 (veg); 0.787 (fr)	1.1911
Eggs	0.610	0.570	0.734		0.630	0.602		1.950	0.6583
Milk	(0.233-0.2348)	0.330 (d. p.)	0.146	0.178 (milk); 0.19164 (d.p.)	0.096	0.1988		0.489 (d.p.)	0.4490
Fish	2.6275	0.910	3.110	0.25467	0.730	0.6015		1.280	1.1390
Cr									
Vo	(0.067-0.1933)	0.020 ***	0.1000		< 0.009 ***		< 0.020 (LOD)	1.500 ***	0.1499
Mw	(0.0004-0.0017)	< 0.003 (LOD)	0.019						0.0000
Ab	(0.0135-0.0197)		0.078		0.017		0.012		0.0107
nAb	(0.0091-0.015)		0.102		< 0.001**		< 0.0011 (LOD)		0.0239
Meat	(0.0963-0.1036)	0.037 (m. p.)	0.299		0.019		0.060 (poultry); 0.117 (pork); 0.056 (beef)	0.870	0.2384
Cereal	(0.0598-0.1127)	< 0.030 (LOQ) (c. p.)	0.286 (c. p.)		0.021 (c. p.)		0.009 (ce); 0.225 (bread)	1.020 (cereals)	0.0992
Pd	(0.0715-0.083)		0.251				0.025		0.1457
Sc	(0.0867-0.1449)	0.080 (swee)	0.574 (swee); 0.345 (cond)		0.100 (swee)		0.799 (swee); < 0.020 (LOD) (salt)		0.7925
Fv	(0.0325-0.0515)	< 0.008 (LOQ) (veg); < 0.007 (LOQ)(fr)	0.119		< 0.005 (veg);0.011 (fr)		0.048 (veg); 0.016 (fr)	0.162 (veg); 0.226 (fr)	0.0598
Eggs	(0.030-0.043)	0.010	0.220		< 0.005			1.150	0.0498
Milk	(0.0367-0.0732)	< 0.010 (LOQ) (d. p.)	0.173		< 0.003		0.024 (d. p.)	0.748 (d.p.); 0.272 (m)	0.0575
Fish	(0.0603-0.075)	0.040	0.272		0.025		0.025	0.784	0.1016
Sn									
Vo		< 0.020 (LOD) ***							0.0000
Mw		< 0.003 (LOD)							0.0000
Ab									0.0178
nAb									0.01841
Meat		0.040 (m. p.)							0.1151
Cereal		< 0.020 (LOD) (c. p.)							0.0551
Pd									0.0776
Sc		< 0.020 (LOD) (swee)							1.8724
Fv		< 0.003 (LOD) (veg); < 0.005 (LOQ) (fr)							0.0091
eggs		< 0.010 (LOD)							0.0389
Milk		< 0.020 (LOQ) (d. p.)							0.0667
Fish		< 0.021 (LOQ)							0.0642

ND: not detected; f: fish; s: shellfish; c:crustaceans; m: molluscs; veg: vegetables; fr: fruits; m. p.: meat products; ce: cereals; c. p.: cereal products; d. p.: dairy products; d.f.: dried fruits; swee: sweeteners; cond: condiments; [a] in braquets,own value calculated from the data of the author (LB-UB); [b] (UB); [c] (MB) ;* median; ** include soft drinks,light beer and mineral water; *** include animal fats.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.toxrep.2018.05.005.

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