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The first harmonised total diet study in Portugal: Arsenic, cadmium and lead exposure assessment

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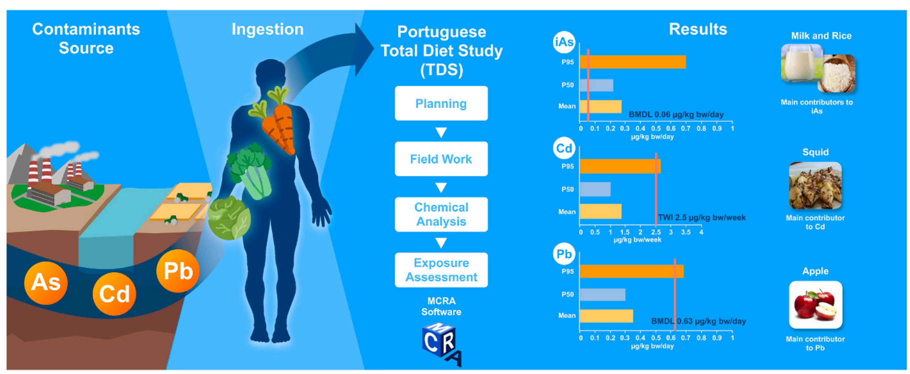
HIGHLIGHTS

- Portuguese population's mean baselines exposure to iAs, Cd and Pb were assessed.
- Mean exposures to iAs and Pb were estimated at 0.28 and 0.35 μg kg^{-1} bw/day.
- Estimated mean exposure to Cd was 1.35 μg kg^{-1} bw/week.
- Potential adverse health effects from exposure to iAs, Cd and Pb cannot be excluded.
- Bread was the common main contributor to iAs, Cd and Pb exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to estimate the 18–74 years old Portuguese population's baseline exposure to inorganic arsenic, cadmium and lead and the risk of exceeding the respective Health Based Guidance Value, using a harmonised Total Diet Study (TDS) methodology. TDS food samples representative of the whole diet were prepared as consumed and analysed for total arsenic, cadmium and lead. European Food Safety Authority's conservative approach was used to estimate inorganic arsenic. Exposure was assessed using the Monte Carlo Risk Assessment software. At upper bound approach, the mean baseline exposure was estimated at 0.28 and 0.35 μg kg^{-1} body weight day^{-1} for inorganic arsenic and lead, respectively, and 1.36 μg kg^{-1} body weight $week^{-1}$ for cadmium. Margins of exposure of below or close to one were found for inorganic arsenic and lead, whereas 5.4 % of individuals exceeded the Tolerable Weekly Intake for cadmium. These results indicate that adverse health effects cannot be ruled out. Bread was the common main contributor for the exposure to all three elements.

1. Introduction

Human exposure to arsenic, cadmium, and lead is concerning due to their widespread presence and potential health risks. Arsenic, naturally found in the Earth's crust, exists in various chemical forms, with inorganic arsenic being the most toxic and prevalent in contaminated environments (European Food Safety Authority [EFSA], 2009). Cadmium, commonly utilized in industrial processes (batteries, pigments, coatings,

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plating, plastics), exists in different chemical forms as well, and poses significant health risks upon exposure (World Health Organization [WHO], 2019). Lead, found in various sources including old paint, gasoline, and some workplaces (batteries, jewelry, soldering and ceramics), exists in different chemical forms and is toxic to human health, especially in its inorganic form (World Health Organization [WHO], 2023).

The exposure routes for arsenic, cadmium and lead vary. Arsenic exposure typically occurs through contaminated water sources and foods, like rice and seafood (European Food Safety Authority [EFSA], 2009), while cadmium exposure primarily originates from food, tobacco, and occupational settings (European Food Safety Authority [EFSA], 2012), (Agency for Toxic Substances and Disease Registry [ATSDR], 2012). Lead exposure can occur through multiple pathways, including contaminated food, water, and inhalation of lead-containing dust or fumes (European Food Safety Authority [EFSA], 2010a). Diet is a common major source of exposure for these elements.

Arsenic, cadmium, and lead have varied adverse effects on human health. Chronic exposure to inorganic arsenic has been linked to skin cancer, cardiovascular diseases, neurotoxicity and immunotoxicity (European Food Safety Authority [EFSA], 2009), (Hughes et al., 2011), (Dangleben et al., 2013), (European Food Safety Authority [EFSA], 2024). Cadmium exposure is associated with kidney damage, bone disorders, and potentially increased cancer risk (World Health Organization [WHO], 2019). Lead exposure can cause developmental delays, cognitive impairment, and behavioral issues in children; and cardiovascular and nephrological problems in adults (European Food Safety Authority [EFSA], 2010a). Health-based guidance values (HBGV) are crucial for managing exposure. Values like Tolerable Weekly Intake (TWI) for cadmium and Benchmark Dose Limits (BMDL) for inorganic arsenic and lead set thresholds to limit exposure and prevent adverse health effects (European Food Safety Authority [EFSA], 2009), (European Food Safety Authority [EFSA], 2012), (European Food Safety Authority [EFSA], 2010a), (European Food Safety Authority [EFSA], 2024).

Total Diet Studies (TDSs) are a cost-effective public health tool for assessing a population's exposure to various chemicals, including contaminants and nutrients (World Health Organization [WHO], 2005), (Food and Agriculture Organization of the United Nations [FAO], 2014). TDSs provide insights into dietary habits, aiding in understanding eating patterns and potential health risks by studying the whole population's diet to assess exposure levels TDSs are useful for both screening and detailed exposure assessment by measuring chemical levels in a broad range of representative foods, prepared as consumed by the population, and are considered a good complement to monitoring programs that focus on individual highly contaminated items. Overall, a TDS can improve food safety programs by offering a thorough assessment of dietary exposure to both beneficial and harmful substances for risk assessment and management (W. H. O. [WHO] European Food Safety Authority [EFSA], 2011). International and European initiatives to harmonise TDS methodologies were carried out with the purpose to facilitate trend analysis, comparisons and understanding of exposure risks across populations and countries (World Health Organization [WHO], European Food Safety Authority [EFSA], Food and agriculture organization of the united Nations [FAO], 2011), (Pité et al., 2018).

No information have been reported on the dietary exposure of the population in Portugal to arsenic, cadmium, and lead, covering the entire diet with food analysed in its consumed form. The study aimed to assess the baseline chronic dietary exposure of the Portuguese population to arsenic, cadmium and lead using a harmonised TDS methodology and to estimate the risk of exceeding the HBGVs or BMDL as appropriate.

2. Material and methods

2.1. Food consumption data and food sampling

The planning and field work harmonised methodology used in this TDS was described in detail by Vasco (Vasco et al., 2021). In summary, food consumption survey data derived from a one day 24 h recall conducted in 2009 (Poínhos et al., 2009) were used to create a core food list consisting of 1072 food items representing the general Portuguese population diet (males and females, 18–93 years old, n = 3470). This food list was then classified and mapped into 20 food groups using the FoodEx2 food classification and description system (European Food Safety Authority [EFSA], 2011). A TDS food list consisting of 528 food items was created for the called 'overall population' ('adults and elderly' 'male and female', 18–74 years old, n = 3272). This list covered 95 % of the total diet and was created by applying two selection criteria, by food group, to determine the food items to include: the most consumed foods contributing to at least 90 % of the overall food consumption, and foods known or suspected to be main contributors to exposure or intake of specific substances of interest. The food items from the TDS food list were aggregated using either the individual or mixed food approach (Dofkova et al., 2016), resulting in 164 different TDS samples. Of these, 21 were identified as having potential for seasonal variation (four samples one per season), bringing the total to 227 TDS samples. Each TDS sample was composed of twelve subsamples (Julious, 2005) and was classified according to FoodEx2. The collection of food items took place between April 2014 and March 2016 in selected shops of the Greater Lisbon region. Subsamples were prepared 'as consumed', reflecting people's behaviour based on national culinary reference books and or package information, and their edible parts were pooled, homogenized, aliquoted, and stored at −20 °C until analysis.

2.2. Arsenic (As), cadmium (Cd) and lead (Pb) quantification

Determination of total arsenic (tAs), Cd and Pb occurrence in TDS samples was performed by a ISO/IEC 17025 accredited method using quadrupole inductively coupled plasma mass spectrometry (ICP-MS; Thermo Elemental, X-series 2, UK) (Coelho et al., 2013; Ventura et al., 2018; Ventura et al., 2019). Instrumental operation conditions were set according to manufacturer guidelines. TDS samples were acid digested in a closed vessel microwave digester (Ethos 1, Milestone) with concentrated nitric acid, hydrogen peroxide and ultrapure water. Internal standards ^{115}In and ^{72}Ge (1000 mg L^{-1}; Merck, Darmstadt, Germany) were used. Two different batches of highest purity multi element standard solutions containing 100 mg L^{-1} of As, Cd, Pb (Merck Multi XVI, VWR, Portugal), were used to prepare calibration standards. Quantification by external standard method was performed with six point equally distributed calibration curves (working range: 0.25–2.5 μg L^{-1} for As and Cd; 0.5–5 μg L^{-1} for Pb). Limits of detection (LOD) and quantification (LOQ) were estimated, respectively, as three and ten times the mean blank tests standard deviation, after correction for sample weight and dilution. LODs ranged from 0.08 to 12 μg kg^{-1} for As and Cd and from 0.17 to 24 μg kg^{-1} for Pb. LOQs ranged from 0.25 to 40 μg kg^{-1} for As and Cd and from 0.5 to 70 μg kg^{-1} for Pb. Results were expressed in μg kg^{-1}. Measurement precision expressed by relative standard deviation was below 10 % for repeatability and 13 % for intermediate precision, for all elements. Recovery of spiked samples ranged between 80 and 120 %. Laboratory performance was assessed by participating in Proficiency Test Schemes (Fapas®, York, UK), with z-scores $\leq |2|$, and analysis of reference materials (DORM-3). Relative uncertainty of the results was 22 % for As and 33 % for Cd and Pb (Coelho et al., 2013), (Ventura et al., 2018), (Ventura et al., 2019).

2.3. Occurrence, exposure assessment and risk characterization

Occurrence results reported below the LOD and/or LOQ (left-censored data) were managed using the upper bound (UB) and lower bound (LB) substitution methods according to World Health Organization (WHO) and European Food Safety Authority (EFSA) (GEMS/Food-EURO, 1995), (European Food Safety Authority [EFSA], 2010b). In the UB approach, non-detected results were replaced with the LOD, while values falling between LOD and LOQ were substituted with the LOQ. In the LB approach, non-detected results were substituted with zero, and values within LOD and LOQ were replaced with the LOD. The TDS samples collected across four seasons were analysed individually and the mean occurrence value, calculated after applying the left-censored substitution method to each sample, was used for exposure assessment.

Total arsenic, cadmium, and lead, minimum, maximum, mean, and median values were computed for each FoodEx2 group. Cd and Pb occurrence values were used directly to estimate the dietary exposure; whereas, for As, as speciation analysis was not performed, EFSA's approach was used to estimate the occurrence of inorganic arsenic (iAs) from tAs. Therefore, iAs in TDS samples was assumed as 0.03 mg kg^{-1} for fish, 0.1 mg kg^{-1} for seafood and 70 % of tAs for samples other than fish and seafood (European Food Safety Authority [EFSA], 2009). To perform the dietary exposure assessment, a semiprobabilistic approach was used combining the estimated UB and LB occurrence data with the corresponding consumption recorded in the food consumption survey. The linkage between occurrence and consumption data was established by the FoodEx2 codes. The exposure calculation was performed utilizing the Monte Carlo Risk Assessment (MCRA) software (Boon et al., 2018), employing the Observed Individual Mean model (van der Voet and Boon, 2015). Exposure estimates for food as measured (TDS samples) and for food as eaten (TDS subsamples), expressed in μg kg^{-1} body weight (bw) day^{-1} or μg kg^{-1} bw $week^{-1}$, were performed for the 'overall population' and segmented by sex and age groups, 'adults' (18–64 years old, n = 2752) and 'elderly' (65–74 years old, n = 520). Results were presented as mean, median (P50) and 95th percentile (P95), representing the average and highly exposed population. A deterministic method assessed the overall population's exposure to As, Cd, and Pb from tap water consumption. Mean tap water consumption was multiplied by average maximum occurrence values (0.43 μg kg^{-1} for As, 0.26 μg kg^{-1} for Cd, 1.25 μg kg^{-1} for Pb), obtained through water quality regulatory checks (2018 and 2022) in Greater Lisbon.

Risk was assessed by comparing the estimated exposure against the respective HBGV and BMDL. The calculated exposure levels of Cd were compared with the TWI of 2.5 μg kg^{-1} bw $week^{-1}$ (European Food Safety Authority [EFSA], 2012). Additionally, the estimated percentages of individuals surpassing the TWI were determined. For As and Pb lower BMDL were used as reference point to calculate the margin of exposure (MOE) (European Food Safety Authority [EFSA], 2005). MOEs were calculated by dividing the BMDL by the mean and P95 exposure values. For iAs the BMDL01 range between 0.3 and 8 μg kg^{-1} bw day^{-1} (lung, skin and bladder cancers, and skin lesions) (European Food Safety Authority [EFSA], 2009) and BMDL05 of 0.06 μg kg^{-1} bw day^{-1} skin cancer (squamous cell carcinoma) (European Food Safety Authority [EFSA], 2024) were used. No iAs MOE of low concern was identified by EFSA as the BMDLs were derived from human cancer data (European Food Safety Authority [EFSA], 2024). BMDL01 of 1.5 μg kg^{-1} bw day^{-1} (cardiovascular effects) and BMDL10 of 0.63 μg kg^{-1} bw day^{-1} (nephrotoxicity) were used for Pb (European Food Safety Authority [EFSA], 2010a). EFSA's CONTAM Panel considered Pb MOEs of 10 or greater sufficient to ensure no appreciable risk of a clinically significant effect, and that the risk would be very low even with MOEs between 10 and 1.0 (European Food Safety Authority [EFSA], 2010a). The contribution of each food item to the total exposure was estimated and the main contributors were highlighted.

3. Results and discussion

The exposure estimates obtained from both the upper and lower bound approaches generally led to the same conclusions regarding health risks. Therefore, unless specified otherwise, all results and considerations presented refer to the upper bound approach, portraying the worst-case scenario. For comparisons with other published results, upper bound estimates were used when specified; otherwise, the available results were applied.

3.1. Arsenic

Total arsenic occurrence levels below LOD and LOQ represented 56 % of the analysed samples. Data on tAs occurrence is summarised in Table 1. The food group 'Fish, seafood, amphibians, reptiles, and invertebrates' had the highest mean occurrence at 2.4 mg kg^{-1} (LB and UB), with European Conger showing the highest tAs level at 14 mg kg^{-1} 'Composite dishes' followed with a mean occurrence of 0.33 mg kg^{-1}. All other food groups had mean occurrences at or below 0.029 mg kg^{-1}.

Table 2 summarises the inorganic arsenic (iAs), LB and UB exposure results by food group and the risk assessment for the Portuguese 'overall population' segmented by sex. Supplementary information includes data for 'adults' and 'elderly' population groups (Tables 1S and 2S). The estimated 'overall population' UB mean exposure to iAs was 0.28 μg kg^{-1} bw day^{-1} being lower for the 'female' 'elderly' group (0.24 μg kg^{-1} bw day^{-1}). P95 (UB) revealed the same exposure tendency, 0.70 μg kg^{-1} bw day^{-1} for 'overall population' and 0.62 μg kg^{-1} bw day^{-1} for the 'female' 'elderly'. Considering the BMDL01 range of 0.3–8 μg kg^{-1} bw day^{-1} (European Food Safety Authority [EFSA], 2009), overall population's mean exposure MOEs ranged from 1.1 to 29 (UB), respectively, whereas at P95, MOEs ranged from 0.4 to 11 (UB). When BMDL05 of 0.06 μg kg^{-1} bw day^{-1} (European Food Safety Authority [EFSA], 2024) was considered, 'overall population' MOEs (UB) were 0.2 at mean exposure and 0.1 at P95. Based on these findings, increased risk of lung, skin, and bladder cancer, as well as skin lesions, cannot be dismissed. The food groups 'composite dishes', 'fish, seafood, amphibians, reptiles and invertebrates, 'grains and grain-based products' and 'milk and dairy products' were the main contributors to iAs exposure of all sex and age groups, except for 'female' 'elderly' with 'milk and dairy products' as third contributor at UB (Table 2). Detailed data on each TDS sample's contribution to iAs exposure are provided in supplementary information (Tables 3S, 4S, 5S). Semi-skimmed milk and rice contributed the most to iAs exposure of the 'overall population', followed by octopus rice and bread at UB. Supplementary information (Table 6S) details the contribution of food as eaten to iAs exposure.

Arnich (Arnich et al., 2012) reported a mean exposure to iAs of 0.28 μg kg^{-1} bw day^{-1} (UB) and a P95 of 0.51 μg kg^{-1} bw day^{-1} (UB) for the adult French population (18–79 years old), corresponding to MOEs of 1.07 (mean) and 0.59 (P95) for BMDL01 of 0.3 μg kg^{-1} bw day^{-1}, and 29 (mean) and 16 (P95) for BMDL01 of 8 μg kg^{-1} bw day^{-1}. The main contributors to iAs exposure were water and coffee.

Mean exposure of Valdivia (Chile) adult population (18–65 years old) to iAs was estimated by Munoz (Muñoz et al., 2017) as 0.28 μg kg^{-1} bw day^{-1}. The main contributors to iAs exposure were non-alcoholic beverages (20 %), bread (16 %) and vegetables (11 %).

Marín (Marin et al., 2017) reported an iAs mean exposure of 0.09 μg kg^{-1} bw day^{-1} (UB) estimated for the adult population (16–95 years old) of Valencia region, Spain. Cereals food group was the main contributor to iAs exposure (50 %, LB).

Exposure of Italian's adult population (18–64 years old) to iAs was estimated by Cubadda (Cubadda et al., 2016) as 0.081 μg kg^{-1} bw day^{-1} for the mean and as 0.284 μg kg^{-1} bw day^{-1} for P95. Considering the range of BMDL01 values of 0.3–8 μg kg^{-1} bw day^{-1} MOEs ranged from 4 to 99 for mean exposure and from 1 to 28 at P95. The main contributors to iAs exposure were cereals and cereal products (33 %), especially rice and bread; water and other non-alcoholic beverages (27 %), especially bottled water; vegetables (11 %); and fruit (7 %).

Hackethal (Hackethal et al., 2023) reported a mean exposure to iAs of 0.09 μg kg^{-1} bw day^{-1} (UB) and a P95 of 0.15 μg kg^{-1} bw day^{-1} (UB)

Table 1
Total arsenic levels (minimum, maximum, mean and median) in TDS samples expressed by FoodEx2 food group.

FoodEx2 food group Nr	FoodEx2 Level 1 food Groups	Total Nr of food items[a]	Nr of food items selected for the TDS food list	Nr of different TDS Samples	Total Nr of TDS Samples [b]	Nr of TDS Samples			Total arsenic (mg kg-1)							
									Minimum		Maximum		Mean		Median	
						≤LOD	>LOD and ≤LOQ	>LOQ	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	28	7	2	2	0	0	2	0.0024	0.0024	0.005	0.005	0.0037	0.0037	0.0037	0.0037
3	Animal and vegetable fats and oils	7	3	2	2	2	0	0	0	0.012	0	0.012	0	0.012	0	0.012
4	Coffee, cacao and tea and infusions	12	9	4	4	0	2	2	0.0005	0.0016	0.011	0.011	0.0039	0.0045	0.0021	0.0026
5	Composite dishes	360	191	34	34	3	12	19	0	0.00025	8	8	0.3316	0.3353	0.0155	0.02
6	Eggs and egg product	2	1	1	1	0	1	0	0.0043	0.013	0.0043	0.013	0.0043	0.013	0.0043	0.013
7	Fish, seafood, amphibians, reptiles and invertebrates	68	41	25	31	0	0	31	0.029	0.029	14	14	2.394	2.394	1.4	1.4
9	Fruit and fruit products	65	15	14	32	25	2	5	0	0.004	0.032	0.032	0.0044	0.0087	0	0.0041
10	Fruit and vegetable juices and nectars	43	38	2	2	0	1	1	0.0007	0.002	0.0038	0.0038	0.0023	0.0029	0.0023	0.0029
11	Grains and grain-based products	158	67	21	21	0	9	12	0.003	0.009	0.06	0.06	0.0149	0.0182	0.014	0.014
12	Legumes, nuts, oilseeds and spices	21	11	8	8	1	5	2	0	0.008	0.042	0.042	0.012	0.022	0.008	0.024
13	Meat and meat products	68	49	11	11	0	9	2	0.0041	0.012	0.03	0.03	0.0116	0.0238	0.008	0.024
14	Milk and dairy products	89	30	6	6	0	5	1	0.0029	0.009	0.009	0.025	0.0048	0.0118	0.0031	0.009
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	9	4	2	2	0	0	2	0.008	0.008	0.05	0.05	0.029	0.029	0.029	0.029
16	Seasoning, sauces and condiments	22	14	5	5	0	0	5	0.004	0.004	0.04	0.04	0.0168	0.0168	0.011	0.011
17	Starchy roots or tubers and products thereof, sugar plants	7	1	1	4	2	2	0	0	0.013	0	0.013	0	0.013	0	0.013
18	Sugar, confectionery and water-based sweet desserts	23	12	3	3	1	0	2	0	0.0022	0.011	0.011	0.0044	0.0057	0.0022	0.004
19	Vegetables and vegetable products	46	22	19	52	13	29	10	0	0.0038	0.08	0.08	0.0083	0.0142	0.0033	0.011
20	Water and water-based beverages[c]	40	13	4	7	0	2	5	0.0005	0.0016	0.004	0.004	0.0020	0.0025	0.0020	0.0025
Total		1068	528	164	227	47	79	101								
	%					21	35	44								

TDS - Total Diet Study, LOD - Limit of Detection, LOQ - Limit of Quantification, LB - Lower Bound approach and UB - Upper Bound approach.
[a] Core food list identified from the Food Consumption Survey.
[b] Including four samples for each of the 21 seasonal TDS Samples (one per season).
[c] Tap water not included.

Table 2
Estimated dietary exposure of the Portuguese population to inorganic arsenic.

FoodEx2 Food Group Nr	Age group	18–74											
	Sex	Male and Female				Male				Female			
	Nr Individuals	3272				1583				1689			
	FoodEx2 Level 1 Food Group	Mean exposure (μg kg-1 bw day-1)		Contribution (%)		Mean exposure (μg kg-1 bw day-1)		Contribution (%)		Mean exposure (μg kg-1 bw day-1)		Contribution (%)	
		LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	0.00	0.00	2.19	1.62	0.01	0.01	3.82	2.90	0.00	0.00	0.54	0.39
3	Animal and vegetable fats and oils	0.00	0.00	0.00	0.29	0.00	0.00	0.00	0.26	0.00	0.00	0.00	0.32
4	Coffee, cacao and tea and infusions	0.00	0.00	2.04	1.79	0.00	0.00	1.90	1.56	0.00	0.01	2.18	2.01
5	Composite dishes	0.09	0.11	42.43	38.02	0.09	0.11	43.43	38.99	0.08	0.10	41.41	37.08
6	Eggs and egg product	0.00	0.00	0.14	0.31	0.00	0.00	0.14	0.32	0.00	0.00	0.14	0.31
7	Fish, seafood, amphibians, reptiles and invertebrates	0.04	0.04	18.19	13.50	0.04	0.04	17.97	13.65	0.04	0.04	18.43	13.36
9	Fruit and fruit products	0.00	0.01	0.16	5.01	0.00	0.01	0.09	3.92	0.00	0.02	0.23	6.06
10	Fruit and vegetable juices and nectars	0.00	0.00	0.50	0.65	0.00	0.00	0.44	0.59	0.00	0.00	0.57	0.71
11	Grains and grain-based products	0.03	0.04	15.89	12.71	0.03	0.04	15.20	12.42	0.03	0.04	16.58	12.99
12	Legumes, nuts, oilseeds and spices	0.00	0.00	0.59	0.75	0.00	0.00	0.76	0.94	0.00	0.00	0.41	0.56
13	Meat and meat products	0.01	0.02	4.54	7.89	0.01	0.02	4.78	8.53	0.01	0.02	4.30	7.26
14	Milk and dairy products	0.02	0.03	10.57	10.09	0.02	0.02	8.80	8.56	0.02	0.03	12.36	11.57
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	0.00	0.00	0.16	0.12	0.00	0.00	0.13	0.10	0.00	0.00	0.19	0.14
16	Seasoning, sauces and condiments	0.00	0.00	0.22	0.16	0.00	0.00	0.30	0.23	0.00	0.00	0.14	0.10
17	Starchy roots or tubers and products thereof, sugar plants	0.00	0.01	0.00	3.27	0.00	0.01	0.00	3.44	0.00	0.01	0.00	3.10
18	Sugar, confectionery and water-based sweet desserts	0.00	0.00	0.34	0.37	0.00	0.00	0.32	0.39	0.00	0.00	0.36	0.35
19	Vegetables and vegetable products	0.00	0.01	1.23	2.55	0.00	0.01	1.01	2.16	0.00	0.01	1.45	2.93
20	Water and water-based beverages[a]	0.00	0.00	0.79	0.85	0.00	0.00	0.87	1.00	0.00	0.00	0.70	0.71
	Mean	0.21	0.28	–		0.22	0.28	–		0.20	0.27	–	
	MOE Mean (BMDL05 of 0.06)	0.3	0.2	–		0.3	0.2	–		0.3	0.2	–	
	MOE Mean (BMDL01 of 0.3)	1.5	1.1	–		1.4	1.1	–		1.5	1.1	–	
	MOE Mean (BMDL01 of 8)	39	29	–		37	28	–		40	29	–	
	P50	0.14	0.22	–		0.15	0.23	–		0.13	0.22	–	
	MOE P50 (BMDL05 of 0.06)	0.4	0.3	–		0.4	0.3	–		0.4	0.3	–	
	MOE P50 (BMDL01 of 0.3)	2.1	1.4	–		2.0	1.3	–		2.2	1.4	–	
	MOE P50 (BMDL01 of 8)	57	36	–		54	35	–		60	37	–	
	P95	0.64	0.70	–		0.67	0.70	–		0.60	0.70	–	
	MOE P95 (BMDL05 of 0.06)	0.1	0.1	–		0.1	0.1	–		0.1	0.1	–	
	MOE P95 (BMDL01 of 0.3)	0.5	0.4	–		0.4	0.4	–		0.5	0.4	–	
	MOE P95 (BMDL01 of 8)	13	11	–		12	11	–		13	11	–	

LB - Lower bound approach, UB - Upper bound approach, Mean - Mean exposure (μg kg-1 bw day-1), MOE - Margin of Exposure, BMDL - Benchmark Dose Limit (μg kg-1 bw day-1), P50 - Percentile 50 exposure (μg kg-1 bw day-1) and P95 - Percentile 95 exposure (μg kg-1 bw day-1).

[a] Tap water not included.

for the adolescent/adult German population (14–64 years old). The food group 'Grains and grain-based products' was the main contributor to iAs exposure (26.4 %).

The estimated UB iAs mean exposure (0.28 μg kg^{-1} bw day^{-1}) of the overall Portuguese population (18–74 years old) was similar to those reported by Arnich (Arnich et al., 2012) and Munoz (Muñoz et al., 2017), and approximately three fold higher than those reported by Cubadda (Cubadda et al., 2016), Marin (Marin et al., 2017) and Hackethal (Hackethal et al., 2023). P95 exposures showed a similar pattern.

The higher exposure values observed in this study, compared to those reported by Cubadda (Cubadda et al., 2016), Marin (Marin et al., 2017), and Hackethal (Hackethal et al., 2023), could be attributed to their use of a speciation method to directly measure iAs in food samples, rather than estimating iAs from total arsenic (tAs) based on assumptions. Accurately assessing dietary exposure to inorganic arsenic (iAs) is challenging because many foods, particularly fish and seafood, contain less toxic organic arsenic species. In seafood, the proportion of iAs is typically small and decreases as total arsenic levels rise, with variations depending on the seafood type. Relying on assumptions to estimate iAs from tAs, which involves high uncertainty, often results in overestimated exposure. This highlights the importance of speciation analysis to provide actual iAs levels for more reliable risk assessments. Cereal products were reported in most publications as main contributors to iAs exposure.

3.2. Cadmium

Cadmium occurrence levels below the LOD and LOQ represented approximately 87 % of the analysed samples. Data on Cd occurrence is summarised in Table 3. The highest Cd level (Common Squid, 0.5 mg kg^{-1}) was found in the 'Fish, seafood, amphibians, reptiles and invertebrates' food group, which had a mean value of 0.049 mg kg^{-1} (LB approach) and 0.062 mg kg^{-1} (UB approach). Other food groups showed mean occurrence values equal or below 0.012 and 0.015 mg kg^{-1} for LB and UB approach, respectively.

Table 4 summarises the Cd exposure results by food group and the risk assessment for the 18–74 years old Portuguese population segmented by sex. Supplementary information includes data for 'adults' and 'elderly' population groups (Tables 7S and 8S). For the 'overall population' the estimated UB mean weekly exposure to Cd was 1.36 μg kg^{-1} bw $week^{-1}$ ('male' 1.38 and 'female' 1.33) and P95 (UB) was 2.63 μg kg^{-1} bw $week^{-1}$ ('male' 2.86 and 'female' 2.53). The 'overall population' UB mean weekly exposure to Cd was 54.3 % of the TWI, with

Table 3
Cadmium levels (minimum, maximum, mean and median) in TDS samples expressed by FoodEx2 food group.

FoodEx2 food group Nr	FoodEx2 Level 1 food Groups	Total Nr of food items[a]	Nr of food items selected for the TDS food list	Nr of different TDS Samples	Total Nr of TDS Samples[b]	Nr of TDS Samples			Cadmium (mg kg^{-1})							
									Minimum		Maximum		Mean		Median	
						≤LOD	>LOD and ≤LOQ	>LOQ	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	28	7	2	2	2	0	0	0	0.0005	0	0.0005	0	0.0005	0	0.0005
3	Animal and vegetable fats and oils	7	3	2	2	2	0	0	0	0.012	0	0.012	0	0.012	0	0.012
4	Coffee, cacao and tea and infusions	12	9	4	4	2	0	2	0	0.0005	0.016	0.016	0.0047	0.0050	0.0015	0.0017
5	Composite dishes	360	191	34	34	14	15	5	0	0.00008	0.027	0.027	0.0044	0.0099	0.004	0.012
6	Eggs and egg product	2	1	1	1	1	0	0	0	0.004	0	0.004	0	0.004	0	0.004
7	Fish, seafood, amphibians, reptiles and invertebrates	68	41	25	31	17	7	7	0	0.006	0.5	0.5	0.049	0.062	0	0.012
9	Fruit and fruit products	65	15	14	32	28	4	0	0	0.004	0.004	0.012	0.0003	0.0049	0	0.004
10	Fruit and vegetable juices and nectars	43	38	2	2	2	0	0	0	0.0007	0	0.0007	0	0.0007	0	0.0007
11	Grains and grain-based products	158	67	21	21	5	12	4	0	0.004	0.018	0.018	0.0050	0.0107	0.004	0.012
12	Legumes, nuts, oilseeds and spices	21	11	8	8	7	0	1	0	0.004	0.06	0.06	0.0075	0.0140	0	0.008
13	Meat and meat products	68	49	11	11	11	0	0	0	0.004	0	0.008	0.0000	0.0076	0	0.008
14	Milk and dairy products	89	30	6	6	6	0	0	0	0.001	0	0.008	0.0000	0.00345	0	0.0029
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	9	4	2	2	0	1	1	0.004	0.006	0.006	0.012	0.005	0.009	0.005	0.009
16	Seasoning, sauces and condiments	22	14	5	5	3	2	0	0	0.0005	0.003	0.01	0.0012	0.00492	0	0.004
17	Starchy roots or tubers and products thereof, sugar plants	7	1	1	4	0	1	3	0.012	0.015	0.012	0.015	0.012	0.015	0.012	0.015
18	Sugar, confectionery and water-based sweet desserts	23	12	3	3	2	0	1	0	0.0007	0.02	0.02	0.0067	0.0096	0	0.008
19	Vegetables and vegetable products	46	22	19	52	30	16	6	0	0.0028	0.011	0.025	0.0016	0.0087	0	0.008
20	Water and water-based beverages[c]	40	13	4	7	7	0	0	0	0.00008	0	0.00008	0.0000	0.0004	0	0.0004
Total		1068	528	164	227	139	58	30								
	%					61	26	13								

TDS - Total Diet Study, LOD - Limit of Detection, LOQ - Limit of Quantification, LB - Lower Bound approach and UB - Upper Bound approach.
[a] Core food list identified from the Food Consumption Survey.
[b] Including four samples for each of the 21 seasonal TDS Samples (one per season).
[c] Tap water not included.

Table 4
Estimated dietary exposure of the Portuguese population to cadmium.

FoodEx2 Food Group Nr	Age group	18–74											
	Sex	Male and Female				Male				Female			
	Nr Individuals	3272				1583				1689			
	FoodEx2 Level 1 Food Group	Mean exposure (μg kg^{-1} bw week^{-1})		Contribution (%)		Mean exposure (μg kg^{-1} bw week^{-1})		Contribution (%)		Mean exposure (μg kg^{-1} bw week^{-1})		Contribution (%)	
		LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	0.00	0.01	0.00	0.42	0.00	0.01	0.00	0.73	0.00	0.00	0.00	0.12
3	Animal and vegetable fats and oils	0.00	0.01	0.00	0.42	0.00	0.01	0.00	0.37	0.00	0.01	0.00	0.47
4	Coffee, cacao and tea and infusions	0.01	0.01	0.98	1.09	0.00	0.01	0.50	0.75	0.01	0.02	1.51	1.42
5	Composite dishes	0.10	0.29	15.26	21.20	0.11	0.29	14.40	20.72	0.10	0.29	16.21	21.67
6	Eggs and egg product	0.00	0.00	0.00	0.20	0.00	0.00	0.00	0.20	0.00	0.00	0.00	0.20
7	Fish, seafood, amphibians, reptiles and invertebrates	0.34	0.45	50.02	33.16	0.40	0.49	53.35	35.74	0.29	0.41	46.33	30.65
9	Fruit and fruit products	0.00	0.09	0.00	6.90	0.00	0.08	0.00	5.42	0.00	0.11	0.00	8.35
10	Fruit and vegetable juices and nectars	0.00	0.01	0.00	0.39	0.00	0.00	0.00	0.35	0.00	0.01	0.00	0.43
11	Grains and grain-based products	0.12	0.18	18.09	13.38	0.12	0.18	16.61	13.02	0.12	0.18	19.74	13.74
12	Legumes, nuts, oilseeds and spices	0.00	0.01	0.37	0.65	0.00	0.01	0.53	0.85	0.00	0.01	0.19	0.46
13	Meat and meat products	0.00	0.07	0.00	5.28	0.00	0.08	0.00	5.69	0.00	0.06	0.00	4.88
14	Milk and dairy products	0.00	0.05	0.00	3.83	0.00	0.04	0.00	3.19	0.00	0.06	0.00	4.45
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	0.00	0.00	0.23	0.13	0.00	0.00	0.09	0.07	0.00	0.00	0.38	0.18
16	Seasoning, sauces and condiments	0.00	0.00	0.03	0.07	0.00	0.00	0.03	0.08	0.00	0.00	0.02	0.05
17	Starchy roots or tubers and products thereof, sugar plants	0.08	0.11	12.45	7.82	0.09	0.11	12.24	8.20	0.08	0.10	12.69	7.46
18	Sugar, confectionery and water-based sweet desserts	0.01	0.02	1.57	1.14	0.01	0.02	1.46	1.20	0.01	0.01	1.69	1.09
19	Vegetables and vegetable products	0.01	0.05	0.99	3.52	0.01	0.04	0.78	2.94	0.01	0.05	1.23	4.08
20	Water and water-based beverages[a]	0.00	0.00	0.00	0.36	0.00	0.01	0.00	0.45	0.00	0.00	0.00	0.27
	Mean	0.68	1.36	–		0.74	1.38	–		0.63	1.33	–	
	% o TWI (Mean)	27.3	54.3	–		29.7	55.4	–		25.0	53.2	–	
	P50	0.31	0.99	–		0.31	0.96	–		0.30	1.00	–	
	% of TWI (P50)	12.3	39.4	–		12.3	38.6	–		12.2	40.1	–	
	P95	1.62	2.63	–		1.97	2.86	–		1.44	2.53	–	
	% of TWI (P95)	64.7	105.2	–		78.7	114.4	–		57.5	101.1	–	
	% of persons above the TWI	3.67	5.39	–		4.34	5.61	–		3.02	5.18	–	

LB - Lower bound approach, UB - Upper bound approach, TWI - Tolerable Weekly Intake (μg kg^{-1} bw week^{-1}), Mean - Mean exposure (μg kg^{-1} bw week^{-1}), P50 - Percentile 50 exposure (μg kg^{-1} bw week^{-1}) and P95 - Percentile 95 exposure (μg kg^{-1} bw week^{-1}).
[a] Tap water not included.

'male' 'adults' showing the highest value (56.9 %). Considering the UB approach, all population groups exceeded the TWI at P95, whereas none did so at the LB. The TWI was exceeded by 3.7 % and 5.4 % individuals of the 'overall population' at LB an UB approaches, respectively, therefore increased risk due to Cd exposure cannot be excluded and should be further investigated. The main contributors to Cd exposure for all sex and age groups were 'fish, seafood, amphibians, reptiles and invertebrates', 'composite dishes' and 'grains and grain-based products' food groups, except for 'female' 'elderly' where 'composite dishes' alternates with the fish group at UB. Detailed data on each TDS sample's contribution to Cd exposure is provided in supplementary information (Tables 9S, 10S, 11S). For the overall population, squid was the main contributor followed by bread (LB) and boiled potato (UB). Supplementary information (Table 12S) details the contribution of food as eaten to Cd exposure.

Nasreddine estimated a 1.2 μg kg^{-1} bw week^{-1} (Nasreddine et al., 2006) and 1.52 μg kg^{-1} bw week^{-1} (Nasreddine et al., 2010) Cd mean total dietary exposure of an urban adult Lebanese population (corresponding to 48 % and 60.8 % of TWI of 2.5 μg kg^{-1} bw week^{-1}, respectively). In the 2006 study, the main contributors to Cd exposure were cereals and cereal-based products (36 %), vegetables and potatoes (28.5 %) and drinking water (24.3 %), whereas in the 2010 study the main contributors reported were vegetables (46.8 %) and bread and cereal-based products (30.9 %).

French adult population's exposure to Cd was estimated at 1.12 μg kg^{-1} bw week^{-1} (mean) and 1.89 μg kg^{-1} bw week^{-1} (P95). The main contributors to Cd exposure were bread and dried bread products (22 % and 13 %, respectively) and potato and potato products (12 % and 14 %, respectively). The TWI was exceeded by 0.6 % of the individuals (Arnich et al., 2012).

Mean exposure to Cd of Chile's adult population was estimated by Munoz et al. as 2.16 μg kg^{-1} bw week^{-1} in Santiago city (Muñoz et al., 2005) and 1.81 μg kg^{-1} bw week^{-1} in Valdivia city (Muñoz et al., 2017). For Santiago's population the main contributor food groups were fish and shellfish (43.8 %), spices (14.3 %) and cereals (6.7 %), whereas for Valdivia's population the bread group (27.4 %) was the main contributor followed by non-alcoholic beverages (21.2 %) and cereals groups (11.1 %).

Czechia's adult population mean exposure to Cd was reported as 42.1 % of the 2.5 μg kg^{-1} bw week^{-1} TWI corresponding to 1.05 μg kg^{-1} bw week^{-1} and the main contributors were potatoes and derivatives followed by cereals (Sokolov et al., 2022).

The estimated UB Cd mean exposure (1.36 μg kg^{-1} bw week^{-1}) of the overall Portuguese population was within the range found in the previously referred studies (1.05–2.16 μg kg^{-1} bw week^{-1}). Cereal based products, and potatoes appear to be common contributors in all studied populations.

3.3. Lead

Lead occurrence values were lower than LOD and LOQ for 92 % of the analysed samples. Data on Pb occurrence is summarised in Table 5.

Table 5
Lead levels (minimum, maximum, mean and median) in TDS samples expressed by FoodEx2 food group.

FoodEx2 food group Nr	FoodEx2 Level 1 food Groups	Total Nr of food items[a]	Nr of food items selected for the TDS food list	Nr of different TDS Samples	Total Nr of TDS Samples[b]	Nr of TDS Samples			Lead (mg kg^{-1})							
									Minimum		Maximum		Mean		Median	
						≤LOD	>LOD and ≤LOQ	>LOQ	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	28	7	2	2	1	0	1	0	0.001	0.022	0.022	0.011	0.012	0.011	0.0115
3	Animal and vegetable fats and oils	7	3	2	2	2	0	0	0	0.024	0	0.024	0	0.024	0	0.024
4	Coffee, cacao and tea and infusions	12	9	4	4	0	1	3	0.001	0.0031	0.012	0.012	0.0073	0.008	0.0080	0.0080
5	Composite dishes	360	191	34	34	21	13	0	0	0.0005	0.016	0.05	0.0036	0.016	0	0.009
6	Eggs and egg product	2	1	1	1	1	0	0	0	0.009	0	0.009	0	0.009	0	0.009
7	Fish, seafood, amphibians, reptiles and invertebrates	68	41	25	31	17	11	3	0	0.011	0.26	0.26	0.020	0.047	0	0.024
9	Fruit and fruit products	65	15	14	32	27	5	0	0	0.008	0.008	0.05	0.001	0.016	0	0.008
10	Fruit and vegetable juices and nectars	43	38	2	2	2	0	0	0	0.0014	0	0.0014	0	0.0014	0	0.0014
11	Grains and grain-based products	158	67	21	21	13	8	0	0	0.006	0.016	0.05	0.003	0.016	0	0.008
12	Legumes, nuts, oilseeds and spices	21	11	8	8	7	1	0	0	0.006	0.016	0.05	0.002	0.018	0	0.016
13	Meat and meat products	68	49	11	11	10	1	0	0	0.008	0.016	0.05	0.001	0.018	0	0.016
14	Milk and dairy products	89	30	6	6	5	1	0	0	0.002	0.016	0.05	0.003	0.013	0	0.006
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	9	4	2	2	0	0	2	0.006	0.006	0.036	0.036	0.021	0.021	0.021	0.021
16	Seasoning, sauces and condiments	22	14	5	5	0	2	3	0.0033	0.006	0.02	0.02	0.009	0.012	0.006	0.012
17	Starchy roots or tubers and products thereof, sugar plants	7	1	1	4	4	0	0	0	0.009	0	0.009	0	0.009	0	0.009
18	Sugar, confectionery and water-based sweet desserts	23	12	3	3	1	2	0	0	0.004	0.004	0.013	0.002	0.008	0.0014	0.008
19	Vegetables and vegetable products	46	22	19	52	31	14	7	0	0.006	0.032	0.036	0.005	0.015	0	0.013
20	Water and water-based beverages[c]	40	13	4	7	5	2	0	0	0.0005	0.001	0.0005	0.0003	0.0014	0.00025	0.0014
Total		1068	528	164	227	147	61	19								
	%					65	27	8								

TDS - Total Diet Study, LOD - Limit of Detection, LOQ - Limit of Quantification, LB - Lower Bound approach and UB - Upper Bound approach.
[a] Core food list identified from the Food Consumption Survey.
[b] Including four samples for each of the 21 seasonal TDS Samples (one per season).
[c] Tap water not included.

Table 6
Estimated dietary exposure of the Portuguese population to lead.

FoodEx2 Food Group Nr	Age group	18–74											
	Sex	Male and Female				Male				Female			
	Nr Individuals	3272				1583				1689			
	FoodEx2 Level 1 Food Group	Mean exposure (μg kg^{-1} bw day^{-1})		Contribution (%)		Mean exposure (μg kg^{-1} bw day^{-1})		Contribution (%)		Mean exposure (μg kg^{-1} bw day^{-1})		Contribution (%)	
		LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	0.02	0.02	20.16	5.74	0.03	0.03	29.45	9.57	0.01	0.01	8.92	2.17
3	Animal and vegetable fats and oils	0.00	0.00	0.00	0.47	0.00	0.00	0.00	0.42	0.00	0.00	0.00	0.51
4	Coffee, cacao and tea and infusions	0.01	0.02	14.56	4.48	0.01	0.01	11.36	3.76	0.02	0.02	18.42	5.16
5	Composite dishes	0.01	0.07	10.16	19.52	0.01	0.07	9.73	19.62	0.01	0.07	10.67	19.43
6	Eggs and egg product	0.00	0.00	0.00	0.25	0.00	0.00	0.00	0.26	0.00	0.00	0.00	0.25
7	Fish, seafood, amphibians, reptiles and invertebrates	0.03	0.05	27.16	14.65	0.03	0.05	23.89	14.11	0.03	0.05	31.11	15.15
9	Fruit and fruit products	0.00	0.06	0.28	17.40	0.00	0.05	0.24	13.89	0.00	0.07	0.32	20.68
10	Fruit and vegetable juices and nectars	0.00	0.00	0.00	0.44	0.00	0.00	0.00	0.40	0.00	0.00	0.00	0.48
11	Grains and grain-based products	0.02	0.06	18.63	16.97	0.02	0.06	16.95	17.32	0.02	0.06	20.67	16.65
12	Legumes, nuts, oilseeds and spices	0.00	0.00	0.00	0.53	0.00	0.00	0.00	0.65	0.00	0.00	0.00	0.41
13	Meat and meat products	0.00	0.02	0.78	6.30	0.00	0.02	0.97	7.10	0.00	0.02	0.55	5.55
14	Milk and dairy products	0.00	0.02	3.14	6.16	0.00	0.02	3.26	5.85	0.00	0.02	3.00	6.46
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	0.00	0.00	0.34	0.09	0.00	0.00	0.24	0.08	0.00	0.00	0.45	0.11
16	Seasoning, sauces and condiments	0.00	0.00	0.27	0.10	0.00	0.00	0.28	0.12	0.00	0.00	0.25	0.08
17	Starchy roots or tubers and products thereof, sugar plants	0.00	0.01	0.00	2.63	0.00	0.01	0.00	2.82	0.00	0.01	0.00	2.45
18	Sugar, confectionery and water-based sweet desserts	0.00	0.00	0.41	0.54	0.00	0.00	0.35	0.58	0.00	0.00	0.49	0.51
19	Vegetables and vegetable products	0.00	0.01	3.66	3.00	0.00	0.01	2.77	2.56	0.00	0.01	4.74	3.42
20	Water and water-based beverages[a]	0.00	0.00	0.32	0.60	0.00	0.00	0.34	0.75	0.00	0.00	0.28	0.46
	Mean	0.09	0.35	–		0.11	0.35	–		0.08	0.35	–	
	MOE Mean (BMDL10 of 0.63)	6.6	1.8	–		5.9	1.8	–		7.6	1.8	–	
	MOE Mean (BMDL10 of 1.5)	15.8	4.3	–		14.0	4.3	–		18.0	4.3	–	
	P50	0.07	0.31	–		0.08	0.31	–		0.06	0.31	–	
	MOE P50 (BMDL10 of 0.63)	9.6	2.0	–		8.0	2.0	–		11.0	2.0	–	
	MOE P50 (BMDL10 of 1.5)	22.8	4.8	–		19.1	4.8	–		26.2	4.8	–	
	P95	0.26	0.67	–		0.29	0.65	–		0.22	0.68	–	
	MOE P95 (BMDL10 of 0.63)	2.4	0.9	–		2.1	1.0	–		2.8	0.9	–	
	MOE P95 (BMDL10 of 1.5)	5.8	2.2	–		5.1	2.3	–		6.7	2.2	–	

LB - Lower bound approach, UB - Upper bound approach, Mean - Mean exposure (μg kg^{-1} bw day^{-1}), MOE - Margin of Exposure, BMDL - Benchmark Dose Limit (μg kg^{-1} bw day^{-1}), P50 - Percentile 50 exposure (μg kg^{-1} bw day^{-1}) and P95 - Percentile 95 exposure (μg kg^{-1} bw day^{-1}).
[a] Tap water not included.

'Fish, seafood, amphibians, reptiles and invertebrates' food group, with mean occurrence values of 0.020 (LB) and 0.047 (UB) mg/kg, included the sample with highest Pb level (Bivalve molluscs, 0.26 mg kg^{-1}). All other food groups had mean occurrence values equal or below 0.021 (LB) and 0.024 (UB) mg kg^{-1}.

Table 6 summarises the Pb exposure results by food group and the risk assessment for the 18–74 years old Portuguese segmented by sex. Supplementary information includes data for 'adults' and 'elderly' population groups (Tables 13S, 14S). The estimated UB mean exposure to Pb for the 'overall population' was 0.35 μg kg^{-1} bw day^{-1} (both 'male' and 'female') and P95 (UB) was 0.67 μg kg^{-1} bw day^{-1} ('male' 0.65 and 'female' 0.68). Considering the BMDL01 of 1.5 μg kg^{-1} bw day^{-1}, defined for cardiovascular effects (European Food Safety Authority [EFSA], 2010a), the 'overall population' UB MOEs were 4.3 for mean exposure and 2.2 for P95. On the basis of the BMDL10 of 0.63 μg kg^{-1} bw day^{-1} defined for nephrotoxic effects, UB MOEs were 1.8 to mean exposure, and 0.9 to P95. Based on these findings the possibility of cardiovascular and nephrotoxic effects of Pb cannot be excluded. The main contributors to Pb exposure for the 'overall population' were 'composite dishes', 'fruit and fruit products' and 'grains and grain-based products' food groups at UB. Detailed data on each TDS sample's contribution to Pb exposure is provided in supplementary information (Tables 15S, 16S, 17S). At UB apple was the main contributor, followed by bread, except for the 'elderly' 'male' group where cod was the second contributor. Other foods that mostly contributed to Pb exposure were coffee and wine. Supplementary information (Table 18S) details the contribution of food as eaten to Pb exposure.

Arnich (Arnich et al., 2012) reported a mean exposure to Pb of 0.20 μg kg^{-1} bw day^{-1} and a P95 of 0.35 μg kg^{-1} bw day^{-1} for the adults French population (18–79 years old). MOEs for cardiovascular effects (BMDL01, 1.5 μg kg^{-1} bw day^{-1}) were 8 for mean exposure and 4 for P95. For nephrotoxic effects (BMDL10, 0.63 μg kg^{-1} bw day^{-1}), MOEs were 3 for mean exposure and 2 for P95. The main contributors to Pb exposure were alcoholic beverages (14 %), bread and dried bread products (13 %) and water (11 %).

Mean exposure of Santiago's (Chile) adult population (18–65 years old) to Pb was estimated by Muñoz (Muñoz et al., 2005) as 3.03 μg kg^{-1} bw day^{-1}. The main contributors to Pb exposure were milk products (17.9 %), fruits (17 %), bread (16.1 %) and sugar (12.6 %).

Nasreddine (Nasreddine et al., 2006) estimated a mean Pb exposure of 0.26 μg kg^{-1} bw day^{-1} for the urban adult Lebanese population (25–54 years old). The main contributors to Pb exposure were cereals and cereal-based products (45.3 %), vegetables and potatoes (17.6 %), drinking water (16.2 %), fruits and fruit juices (9.9 %), dairy products (6.4 %) and meat and poultry (4.6 %).

Marín (Marin et al., 2017) reported a Pb mean exposure of 0.21 (LB) and 0.44 (UB) μg kg^{-1} bw day^{-1} estimated for the adult population (16–95 years old) of Valencia region, Spain. Cereals food group was the main contributor to Pb exposure (49 %, LB), mainly due to high bread consumption.

Lead mean exposure by the Czechia's adult population was reported as 0.11 μg kg^{-1} bw day^{-1}. MOEs were 13.9 for cardiovascular effects (BMDL01, 1.5 μg kg^{-1} bw day^{-1}) and 3 for nephrotoxic effects (BMDL10, 0.63 μg kg^{-1} bw day^{-1}). Coffee, tea, fine bakery, rice, pasta, wine and beer were the main contributors to Pb exposure (Sokolov et al., 2022).

Exposure of Germany's adult population (14–79 years old) to Pb was estimated by Kolbaum (Kolbaum et al., 2019) as 0.07 (LB) and 0.08 (UB) μg kg^{-1} bw day^{-1} for the mean and as 0.16 (LB) and 0.17 (UB) for the P95. Wine, vegetable salad and multigrain bread were the main contributors to Pb exposure.

The exposure of Sub-Saharan Africa populations to Pb was estimated by Ingenbleek (Ingenbleek et al., 2020). Lead exposure (μg kg^{-1} bw day^{-1}) ranged from 0.20 (Mali, Sikasso) to 1.24 (Nigeria, Kono) for the mean, and from 0.53 (Cameroon, North region) to 2.65 (Nigeria, Kono) at P95. The main contributors to Pb dietary exposure were sorghum, millet, and cassava.

The estimated mean Pb exposure for the overall Portuguese population (0.35 μg kg^{-1} bw day^{-1}) was within the range of previous studies (0.08–3.03 μg kg^{-1} bw day^{-1}), as was the P95 exposure. Across all studied populations, cereal-based products, particularly bread, were the primary contributors to Pb exposure.

3.4. Final considerations

In summary, the results of this study are consistent with other international assessments of Cd and Pb exposure. However, for iAs, while two studies reported similar findings, three others (Italy, Spain, Germany) showed mean exposures approximately three times lower. MOEs lower or close to one found for the Portuguese population regarding iAs emphasizes the need to refine exposure assessments by using arsenic speciation data, rather than relying on EFSA's assumptions to convert tAs into iAs. This is further supported by the lower exposure levels observed in studies from Italy, Valencia (Spain), and Germany, which utilized arsenic speciation data.

3.5. Contribution of tap water

The contribution of tap water to the Portuguese 'overall population' exposure to iAs, Cd, and Pb was estimated as 0.0037 μg kg^{-1} bw day^{-1}, 0.016 μg kg^{-1} bw $week^{-1}$, and 0.011 μg kg^{-1} bw day^{-1}, respectively, accounting for roughly 1 %, 0.2 %, and 3 % of total exposure. Thus, tap water's contribution is negligible.

3.6. Strengths and limitations

TDSs are highly effective for assessing chronic exposure to chemicals, as they provide comprehensive coverage of the entire diet at relatively low costs through the pooling of food samples. Additionally, the foods are analysed after being prepared in line with typical consumption habits, ensuring that the detected chemical levels closely reflect what consumers actually ingest. Furthermore, using a harmonised TDS methodology generates data that enables the assessment of time trends and cross-country comparisons of dietary exposure to chemical substances in populations. However, uncertainties at various stages of this study, potentially led to overestimation or underestimation of exposure. Since the consumption data was collected in 2009, it is possible that food habits have changed over time. Additionally, another source of uncertainty stemmed from the fact that only one day 24 h recall was recorded, which may have led to an overestimation of the higher percentiles of chronic exposure. Uncertainties such as the high left-censored data rate for Cd and Pb and relying on assumptions to estimate iAs from tAs may have affected the accuracy of the exposure assessment or of the contribution of specific foods to exposure.

4. Conclusions

This study established baseline dietary exposure to iAs, Cd, and Pb in the Portuguese population aged 18 to 74. The findings indicate that potential adverse health effects cannot be excluded and should be carefully evaluated by public health risk managers considering the mean exposure obtained for iAs and P95 exposure for all elements. The food groups 'composite dishes', 'grains and grain-based products' and 'fish, seafood, amphibians, reptiles and invertebrates' each contributed with more than 12 % to the UB exposure of the 'overall population' to iAs, Cd and Pb. Of the top five food contributors, bread was the only common source of exposure to all three elements. Implementing a regular national TDS using Vasco's harmonised methodology (Vasco et al., 2021), based on current national food consumption survey data, would enable accurate trend analysis of exposure to iAs, Cd, and Pb to inform risk management efforts for protecting public health. This regular TDS should also cover younger age groups, given their higher food intake relative to body weight and the concerns raised about their exposure levels in other studies (Jean et al., 2018) (Sirot et al., 2018) (Mhungu et al., 2023). Additionally, efforts should be made to reduce uncertainties in the assessment of exposure to Cd, Pb, and As in TDS by lowering analytical limits for Cd and Pb, conducting speciation analysis for As, and utilizing two-day 24 h recall food consumption data.

CRediT authorship contribution statement

Elsa Vasco: Writing – original draft, Visualization, Investigation, Formal analysis, Conceptualization. **M. Graça Dias:** Writing – review & editing, Conceptualization. **Luísa Oliveira:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2024.144003.

Data availability

Data will be made available on request.

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