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Dietary exposure estimates of twenty-one trace elements from a Total Diet Study carried out in Pavia, Northern Italy

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The significant role of trace elements in human health is well documented. Trace elements are those compounds that need to be present in the human diet to maintain normal physiological functions. However, some microelements may become harmful at high levels of exposure, or, on the other hand, may give rise to malnutrition, when their exposure is too low. The aim of the present study was to provide a reliable estimate of the dietary exposure of twenty-one trace elements in a Northern Italian area. For this purpose, trace element analyses were undertaken on total diet samples collected from a university cafeteria in Pavia, Northern Italy. The average daily exposure for the adult people was calculated on the basis of food consumption frequency, portion size and trace element levels in foodstuffs. The mean exposure values satisfy the Italian RDA for all the essential trace elements, except for Fe exposure in females, and are well below the Provisional Tolerable Daily Intake for all the toxic compounds, showing that the probability of dietary exposure to health risks is overall small. As far as Fe exposure is concerned, a potential risk of anaemia in the female adult population should be considered, then studies aimed at evaluating the Fe nutritional status of adult Italian women should be addressed. In conclusion, while not excluding the possibility that the daily exposure determined in the present study may not be representative of the population as a whole, this study provides a good estimate of the Italian adult consumer exposure to twenty-one trace elements.

Trace elements: Total Diet Study: Market basket: Northern Italy

The very important and significant role of trace elements in human health is well documented[1]. Trace elements are those compounds that need to be present in the human diet in order to maintain normal physiological functions. However, microelements may become harmful at high levels of exposure (e.g. Se, Cr, Cu, Zn)[1], or, on the other hand, may give rise to malnutrition, when their exposure is too low. While some elements (e.g. Cr, Co, Cu, Fe, Mo, Mn, Zn etc.) are essential to health, other elements are likely to be essential (e.g. Ni, B, Va), although their positive role in human nutrition remains to be confirmed. In addition, other elements have no proven essential functions in man and are likely to have adverse physiological effects (e.g. Al, As, Li, Sn)[1].

Furthermore, some elements (e.g. Pb, As, Hg, Cd) are well known to be toxic if their exposure through diet and/or inhalation is excessive. For example, exposure to Pb can be harmful to neuropsychological development, inorganic As has been associated with human cancer and other adverse health effects, organic Hg compounds are neurotoxins and Cd may affect renal function[1]. Other elements can cause short-term health effects from one incidence of high-level exposure. For example, high concentrations of Sn in foods can cause stomach upsets. Finally, a metal can be a nutrient or be a toxic compound depending on its chemical form (e.g. Cr^{3+} and Cr^{6+}).

Indeed imbalance, as well as deficiency or excess in trace element dietary exposure, can have deleterious influences on human health[1,2].

It is well known that the amount of trace elements ingested by man depends on dietary habits as well as on their quantities in foodstuffs that are related to soil characteristics which can affect the bioavailability of elements.

Therefore, estimation of dietary exposure to trace elements in population groups is of great relevance for assessing the adequacy of the diet with respect to essential elements and for evaluating a possible risk for high levels of exposure to those that are toxic.

The Total Diet Study (TDS), consisting of analysing a representative 'market basket' of foods usually prepared for normal consumption, is the best method widely used in several European and extra European countries, aimed at estimating dietary exposure of nutrient and toxic compounds and it represents the most cost-effective and reliable method[3–8]. It has the advantage of yielding more refined exposure data, in that foods are analysed 'as consumed' by the eater.

Abbreviations: ICP-MS, inductively coupled plasma MS; ICP-OES, inductively coupled plasma emission spectrometry; NIST, National Institute of Standards & Technology; PTDI, Provisional Tolerable Daily Intake; SRM, Standard Reference Materials; TDS, Total Diet Study.

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The 'market basket' method is indeed considered an important tool for monitoring nutrient and toxic compound dietary exposure and then assessing compliance with nutritional requirements and with provisional acceptable exposure for toxic elements.

To carry out TDS, samples of food are usually collected in representative centres and weighed out in the proportions in which they are consumed in the total diet. This approach is particularly important because it determines levels of the analytes in foods as they would be consumed, so that they resemble as much as possible the usual pattern of the total diet. Moreover, it is suitable for estimating the dietary exposure of compounds which may be changed as a result of washing, peeling and cooking.

The aim of the present study was to provide a reliable estimate of the dietary exposure to twenty-one trace elements in a Northern Italian area. For this purpose, trace element analyses were undertaken on total diet samples collected from a university cafeteria in Pavia, Northern Italy. The average daily exposure for adults was calculated on the basis of food consumption frequency, portion size and trace element levels in foodstuffs. The estimated ingestion of essential trace elements was then compared with the average requirement values of the Italian RDA[9], while exposure to toxic elements was compared with the Provisional Tolerable Daily Intake (PTDI), as established by the FAO/WHO[1]. Comparisons were also made with results of previous world-wide studies. In addition, the contribution of each food group to the mean trace element daily exposure was evaluated.

Materials and methods

Food choice and dietary consumption data

In the present study, the choice of the foods and their consumption data utilized for the formulation of the 'market basket' of this Italian TDS were drawn from the Italian Household National Survey carried out in 1994–6 by the Italian National Institute for Food and Nutrition Research[10].

This survey is the most representative Italian food consumption study since it covered 1978 subjects randomly selected to be representative of the four main geographical areas in Italy (Northwest, Northeast, Centre, South).

Foods selected representing the same food category were aggregated into six main different food groups using the same aggregation used in the Italian Household National Survey[10]: bread, cereals and potatoes; meat and meat products, fish and fish products, eggs (protein-rich foods); milk and dairy products; fruit and vegetables; sweet foods; beverages.

The cereals food group was composed of four items (bread and pizza, pasta, rice, potatoes); the protein-rich foods of six items (beef, pork, poultry, ham and salami, fish and sea foods, eggs); the milk and dairy products group of three items (milk, yogurt, cheese); the fruit and vegetables group of four items (vegetables, legumes, citrus fruit, fresh fruit); the sweet food group of two items (dessert, ice cream) and the beverage group of four items (mineral water, soft drinks, wine, beer).

Food sampling

The 'market basket' method was slightly modified. Because our laboratory does not provide a kitchen in which to prepare and cook all the food items purchased in local markets following the Italian traditional recipes as the TDS approach suggests, we collected most samples of foods in a cafeteria. Therefore, raw, cooked and ready-to-eat foods were collected over 2 consecutive weeks in July 2004, from a university cafeteria located in Pavia, Northern Italy. This large cafeteria distributes about 2000 meals/d. When necessary, some foods (pasta, rice, boiled meat, fish, eggs and vegetables) were prepared using local drinking tap water. A sample of each food that was prepared daily was collected. Since the food collection was performed only in July, our dietary estimations did not take into account seasonal variation.

Some traditional breakfast foods, such as biscuits, sweets, cereals and milk, which were not served at the cafeteria, were purchased at three local supermarkets as well as a few foods that were included in the Italian Household National Survey[10], but not served at the cafeteria.

All the foodstuffs (226 from the cafeteria and twenty-two from the supermarkets) are listed in Table 1.

Each sample was weighted, given a name and numerical code, and the list was kept on record. Collected items were immediately transferred to the laboratory where the pre-analytical treatment was performed.

Standards and reagents

Multielemental standard solutions were obtained from 10 mg/l and inductively coupled plasma MS (ICP-MS) calibration standard numbers 3, 4, MS3, MS1 (CPI International, Amsterdam, The Netherlands), by dilution with water containing the same amount of acids as the samples.

Distilled water was produced using a Milli-Q™ deionizing system (Millipore, Bedford, MA, USA).

Suprapur reagents were used: HNO_3 (65 % m/v), HCl (37 % m/v) and HF (40 % m/v) (Merck, Darmstadt, Germany).

Certified reference materials

National Institute of Standards & Technology (NIST) Standard Reference Materials (SRM) 1573 *Tomato Leaves*, NIST SRM 1547 *Peach Leaves*, NIST SRM 1567 *Wheat Flour* and NIST SRM 1575 *Pine Needles* were used to evaluate the accuracy of the analytical procedures.

Sample preparation

Food samples, except for dairy products and beverages, were slightly thawed and cut into pieces. They were then pooled and homogenized with double-distilled and deionized water until completely pureed, by a STERILMIXER blender (PBI International, Milan, Italy). The samples were stored in polypropylene containers and frozen at −18°C until analysis.

A microwave CEM model MARS-Xpress (CEM Corp., Matthews, NC, USA) was used to digest all the samples. Digestion vessels were cleaned with 10 ml HNO_3 using the microwave cleaning programme and were rinsed with deionized water supplied by a Milli-Q™ Laboratory Water System (Millipore). Homogenized samples (800 mg) were added to the digestion vessels with 4 ml HNO_3, 4 ml HCl and 200 μl HF. After complete digestion and cooling, the

Table 1. Food products collected from the cafeteria and bought at the supermarkets

Sample	No. of samples collected from the cafeteria	No. of samples bought at the supermarkets
Cereals and tubers		
Bread	10	
Pizza margherita		1
Pizza (wurstel, ham, onions)	1	
Crackers, breadsticks		1
Cornflakes		1
Polenta (thick corn mush)	2	
Pasta with eggplants and peppers	1	
Pasta with tomato and ricotta cheese	2	
Pasta with tomato	9	
Pasta amatriciana (tomato and smoked bacon)	2	
Pasta with cream, peas, carrots, potatoes	1	
Pasta with zucchini, eggplants and peppers	3	
Pasta with tuna and tomato	1	
Pasta with gravy	2	
Pasta with tomato and rocket	1	
Pasta with cream and gorgonzola	1	
Pasta with seafood	1	
Pasta with cauliflower	1	
Risotto with spinach	2	
Risotto with pork meat	1	
Risotto with carrots, peas and potatoes	2	
Risotto with mushrooms	1	
Risotto with peas	1	
Risotto with artichokes	1	
Fried rice balls	1	
Potato dumplings with gravy	1	
Potato dumplings with cream and gorgonzola	1	
Boiled potatoes	10	
Fried potatoes	9	
Meat and meat products, fish and fish products, eggs, legumes		
Beef steak	4	
Beef shank	1	
Beef stew with peas and potatoes	2	
Meatballs (pork, poultry, eggs)	3	
Pepper stuffed with minced meat	2	
Pork cutlet	1	
Pork cutlet with mushrooms	1	
Roasted turkey	6	
Grilled turkey breast	1	
Roasted chicken (wing, breast)	2	
Breaded chicken breast	2	
Breaded hamburger	1	
Hamburger (pork and turkey)	3	
Tripe	1	
Parma ham	4	
Ham steak	10	
Dried beef	4	
Speck ham	2	
Breaded cod fillet	2	
Fried cod sticks	1	
Boiled shrimps		1
Flounder		1
Tuna fish in olive oil		1
Hard-boiled eggs	4	
Omelette with spinach and ricotta cheese	2	
Boiled green beans	6	
Boiled peas	2	
Stewed peas with tomato sauce	1	
Boiled beans (French, haricot, white)		3
Green olives		1
Milk and dairy products		
Whole milk		1
Cream		1
Cheese (fontina, smoked provola, asiago, taleggio, primo sale)	10	
Cheese (mozzarella, Philadelphia, caprino)	10	
Yogurt (apricot)	1	
Yogurt (banana)	1	
Yogurt (strawberry)	1	
Yogurt (raspberry, blackcurrant and blueberry)	1	
Fruit and vegetables		
Boiled carrots	4	
Green salad	7	
Boiled cauliflower	3	
Boiled beetroots	4	
Boiled fennel	5	
Boiled spinach	3	
Boiled Brussels sprouts	3	
Tomato salad	6	
Mixed salad	3	
Mixed big salad (tuna fish, potatoes, peas, carrots, olives, tomatoes, green beans, corn, green salad)	2	
Russian salad (mayonnaise, peas, carrots, potatoes)	1	
Fruit (kiwi, peach, apple, banana)	10	
Citrus fruit (mandarin, orange, grapefruits)		3
Nuts, peanuts		2
Dried apricots		1
Cakes and desserts		
Creamy tiramisù	1	
Chocolate pudding	1	
Crème-caramel pudding	1	
Vanilla pudding	1	
Lemon ice-cream	1	
Vanilla and chocolate ice-cream	1	
Beverages		
Bottled mineral water	1	
Bottled mineral water (sparkling)	1	
Red wine	1	
White wine	1	
Beer	1	
Cola-flavoured carbonated beverage	1	
Orange-flavoured carbonated beverage	1	
Lime-flavoured carbonated beverage	1	
Fats and oils		
Olive oil		1
Seed oil		1
Butter		1
Margarine		1
Total	226	22

samples were filtered and transferred to 50 ml graduated polypropylene tubes and diluted to volume with deionized water.

SRM were handled according to NIST's specifications and were treated as previously described for food samples.

Liquid samples (beverages; 10 ml) were treated with 4 ml HNO_3 and 4 ml HCl, heated at 90°C for about 1 h and, after cooling, diluted to 50 ml with deionized water.

Inductively coupled plasma emission spectrometry (ICP-OES) and ICP-MS were used to determine the following twenty-one trace elements: Al, B, Ba, Be, Cd, Ce, Co, Cs, Cu, Fe, Ge, Li, Mo, Mn, Ni, Pb, Rb, Sn, Sr, V, Zn.

Apparatus and analytical conditions

Measurements of trace elements were carried out using an inductively coupled plasma mass spectrometer (ELAN 6100 DRCII ICP-MS; Perkin-Elmer SCIEX Instruments, Concord, Ontario, Canada) equipped with a cyclonic spray chamber with a concentric nebulizer, a dynamic reaction cell, a quadrupole mass filter and AS 90plus auto-sampler (Perkin-Elmer). The dynamic reaction cell can be pressurized with reactive gases (CH_4, NH_3) and is vented and pressurized under computer control. The dynamic reaction cell provides online chemical modification of the ion beam to eliminate interferences. The specificity of interference rejection is obtained through the selection of the reaction gas and the operating conditions. In the vented 'standard mode' no reaction gas is present in the cell.

B, Be, Li, Ce, Cs, Ge, Mo and Pb were determined in standard mode while Al, Cd, Ba, Cu, Co, Ni, Mn, Rb, Sn, V and Zn were determined in enhanced mode (CH_4, NH_3).

The ICP-OES analyses were performed on a Perkin-Elmer Optima 4300DV, equipped with a standard torch, a Scott-type spray chamber in Ryton, a Gem-Tip cross-flow nebulizer and an AS-90plus auto-sampler; the detector is a custom, two-dimensional CCD array; the plasma source is a dual-view (can be viewed either axially or radially) RF generator. The elements determined by ICP-OES were Al, Mn, Fe and Sr. All the measurements were carried out in axial mode.

Estimation of trace element dietary exposure

The mean, minimum and maximum trace element daily exposures (mg/d, μg/d or ng/d) were estimated by multiplying the mean, minimum and maximum concentrations (μg/g or ng/g) in any one food item by the average amount of that food consumed by the North Western Italian adult population[10] expressed as g food/d or ml beverage/d. Total exposure of each trace element was then obtained by summing the exposure from all food items, after which they were compared with the average requirement values of the Italian RDA[9] for essential trace elements, while toxic element exposures were compared with PTDI, as established by the FAO/WHO[1].

In addition, the contribution of each food group to the total daily exposure of each element was evaluated. Food items were aggregated according to the same aggregation in food groups used in the Italian Household National Survey[10] as above earlier. The exposure estimates were based on the average food amounts consumed by the entire population (consumer and non-consumer alike).

Results

The validation of the analytical methods was carried out using the certified reference materials mentioned earlier.

Accuracy was ascertained by adding to each series of samples one of the certified reference materials; precision was monitored by replicating the entire analytical procedure five times in all cases.

Recovery is acceptable: percentages range from 80 to 120 % depending on the element. Only for elements whose measured concentrations are close to detection limit (V in NIST 1547), or for B in NIST 1547 the recovery is 140 and 70 %, respectively.

Precision is also satisfactory: relative standard deviation values are always less than 20 % with the exception of Cu in NIST 1567 (35 %). Higher relative standard deviation values are found only in the case of elements whose concentrations are less than 0·1 μg/g.

The results for NIST SRM 1547 *Peach Leaves*, NIST SRM 1573 *Tomato Leaves*, NIST SRM 1575 *Pine Needles* and NIST SRM 1567 *Wheat Flour* are summarized in Table 2.

The detection limits are 0·05 μg/l for the elements of liquid samples analysed by ICP-MS and 5 μg/l for those determined by ICP-OES. As for the solubilized samples, the detection limits are 0·001 μg/g for ICP-MS and 0·1 μg/g for ICP-OES.

Table 3 shows the mean, minimum and maximum daily exposure to twenty-one trace elements in the TDS in a Northern Italian town, Pavia, compared with the Italian RDA[9] for the essential trace elements and with PTDI for the toxic compounds, where these values are provided by FAO/WHO[1]. The percentage contribution of each estimate compared with the Italian RDA or PTDI is also reported. In addition, results from the literature of various TDS are reported in the same table, so that our data can be easily compared with those of other studies.

Table 4 shows the percentage contribution of each food group to the total daily exposure of twenty-one trace elements. Cereals and tubers is the food category that, in general, mostly contributes to the whole trace element total daily exposure, except for Ce; indeed it provides 97 % of the total Sn daily exposure. Protein-rich foods are a good supply of Cs, Cu, Fe, Rb and Zn; milk and dairy products are the foods that mostly contribute Al, Ba, Ce, Ni and Zn; fruit and vegetables are good suppliers of all the elements, except for Ce and Sn; finally, the contributions of sweet foods and beverages are negligible.

The Al exposure is 3·58 mg/d (range 0·28–5·15 mg/d), which is well below the Provisional Tolerable Weekly Intake (7 mg/kg body weight per week for an adult man of 65 kg body weight)[1] and it falls in the range of the values reported in the literature (1·9–12 mg/d)[6,7,11–17].

The B exposure is 1·65 mg/d (range 0·54–3·19 mg/d) that is slightly higher than the mean basal requirements (range 0·5–1·35 mg/d), but below the toxic level of 13 mg/d[1]. The B exposure is very similar to the data reported in the literature, ranging from 0·89 to 2·12[6,11,12,18–21] and our estimation is consistent with these reports stating that normal adult daily B exposures are approximately 1 mg. The Ba exposure is 366·6 μg/d (range 53·1–398·3 μg/d). There is no reference value with which to compare it. The Be exposure is 12·0 ng/d (range 4·0–14·0 ng/d) and also for this element there is no reference value with which to compare it, nor

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Table 2. Concentrations (μg/g) of elements in National Institute of Standards & Technology (NIST) Standard Reference Materials (SRM) after microwave digestion and inductively coupled plasma MS analysis (*n* 5)*

(Mean values and standard deviations)

	NIST SRM 1547				NIST SRM 1573				NIST SRM 1567				NIST SRM 1575			
	Certified		Found		Certified		Found		Certified		Found		Certified		Found	
Element	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Al	249	8	230	14	–		–		–		–		545	30	490	27
B	29	2	22	2	–		–		–		–		17‡	3	16·5	3
Ba	124	4	120	6	–		–		–		–		6·2‡	1·7	5·8	1·2
Cd	0·026	0·003	0·024	0·002	2·5‡	0·2	1·9	0·3	0·032	0·007	0·027	0·008	<0·5	–	<0·5	–
Co	0·07†	–	0·16	0·03	0·53‡	0·05	0·48	0·05	0·021‡	–	0·025	0·006	0·1†	–	0·14	0·06
Cu	3·7	0·4	3·2	0·5	11	1	10	1	2·0	0·3	1·8	0·7	3·0	0·3	2·8	0·4
Fe	218	14	205	15	690	25	695	28	18·3	1·0	17·8	1·9	200	10	199	22
Mn	98	3	96	4	238	7	225	10	8·5	0·5	8·8	0·8	675	15	690	11
Ni	0·69	0·09	0·79	0·10	1·17‡	0·08	1·18	0·05	0·180†	–	0·170	0·11	3·5†	–	2·9	0·6
Pb	0·87	0·03	0·77	0·05	6·3	0·3	5·2	0·5	0·02	0·01	0·02	0·01	10·8	0·5	11·2	1·1
Sr	53	4	45	5	44·9	0·3	44	10	0·97‡	0·11	1·0	0·20	4·8	0·2	4·5	0·3
V	0·37	0·03	0·52	0·05	1·3‡	0·3	1·1	0·2	11·25‡	–	11·3	1·1	0·4‡	0·05	0·38	0·04
Zn	17·9	0·4	19·3	0·5	62	6	61	8	10·6	1·0	12·1	1·8	67‡	9	66	7

* For details of procedures, see the Materials and methods.
† Values reported but not certified.
‡ Consensus values reported by Gladney *et al.*[60].

any study of Be exposure in the literature in order to compare our data. The Cd exposure is 13·6 μg/d (range 1·4–32·7 μg/d) which is well below the PTDI of 65 μg/d (7 μg/kg body weight per week for an adult man of 65 kg body weight)[1]. This estimated value is in the range of those reported in various European and extra European countries (10·0–50·0 μg/d)[7,16,22–34]. The Ce exposure is 154·8 μg/d (range 124·5–197·2 μg/d). There is no reference value with which to compare it, nor any study of Ce exposure in the literature in order to compare our data. The Co exposure is 29·0 μg/d (range 15·0–57·0 μg/d). There are no recommended dietary allowances for this element. Our value is in the range of those reported in various European and extra European countries (4·0–29·0 μg/d)[11–13,15,35]. The Cs exposure is 4·91 mg/d (range 0·54–7·93 mg/d) and it falls within the range of values reported in the literature (1·46–9·0 mg/d)[12,35–37]. There is no reference value with which to compare it. The Cu exposure is 1·14 mg/d (range 0·73–1·96 mg/d) and it adequately satisfies the Italian RDA[9] (1·2 mg/d). Our estimated value is in agreement with those reported in a wide recent review obtained from twenty-four European countries (0·54–1·77 mg/d)[38,39]. The Fe exposure is 11·0 mg/d (range 6·41–19·45 mg/d) and it satisfies the Italian RDA[9] for a male (10 mg/d), but it is below the recommended value for an adult woman (18 mg/d). It is in the range of the value reported in the literature (9·0–15·9 mg/d)[6,12,13,35,39–41]. The Ge exposure is 10·7 μg/d (range 1·1–17·5 μg/d). There is no reference value with which to compare it. Our value is slightly higher than the one reported by Ysart *et al.*[6] in the UK, equal to 7·0 μg/d. The Li exposure is 29·9 μg/d (range 8·2–42·4 μg/d). This element is known to be essential for laboratory animals, but there is no scientific evidence on its essentiality for man[1]. Our value is in the range of those of the literature (11·0–31·0 μg/d)[6,12,13,15]. The Mn exposure is 1·38 mg/d (range 0·45–2·19 mg/d) and it satisfies the Italian RDA[9] (1–10 mg/d) at the lower value of recommendation. Our value is slightly lower than those reported in the literature (2·15–2·72 mg/d)[11,16,40,42]. The Mo exposure is 79·6 μg/d (range 32·6–106·2 μg/d) and it clearly satisfies the Italian RDA[9] (50–100 μg/d). Nevertheless, it is below the values reported in the literature (112–131 μg/d)[6,12,13,15,16]. The Ni exposure is 361·1 μg/d (range 45·0–764·2 μg/d) and it is below the PTDI[1] (600 μg/d), even though the maximum exposure is higher than 600 μg/d, a value that gives rise to allergic reactions in predisposed subjects. Our value is in the range of those reported in the literature (74·0–438·0 μg/d)[6,7,12–15,33,42–44]. The Pb exposure is 55·2 μg/d (range 25·8–66·6 μg/d) that is below the PTDI of 232 μg/d for an adult man of 65 kg body weight (25 μg/kg body weight per week)[1] and it is in the range of those reported from various European and extra European countries (16·0–280·0 μg/d)[6,7,14,22–25,37,43,45–47]. The Rb exposure is 1·37 mg/d (range 0·66–2·27 mg/d). There is no reference value with which to compare it. Our value is lower than that reported by Iyengar *et al.*[12] in the USA, equal to 2·44 mg/d. The Sn exposure is 1·57 mg/d (range 0·01–4·44 mg/d) that is well below the Provisional Tolerable Weekly Intake of 14 mg/week (2 mg/kg body weight per d for an adult man of 65 kg body weight)[1]. Our value is in the range of those reported in the literature (0·03–3·2 mg/d)[6,7,12,13,16,42].

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Table 3. Mean, minimum and maximum daily exposure to twenty-one trace elements in the Northern Italian adult population*

Trace elements	Mean value	Minimum value	Maximum value	PTDI†	Italian RDA‡	% of PTDI or Italian RDA	TDS data from the literature (range)
Al (mg)	3·58	0·28	5·15	65		6	1·9–12·0[6,7,11–17]
B (mg)	1·65	0·54	3·19	13		13	0·89– 2·12[6,11,12, 18–21]
Ba (μg)	366·6	53·1	398·3	–		–	530·0[6]§
Be (ng)	12·0	4·0	14·0	–		–	–
Cd (μg)	13·6	1·4	32·7	65		21	10·0–50·0[7,16,22–34]
Ce (μg)	154·8	124·5	197·2	–		–	–
Co (μg)	29·0	15·0	57·0	–		–	4·0–29·0[11–13,15,35]
Cs (mg)	4·91	0·54	7·93	–		–	1·46–9·0[12,35–37]
Cu (mg)	1·14	0·73	1·96		1·2	95	0·54–1·77[38,39]
Fe (mg)	11·0	6·41	19·45		10/18	110/61	9·0–15·9[6,12,13,35,39–41]
Ge (μg)	10·7	1·1	17·5	–		–	7·0[6]§
Li (μg)	29·9	8·2	42·4	–		–	1·0–31·0[6,12,13,15]
Mn (mg)	1·38	0·45	2·19		1–10	138/14	2·15–2·72[11–16,40,42]
Mo (μg)	79·6	32·6	106·2		50–100	159/79	112–131[6,12,13,15,16]
Ni (μg)	361·1	45·0	764·2	600		60	74·0–438·0[6,7,12–15,33,42–44]
Pb (μg)	55·2	25·8	66·6	232		24	16·0–280·0[6,7,14,22–25,37,43,45–47]
Rb (mg)	1·37	0·66	2·27	–		–	2·44[12]§
Sn (mg)	1·57	0·01	4·44	140		1	0·03–3·2[6,7,12,13,16,42]
Sr (mg)	1·14	0·67	1·88	–		–	1·40–1·65[6,12,35,37]
V (μg)	12·2	3·3	17·9	–		–	16·0–28·9[12,42]
Zn (mg)	12·0	7·5	19·8		10/7	120/172	4·8–16·0[6,7,11–16,35,39–41,48]

PTDI, Provisional Tolerable Daily Intake; TDS, Total Diet Study.
* For details of procedures, see the Materials and methods.
† From WHO[1].
‡ From Società Italiana di Nutrizione Umana[9]. Where two values are given they are for males/females.
§ Only one study.

The Sr exposure is 1·14 mg/d (range 0·67–1·88 mg/d), falling within the range of the values reported in the literature (1·40–1·65 mg/d)[6,12,35,37]. There is no reference value with which to compare it. The V exposure is 12·2 μg/d (range 3·3–17·9 μg/d) and it is lower than the values in the literature (16·0–28·9 μg/d)[12,42]. There is no reference value with which to compare it. The Zn exposure is 12·0 mg/d (range 7·5–19·8 mg/d) and it clearly satisfies the Italian RDA[9] both for males (10 mg/d) and females (7 mg/d). Our value is within the range of the estimated values reported in the literature (4·8–16·0 mg/d)[6,7,11–16,35,39–41,48].

Discussion

A TDS is a tool aimed at achieving the most accurate exposure estimates of a population's diet, either as exposure levels of nutrients or toxic compounds, and which can then offer important information about the nutritional quality of the diet as well as about potential nutritional problems. Therefore, it is used in many countries to evaluate food safety risks and offers several advantages to managers involved in food safety for guiding and orienting suitable programmes. First, it has the advantage of providing more realistic exposure data since foods are analysed 'as consumed' by the eater. Secondly, it provides a good tool for identifying the population or age groups most exposed (children, elderly, etc.) and can identify the main foods or food groups that contribute most greatly to the exposure. Finally, it provides data usable for international comparison among TDS.

Nevertheless, many factors may be considered when estimating the exposure of any dietary component, including the concentration of the component in foods, the amount of food consumed, the frequency of consumption and the proportion of population that consumes the food.

Estimated exposure of compounds from the TDS are based on average per-capita food consumption; that is, the diet reflects the average amounts consumed by the entire population (consumer and non-consumer alike). While the TDS provides a rich source of information on the major dietary sources and average dietary exposure of nutrients as well as of toxic compounds, it is not designed for estimating exposure at the extremes (upper or lower percentiles) or those population subgroups that may have specific nutritional needs (e.g. pregnant and lactating women).

The present study focuses on twenty-one trace elements in a TDS conducted in a Northern area of Italy and provides information on their daily average exposure by the adult population, increasing the contribution to the scientific literature in this field.

The 'market basket' method was slightly modified. We collected most samples of foods, just ready to eat, in a cafeteria, instead of purchasing them in local markets and then preparing and cooking the foods following the Italian traditional recipes. We chose to follow this method since the funding obtained for this research was sufficient only for carrying out the analytical analyses. Indeed, we are confident that the foods prepared in the cafeteria are not different from those purchased in the markets.

In Italy, a few studies were carried out recently on trace element exposure[33,39], but they did not investigate so many trace elements. Other studies have been conducted previously[34,44,47,49–51], especially on heavy metal exposure.

Mean exposure values satisfy the Italian RDA[9] for all the essential trace elements, except for Fe exposure in females, and they are well below the PTDI[1] for all the toxic compounds. As far as Fe exposure is concerned, a potential risk of anaemia in the female adult population should be

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Table 4. Percentage of exposure to twenty-one trace elements according to food categories*

Food categories	Al	B	Ba	Be	Cd	Ce	Co	Cs	Cu	Fe	Ge	Li	Mn	Mo	Ni	Pb	Rb	Sn	Sr	V	Zn
Cereals and tubers	32	12	17	24	55	2	41	42	25	42	17	31	62	45	34	51	24	97	25	37	26
Meat, fish and seafood, eggs	13	10	8	24	15	16	22	27	31	28	12	20	10	16	20	17	31	2	17	18	28
Milk and dairy products	33	16	47	10	0	81	5	4	2	4	1	6	3	8	21	4	6	0	14	5	21
Fruit and vegetables	18	47	18	32	30	1	27	25	39	22	69	33	20	25	15	17	28	1	37	23	20
Sweet foods	2	0	1	3	0	0	2	2	3	3	0	3	2	3	0	4	4	0	3	2	3
Beverages	2	15	9	7	0	0	3	0	0	1	1	7	3	3	10	7	7	0	4	15	2

* For details of procedures, see the Materials and methods.

considered, since its total daily exposure is derived mostly from plant foods, so most Fe exposure is non-haem Fe. In some previous studies conducted on trace elements and iron exposure in European and extra European countries[52–59], Fe exposure was the major health concern, since it appeared to be inadequate in several countries in groups of the population. It would be considered that in this TDS most Fe exposure is derived from plant foods, and then it is non-haem Fe. Therefore, studies aimed at evaluating the Fe nutritional status of adult Italian women should be addressed.

The high exposure of Ni, but below the PTDI[1], is in agreement with the data found previously in a study conducted in the same area as ours by Roggi *et al.*[44] and may be explained by its release from steel cookeries.

Concerning those trace elements for which no RDA or PTDI were provided, we cannot give any evaluation on their exposure, but only report their values of exposure. In addition, we cannot compare Be and Ce exposure with other data, since there are no TDS data about these trace elements in the literature.

Furthermore, the present findings may identify the specific food groups that contribute to a major or minor extent to the dietary exposure of these elements. As shown by Turrini *et al.*[10] in their Italian Household National Survey, the Italian diet is a plant-centred diet, with cereal, fruit and vegetable food groups having the highest daily consumption. Indeed, cereal, fruit and vegetable food groups contribute highly to most of the trace element exposure. Also protein-rich foods are good contributors, while milk and dairy products generally give a small contribution, except in the case of Al, Ba, Ce, Ni and Zn; finally, sweet foods and beverages are negligible contributors.

The present study has some limitations that must be considered. The food samples were only collected in a selected area in Northern Italy and so the results cannot be transferred to the entire Italian population. Furthermore, the samples were collected only once, during summer, and so the data are representative of the element exposure only for a selected period of the year. As far as the sample collection goes, we have in mind to repeat the food collection and analyses during another season of the year, such as in winter, in order to study seasonal variation. Finally, the present study was confined only to the adult fraction of the population. Thus, while not excluding the possibility that the daily exposure determined in the present study may not be representative of the population as a whole, the present study provides a good estimate of the Italian adult consumer exposure to twenty-one trace elements. It would be of interest also to evaluate the exposure for maximum food consumption data, especially for consumers with specific diet behaviour, such as vegetarians and vegans, who may be at a higher risk of exceeding the PTDI/Provisional Tolerable Weekly Intake of heavy metals.

Conclusions

The present study reveals that, for an adult population living in a northern area of Italy, the observed levels of the essential and probably essential trace elements estimated with the ‘market basket’ TDS are on the whole satisfactory compared with the Italian RDA, where recommendations are provided. The only critical trace element exposure is that of

Fe, regarding which suggestions were addressed. Concerning potentially toxic elements, the study shows that, for the population investigated, the probability of dietary exposure to health risks from such elements is overall small.

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