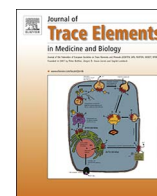




Contents lists available at ScienceDirect

## Journal of Trace Elements in Medicine and Biology

journal homepage: [www.elsevier.com/locate/jtemb](http://www.elsevier.com/locate/jtemb)

## Nutrition

## Dietary intake of cadmium, chromium, copper, manganese, selenium and zinc in a Northern Italy community

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## ARTICLE INFO

## Keywords:

Trace elements  
Selenium  
Cadmium  
Chromium  
Manganese  
Copper  
Zinc  
Dietary intake  
Nutritional epidemiology  
Food analysis

## ABSTRACT

This study provides the dietary intakes of six trace elements (cadmium, chromium, copper, manganese, selenium and zinc), generally characterized by both nutritional and toxicological features depending on their exposure. Being diet the most relevant source of exposure to trace elements in non-professionally exposed subjects, we measured content of these trace elements in foods composing the typical Italian diet using inductively coupled plasma-mass spectrometry, and assessing dietary habits using a validated semi-quantitative food frequency questionnaire we eventually estimated dietary daily intake of trace elements in a Northern Italian community.

In the 890 analyzed food samples, the main contributors to cadmium intake are cereals, vegetables and sweets, while cereals, beverages and vegetable are to primary source of manganese. The primary contributors for copper are cereals, fresh fruits and vegetables, while for chromium are beverages, cereals and meat. The main source of selenium intake are cereals and meat, followed by fish, seafood and milk and dairy products, while of zinc intake are meat, cereals, milk and dairy products. In our Italian population sample, the estimated median (interquartile range) dietary daily intakes are 5.00 (3.17–7.65), 56.70 (36.08–86.70) and 66.53 (40.04–101.32) µg/day for cadmium, chromium and selenium, and corresponding figures are 0.98 (0.61–1.49), 2.34 (1.46–3.52) and 8.50 (5.21–12.48) mg/day for copper, manganese and zinc.

The estimated intakes are generally within the average intake reported in other European populations, and in such cases well above the daily dietary intakes recommended by national international agencies, avoiding the risk of excess or deficiency. The present estimated intake data can be used to examine a specific trace element of interest and would afford enhanced health protection from those trace elements characterized by both nutritional and toxicological effects.

## 1. Introduction

The relevance of trace elements in human health and disease is well documented [1]. Depending of their role within the metabolism, trace elements are generally classified in essential and non-essential. In particular, six trace elements, i.e. cadmium, chromium, copper, manganese, selenium and zinc, were selected due to their intriguing relation with human health showing either nutritional and toxicological effects [2–9]. Two of these elements are also included as prioritized substances within the Human Biomonitoring Initiative at the European level [10].

Diet is generally recognized as being the main source of exposure to

trace elements. Some exceptions are the high exposure in occupationally exposed workers, e.g. to cadmium [11,12] and chromium [13,14], and smoking. Medical devices including prostheses may also represent a source of trace element exposure [15], and it is certainly true also for outdoor and indoor pollution, particularly for elements such as cadmium, manganese and selenium [16–21]. Being diet most relevant source of exposure to the above mentioned trace elements in non-occupationally exposed subjects, the systematic, periodic and updated evaluation of their content in foods composing population diet is a key element for a comprehensive assessment of their intake levels and of possible consequent health risks [22].

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<https://doi.org/10.1016/j.jtemb.2018.03.001>

Received 15 January 2018; Received in revised form 26 February 2018; Accepted 2 March 2018

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In the present study, we assessed the dietary intake of these trace elements in a Northern Italian community, taking into account their concentrations in foods and the dietary habits of the community.

## 2. Methods

### 2.1. Food collection and analysis

We determined the trace element content in the food items characterizing the usual diet of a Northern Italian population. Relevant food items characterizing the diet of this community were selected from previous population-based studies addressing the dietary habits of subjects from Northern Italy, with particular reference of Emilia-Romagna Region [23]. We purchased samples of the food items in local markets, large supermarkets, and grocery stores as well as in community canteens from Reggio Emilia and Modena provinces. Food collection started in October 2016 and ended in February 2017. In order to avoid contamination of metals from food containers, plastic tubes or jars were used and plastic cutlery implemented during the collection.

Food samples were homogenised in a food blender (equipped with a stain less-steel blade) to ensure the homogeneity and for each food sample, specimen in six different points from the plate were collected for subsequent analyses.

We placed portions of the samples (0.5–1.0 g) in quartz containers previously washed with MilliQ water and HNO<sub>3</sub>. Food samples were liquid-ashed (5 ml HNO<sub>3</sub> + 5 ml H<sub>2</sub>O<sub>2</sub>) in a microwave digestion system, then we stored them in a plastic tube and diluted to 50 ml with deionised water before analyses. We performed trace element determination using inductively coupled plasma-mass spectrometer 7500 Agilent. All reagents were of analytical grade and deionised water was used throughout. We performed all the analyses in duplicate and quality controls including both blanks (solution of MilliQ water) and a control solution of tap-water additionally enriched with 22 ppb of each element under investigation. Limit of quantification was 0.02 µg/kg for cadmium, chromium and selenium, while 0.5 µg/kg for copper, manganese and zinc. Corresponding limit of detection (LOD) was quantified in 0.007 µg/kg and 0.17 µg/kg.

Therefore, we reported the concentrations of investigated trace elements according to the food consumption pattern typical of this Italian population, as previously described [24,25]. The final list of foods included cereals (namely pasta and other grain, rice, bread, crispbread rusks and salty biscuits), meat (namely red, white, processed meat and offal), milk and dairy products (including yogurt and cheese), eggs, fish and seafood (namely fish of big and little size, bivalves, crustaceans and cephalopods), vegetables (namely leafy vegetables, tomatoes, cabbages, onions, mushrooms, root and other vegetables), legumes, potatoes, fresh fruits, dry fruits, sweets (namely chocolate sweets, cakes, pastries and puddings, dry cakes and biscuits, and other sweets products) and beverages (namely non-alcoholic beverages, e.g. fruits and vegetables juices, carbonated and soft drinks, coffee and tea, and alcoholic beverages, e.g. wine, beer and spirits).

### 2.2. Study population and estimation of daily element intake

We carry out the evaluation of dietary habits of Northern Italy community in a representative sample of the Emilia-Romagna Region population [26,27]. Briefly, the large sample population was randomly selected from the database of the Emilia-Romagna region National Health Service directory, namely from the provinces of Bologna, Ferrara, Modena, Parma, and Reggio Emilia. The final population was made up by 719 subjects, 319 males and 400 females with mean age of 55.3 years, range 18–87 years. Body mass index was 25.1 (interquartile range – IQR: 22.7–27.8) in all subjects, with corresponding Figs. 26.1 (IQR: 24.2–28.7) and 24.1 (IQR: 22.6–7.0), respectively in males and in females. Median total energy intake estimated through the FFQ was 1906 kcal/day (IQR: 1538–2364), 2024 kcal/day (IQR: 1649–2462)

and 1800 kcal/day (IQR: 1455–2296) in male and female participants, respectively.

In this population, we assessed the total daily intake and for food categories through the assessment of food intake by using a validated semi-quantitative food frequency questionnaire (FFQ) specifically developed for the Central-Northern Italian population [28,29]. Briefly, the EPIC (European Prospective Investigation into Cancer and Nutrition) questionnaire assessed the frequency and amount of consumption of 188 food items over the previous year, and allowed the frequency and quantity of consumption of foodstuffs and the related intake of nutrients and contaminants to be calculated using an *ad hoc* software [27,30].

By combining the analytical results of trace elements determination in foods and dietary assessment performed through with the EPIC FFQ, we assessed daily trace element intake using the equation presented below:

$$\begin{aligned} \text{Total daily dietary exposure} & \left( \frac{\mu\text{g}}{\text{day}} \right) \\ & = \sum \frac{\text{food intake} \left( \frac{\text{g}}{\text{day}} \right) \times \text{element food content} \left( \frac{\mu\text{g}}{\text{kg}} \right)}{1000} \end{aligned}$$

We estimated daily dietary exposure for each category and for total dietary exposure, reporting median and interquartile range of intake.

## 3. Results

We determined trace element content of 890 food samples. Table 1 shows the distribution of investigated trace elements in food categories. All concentrations were found to be above the LOD for chromium, copper, and zinc, while four samples were below the LOD for manganese and selenium, and 25 for cadmium. For cadmium, the higher median content was detected in cereals followed by potatoes, while the highest concentrations were found in dry fruits, totally driven by pine nuts levels, in sweets (due to high content in dark chocolate) and eventually in seafood, due to high levels in samples of shellfish and cephalopods, as shown in Supplementary Fig. S1. The high cadmium concentration in meat were almost totally driven by levels in offal samples (Supplementary Fig. S2).

Chromium showed high median content in three food categories: oils and fats, due to high concentrations in seed oils (i.e. mixed, sunflower and peanut), sweets, which showed the maximum levels in dark chocolate, and meat. The highest chromium levels were found in dry fruits, especially in nuts, and in beverages, due to high concentration in such spirits and liqueurs.

The highest median content of selenium was found in fish and seafood, with elevated levels in fish of big size, e.g. fresh and canned tuna, showing values generally above 1 mg/kg, mackerel and perch fish, and crustaceans (Supplementary Fig. S1). High selenium levels were also found in eggs, due to higher concentrations in the yolk, and in meat samples, with higher content in offal, followed by processed and red meat (Supplementary Fig. S2). Vegetables generally showed a low content of this trace element, which in turn was higher in Se-accumulator vegetables (i.e. cabbage, onion and garlic) that revealed four- to ten-fold higher levels, with median content of 18.87 µg/kg (IQR: 5.62–65.90), and especially in mushrooms with a median content of 106.37 µg/kg (IQR: 129.00–151.64) (Supplementary Fig. S3).

Copper showed the both median and maximum levels in dry fruits, due to pine nuts, hazelnuts and nuts content, followed by legumes, where copper-rich samples were soy beans, black-eyed beans/peas and lentils. Sweets were generally at low content of copper, except for high concentration in dark chocolate (Supplementary Fig. S4).

Similarly, manganese levels were high in almost all dry fruits, especially pine nuts, followed by nuts, hazelnuts, nuts and in addition almonds, in legumes, with the highest levels in chickpeas, followed by soybeans, beans in general (black-eyed, black peas, ‘borlotto’ and ingot/

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