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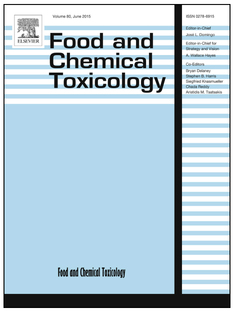

Intake of arsenic and mercury in a northern Italy community from fish and seafood

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Title: Intake of arsenic and mercury in a Northern Italy community from fish and seafood

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1.1 Introduction

During the last few decades, the consumption of seafood has increased considerably, mainly due to growing consciousness that seafood is an important source of protein, omega-3 polyunsaturated fatty acids (n-3 PUFAs), vitamins, and minerals (Domingo et al., 2007; Matos et al., 2015). The European Food Safety Authority (EFSA) has recently reviewed the role of seafood in European diets, assessing the beneficial effects of its consumption on health outcomes, and recommending fish consumption of 1-2 servings per week (European Food Safety Authority, 2014), similarly to World Health Organization and Food and Agriculture Organization (FAO/WHO, 2010b). However, fish and seafood also represent a source of environmental contaminants (Adel et al., 2016; Copat et al., 2014; Gonzalez-Alzaga et al., 2017; Vilavert et al., 2017). In fact, urbanization, globalization and human activity have adversely influenced water quality during the last decades, due to increased pollution in global aquatic ecosystem and the decreased capacity of freshwater ecosystems to dilute anthropogenic contaminants (Alava et al., 2017; Delpla et al., 2009; Duh et al., 2008; Pal et al., 2014; WHO, 2017). Therefore, fish and seafood are foods with the highest contribution to total dietary uptake of chemical contaminants by humans (Bae et al., 2017; Bosch et al., 2016; Cano-Sancho et al., 2015b; Copat et al., 2013; Perello et al., 2014; Taylor et al., 2016). In particular, fish and seafood are a relevant source of heavy metals and metalloids, which can spread though the food chain and accumulate in human body reaching levels which could represent a potential risk to human health (Arslan et al., 2017; Chiocchetti et al., 2017; Copat et al., 2013; Kaya and Turkoglu, 2017; Vinceti et al., 2013). Arsenic (As) and mercury (Hg) are two of the heavy metals of major environmental and toxicological concern (ATSDR, 2012a, b; Donaldson et al., 2010; Papadopoulos et al., 2015).

In this study, we used validated semi-quantitative food frequency questionnaires to assess the dietary habits of a representative sample of individuals aged 18-87 years from Northern Italy.

We estimated weekly dietary intakes of Arsenic (As) and Mercury (Hg), and we compared them with safety standards set by the European Food Safety Authority.

2.1 Methods

2.1.1 Food collection and analysis

We determined the arsenic and mercury content of foods by characterizing the usual diet of a sample of the population of the Emilia-Romagna region, Northern Italy. As previously described, we selected the most frequently consumed foods in a random sample of this population (Bottecchi and Vinceti, 2012; Cilloni et al., 2017), and we bought samples of these food items in markets and community canteens during the period from October 2016 to February 2017, as previously described (Filippini et al., 2018). To summarize, we strove to avoid cross-contamination with metals from food containers using plastic tubes or jars as well plastic cutlery or stainless-steel knife during food collection and handling. We then cut with a clean stainless-steel knife solid food samples collecting specimens in six different points from the plate. These samples were homogenised separately in a food blender, equipped with a stainless-steel blade, for subsequent analyses. We placed portions of the samples (0.5-1.0 g) in quartz containers previously washed with MilliQ water (MilliQPlus, Millipore, MA, USA) and HNO_3. Food samples were liquid-ashed (5 ml HNO_3 + 5 ml H_2O_2) in a microwave digestion system (Discover SP-D, CEM Corporation, NC, USA) and then stored in plastic tubes and diluted to 50 ml with deionised water before analyses. We performed trace element determination using an inductively coupled plasma-mass spectrometer (Agilent 7500ce, Agilent Technologies, CA, USA). We implemented quality controls including both blank solution (MilliQ water, Millipore, MA, USA), and using a control solution of tap-water additionally enriched with 22 ppb of each element under investigation. Limit of quantification was 0.05 µg/kg for arsenic and 0.02 µg/kg for mercury, corresponding limit of detection (LOD) was 0.012 µg/kg for arsenic and 0.005 µg/kg for mercury. Values below the LOD were set equal to LOD/2. We performed all the analyses in duplicate.

We then reported the concentrations of investigated trace elements according to the food consumption patterns and food categories typical of this Italian population, as assessed through a food frequency questionnaire (FFQ), as explained in detail (Filippini et al., 2017; Turrini et al., 2001). Our final list of food categories included cereals, meat, milk and dairy products, eggs, fish and seafood, vegetables, legumes, potatoes, fresh fruits, dry fruits, sweets and beverages. We further divided fish and seafood according to detailed food consumption patterns (Pala et al., 2003) into seafood (i.e. crustaceous, cephalopods, bivalves) and fish (i.e. preserved fish, tinned fish, and fresh fish). In particular group of fresh fish was further divided in codfish, sole or founder, sardine, trout, swordfish, and other fish (including tuna and salmon).

2.1.2. Study population and estimation of daily element intake

We carried out the evaluation of dietary habits of a representative sample of a Northern Italy population, as previously described (Malagoli et al., 2015; Vinceti et al., 2011). Briefly, we identified a list of 2825 potentially eligible subjects from the population database of the Emilia-Romagna Region residents, using the National Health Service directory (namely from the provinces of Bologna, Ferrara, Modena, Parma, and Reggio Emilia). The eligible subjects were selected among all residents in the study provinces having the same sex, year of birth (±5 years) and province of residence of 572 melanoma patients diagnosed during 2005-2006 in the same provinces. A total of 747 (26.4%) controls agreed to participate in the study, after giving their informed written consent, and returned the lifestyle and FFQ which we had mailed to them. Twenty-eight individuals were later excluded from subsequent analysis because of incomplete data or extreme values (ratio of total energy intake:calculated basal metabolic rate lower than the

0.5th percentile or higher than the 99.5th percentile) derived from the FFQ. The final population sample comprised 719 subjects, 319 males and 400 females, aged from 18 to 87 years (median 57, interquartile range (IQR) years in all subjects, with corresponding figures 60 (IQR: 50-69) and 53 (IQR: 41-63) for males and females, respectively. Median energy intake was 1906 kcal/day (IQR: 1538-2364) in all subjects, with 2024 kcal/day (IQR: 1649-2462) and 1800 kcal/day (IQR: 1455-2296) in male and female participants, respectively.

In this population, we assessed food and nutrient intake using a validated semi-quantitative FFQ specifically developed for the Central-Northern Italy population (Pala et al., 2003; Pasanisi et al., 2002) 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, using also photos of serving sizes to help proper completion by participants. Dietary intake of individual nutrients and contaminants were calculated using *ad hoc* software (Malagoli et al., 2015; Malavolti et al., 2013).

We combined data on the estimated trace elements in foods and the EPIC FFQ to compute total weekly trace element intake using the equation presented below.

$$Weekly\ dietary\ exposure\ \left(\frac{\mu g}{kg\ bw/week}\right) = \sum \frac{element\ food\ content\ \left(\frac{\mu g}{kg}\right) \times food\ intake\ \left(\frac{g}{day}\right) \times 7}{body\ weight\ (kg) \times 1000}$$

We multiplied the element content measured in food (µg/kg) with the intake as estimated with the FFQ (g/day) and we divided by the body weight (kg) of each participant. Accordingly, we estimated weekly dietary As and Hg intake for the total diet and for each food category by reporting median and interquartile ranges of intake. Finally, we estimated inorganic As, inorganic Hg and methylmercury (methyl-Hg) intake according to the conservative EFSA approach (European Food Safety Authority, 2009, 2012). In particular, for the Hg species, we assumed that 100% of Hg in fish is in the organic form (i.e. methyl-Hg). However, in order to ensure that dietary exposure to inorganic was not underestimated, 20% of total mercury from fish was assumed to be inorganic (See Supplemental Table S1 for details).

3.1 Results

Table 1 shows the distribution of As and Hg in our study population, taking into account the 890 food items we assessed and their percentage contribution to each food category. Levels were below the LOD in 4 and 90 samples for arsenic and mercury, respectively.

We report in table 2 the average As and Hg intake according to food category consumption for all participants and by sex.

Table 3 summarizes the estimated levels of weekly dietary intake of total and inorganic As, , respectively of 5.34 µg/kg of body weight (bw) per week (IQR: 2.75-8.84) and 0.58 µg/kg bw/week (IQR:0.44-0.75), and total and inorganic Hg, respectively of 0.39 µg/kg bw/week (IQR: 0.25-0.59) and 0.14 µg/kg bw/week (IQR: 0.10-0.19). For both elements, the highest contribution to total dietary intake was driven by fish and seafood intake. Concerning inorganic trace element intake, aside from fish and seafood, major contributors to inorganic As intake were cereals and beverages, while for inorganic Hg, they were cereals and vegetables. On the converse, estimated intake of methyl-Hg corresponded to total contribution of Hg by fish and seafood, namely 0.32 (IQR: 0.18-0.51) µg/kg bw/week, on the basis of the most conservative value of 100% as suggested in the EFSA exposure assessment (European Food Safety Authority, 2012). Sex-specific estimates showed slightly higher values of total As among females, due to higher contribution from fish and seafood, while results for inorganic As were substantially comparable (Tables 4-5). Regarding total and inorganic Hg, dietary exposure was higher in females, due to a higher contribution from fish and vegetable intake. Methyl-Hg intake was 0.287 µg/kg bw/week (IQR: 0.159-0.457) and 0.345 µg/kg bw/week (IQR: 0.192-0.556) in males and females, respectively.

Since fish and seafood showed the highest contamination levels and contributed to the highest amount of exposure in the study population, we further investigated their concentrations (white bars) in different types of fish (Figure 1). Interestingly, the highest As values were found in sardine, sole/flounder and cephalopods (i.e. octopus, cuttlefish and squid). On the converse, for Hg, the highest concentrations were found in typical big and predatory fishes, i.e. sword fish, and both fresh and tinned tuna. Supplemental Figures S1-S3 show the concentration of each element and relative contribution to estimated dietary intake, respectively, for cereals, vegetables, and beverages. Among cereals, rice showed the highest concentration and contribution (60%) to total As intake, followed by bread and pasta (around 17% each). Bread demonstrated the highest contribution to total Hg intake (55%), followed by crispbread/rusks (20%) and rice (16%). With regard to vegetables, the highest concentrations for both total As and total Hg were found in mushrooms, followed by leafy vegetables. Similarly, leafy vegetables (42%), root vegetables (13%) and mushrooms (12%) made large contributions to total As, while leafy vegetables (35%), mushrooms (25%) and tomatoes (21%) made large contributions to total Hg. Finally, among beverages, the major contributors to As and Hg were coffee and wine, accounting for 46% and 35% for total As and 90% and 6% for total Hg, respectively, within this category.

4.1 Discussion

In this study, we assessed total As and Hg intake within a Northern Italian community by measuring trace element content in foods and assessing usual dietary intake of foods containing these elements. While our findings are new for the northern part of Italy due to the absence of previous studies carried out in this region, they agree with previous studies from other European countries (Cirillo et al., 2010; Marti-Cid et al., 2008; Martorell et al., 2011; Miklavcic et al., 2013; Perello et al., 2015), especially for fish and seafood, which showed the highest levels of contamination (Brambilla et al., 2013; Calatayud et al., 2012; Cano-Sancho et al., 2015a). Therefore, our findings indicate that, in industrialized nations, fish and seafood are likely to be the most important contributors to total As and Hg intake.

Both As and Hg are environmental contaminants that have both natural and anthropogenic origin. For both elements, their toxicity is strongly related not only to the environmental levels but also to their species, either elemental or inorganic or organic (ATSDR, 2012a, b). As is most toxic in its inorganic form (Sattar et al., 2016). In our population, the estimated weekly intake of inorganic arsenic of 0.58 µg/kg bw/ week (IQR: 0.44-0.75) is consistent with previous findings (Barone et al., 2015; Copat et al., 2014; Storelli and Barone, 2013), and it is within the range values of benchmark dose suggested by EFSA (European Food Safety Authority, 2009).

For Hg, its organic compounds, including the most abundant methyl-Hg form, are the most common species entering the food chain, and they show the highest toxicity to humans (Bjorklund et al., 2017; European Food Safety Authority, 2012). Based on our estimates, weekly intake of inorganic Hg and methyl-Hg are 0.14 (IQR: 0.10-0.19) and 0.32 (IQR: 0.18-0.51), respectively, below the tolerable weekly intake (TWI) of 4 µg/kg bw and 1.6 µg/kg bw established by both EFSA and the Joint FAO/WHO Expert Committee on Food Additives (European Food Safety Authority, 2012; FAO/WHO, 2010a).

Since large, predator fish showed the highest Hg concentrations (Burger and Gochfeld, 2005; Donald et al., 2015), as expected, subjects consuming a large portion of their diet from these type of fishes, such as swordfish or tuna, were at higher risk of a relevant exposure (Adel et al., 2016; Hightower and Moore, 2003). Interestingly, we did not find appreciable differences between tinned and fresh tuna in Hg levels, while for As, similarly with previous findings, slightly lower concentrations were detected in tinned tuna as compared with fresh tuna (Nunez et al., 2018).

Sex-related differences in dietary As and Hg exposure have already been described (Llobet et al., 2003; Marzec and Schlegel-Zawadzka, 2004). Our analysis showed a higher intake of total As in females, due to the higher contribution of fish category in females, while intake of inorganic As was substantially similar between males and females. Consistent with previous findings (Liu et al., 2018), we found higher levels of total intake of Hg, inorganic Hg and methyl-Hg in females, entirely driven by higher fish and seafood consumption.

Previous studies suggested that only a fraction of total heavy metal intake is absorbed after ingestion (Calatayud et al., 2012; Cano-Sancho et al., 2015a; Maulvault et al., 2011).

Study strengths include collection of data on a large number of foods, which were divided in categories based on food consumption patterns derived from a validated dietary assessment in the same population (Filippini et al., 2018; Malagoli et al., 2015; Pala et al., 2003; Pasanisi et al., 2002; Turrini et al., 2017; Vinceti et al., 2011). We could therefore obtain both a weighted average of As and Hg food category content and an estimate of overall and food-specific intake in our study population, allowing comparability of our results with other studies. Moreover, use of local groceries and supermarkets for purchasing foodstuffs provided a close-to-real estimation of dietary intake in our Northern Italy community. Finally, we further divided food categories by sub-categories reported in the FFQ, thereby allowing estimation of As and Hg intake in individual species of fish and seafood.

Study limitations include misclassification of elemental species. While speciation analysis is of increasing scientific interest (Piras et al., 2015; Ruiz-de-Cenzano et al., 2017; Ruzik, 2012; Solovyev et al., 2017) due to the heterogeneous and sometimes opposite health effects of the various species (Karri et al., 2016; Michalke et al., 2017; Vinceti et al., 2012), we attempted to estimate intake of both inorganic and organic compounds. Unfortunately, we were unable to directly determine these single element species in foods, thus hampering their direct estimation in food samples. We therefore used an indirect and conservative approach to assess elements fraction from total concentration according to the EFSA methodology (European Food Safety Authority, 2009, 2012), which may have induced misclassification (Bradley et al., 2017).

Since food collection was carried out mainly during winter and autumn, we could only partially account for possible seasonal variation of heavy metal content in food (Turrini et al., 2017; Vincevica-Gaile and Klavins, 2013). However, conflicting results have been reported about the existence of seasonal effects for several elements, specifically for As and Hg (Biswas et al., 2014; Copat et al., 2012; Nacano et al., 2014; Nevarez et al., 2015; Piras et al., 2015), and they have not been shown to occur for other elements (Agudo et al., 2002; Halkjaer et al., 2009; Olsen et al., 2009; Welch et al., 2009).

Finally, we assessed dietary intake in an adult population only, and our estimates may not apply to adolescents and children, due to differences in dietary habits, heavy metal metabolism, and toxic effects.

5.1 Conclusions

In a population-based study from Northern Italy, we provided detailed information about the foods most likely to contain appreciable amounts of As and Hg. In agreement with previous studies of individuals from industrialized nations, we found that fish and seafood contributed the highest levels of As and Hg to the human diet. Weekly dietary intake of As and Hg in our population did not appear to exceed the recommended safety limits set by EFSA.

Declaration of interest

All authors declare that they have no conflict of interest.

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Table 1. Distribution of arsenic (As) and mercury (Hg) concentrations in foods composing the usual diet of the study population.

Food (N)	As (µg/kg)			Hg (µg/kg)		
	50th	IQR	<LOD n (%)	50th	IQR	<LOD n (%)
Cereals (126)	6.01	(3.63 - 9.90)	1 (0.8)	0.78	(0.16 - 3.51)	17 (13.7)
Meat (86)	3.97	(2.73 - 8.23)	0 (0.0)	0.65	(0.18 - 1.80)	12 (14.0)
Milk and dairy products (72)	3.92	(1.43 - 8.64)	0 (0.0)	0.37	(0.14 - 0.92)	6 (8.3)
Eggs (9)	2.58	(1.45 - 8.29)	1 (11.1)	0.99	(0.12 - 2.79)	0 (0.0)
Fish and seafood (62)	1346.95	(686.1 - 3831.32)	0 (0.0)	55.03	(25.27 - 142.26)	0 (0.0)
Vegetables (193)	2.51	(0.94 - 6.59)	1 (0.5)	0.63	(0.18 - 2.06)	14 (7.3)
Legumes (42)	4.32	(3.74 - 8.24)	0 (0.0)	0.66	(0.14 - 3.65)	3 (7.1)
Potatoes (14)	2.61	(1.08 - 4.68)	0 (0.0)	0.36	(0.15 - 0.91)	1 (7.1)
Fresh fruits (65)	1.20	(0.62 - 3.33)	0 (0.0)	0.28	(0.06 - 0.60)	7 (10.8)
Dry fruits (39)	10.82	(3.81 - 30.67)	0 (0.0)	0.82	(0.33 - 3.71)	5 (12.8)
Sweets (64)	5.76	(2.50 - 8.90)	0 (0.0)	0.27	(0.01 - 0.99)	15 (23.4)
Oils and fats (22)	2.59	(1.76 - 10.82)	1 (4.5)	0.03	(0.00 - 0.19)	10 (45.5)
Beverages (96)	2.70	(0.89 - 7.77)	0 (0.0)	0.01	(0.00 - 0.04)	0 (0.0)

Table 2. Median (interquartile range - IQR) of daily dietary intake (g/day) for main food categories in the study population.

Food	Total (n=719)		Males (N=319)		Females (N=400)	
	50th	IQR	50th	IQR	50th	IQR
Cereals	175.4	(120.6 - 245.5)	194.7	(129.5 - 267.4)	165.9	(113.9 - 221.5)
Meat	119.3	(79.5 - 164.3)	136.6	(92.1 - 174.3)	107.5	(71.8 - 150.6)
Milk and dairy products	185.0	(85.2 - 301.7)	161.3	(63.9 - 286.1)	209.8	(120.7 - 309.6)
Eggs	13.5	(7.3 - 21.2)	12.4	(7.0 - 20.0)	14.0	(7.6 - 21.4)
Fish and seafood	28.7	(15.9 - 45.7)	29.1	(16.1 - 48.2)	28.6	(15.7 - 44.8)
Vegetables	140.8	(96.3 - 210.8)	137.2	(99.2 - 204.3)	143.6	(92.2 - 212.6)
Legumes	13.6	(6.3 - 25.3)	14.2	(6.2 - 25.7)	13.5	(6.4 - 23.9)
Potatoes	18.0	(10.7 - 32.0)	18.1	(10.7 - 30.8)	17.9	(10.1 - 32.3)
Fresh fruits	302.0	(206.1 - 436.9)	284.0	(202.8 - 426.9)	318.7	(208.8 - 452.1)
Dry fruits	0.3	(0.2 - 1.6)	0.3	(0.2 - 1.8)	0.3	(0.2 - 1.6)
Sweets	68.7	(38.6 - 112.5)	65.5	(32.6 - 108.8)	74.2	(43.0 - 114.0)
Oils and fats	24.7	(18.4 - 33.4)	25.6	(18.9 - 34.5)	24.1	(17.9 - 32.7)
Beverages	290.7	(159.0 - 483.1)	360.7	(185.4 - 574.6)	239.9	(141.2 - 413.5)

Table 3. Weekly dietary intake of estimated total and inorganic arsenic (As) and mercury (Hg) in the study population. Values in µg/kg of body weight per week. Median and interquartile range (IQR) are reported. Range of benchmark dose lower confidence limit (BMDL) values for 1% excess risk of cancers of the lung, skin and bladder, and skin lesions (BMDL_{01}) of 2.1-24 µg/kg bw/week corresponding to 0.3-8 µg/kg bw/day. Tolerable weekly intake (TWI) for inorganic mercury 4 µg/kg bw/week.

Food	**Total As**		**Inorg As**	
	50^{th}	**IQR**	**50^{th}**	**IQR**
Cereals	0.195	(0.120 - 0.289)	0.195	(0.120 - 0.289)
Meat	0.046	(0.031 - 0.064)	0.023	(0.016 - 0.032)
Milk and dairy products	0.036	(0.021 - 0.053)	0.018	(0.011 - 0.027)
Eggs	0.003	(0.002 - 0.005)	0.002	(0.001 - 0.003)
Fish and seafood	4.686	(2.163 - 8.128)	0.094	(0.043 - 0.163)
Vegetables	0.036	(0.023 - 0.053)	0.018	(0.011 - 0.027)
Legumes	0.006	(0.003 - 0.011)	0.003	(0.001 - 0.005)
Potatoes	0.005	(0.003 - 0.008)	0.002	(0.001 - 0.004)
Fresh fruits	0.03	(0.018 - 0.044)	0.015	(0.009 - 0.022)
Dry fruits	0	(0.000 - 0.002)	0	(0.000 - 0.001)
Sweets	0.032	(0.017 - 0.057)	0.016	(0.009 - 0.028)
Oils and fats	0.007	(0.004 - 0.012)	0.003	(0.002 - 0.006)
Beverages	0.121	(0.071 - 0.187)	0.121	(0.071 - 0.187)
Total	**5.337**	**(2.749 - 8.835)**	**0.578**	**(0.435 - 0.749)**

	Total Hg		**Inorg Hg**	
	50^{th}	**IQR**	**50^{th}**	**IQR**
Cereals	0.024	(0.015 - 0.035)	0.024	(0.015 - 0.035)
Meat	0.005	(0.003 - 0.008)	0.005	(0.003 - 0.008)
Milk and dairy products	0.004	(0.002 - 0.006)	0.004	(0.002 - 0.006)
Eggs	0.001	(0.001 - 0.002)	0.001	(0.001 - 0.002)
Fish and seafood	0.319	(0.178 - 0.511)	0.064	(0.036 - 0.102)
Vegetables	0.017	(0.011 - 0.026)	0.017	(0.011 - 0.026)
Legumes	0.001	(0.000- 0.002)	0.001	(0.000 - 0.002)
Potatoes	0.001	(0.000 - 0.001)	0.001	(0.000 - 0.001)
Fresh fruits	0.006	(0.004 - 0.01)	0.006	(0.004 - 0.01)
Dry fruits	0	-	0	-
Sweets	0.002	(0.001 - 0.004)	0.002	(0.001 - 0.004)
Oils and fats	0	-	0	-
Beverages	0.003	(0.002 - 0.004)	0.003	(0.002 - 0.004)
Total	**0.391**	**(0.248 - 0.588)**	**0.139**	**(0.102 - 0.188)**

Table 4. Weekly dietary intake of estimated total and inorganic arsenic (As) and mercury (Hg) in male study population. Values in µg/kg of body weight (bw) per week. Median and interquartile range (IQR) are reported. Range of benchmark dose lower confidence limit values for 1% excess risk of cancers of the lung, skin and bladder, and skin lesions of 2.1-24 µg/kg bw/week corresponding to 0.3-8 µg/kg bw/day. Tolerable weekly intake for inorganic Hg=4 µg/kg bw/week.

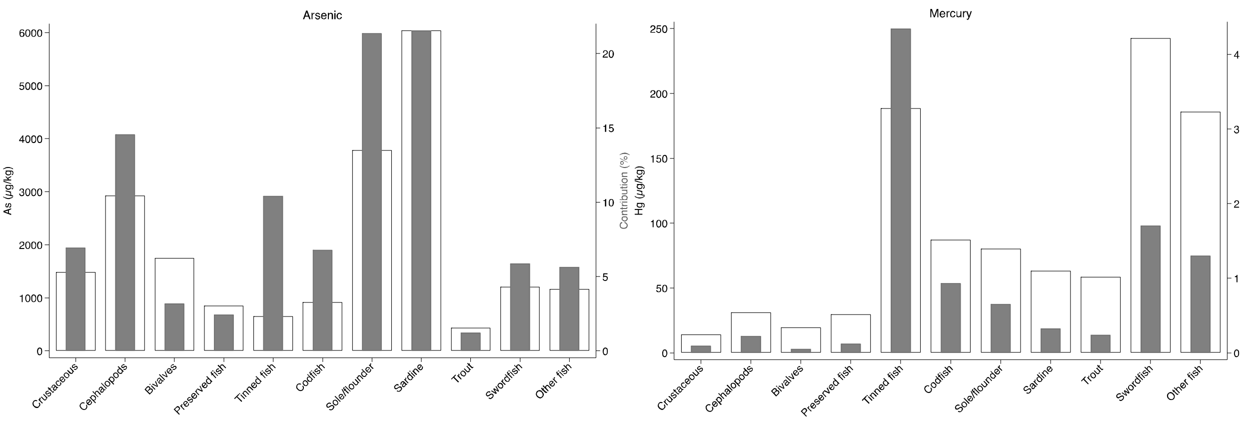
Food	**Total As**		**Inorganic As**	
	50th	**IQR**	**50th**	**IQR**
Cereals	0.191	(0.120 - 0.290)	0.191	(0.120 - 0.290)
Meat	0.047	(0.032 - 0.064)	0.023	(0.016 - 0.032)
Milk and dairy products	0.030	(0.017 - 0.046)	0.015	(0.008 - 0.023)
Eggs	0.003	(0.002 - 0.005)	0.001	(0.001 - 0.002)
Fish and seafood	4.279	(1.852 - 7.684)	0.086	(0.037 - 0.154)
Vegetables	0.030	(0.020 - 0.046)	0.015	(0.010 - 0.023)
Legumes	0.005	(0.002 - 0.010)	0.003	(0.001 - 0.005)
Potatoes	0.004	(0.002 - 0.007)	0.002	(0.001 - 0.004)
Fresh fruits	0.025	(0.016 - 0.037)	0.013	(0.008 - 0.019)
Dry fruits	0	(0.000 - 0.002)	0	(0.000 - 0.001)
Sweets	0.027	(0.012 - 0.049)	0.013	(0.006 - 0.024)
Oils and fats	0.007	(0.004 - 0.011)	0.003	(0.002 - 0.006)
Beverages	0.127	(0.077 - 0.199)	0.127	(0.077 - 0.199)
Total	**4.793**	**(2.345 - 8.364)**	**0.571**	**(0.423 - 0.725)**
	Total Hg		**Inorganic Hg**	
	50th	**IQR**	**50th**	**IQR**
Cereals	0.023	(0.013 - 0.033)	0.023	(0.013 - 0.033)
Meat	0.005	(0.003 - 0.007)	0.005	(0.003 - 0.007)
Milk and dairy products	0.003	(0.002 - 0.005)	0.003	(0.002 - 0.005)
Eggs	0.001	(0.001 - 0.002)	0.001	(0.001 - 0.002)
Fish and seafood	0.287	(0.158 - 0.457)	0.057	(0.032 - 0.091)
Vegetables	0.015	(0.010 - 0.024)	0.015	(0.010 - 0.024)
Legumes	0.001	(0.000 - 0.002)	0.001	(0.000 - 0.002)
Potatoes	0.001	(0.000 - 0.001)	0.001	(0.000 - 0.001)
Fresh fruits	0.006	(0.003 - 0.008)	0.006	(0.003 - 0.008)
Dry fruits	0	-	0	-
Sweets	0.002	(0.001 - 0.004)	0.002	(0.001 - 0.004)
Oils and fats	0	-	0	-
Beverages	0.003	(0.001 - 0.004)	0.003	(0.001 - 0.004)
Total	**0.352**	**(0.222 - 0.54)**	**0.128**	**(0.096 - 0.167)**

Table 5. Weekly dietary intake of estimated total and inorganic arsenic (As) and mercury (Hg) in female study population. Values in µg/kg of body weight (bw) per week. Median and interquartile range (IQR) are reported. Range of benchmark dose lower confidence limit values for 1% excess risk of cancers of the lung, skin and bladder, and skin lesions of 2.1-24 µg/kg bw/week corresponding to 0.3-8 µg/kg bw/day. Tolerable weekly intake for inorganic Hg=4 µg/kg bw/week.

Food	Total As		Inorganic As	
	50th	IQR	50th	IQR
Cereals	0.201	(0.120 - 0.288)	0.201	(0.120 - 0.288)
Meat	0.046	(0.031 - 0.064)	0.023	(0.015 - 0.032)
Milk and dairy products	0.043	(0.027 - 0.058)	0.022	(0.013 - 0.029)
Eggs	0.004	(0.002 - 0.006)	0.002	(0.001 - 0.003)
Fish and seafood	5.088	(2.422 - 8.531)	0.102	(0.048 - 0.171)
Vegetables	0.041	(0.026 - 0.061)	0.020	(0.013 - 0.031)
Legumes	0.006	(0.003 - 0.011)	0.003	(0.002 - 0.006)
Potatoes	0.005	(0.003 - 0.008)	0.003	(0.001 - 0.004)
Fresh fruits	0.035	(0.021 - 0.049)	0.017	(0.011 - 0.025)
Dry fruits	0	(0.000 - 0.002)	0	(0.000 - 0.001)
Sweets	0.036	(0.020 - 0.062)	0.018	(0.010 - 0.031)
Oils and fats	0.007	(0.004 - 0.012)	0.003	(0.002 - 0.006)
Beverages	0.116	(0.067 - 0.180)	0.116	(0.067 - 0.180)
Total	**5.761**	**(2.957 - 9.159)**	**0.583**	**(0.454 - 0.781)**

	Total Hg		Inorganic Hg	
	50th	IQR	50th	IQR
Cereals	0.025	(0.017 - 0.036)	0.025	(0.017 - 0.036)
Meat	0.005	(0.003 - 0.008)	0.005	(0.003 - 0.008)
Milk and dairy products	0.004	(0.003 - 0.006)	0.004	(0.003 - 0.006)
Eggs	0.001	(0.001 - 0.002)	0.001	(0.001 - 0.002)
Fish and seafood	0.345	(0.192 - 0.556)	0.069	(0.038 - 0.111)
Vegetables	0.019	(0.013 - 0.029)	0.019	(0.013 - 0.029)
Legumes	0.001	(0.000 - 0.002)	0.001	(0.000 - 0.002)
Potatoes	0.001	(0.000 - 0.001)	0.001	(0.000 - 0.001)
Fresh fruits	0.008	(0.005 - 0.011)	0.008	(0.005 - 0.011)
Dry fruits	0	-	0	-
Sweets	0.003	(0.001 - 0.005)	0.003	(0.001 - 0.005)
Oils and fats	0	-	0	-
Beverages	0.003	(0.002 - 0.005)	0.003	(0.002 - 0.005)
Total	**0.423**	**(0.264 - 0.621)**	**0.149**	**(0.111 - 0.200)**

Figure 1. Trace elements content (white bars) according to fish and seafood category and % contribution to trace element daily intake (gray bars) in study population.



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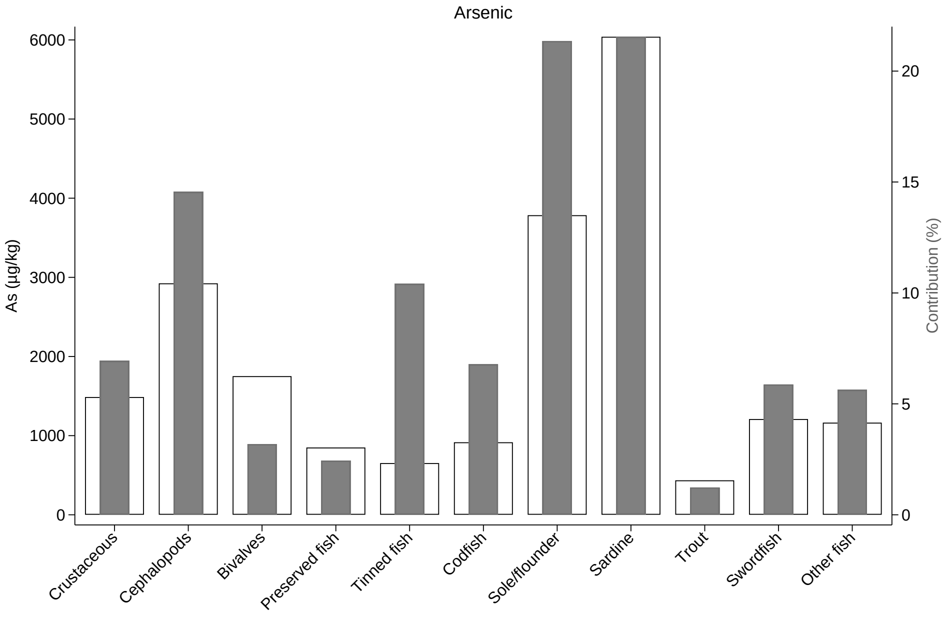
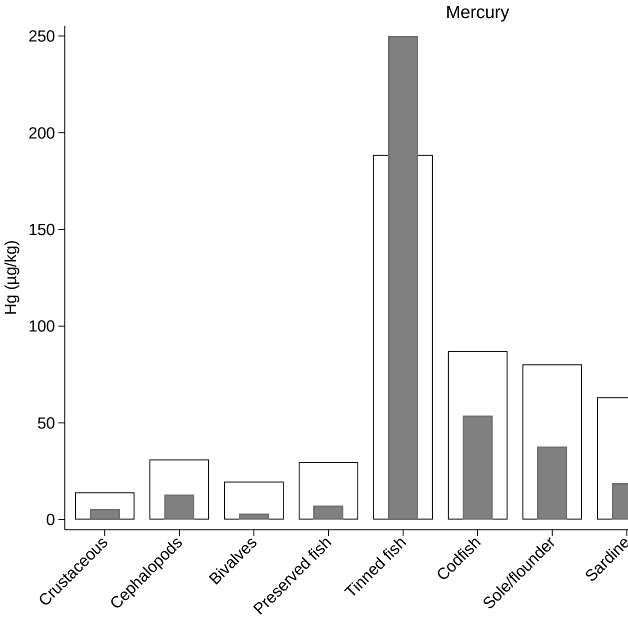

Arsenic
As (µg/kg)
0
1000
2000
3000
4000
5000
6000
Contribution (%)
0
5
10
15
20
Crustaceous
Cephalopods
Bivalves
Preserved fish
Tinned fish
Codfish
Sole/flounder
Sardine
Trout
Swordfish
Other fish


Mercury
Hg (µg/kg)
0
50
100
150
200
250
Crustaceous
Cephalopods
Bivalves
Preserved fish
Tinned fish
Codfish
Sole/flounder
Sardine

Highlights:

- We assessed arsenic and mercury contamination levels in foods composing dietary habits of Northern Italy community
- Sardine and sole species showed higher concentration of arsenic, while swordfish and tuna of mercury
- Estimated dietary intake of both trace elements were below the EFSA recommended tolerable intake