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Dietary exposure to trace elements and radionuclides: the methodology of the Italian Total Diet Study 2012-2014

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Abstract

This article presents the methodology of the Italian Total Diet Study 2012-2014 aimed at assessing the dietary exposure of the general Italian population to selected non-essential trace elements (Al, inorganic As, Cd, Pb, methyl-Hg, inorganic Hg, U) and radionuclides (^{40}K, ^{134}Cs, ^{137}Cs, ^{90}Sr). The establishment of the TDS food list, the design of the sampling plan, and details about the collection of food samples, their standardized culinary treatment, pooling into analytical samples and subsequent sample treatment are described. Analytical techniques and quality assurance are discussed, with emphasis on the need for speciation data and for minimizing the percentage of left-censored data so as to reduce uncertainties in exposure assessment. Finally the methodology for estimating the exposure of the general population and of population subgroups according to age (children, teenagers, adults, and the elderly) and gender, both at the national level and for each of the four main geographical areas of Italy, is presented.

Key words
- metals
- arsenic
- radioisotopes
- exposure assessment
- food safety
- total diet study

INTRODUCTION

Trace elements are chemical substances taken up at trace levels from the diet. Whereas essential trace elements are nutrients needed in very minute quantities for the proper growth, development, and physiology of the organism (*e.g.* iron, copper, zinc, iodine, selenium, molybdenum), dietary exposure to non-essential elements such as cadmium, lead or mercury is of concern [1-3]. Environmental sources are the main contributors to contamination of food with metals and other non-essential elements. Even though they are ubiquitous and thus naturally present in the diet, higher levels may occur as a result of environmental pollution from industrial and other anthropogenic activities.

Non-essential elements may enter the food chain at any point during growth and harvesting, through to storage and processing, including packaging. Food is the major contributor to exposure of the general (non-occupationally exposed) population, although other routes may also be significant for specific elements. Certain food groups are known to accumulate some trace elements naturally and, consequently, they can contain relatively high concentrations of these ones. For example, fish and shellfish are known to accumulate mercury in the toxic form of methylmercury. Wheat takes up cadmium whereas rice accumulates arsenic largely in the toxic form of inorganic arsenic. It is to be noted that other food items such as fish and seafood contain very high concentrations of arsenic, but it occurs as organic species of lower or negligible toxicity. Therefore, for risk assessment of arsenic speciation data are needed in order to characterize the presence of the toxic inorganic form [4, 5]. Also in the case of mercury speciation is important, since methylmercury is considerably more toxic than inorganic mercury [5, 6].

Another element of concern is aluminium, which is found in food as a result of its natural occurrence in the environment, contamination from various sources, leaching from food contact materials and the use of aluminium-containing food additives [7, 8]. As regards

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uranium, it can be present in food and feed in varying concentrations through leaching from natural deposits such as soil or rocks, dissolution in fertilizers and emission from mining and milling (uranium mines, production of coal and other fuels) [9]. Uranium may be present at relatively high concentrations in the water supply as well as in mineral water, and the contribution from water and food sum up leading to an overall dietary exposure that needs be thoroughly assessed [10]. Besides chemical toxicity, there is also a radiological risk associated to uranium exposure, even though the latter is much less important compared to chemical toxicity when ingestion of uranium is considered.

Although radioactivity in food chain has been decreasing since the late 1960s and the effect of Chernobyl accident is not detectable anymore in most food categories, other nuclear emergencies as the recent Fukushima accident have raised public health concerns. The primary factor contributing to the internal effective dose in the human organism, apart from radon inhalation, is contaminated food [11]. Background levels of radionuclides in foods vary and depend on several factors, including the type of food and the geographic region where the food has been produced. The concentration of natural radionuclides varies because of differences in background levels related to soil, climate, agricultural conditions and contamination due to the NORM (Naturally Occurring Radioactive Material) industries. Among naturally occurring radioisotopes entering the human body primarily by ingestion of foods, ^{40}K is usually the one contributing the most to the internal effective dose. Artificial radioisotopes potentially significant in terms of food safety include long-lived radionuclides such as ^{137}Cs and ^{90}Sr, and short-lived gamma-emitting radionuclides such as ^{134}Cs.

Total Diet Studies (TDS) are national surveys carried out to assess dietary exposure of the general population to contaminants and characterize the associated public health risks [12]. A TDS consists in the selection, collection and analysis of commonly consumed food purchased at retail level, processed as for consumption, and pooled into representative food groups. TDSs are designed to cover the whole diet and to measure the amount of each contaminant ingested by the population living in a country using average and high level consumption data, the latter being referred to those individuals who consistently consume high amounts of specific foods (high percentile consumers). The TDS method is a complementary approach to traditional monitoring and surveillance activities, but instead of focusing on compliance it is designed to provide a solid basis for calculating population dietary exposure and assess the impact on public health.

The aim of this article is to present the methodology of the 2012-2014 Italian TDS, launched by the Italian Ministry of Health and run under the supervision of the Istituto Superiore di Sanità (ISS), the Italian National Health Institute. The goal of this survey is to estimate the exposure of the general population and of selected population subgroups to aluminium, inorganic arsenic, cadmium, lead, methylmercury, inorganic mercury, uranium, ^{40}K, ^{134}Cs, ^{137}Cs, and ^{90}Sr both at the national level and for each of the four main geographical areas of Italy, *i.e.* North-West, North-East, Centre, South and Islands.

FOOD LIST

Core foods were selected in order to be representative of the diet of the Italian population, similarly to the two previous TDSs [13, 14]. The selection was based on the results of the National Food Consumption Survey INRAN-SCAI 2005-06 [15]. This was a cross-sectional study where 1329 households were randomly selected after geographical stratification of the national territory. Food consumption of 3323 subjects was assessed on three consecutive days through individual estimated dietary records. The mean, the standard deviation, and three percentiles (50th, 95th, and 99th) of individual daily consumption (3 d average) by food category in the total population and (50th and 95th) in consumers only were obtained. These statistics were calculated for the two genders and four age classes, *i.e.* children (3-9.9 years), teenagers (10-17.9 years), adults (18-64.9 years) and the elderly (≥ 65 years). Furthermore, food consumption for the four main geographical areas of Italy, namely North-West, North-East, Centre, South and Islands, was characterized.

For the TDS food list, the most widely consumed foods by adults and/or children (consumer rate of at least 5%) were selected *(Table 1)*. Foods were grouped so that commodities known to be susceptible to contamination (*e.g.* offal, crustaceans and molluscs, spices and herbs) or to represent major exposure sources (*e.g.* fish for methylmercury) are kept separate, as are foods which are consumed in large quantities (*e.g.* bread, pasta, potatoes, milk, cheese). Food categories contributing less than 0.25% to the diet in terms of quantity where ignored as well as food with a daily consumption < 1.5 g, unless potentially containing high levels of the analytes (*e.g.* offal). In the end, the core foods (n = 51) covered about 99.7% of the whole diet of adults and children.

FOOD SAMPLING

The food items making up the 51 core foods are being bought at retail in 4 cities, selected to represent the four main geographical areas of Italy. They were Milano (North-West), Bologna (North-East), Rome (Centre), Bari (South and Islands). The premises for sample collection were chosen in order to ensure representativeness of the TDS results [16]. Specific retail outlets have been selected for each core food according to consumer habits. For instance bread is bought both at supermarkets and at bakeries, pizza mainly at pizzerias, fruits both at supermarkets and at traditional markets. Hyper and supermarket supplied by different distribution centres have been chosen so as to reflect the structure of food retailing in Italy. Market share data provided by food manufacturers (trade associations) allowed to identify types and brands of the food items to be sampled at large-scale retail trade. Based on above information, a food shopping list and a comprehensive sampling plan have been designed.

Sampling has started in November 2012 and is

scheduled to be completed within July 2014. Three sampling campaigns will be undertaken according to the plan shown in *Table 1*. Shoppers are provided with a food collection protocol specifying the retail market places and including detailed descriptions of every food product to be purchased (type, brand, and sample size). The shoppers draw up a collection report with data on the individual food items that are bought, which are being stored in a specifically developed section of the SINSVA database of the Ministry of Health.

Overall 944 individual samples are being collected and pooled in 204 samples, *i.e.* the 51 core foods representative of the population diet, obtained for each of the four main geographical areas *(Table 1)*. In order to be as representative as possible of the Italian food consumption habits, each core food corresponds to a pooled sample composed of up to 8 different foods ("individual samples"), selected according to market share and processing (packed food), origin and species (fresh food). The relative proportion of each individual sample within the pooled sample reflects its importance in the average Italian diet. In turn, each individual sample is composed by a fixed number of elementary samples; in *Figure 1* the core food "Bread" is shown as an example. Altogether, > 3000 products (elementary samples) are being collected. Fruits and vegetables are being sampled during two different seasons.

SAMPLE PREPARATION

Individual food samples are prepared and cooked according to normal consumer practices. Fruits and vegetables are washed and peeled, pasta and rice are boiled, meat and seafood are grilled, pan-fried, etc., according to standard recipes for each geographical area. Then composite samples are prepared and ~1.2 kg (fresh weight) are destined to radionuclide determination, whereas ~400 g are freeze-dried and stored in amber glass bottles with screw caps for subsequent trace element determination. Water loss as a consequence of freeze-drying is measured and fresh/dry weight ratios calculated. Freeze-drying has been chosen in order to enable long-term storage and successive analysis for other chemicals later on. In particular, long-term storage gives the possibility to survey novel/emerging contaminants years after the completion of the TDS and evaluate trends by comparison with occurrence level determined afterwards.

Great care was taken in the selection of the water for preparing beverages (*e.g.* coffee, tea) and boiling foods such as pasta and rice. Water is often neglected in TDSs because tap water is not always included in food consumption or supply data, but it can be a significant source of contaminants in the diet [12]. Tap water affects the intake of trace elements and other contaminants both directly, as drinking water, and indirectly, being the medium in which many foods are cooked. Whereas water with low levels of trace elements and radionuclides reduces the concentrations of these contaminants in food during boiling [17, 18], water with higher than background levels may enrich the cooked food compared to the raw (uncooked) counterpart.

In this study, a detailed survey of tap water from the four main sampling areas was carried out and the individual samples from each location were analysed separately (*i.e.* without pooling) in order to get an insight into the magnitude and variability of concentrations levels. The results shown in *Table 2* highlight that especially aluminium, arsenic (present in water as inorganic arsenic), and uranium are highly variable both within sampling locations (coefficients of variation up to 61%, 101%, and 87%, respectively) and among sampling locations (coefficients of variation of 107%, 107%, 90%, respectively). Therefore an "average water" has been used in kitchen preparation of TDS foods and its composition varied for the four main geographical areas to reflect the mean occurrence level in each of them. Salt was always added to boiling water in order to closely simulate actual household conditions, and considering aspects such as osmotic pressure, minimization of the loss of the analytes into cooking water, and the intention to capture full exposure [12]. Standardized conditions for salt use in cooking were

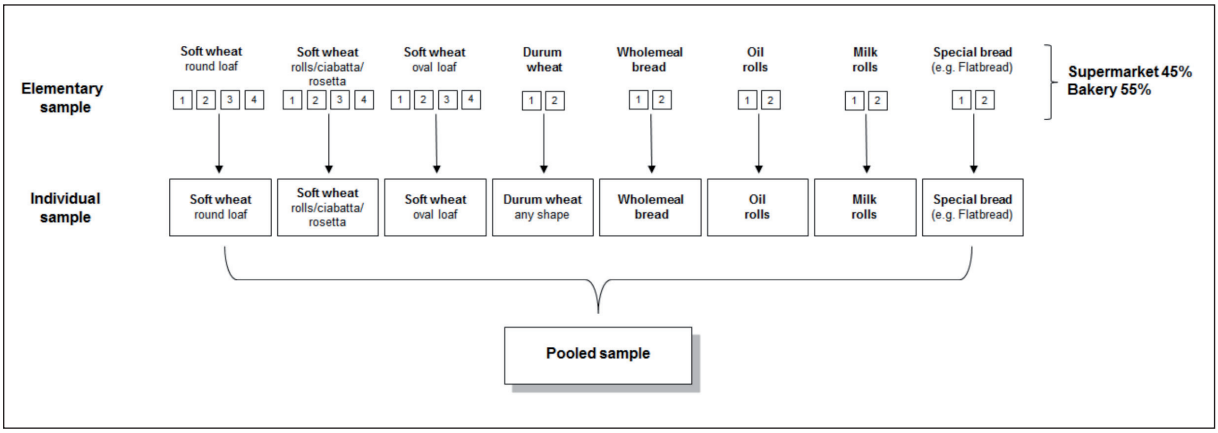

Figure 1
Sampling scheme for the preparation of the pooled sample "bread".

Table 1
TDS Food List showing the average daily consumption in g/d by food category in the total population (all ages, males and females), the percentage contribution of each food (in parenthesis), the percentage of consumers of each food and food category, the TDS sampling year, the number of TDS samples analysed (pooled samples) and collected at retail (individual sample)

Food categories	Consumption	Consumers (%)	Sampling year	Pooled samples	Individual samples
Cereals, cereal products and substitutes	**258.4**	**99.8**			
Bread	(40)	92.1	1	4	32
Pasta	(21)	91.1	1	4	32
Pizza	(3)	13.9	1	4	16
Rice	(6)	41.2	1	4	16
Wheat, other cereals and flours	(14)	84.1	1	4	16
Breakfast cereals	(1)	10.1	1	4	16
Biscuits	(5)	50.6	1	4	16
Savoury fine bakery products	(3)	38.0	1	4	16
Cakes and sweet snacks	(7)	44.4	1	4	16
Pulses, fresh and processed	**11.3**	**34.6**	1	4	16
Vegetables, fresh and processed	**211.2**	**99.6**			
Leafy vegetables, fresh	(20)	84.0	3	4	16
Tomatoes, fresh	(20)	83.6	3	4	16
Other fruiting vegetables, fresh	(15)	64.3	3	4	16
Roots and onions, fresh	(9)	97.8	3	4	16
Other vegetables, fresh	(18)	82.9	3	4	16
Vegetables, processed	(17)	78.0	3	4	16
Spices and herbs	(1)	83.1	3	4	16
Potatoes, tubers and their products	**50.9**	**69.2**	1	4	16
Fruit, fresh and processed	**208.5**	**93.7**			
Citrus fruit, fresh	(22)	46.9	3	4	16
Exotic fruit, fresh	(8)	38.9	3	4	16
Other fruit, fresh	(68)	83.1	3	4	32
Nuts, seeds, olives and their products, dried fruit	(1)	27.1	3	4	16
Meat, meat products and substitutes	**110.1**	**99.0**			
Beef and veal, not preserved, excl. offal	(39)	75.2	3	4	16
Pork, not preserved, excl. offal	(12)	31.4	3	4	16
Poultry and game, not preserved, excl. offal	(19)	42.4	3	4	16
Other meats, not preserved, excl. offal	(5)	10.2	3	4	16
Ham, salami, sausages and other preserved meats, excl. offal	(25)	81.3	3	4	16
Offal, blood and their products	(1)	3.3	3	4	16
Fish, seafood and their products	**44.7**	**68.0**			
Fish	(70)	62.0	2	4	16
Crustaceans and molluscs	(30)	21.8	2	4	16
Milk, milk products and substitutes	**198.0**	**99.2**			
Milk, milk-based beverages, infant formula	(60)	78.6	2	4	32
Yoghurt and fermented milk	(10)	86.3	2	4	16
Cheese	(29)	96.7	2	4	32

(continues)

Table 1 (continued)

Food categories	Consumption	Consumers (%)	Sampling year	Pooled samples	Individual samples
Oils and fats	**40.4**	**99.7**			
Olive oil	(81)	99.7	3	4	16
Other vegetable oils	(6)	41.8	3	4	16
Butter and creams	(10)	45.7	3	4	16
Other fats	(2)	17.9	3	4	16
Eggs	**20.9**	**74.3**	2	4	16
Alcoholic beverages	**91.0**	**74.5**			
Regular wine	(70)	69.7	2	4	32
Beer, cider	(27)	16.6	2	4	16
Sweet wine, spumante, wine-based appetizers, spirits and liquors	(3)	13.2	2	4	16
Sweet products and substitutes	**33.1**	**93.2**			
Ice cream and ice lolly	(30)	20.3	3	4	16
Chocolate and substitutes	(8)	22.7	3	4	16
Candies, jam and other sweet products (incl. sugar-free)	(10)	26.6	3	4	16
Sugar, fructose, honey and other nutritious sweeteners	(50)	84.9	3	4	16
Cocoa and cocoa-based powder	(2)	9.6	3	4	16
Water and other non-alcoholic beverages	**836.1**	**99.9**			
Tap water (as such, in beverages or recipes)	(23)	57.1	1	4	16
Bottled water	(54)	76.5	2	4	32
Coffee, tea, and herbal tea	(15)	87.7	2	4	32
Fruit and vegetable juices	(4)	56.2	2	4	16
Other soft drinks	(3)	21.8	2	4	16

used throughout (*e.g.*, a concentration of 9 g/L of salt was used for boiling pasta and rice).

ANALYTICAL TECHNIQUES AND QUALITY ASSURANCE

Analyses are performed by a network of three specialized laboratories of the Italian National Health Service, including ISS that acts as coordinator of the network, in compliance with good laboratory practice, quality control procedures and the ISO/IEC 17025 standard. Inorganic arsenic is determined by HPLC-ICP-MS after chemical extraction [19, 20]. Aluminium, cadmium, lead, and uranium are determined after microwave digestion by ICP-MS (see EN 15763 and [21, 22]), whereas CV-AAS using a Flow Injection Mercury System is the analytical technique for mercury (see EN 13806 and [23, 24]). Regarding radionuclides, ^{40}K, ^{134}Cs, ^{137}Cs are determined by direct gamma-ray spectrometry with a high-purity germanium (HPGe) detector cooled by liquid nitrogen [25]. ^{90}Sr activity concentration is measured by accurate radiochemical methods with ultra-low level liquid scintillation counting [26].

All analytical methods are validated for the food matrices and the analytes under study, and were adopted for the 2012-2014 Italian TDS after scrutiny of the limits of detection (LOD) and limits of quantification (LOQ) achieved. In order to reduce uncertainties in exposure assessment it is important to minimize the number of analytical results that may fall below those limits (*i.e.* left-censored data) due to the dilution caused by the pooling process. For trace elements, the LODs (LOQs) achieved based on the 3σ (6 σ) criterion – where σ is the standard deviation of the measurement of 20 method blanks – expressed as µg/g fresh food are in the order of 0.001 (0.003) for inorganic arsenic, 0.2 (0.6) for aluminium, 0.0003 (0.001) for cadmium, 0.002 (0.006) for lead, and 0.0001 (0.0004) for uranium. These values have been compared with occurrence levels for the different TDS food groups – data produced by the participating laboratories and from recent TDS [27, 28] have been used for this purpose – and it has been assessed that LODs/LOQs achieved are adequate for the quantification of the analytes in the vast majority of cases; thus a small proportion of left-censored data is expected.

For gamma-emitting radionuclides (^{134}Cs, ^{137}Cs, ^{40}K) and ^{90}Sr, a pure beta emitter, lower limits of detection are 0.1 Bq/kg (^{134}Cs, ^{137}Cs), 2.4 Bq/kg (^{40}K) and 8 mBq/kg, respectively. Gamma-emitting radionuclides, with

Table 2
Concentration of trace elements in tap water collected at four different sites in each of the four TDS sampling areas (µg/L). RSD (%) in parentheses

Sampling area	Site	Al	As	Cd	Hg	Pb	U
1	1.1	5.1	0.9	0.009	0.03	0.10	5.5
		(1.0)	(0.2)	(0.1)	(0.1)	(0.3)	(0.3)
	1.2	4.0	0.9	0.004	0.02	0.07	9.6
		(0.3)	(0.3)	(0.1)	(0.1)	(0.1)	(1.9)
	1.3	3.8	0.9	0.003	0.02	0.05	4.6
		(0.1)	(0.4)	(0.1)	(0.1)	(0.1)	(0.5)
	1.4	4.1	0.8	0.003	0.02	0.06	10.5
		(0.4)	(0.2)	(0.1)	(0.1)	(0.1)	(0.8)
2	2.1	2.7	2.7	0.008	0.03	0.49	1.6
		(2.1)	(0.5)	(0.1)	(0.1)	(0.1)	(0.2)
	2.2	0.4	0.7	0.005	0.02	0.25	0.7
		(0.1)	(0.2)	(0.1)	(0.1)	(0.1)	(0.1)
	2.3	1.2	0.6	0.011	0.02	0.66	0.8
		(0.7)	(0.2)	(0.1)	(0.1)	(0.1)	(0.1)
	2.4	1.9	6.1	0.005	0.04	0.85	4.0
		(2.7)	(0.5)	(0.1)	(0.1)	(0.2)	(0.2)
3	3.1	51.4	0.2	0.011	0.02	0.42	0.9
		(0.6)	(0.1)	(0.1)	(0.1)	(0.1)	(0.3)
	3.2	113.8	0.2	0.012	0.01	0.49	0.8
		(2.8)	(0.1)	(0.1)	(0.1)	(0.1)	(0.1)
	3.3	68.6	0.2	0.006	0.01	0.06	0.8
		(0.4)	(0.1)	(0.1)	(0.1)	(0.1)	(0.1)
	3.4	38.0	0.2	0.049	0.01	0.57	0.9
		(0.2)	(0.2)	(0.1)	(0.1)	(0.2)	(0.2)
4	4.1	40.0	0.4	0.007	0.02	0.09	3.3
		(0.6)	(0.1)	(0.1)	(0.1)	(0.1)	(0.1)
	4.2	51.7	0.4	0.008	0.02	0.19	3.1
		(0.3)	(0.1)	(0.1)	(0.1)	(0.1)	(0.1)
	4.3	56.5	0.4	0.005	0.02	0.08	3.2
		(0.8)	(0.1)	(0.1)	(0.1)	(0.1)	(0.1)
	4.4	57.5	0.4	0.040	0.02	0.20	3.0
		(1.0)	(0.1)	(0.1)	(0.1)	(0.1)	(0.1)

the exception of ^{134}Cs, are expected to be detected in most samples, whereas ^{137}Cs is likely to be detectable only in some food categories. Although Cs isotopes, particularly ^{134}Cs, are rarely detected in most foods, it is important to continue monitoring the presence of these radionuclides as it is currently done by several national bodies, including the US-FDA [29-31].

Quality control (QC) in all the participating laboratories includes internal QC, *i.e.*, use of control samples (calibrants, spiked samples, replicate samples) and of certified reference materials (CRMs), and external QC, *i.e.* participation to Proficiency Tests and Interlaboratory Comparisons with consistent good performance. Several CRMs (*e.g.* SRM 1570a, SRM 1567a, CRM 679, SRM 2976) are being used in order to account for the variety of food matrices and analytes investigated. Materials certified for inorganic arsenic (*e.g.* ERM-BC211) and radionuclides (IAEA-414, IAEA-152, IAEA-330) are included. ERM CA021a is being used to check the accuracy of the analyses of water and other non-alcoholic beverages.

EXPOSURE ASSESSMENT AND RISK CHARACTERIZATION

Exposure will be assessed by combining the occurrence data from the analyses with the individual consumption data from the INRAN-SCAI 2005-06 survey. Mean exposure and 95th percentile will be calculated for the total population and for consumers of each specific core food. Exposure of both the

Table 3
Health-based guidance values (HBGVs) for trace elements and their species

Element/ species	Evaluation	Year	HBGV	Value	Unit	Toxicological data	Hazard[a]	Reference
Al	EFSA	2008	TWI	1	mg/kg bw/week	Animal	Neurotoxicity, embryotoxicity, and effects on the male reproductive system and the developing nervous system	[7, 34]
	JECFA	2011	PTWI	2	mg/kg bw/week	Animal	Renal damage and reduced grip strength	[8]
Inorganic As	EFSA	2009	$BMDL_{01}$[b]	0.3-8	µg/kg bw/day	Human	Cancers of the lung, skin and bladder, and skin lesions	[4]
	JECFA	2010	$BMDL_{0.5}$[c]	3	µg/kg bw/day	Human	Lung cancer	[5]
Inorganic Hg	EFSA	2012	TWI	4	µg/kg bw/week	Animal	Kidney weight changes	[6]
	JECFA	2010	PTWI	4	µg/kg bw/week	Animal		[5]
Methyl-Hg	EFSA	2012	TWI	1.3	µg/kg bw/week	Human	Developmental neurotoxicity	[6]
	JECFA	2003	PTWI	1.6	µg/kg bw/week	Human		[3, 35]
Cd	EFSA	2009	TWI	2.5	µg/kg bw/week	Human	Nephrotoxicity. It also causes bone demineralisation, either through direct bone damage or indirectly as a result of renal dysfunction	[1, 36]
	JECFA	2010	PTMI	25	µg/kg bw/month	Human		[37]
Pb	EFSA	2010	$BMDL_{01}$	0.5	µg/kg bw/day	Human	Developmental neurotoxicity in young children	[2]
			$BMDL_{01}$	1.50	µg/kg bw/day	Human	Effects on systolic blood pressure in adults	
			$BMDL_{10}$	0.63	µg/kg bw/day	Human	Effects on prevalence of nephrotoxicity in adults	
U	EFSA	2009	TDI	0.6	µg/kg bw/day	Animal	Nephrotoxicity	[9]

[a] Critical effect, *i.e.* most sensitive toxicological endpoint
[b] Range of benchmark dose lower confidence limit values for 1% excess risk of cancers of the lung, skin and bladder, and skin lesions
[c] Benchmark dose lower confidence limit values for 0.5% increased incidence of lung cancer.

general population and each subgroup of population (according to age and gender) will be estimated both at the national levels and for each of the four main geographical areas of Italy.

For left-censored values, the lower and upper bound approach will be adopted [12]. In case the upper and lower bound values are close to each other, the upper bound value will be used as exposure estimate, whereas if they are significantly different the entire range of dietary exposure will be considered.

Methylmercury is considerably more toxic than inorganic mercury, therefore separate exposure assessment is needed for these two chemical species. Methylmercury exposure occurs through the food category fish and seafood. In order to derive occurrence level from total mercury analytical data for the core foods in this category, the assumption will be made according to EFSA that 100% and 80% of total mercury is present as methylmercury in fish meat and in crustaceans/ mollusks, respectively [6]. For inorganic mercury it will be assumed that the respective percentages are 20% and 50%, whereas for all other food categories total mercury will be regarded as occurring in the form of inorganic mercury only. It has to be noted that because of this conservative approach, total mercury dietary exposure cannot be derived by adding inorganic and methylmercury dietary exposure together.

For risk characterization, the estimated exposure to trace elements is compared with available health-based guidance values *(Table 3)*. The percentages of consumers exceeding the health-based guidance values will be assessed. If a benchmark dose limit (BMDL) is available instead of a Tolerable Intake, the margin of exposure (MOE) is calculated based on the mean and P95 exposure for the general population and each subgroup. In the case of radionuclides, annual effective doses will be estimated according to ICRP methodologies. The exposure dose through ingestion of each radionuclide will be based on nuclide intakes (Bq) and coefficient for conversion to the effective dose (mSv/Bq), taking into account age-dependent factors and consumer patterns [32, 33]. Total annual estimates will be compared those internationally available [10, 29-31].

CONCLUDING REMARKS

The 2012-2014 Italian TDS entails the collection of > 3000 food items, bought at retail in 4 cities selected to represent the four main geographical areas of Italy. The food items will be pooled according to the TDS food list, based on individual food consumption data, into 51 core foods accounting for 99.7% of the whole diet of adults and children. Three sampling campaigns will be undertaken and seasonal aspects will be accounted for by sampling fruits and vegetables in two different seasons.

Dietary exposure to 11 trace elements, element species and radionuclides will be assessed for the general population and for children, teenagers, adults, and elderly of the two sexes, both at the national level and for each of the four main geographical areas of Italy. The lower and upper bound approach will be adopted to deal with left-censored data. For risk characterization, the exposure will be compared with health-based guidance values for trace elements whereas annual effective doses will be estimated for radionuclides.

Proper attention is given to speciation of arsenic and mercury, since in both cases dietary risk characterization requires an understanding of the chemical form present. In the case of arsenic, analytical speciation data are produced in order to selectively determine toxic inorganic arsenic. In the case of mercury, reasonable assumptions have been made to estimate the contribution of the different food groups to methylmercury and inorganic mercury exposure based on analytical data for total mercury.

Tap water from the four main sampling areas has been closely investigated due to its significance both as a direct source of exposure and for its impact on analyte levels during cooking. Water samples from each sampling site have been characterized individually in order to allow the calculation of specific exposure scenarios based on the use of drinking water with widely differing levels of the various contaminants.

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Conflict of interest statement

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