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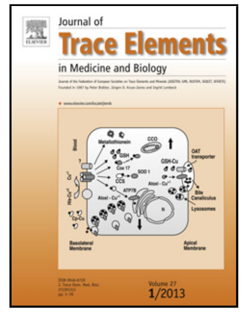

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Title: Aluminum and tin: Food contamination and dietary intake in an Italian population

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Highlights

- We estimated dietary intake of aluminum and tin in an adult Italian population sample.
- Legumes, sweets, and cereals show the highest contents of aluminum, whereas sweets, meat, and fish contain large amounts of tin.
- The estimated median aluminum and tin intake are 4.1 mg/day and 66.8 µg/day, respectively.
- Major intake contributors are beverages, leafy vegetables, and cereals for aluminum, and tomatoes, fruit and processed meat for tin.

Abstract

Aluminum and tin are ubiquitous in the environment. In normal biological systems, however, they are present only in trace amounts and have no recognized biological functions in humans. High exposure to these metals can result in adverse health effects such as neurodegenerative diseases. In non-occupationally exposed subjects, diet is the primary source of exposure. In this study, we aimed at estimating dietary aluminum and tin intake in an Italian adult population.

We measured aluminum and tin concentrations through inductively-coupled plasma mass spectrometry in 908 food samples. We also estimated dietary intake of these two metals, by using a validated semi-quantitative food-frequency questionnaire administered to 719 subjects (319 men and 400 women) recruited from the general population of the Emilia Romagna region, Northern Italy.

We found the highest aluminum levels in legumes, sweets, and cereals, while the highest tin levels were in sweets, meat and seafood. The estimated median daily dietary intake of aluminum was 4.1 mg/day (Interquartile range - IQR: 3.3-5.2), with a major contribution from beverages (28.6%), cereals (16.9%), and leafy vegetables (15.2%). As for tin, we estimated a median intake of 66.8 µg/day (IQR: 46.7-93.7), with a major contribution from vegetables (mainly tomatoes) (24.9%), fruit (15.5%), aged cheese (12.2%) and processed meat (10.4%).

This study provides an updated estimate of the dietary intake of aluminum and tin in a Northern-Italy adult population, based on data from a validated food-frequency questionnaire. The intake determined for this population does not exceed the established thresholds of tolerable intake.

Keywords: aluminum, tin, dietary intake, food contamination, food safety.

Introduction

Aluminum and tin are ubiquitous in the environment. Both metals have no known biological function in humans, while a markedly increased exposure can result in toxic effects involving several systems [1, 2]. Non-occupational exposure to aluminum has been linked to adverse effects, particularly anemia, bone disease, and dialysis encephalopathy [3, 4]. Moreover, overexposure to such trace elements including high chronic aluminum exposure has been related to an increased risk of neurodegenerative diseases, including Alzheimer's Dementia, Parkinson's disease, and amyotrophic lateral sclerosis [5-14].
With reference to tin, its different chemical species play a major role in the assessment of adverse health effects [15]. High exposure to inorganic compounds mainly leads to gastrointestinal problems [15-17]. On the other hand, exposure the organotin compounds, such as trimethyltin and triethyltin, has been related to gastrointestinal, metabolic and neurological adverse effects [16, 18-20].

In non-occupationally exposed individuals, the primary sources of exposure to these metals is diet. In industrialized societies, more specifically, the use of food packaging materials or food additives enhancing food properties has markedly increased exposure to these metals [21-24]. In particular, aluminum compounds may be added during food industrial transformation to produce convenience foods. Aluminum containers are thus widely used to cook, freeze or wrap foods through aluminum foil [25-28], while food additives containing aluminum are used as coloring and/or anticaking agents [29-31].

Similarly, the main use of tin is for the manufacture of cans and containers, typically as tinplate [16, 21, 32]. Another source of exposure to tin, chiefly for organic compounds, is its use as a stabilizer for polyvinyl chloride [33, 34]. As a consequence, direct contact of tin with foods for general populations may result in migration from the inside wall of cans and other packaging materials to edible contents.

For these reasons, dietary intake, particularly through convenience foods produced through food industrialized transformation and/or packaging processes, is generally the most important source of exposure to these metals [16, 29, 35, 36]. In this study, we sought to assess dietary aluminum and tin intake in order to provide an updated estimate of their exposure in the general adult population of the Emilia Romagna Region, Northern Italy.

Methods

Study population and assessment of dietary habits

We assessed the dietary habits of a sample population from Northern Italy. Detailed information of participant identification and recruitment has been previously reported [37, 38]. In brief, we identified 2,825 potentially eligible subjects from the population database of the Emilia-Romagna Region residents. These were from the Bologna, Ferrara, Modena, Parma, and Reggio Emilia provinces. A list was compiled by accessing the National Health Service directory. Of the eligible subjects, 747 (26.4%) agreed to study participation. After providing written informed consent, they returned a lifestyle and food frequency questionnaire (FFQ), which we had mailed to them. We excluded twenty-eight individuals from the final analysis because of incomplete data or extreme values (ratio of total energy intake: basal metabolic rate lower than the 0.5th percentile or higher than the 99.5th percentile) derived from the FFQ. The final sample comprised 719 participants aged 18-87 years (mean 55 years), including 319 men (mean age 59 years) and 400 women (mean age 52 years) [39, 40]. With regard to the evaluation of dietary habits, we assessed food and nutrient intake using a validated semi-quantitative FFQ specifically developed for the Central-Northern Italy population [41, 42] within the EPIC (European Prospective Investigation into Cancer and Nutrition) project. The FFQ was designed to estimate frequency and amount of consumption of 188 food items over the previous year, also using photos of serving sizes to ensure proper completion by participants. The median energy intake was 1906 kcal/day (interquartile range-IQR: 1538-2364) in all subjects, 2024 kcal/day (IQR: 1649-2462) in men and 1800 kcal/day (IQR: 1455-2296) in women, respectively.

Food collection and analysis

We determined aluminum and tin contents of food by characterizing the usual diet of a sample from the Emilia-Romagna population. As previously described in detail, we selected food consumed by a random sample of this population. Furthermore, we bought samples of these food items in markets and community canteens between October 2016 and February 2017 [43]. We strove to avoid cross-contamination with metals and other trace elements from food containers, by using plastic tubes or jars as well as plastic cutlery or stainless-steel knives during food collection and handling. With a clean stainless-steel knife, we then cut solid food samples by collecting specimens from the plate in six different points. For the purpose of subsequent analysis, these samples were homogenised separately in a food blender equipped with a stainless-steel blade. We placed portions of the samples (0.5 g) in quartz containers previously washed with MilliQ water (MilliQPlus, Millipore, MA, USA) and HNO_3. Food samples were liquid-ashed with 10 ml solution (5 ml HNO_3 + 5 ml H_2O) in a microwave digestion system (Discover SP-D, CEM Corporation, NC, USA). We then stored them in plastic tubes and diluted them to 50 ml with deionised water before analysis. We performed trace element determination using an inductively coupled plasma-mass spectrometer (Agilent 7500ce, Agilent Technologies, CA, USA). We performed all analyses in duplicate and implemented quality controls as previously described [44, 45]. Limits of quantification amounted to 0.5 µg/kg for aluminum and 0.02 µg/kg for tin, corresponding to limits of detection (LOD) of 0.17 µg/kg and 0.007 µg/kg, respectively. Values below the LOD were set equal to LOD/2.

We reported aluminum and tin concentration according to food consumption patterns and food categories typical of this Italian population, as assessed through the FFQ [46, 47]. Our final list of main food categories included the following items: cereals and cereal products (pasta and other grains: durum wheat pasta, whole grain pasta, filled pasta, egg pasta, and pasta containing other grains such as barley and spelt; rice: both white (Carnaroli, Arborio) and brown; bread: white bread; whole-wheat bread; multigrain bread; crackers, crispbread and salty snacks, including maltose crackers, whole-wheat crackers and taralli), meat and meat products (red meat: beef, calf, pork, horse and game, including lean and fatty cuts; white meat: chicken, turkey, rabbit; processed meat: several types of salt-cured meat, especially those traditional of Emilia-Romagna including ham, sausage, salami, mortadella, bacon, etc.; offal: liver, heart, spleen); milk and dairy products, e.g. milk and yogurt (both whole milk, reduced-fat or skim milk, and fruit yogurts), fresh cheese including cream cheese, ricotta, stracchino, mozzarella, etc., aged cheese such as Parmigiano-Reggiano, Grana Padano, and Emmental, etc.); eggs, fish and seafood (preserved fish such as smoked salmon, and tinned tuna or mackerel; non-piscivorous fish such as sole and plaice; piscivorous fish including tuna, swordfish and salmon; crustaceans and molluscs such as shrimp, octopus, cuttlefish, mussels and clams); vegetables (leafy vegetables: various types of salad, spinach, chard, etc.; other vegetables: sweet, pepper, fennel, celery, artichoke, etc.; tomatoes; root vegetables: carrot, turnip; cabbage: green, Savoy, red, and Brussels sprouts; mushrooms, both cultivated and wild; onion and garlic); legumes (kidney beans, peas, lentils, etc.); potatoes (mainly regular but also sweet potatoes); fresh fruit (citrus fruit: orange, mandarin, grapefruit; other fruit: apple, pea, apricot, plum, banana, etc.); dried fruit and seeds (raisins, prunes, walnuts, hazelnuts, etc.); sweets including chocolate and cakes (sugar, non-chocolate confectionery with honey, jam and candies; chocolate and chocolate-based candy bars; ice cream: chocolate, non-chocolate and fruit ice cream; cakes, pies and pastries; biscuits and dried cakes); oils and fats (vegetables oils and non-olive oil including margarine; olive oil; butter and other animal fats), and beverages (coffee and tea: caffeinated and decaffeinated coffee made using moka pots as well as espresso machines, black and green tea; wine: several types of red and white wine mainly from Emilia-Romagna but also from other Italian regions; aperitif wines and beers; spirits and liqueurs such as rum, whiskey and limoncello; fruit juices: orange juice and non-citrus juices; soft drinks: cola-type drinks, soda-type drinks, orangeade, etc.).

2.3 Estimate of metal dietary intake

We combined data from the measurement of metals in foods and the dietary habits assessed by the FFQ, in order to compute total daily metal intake by using the equation presented below.

$$Daily\ dietary\ exposure\ \left(\frac{\mu g}{day}\right) = \sum \frac{element\ food\ content\ \left(\frac{\mu g}{kg}\right) \times food\ intake\ \left(\frac{g}{day}\right)}{1000}$$

We multiplied contents measured in food (µg/kg) with intake as estimated through the FFQ (g/day). Accordingly, we estimated daily dietary aluminum and tin intake for the diet as a whole and for each food category, by reporting median and interquartile ranges (IQR). For the estimate of dietary intake by body weight (bw), we divided by participant weight (kg). Compared to other studies, during conversion from µg/day to µg/kg of bw per day or week, we considered a body weight of 60 kg for the purpose of our calculations, in accordance with guidelines from international agencies [29, 36, 48].

Results

Table 1 reports details about dietary habits in the study population. In general, we found a comparable intake of main food categories in both sexes, with the partial exception of a higher intake of cereal products, meat and beverages (mainly due to wine intake) in men. Conversely, women showed higher consumption of milk and yogurt, coffee and tea and slightly higher consumption of citrus fruits.

The analysis of aluminum and tin concentration in food samples is presented in Table 2 according to main food categories and subgroups. All values were above the LOD for aluminum. As far as tin is concerned, in contrast, 61 specimens (6.7% of total sampled foods) showed concentrations below the LOD (Supplemental Table S1). Considering the main food categories, the highest aluminum contents were found in legumes ahead of sweets (mainly in chocolate-based products) and cereals, mainly due to higher contents in bread, crackers and crispbread. Results from the subgroup analysis showed high aluminum concentrations in processed meat, crustaceans/molluscs, leafy vegetables, dried fruit and coffee/tea, compared with the median value of the main food categories. As for tin, likewise, the highest contents were found in sweets and meat, ahead of dried fruit and fish, mainly preserved and canned samples. The subgroup analysis demonstrated high tin contents in bread, mushrooms and leafy vegetables.

The dietary intake of the two metals is presented in Table 3. The median total aluminum intake was 4.1 mg/day (IQR: 3.3-5.2 mg/day), corresponding to 407.43 (IQR: 319.75-524.05) µg/kg of bw/week or 58.20 (IQR: 45.68-74.86) µg/kg bw/day. The food categories which mainly contributed to total intake were vegetables (25.2%), mainly leafy vegetables (15.2%) and tomatoes (3.8%), followed by beverages (28.6%), mainly coffee and tea (24.9%), and finally cereal products (16.9%) (Figure 1). The median tin intake was 66.80 µg/day (IQR: 46.72-93.66 µg/day), corresponding to 6.70 (IQR: 4.85-9.25) µg/kg bw/week or 0.96 (IQR: 0.69-1.32) µg/kg bw/day. The main contribution to total tin intake was made by vegetables (30.3%) – particularly tomatoes (24.9%) – and fresh fruit (15.5%), with a substantial contribution from aged cheese (12.2%), processed meat (10.4%) and other animal fats (4.1%) as well (Figure 1). Estimates stratified by sex generally showed comparable results, except for the slightly higher intake of tin in men related to a higher intake of vegetables, mainly tomatoes (Supplemental Tables S2-S3 and Supplemental Figures S1-S2).

Finally in Table 4, we compared the estimated aluminum and tin intake in our population with dietary intake and tolerable upper intake levels reported by international agencies for European [29, 36] and worldwide populations [24, 49, 50]. While median and upper (95th percentile) values of aluminum intake fall in the range reported by other European and non-European populations, the corresponding figures for tin are below the range of intakes reported in other populations. Finally, Table 4 demonstrated that for both metals, the tolerable levels established by EFSA and WHO are not exceeded even considering the upper estimated intake of the two metals.

Discussion

In the context of a Northern Italian population, we assessed dietary intake of aluminum and tin by determining their concentration in a large number of food samples. Aluminum intake fell within the range of values reported by EFSA for European populations [29], consistent with other studies [26, 51-54]. However, other studies have yielded higher [55-61] or lower values [25, 27, 62-66]. Variations in dietary exposure may be accounted for by differences in soil composition in the regions where food is produced or in individual dietary patterns. The population investigated in our study was characterized by generally high adherence to the Mediterranean diet [38], as compared with other populations with different dietary habits [67]. This may have influenced amount and frequency of consumption of the kind of foods that we observed to be major contributors to aluminum and tin intake. In particular, cereals and vegetables were found to be major contributors to aluminum intake, consistent with other studies on European populations [56, 61, 65, 68, 69]. We found the highest levels of aluminum in legumes, which were shown to naturally accumulate large amount of the metal [56, 70, 71]. We also found high levels of aluminum in cereal products and sweets, especially in bread products (including crackers and rusks), cakes and biscuits. Such high levels might be due both to the use of food additives, e.g. leavening agents such as acid sodium aluminum phosphate in cereal products, or anti-caking agents such as sodium aluminum silicate in cake mixes and dried products [31, 56, 72], and to generally very high aluminum contents in cocoa powder and chocolate [70].

As expected for vegetables, they showed fairly marked variation in aluminum contents. Thus, higher levels were observed in leafy vegetables [57, 69, 70] and tomatoes, especially in canned samples. This may be due to migration from containers and the low pH of foodstuffs as well as higher temperatures during storage time [73-76]. Furthermore, beverages (chiefly coffee and tea) were an important contributor to aluminum intake, consistent with previous studies [60, 69]. Consumption of coffee and tea may influence overall aluminum intake considerably, due to substantial variation in consumption and metal content [77, 78]. Interestingly, the analysis of aluminum distribution in different parts of tea plants also showed higher accumulation in mature leaves. On the other hand, lower values were detected in roots and young leaves [79], thus explaining the high amount of aluminum in tea samples. Consistent with other studies, we found generally high aluminum contents in aged cheese, arguably related to aluminum additives especially in (semi-)hard and industrially processed samples [31, 53, 56, 80].

Low levels of dietary tin intake were generally reported for our population, compared with others [36, 59, 61, 63, 65, 66, 81-83]. Overall, tin intake levels were similar to or slightly higher than values reported for few other countries [32, 84, 85]. Our findings indicated that vegetables and fruits were the main contributors to tin intake. This could be due to contamination from can containers [16, 23], as shown by the high levels found in canned tomatoes. Both the high content and the high consumption of this food item resulted in a major contribution to dietary intake, tomatoes being the highest source among all vegetables. Similarly to aluminum, the migration of this metal from packaging materials could have been favored by the acidity of this type of food [86, 87].

By contrast, the contribution of tinned fish in our population was not relevant compared with that observed in other populations. This may be due to varying concentrations of tin as well as lower daily intake of this type of preserved foodstuff [32, 88]. The relevant role of processed meat and aged cheese to overall tin intake was due to high metal contents in these foods, probably correlated with industrial processing, use of higher amounts of food additives and/or migration from packaging materials [16, 36].

Overall, the risk assessment of both aluminum and tin intake in our study population indicated that safety levels were not exceeded. This view is based on the upper levels established by FAO/WHO and EFSA, considering neither median nor upper levels of estimated intake for both metals.

A strength of our study lies in the collection and analysis of a large number of food samples. In order to identify food relevant to the dietary habits of our population, we organized sample collection according to main categories, as in previous studies on dietary intake of other trace elements [44, 45]. We also followed guidelines from total diet studies, thus improving the validity of our findings and comparability with other studies [89]. Moreover, we aimed at mimicking a close-to-real mode of food collection in the general population through the purchase of food samples in both supermarkets and groceries of two provinces of Emilia Romagna (i.e. Modena and Reggio Emilia). However, we could not assess other kinds of possible contamination between local and imported foodstuff, since we did not evaluate the geographical origin of products.

Another strength of the study is the estimation of dietary habits in a large population sample through a detailed and validated food frequency questionnaire specifically developed for a Northern Italian population [41, 42].

A first limitation is that we assessed dietary habits in an adult population. Therefore, our findings may not be applicable to other population subgroups, such as children and adolescents.

We also carried out no speciation analysis for the two elements, hampering intake evaluation for different species characterized by heterogeneous or sometimes opposite health effects. With reference to tin, however, a study carried out using duplicate diet samples demonstrated relatively low levels of organotin compounds in sampled foods, and estimated daily intake was lower than the established tolerable daily intake [90].

In addition, we did not assess metal bioavailability. The average fraction of aluminum absorbed from a given dose is estimated to be low, subject though it is to wide variation (from less than 1% to 24% in humans). This depends on a number of factors: aluminum intake, intraluminal presence of substances with structural similarity (as for essential metals such as calcium and iron), or with binding potential such as that of citrate, as well as intraluminal pH, iron status and calcium status [72, 91-93]. As regards tin, metallic ions are practically insoluble and do not break through gastrointestinal barriers. As a result, they are mainly absorbed as organotin compounds [90, 94], for the bioavailability of which pH is considered an important factor [21]. In this regard, a bioconcentration study indicated that, as pH increases, the uptake of organotin compounds increases [95].

Furthermore, we did evaluate the health status of the study subjects, including possible malabsorption disease affecting metal uptake. With regard to overall aluminum intake, finally, we did not evaluate the influence of other dietary sources of the metal such as drinking water [96] and aluminum-containing medications, above all antacid drugs likely to increase daily aluminum intake by up to several grams [97, 98]. Moreover, our assessment was limited to the dietary intake of the two metals, while evaluating overall exposure in humans requires that additional sources be considered, such as inhalation and dermal contact. However, with the exception of rare occupational exposure or severe environmental pollution, these sources generally play a negligible role in determining overall exposure, compared with diet [21, 22].

Conclusions

In the Italian area investigated in the present study, legumes, sweets, and cereals presented the highest aluminum contents, whereas the highest tin levels were found in sweets, meat, fish and seafood. Overall, aluminum and tin intake was lower compared with the tolerable intake, as established by international agencies.

Founding

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Declarations of interest: none.

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Table 1. Food intake (g/day) according to different categories for the whole study population and by sex.

Foods	All (N=719)	Men (N=319)	Women (N=400)
	Mean (St. Dev.)	Mean (St. Dev.)	Mean (St. Dev.)
Cereals and cereal products	**188.5 (99.3)**	**206.4 (104.6)**	**174.2 (92.5)**
Pasta, other grain	57.1 (40.7)	70.1 (45.3)	46.8 (33.2)
Rice	5.6 (7.6)	6.2 (8.7)	5.0 (6.7)
Bread	78.3 (72.0)	83.9 (73.4)	73.9 (70.6)
Crackers, crispbread, salty snacks	47.5 (34.2)	46.2 (35.7)	48.5 (33.0)
Meat and meat products	**128.4 (70.9)**	**142.4 (73.5)**	**117.2 (66.8)**
Red meat	67.4 (45.1)	76.8 (49.6)	59.9 (39.7)
White meat	29.7 (26.2)	31.2 (25.6)	28.4 (26.6)
Processed meat	29.5 (24.9)	32.3 (27.2)	27.3 (22.7)
Offal	1.8 (4.6)	2.1 (5.2)	1.6 (4.0)
Milk and dairy products	**229.4 (216.1)**	**201.8 (191.7)**	**251.5 (231.6)**
Milk and yogurt	188.9 (209.3)	159.4 (188.3)	212.5 (222.0)
Cheese	40.5 (33.9)	42.4 (35.5)	39.0 (32.5)
Fresh cheese	14.6 (19.0)	11.9 (15.5)	16.8 (21.1)
Aged cheese	25.9 (24.0)	30.5 (27.6)	22.2 (20.1)
Eggs	**15.1 (11.4)**	**14.6 (11.2)**	**15.4 (11.5)**
Fish and seafood	**35.1 (28.1)**	**35.5 (26.8)**	**34.9 (29.0)**
Fish	27.9 (23.1)	28.5 (22.6)	27.5 (23.6)
Preserved and tinned fish	9.3 (10.7)	10.2 (9.7)	8.5 (11.4)
Non-piscivorous fish	10.4 (12.3)	10.8 (13.1)	10.2 (11.6)
Piscivorous fish	8.2 (11.7)	7.5 (10.7)	8.7 (12.4)
Crustaceans and molluscs	7.2 (10.1)	7.0 (9.4)	7.4 (10.6)
All vegetables	**160.7 (94.7)**	**156.6 (88.8)**	**163.9 (99.2)**
Leafy vegetables	31.6 (25.6)	29.4 (22.8)	33.3 (27.5)
Other vegetables	27.3 (20.0)	24.0 (17.0)	29.9 (21.8)
Tomatoes	63.4 (50.8)	67.0 (52.0)	60.5 (49.7)
Root vegetables	14.2 (19.7)	11.0 (16.7)	16.8 (21.4)
Cabbage	4.2 (7.1)	3.8 (8.0)	4.4 (6.2)
Mushrooms	2.5 (4.0)	2.5 (4.2)	2.5 (3.8)
Onion and garlic	17.5 (21.9)	18.9 (22.0)	16.5 (21.8)
Legumes	**18.7 (18.6)**	**19.5 (19.3)**	**18.1 (18.1)**
Potatoes	**24.5 (24.2)**	**25.5 (26.4)**	**23.7 (22.3)**
Fresh fruit	**279.4 (165.7)**	**270.3 (162.8)**	**286.7 (167.8)**
Citrus fruit	216.3 (135.6)	207.0 (132.9)	223.8 (137.5)
All other fruit	63.1 (50.0)	63.3 (49.5)	62.9 (50.5)
Dried fruit, nuts and seeds	**1.7 (3.0)**	**1.8 (2.9)**	**1.6 (3.0)**
Dried fruit	0.4 (1.1)	0.5 (1.2)	0.3 (1.0)
Nuts and seeds	1.3 (2.5)	1.3 (2.3)	1.2 (2.8)
Sweets, chocolate, cakes, etc.	**86.5 (73.8)**	**82.9 (77.3)**	**89.4 (70.8)**
Sugar, non-chocolate confectionery	18.5 (22.5)	17.3 (18.2)	19.4 (25.4)
Chocolate, candy bars, etc.	5.3 (8.7)	4.9 (8.5)	5.6 (8.9)
Ice cream	13.9 (15.6)	13.6 (16.9)	14.2 (14.5)
Cakes, pies and pastries	35.1 (54.3)	34.0 (61.6)	36.0 (47.7)
Biscuits, dried cakes	13.7 (17.1)	13.1 (17.2)	14.2 (17.1)
Oils and fats	**27.2 (13.5)**	**27.9 (12.7)**	**26.7 (14.1)**
Vegetable fats and oils (non-olive)	2.4 (5.7)	2.6 (6.1)	2.2 (5.4)
Olive oil	22.0 (12.6)	22.0 (12.1)	22.1 (12.9)
Butter and other animal fats	2.8 (3.5)	3.3 (4.1)	2.4 (2.9)
Beverages	**429.5 (341.3)**	**479.5 (322.5)**	**389.7 (350.8)**
Coffee and tea	148.4 (160.3)	120.6 (109.3)	170.6 (188.7)
Wines	126.0 (164.1)	191.4 (190.0)	73.9 (116.3)
Red wine	74.5 (119.0)	116.6 (144.1)	41.0 (80.0)
White wine	51.5 (102.0)	74.8 (123.8)	32.9 (75.8)
Aperitif wines and beers	41.4 (117.5)	49.9 (123.8)	34.6 (126.0)
Spirits and liqueurs	3.1 (11.0)	5.3 (14.9)	1.3 (5.7)
Fruit juices	66.4 (135.0)	67.6 (126.7)	65.5 (141.4)
Soft drinks	44.2 (118.2)	44.7 (100.7)	43.8 (130.6)

Table 2. Aluminum and tin concentrations (μg/kg) in food samples by food category. Median (50th) and interquartile range (IQR) are reported.

Foods	N	Aluminum (μg/kg)		Tin (μg/kg)	
		50th	(IQR)	50th	(IQR)
Cereals and cereal products	**112**	**2470.36**	**(1466.80-5685.81)**	**3.60**	**(0.97-8.58)**
Pasta, other grain	41	1602.21	(960.83-2498.67)	2.97	(0.78-5.53)
Rice	8	276.12	(120.03-439.64)	0.52	(0.13-4.09)
Bread	42	3555.10	(2257.65-7756.52)	6.81	(3.60-11.38)
Crackers, crispbread, salty snacks	21	3515.11	(2607.98-5691.34)	2.66	(1.21-4.91)
Meat and meat products	**86**	**584.59**	**(367.16-1181.77)**	**5.73**	**(3.00-12.37)**
Red meat	28	478.27	(339.87-801.01)	5.90	(3.14-8.48)
White meat	12	386.29	(272.67-493.60)	3.08	(2.32-6.44)
Processed meat	36	1227.92	(750.19-2004.49)	8.54	(4.35-17.99)
Offal	10	334.31	(80.83-494.37)	4.49	(1.91-10.22)
Milk and dairy products	**72**	**442.54**	**(218.02-1117.32)**	**3.28**	**(1.53-6.11)**
Milk and yogurt	13	56.84	(29.30-63.00)	0.37	(0.17-0.88)
Cheese	59	647.04	(322.31-1622.85)	3.74	(2.39-7.56)
Fresh cheese	17	794.34	(312.66-1215.75)	3.00	(1.40-4.89)
Aged cheese	42	613.66	(336.53-1622.85)	5.05	(2.58-8.09)
Eggs	**9**	**127.59**	**(77.54-168.41)**	**0.45**	**(0.26-0.97)**
Fish and seafood	**62**	**973.05**	**(432.43-2948.74)**	**4.31**	**(2.26-14.55)**
Fish	41	530.75	(312.13-1181.82)	4.88	(2.62-16.64)
Preserved and tinned fish	9	589.84	(274.58-1088.94)	10.41	(3.82-15.56)
Non-piscivorous fish	15	568.86	(269.82-1584.56)	3.96	(1.51-11.35)
Piscivorous fish	17	478.79	(393.96-1012.41)	6.74	(2.37-23.37)
Crustaceans and molluscs	21	4258.89	(1633.63-5995.71)	3.50	(1.83-8.92)
All vegetables	**201**	**858.22**	**(283.84-2732.59)**	**3.79**	**(1.26-13.01)**
Leafy vegetables	40	9039.40	(1921.10-40667.56)	7.36	(4.14-18.92)
Other vegetables	63	815.34	(286.17-2121.31)	3.22	(1.14-17.70)
Tomatoes	19	356.47	(155.76-956.53)	2.94	(0.97-12.12)
Root vegetables	14	1358.58	(283.84-2428.15)	2.28	(1.12-5.34)
Cabbage	28	666.84	(290.25-1432.59)	4.03	(2.07-8.65)
Mushrooms	5	1855.89	(1484.48-2339.60)	17.09	(17.02-49.23)
Onion and garlic	32	242.67	(146.24-674.40)	1.68	(0.77-3.31)
Legumes	**43**	**7370.23**	**(1231.42-15515.08)**	**1.65**	**(0.00-4.99)**
Potatoes	**14**	**471.93**	**(336.16-1225.13)**	**2.19**	**(1.32-4.89)**
Fresh fruit	**60**	**353.20**	**(177.85-706.13)**	**1.58**	**(1.04-2.52)**
Citrus fruit	12	102.51	(60.22-187.47)	1.72	(1.30-5.46)
All other fruit	48	437.92	(235.44-846.96)	1.58	(0.92-2.39)
Dried fruit, nuts and seeds	**45**	**1303.11**	**(571.52-3226.65)**	**4.53**	**(2.37-8.21)**
Dried fruit	8	8318.11	(2737.64-26133.22)	5.43	(2.73-34.94)
Nuts and seeds	37	845.03	(367.23-2535.28)	3.65	(2.31-8.10)
Sweets, chocolate, cakes, etc.	**79**	**4387.24**	**(1349.90-7949.51)**	**6.01**	**(3.14-10.21)**
Sugar, non-chocolate confectionery	8	3555.18	(644.19-7552.50)	9.73	(5.70-14.62)
Chocolate, candy bars, etc.	21	14050.13	(7830.60-26330.64)	8.50	(4.89-13.25)
Ice cream	5	368.01	(90.60-627.10)	2.29	(1.84-2.40)
Cakes, pies and pastries	30	2339.53	(866.36-5138.26)	4.20	(1.78-8.33)
Biscuits, dried cakes	15	4436.53	(2584.85-5335.13)	7.19	(4.79-13.85)
Oils and fats	**23**	**308.76**	**(167.71-449.64)**	**2.04**	**(1.00-39.95)**
Vegetable fats and oils (non-olive)	12	284.61	(185.04-496.17)	6.62	(1.21-20.89)
Olive oil	4	238.54	(48.57-589.19)	1.63	(0.99-22.43)
Butter and other animal fats	7	314.81	(193.24-449.64)	1.90	(0.14-5.30)
Beverages	**102**	**364.73**	**(91.96-885.03)**	**1.09**	**(0.45-2.84)**
Coffee and tea	8	2411.92	(597.90-8213.29)	1.81	(0.16-7.49)
Wines	50	564.51	(324.03-978.96)	1.17	(0.60-1.99)
Red wine	27	546.80	(324.03-843.16)	1.72	(1.16-3.30)
White wine	23	807.22	(227.28-1019.64)	0.63	(0.45-1.11)
Aperitif wines and beers	8	102.75	(30.95-523.99)	0.95	(0.56-1.91)
Spirits and liqueurs	21	73.14	(33.44-167.52)	3.58	(0.60-6.23)
Fruit juices	8	71.78	(42.07-99.46)	0.39	(0.08-0.71)
Soft drinks	7	282.42	(21.93-1122.58)	0.21	(0.11-3.38)

Table 3. Estimated aluminum and tin intake (µg/day) by food category. Values of median (50th), interquartile range (IQR), and 95th percentile are reported.

Foods	Aluminum (µg/day) 50th	Aluminum (µg/day) (IQR)	Aluminum (µg/day) 95th	Tin (µg/day) 50th	Tin (µg/day) (IQR)	Tin (µg/day) 95th
Total intake (µg/day)	*4103.98*	*(3309.68-5162.74)*	*7226.66*	*66.80*	*(46.72-93.66)*	*147.67*
Cereals and cereal products	**693.05**	**(430.42-1031.53)**	**1592.24**	**2.46**	**(1.60-3.46)**	**5.26**
Pasta, other grain	194.72	(111.94-302.00)	525.48	1.19	(0.68-1.84)	3.20
Rice	1.14	(0.36-1.99)	6.31	0.02	(0.01-0.04)	0.11
Bread	337.62	(111.98-611.09)	1157.47	0.77	(0.25-1.39)	2.62
Crackers, crispbread, salty snacks	88.53	(51.95-141.85)	263.03	0.19	(0.10-0.34)	0.65
Meat and meat products	**90.95**	**(60.16-127.91)**	**224.74**	**7.67**	**(4.36-12.65)**	**24.88**
Red meat	31.77	(17.70-47.52)	80.68	0.52	(0.29-0.81)	1.40
White meat	10.00	(4.93-18.51)	35.90	0.13	(0.06-0.23)	0.43
Processed meat	40.18	(21.39-67.65)	135.31	6.96	(3.71-11.72)	23.44
Offal	0.00	(0.00-0.57)	2.84	0.00	(0.00-0.02)	0.09
Milk and dairy products	**52.96**	**(31.81-78.90)**	**146.82**	**8.29**	**(4.08-14.78)**	**28.24**
Milk and yogurt	9.37	(2.50-16.12)	34.03	0.09	(0.03-0.15)	0.33
Cheese	40.93	(22.06-67.41)	129.52	8.14	(4.03-14.72)	28.06
Fresh cheese	9.79	(3.26-19.57)	51.32	0.03	(0.01-0.07)	0.18
Aged cheese	26.31	(13.02-47.79)	91.55	8.06	(3.99-14.65)	28.06
Eggs	**1.77**	**(0.96-2.78)**	**4.36**	**0.01**	**(0.00-0.01)**	**0.02**
Fish and seafood	**68.38**	**(31.53-170.46)**	**409.74**	**0.75**	**(0.40-1.22)**	**2.41**
Fish	23.83	(11.54-36.94)	80.34	0.71	(0.35-1.15)	2.35
Preserved and tinned fish	5.63	(2.61-11.27)	22.96	0.48	(0.20-1.01)	2.03
Non-piscivorous fish	8.47	(2.03-18.44)	46.71	0.05	(0.01-0.12)	0.34
Piscivorous fish	3.60	(0.39-10.16)	27.94	0.06	(0.01-0.20)	0.58
Crustaceans and molluscs	39.08	(5.58-130.51)	376.91	0.02	(0.00-0.07)	0.16
All vegetables	**1032.95**	**(671.34-1598.96)**	**2831.48**	**20.26**	**(9.29-38.41)**	**81.36**
Leafy vegetables	624.49	(337.32-1100.17)	2061.89	0.32	(0.17-0.56)	1.05
Other vegetables	95.48	(52.80-163.30)	294.07	1.95	(1.08-3.34)	6.00
Tomatoes	156.48	(60.98-317.06)	670.26	16.66	(6.07-35.89)	77.42
Root vegetables	26.85	(6.32-72.65)	202.94	0.04	(0.01-0.11)	0.29
Cabbage	2.64	(0.00-8.06)	25.64	0.01	(0.00-0.04)	0.12
Mushrooms	2.32	(0.58-7.75)	16.66	0.04	(0.01-0.12)	0.26
Onion and garlic	4.80	(2.15-9.59)	28.15	0.03	(0.01-0.06)	0.18
Legumes	**139.59**	**(64.66-259.68)**	**550.15**	**0.21**	**(0.10-0.40)**	**0.84**
Potatoes	**13.66**	**(8.12-24.29)**	**53.13**	**0.19**	**(0.11-0.33)**	**0.73**
Fresh fruit	**145.18**	**(92.04-210.48)**	**348.22**	**10.32**	**(5.17-14.88)**	**27.02**
Citrus fruit	135.74	(83.75-196.77)	322.08	0.70	(0.43-1.02)	1.67
All other fruit	9.13	(4.36-13.08)	24.43	9.54	(4.56-13.68)	25.54
Dried fruit, nuts and seeds	**2.44**	**(1.44-12.22)**	**56.06**	**0.003**	**(0.002-0.014)**	**0.07**
Dried fruit	1.48	(0.00-1.48)	35.47	0.002	(0.000-0.002)	0.04
Nuts and seeds	0.96	(0.96-6.22)	33.99	0.001	(0.001-0.009)	0.05
Sweets, chocolate, cakes, etc.	**283.39**	**(147.38-480.23)**	**957.61**	**0.54**	**(0.30-0.91)**	**1.77**
Sugar, non-chocolate confectionery	52.96	(17.65-117.31)	235.89	0.13	(0.04-0.29)	0.57
Chocolate, candy bars, etc.	34.04	(0.00-114.14)	450.56	0.02	(0.00-0.06)	0.25
Ice cream	4.24	(1.02-8.08)	17.50	0.03	(0.01-0.05)	0.11
Cakes, pies and pastries	45.95	(10.60-168.05)	385.58	0.11	(0.03-0.42)	0.96
Biscuits, dried cakes	29.28	(0.00-111.55)	224.02	0.06	(0.00-0.21)	0.43
Oils and fats	**8.10**	**(6.00-10.97)**	**17.92**	**4.41**	**(1.37-10.26)**	**25.44**
Vegetable fats and oils (non-olive)	0.18	(0.09-0.41)	5.60	0.25	(0.13-0.51)	15.96
Olive oil	6.41	(4.46-9.02)	14.70	0.24	(0.16-0.33)	0.54
Butter and other animal fats	0.57	(0.17-1.30)	2.78	2.77	(0.31-7.08)	14.78
Beverages	**1175.13**	**(770.85-1676.82)**	**2825.79**	**1.12**	**(0.95-3.44)**	**4.64**
Coffee and tea	1020.53	(656.61-1533.78)	2628.13	0.51	(0.26-0.73)	1.24
Wines	40.77	(1.53-189.86)	380.32	0.22	(0.00-1.40)	3.78
Red wine	12.07	(0.00-86.27)	253.12	0.18	(0.00-1.28)	3.75
White wine	1.62	(0.00-37.05)	192.57	0.00	(0.00-0.04)	0.21
Aperitif wines and beers	1.17	(0.00-3.32)	23.52	0.02	(0.00-0.04)	0.30
Spirits and liqueurs	0.00	(0.00-0.13)	3.08	0.00	(0.00-0.00)	0.06
Fruit juices	2.09	(0.00-9.70)	33.98	0.01	(0.00-0.04)	0.13
Soft drinks	0.00	(0.00-20.88)	125.40	0.00	(0.00-0.04)	0.27

Table 4. Dietary weekly intake (DWI) and tolerable weekly intake (TWI) of aluminum and tin in the study population compared with data reported by international agencies. Values reported in mg/kg of body weight (bw) per week, based on a body weight of 60 kg[a].

Metal	This study		EFSA[b]		WHO[c]	
	50th DWI	95th DWI	DWI	TWI	DWI	TWI
Aluminum	0.41	0.78	0.20-1.50	1.00	0.20-2.26	2.00
Tin	0.007	0.015	0.18-0.63	ND[d]	0.06-0.35	2.00

[a]DI: dietary intake; EFSA: European Food Safety Authority; WDI: Weekly dietary intake; TWI: tolerable weekly intake; WHO: World Health Organization.

[b]Data from ranges of average dietary intake obtained from EFSA Scientific Opinions [29, 36].

[c]Data from ranges of average dietary intake obtained from FAO/WHO [24, 49, 50].

[d]Tolerable upper intake level was not derived due to insufficient data, but regulatory limits of tin contents in canned food (200 mg/kg) and beverages (100 mg/kg) have been established [36].

Figure 1. Aluminum (A) and tin (B) food content (µg/kg), and contribution to dietary intake (% - percentage) by food category in the whole population.