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RESEARCH ARTICLE

Dietary Metals (Pb, Cu, Cd, Zn) Exposure and Associated Health Risks in Baia Mare Area, Northwestern Romania

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ABSTRACT

This study estimated the non-carcinogenic health risk of Pb, Cu, Cd, Zn via dietary intake in the urban and rural areas of Baia Mare city, northwestern Romania, a former mining area. A total of 230 food items grouped in ten food categories (meat and animal organs, meat-derived products, animal fat, eggs, dairy, bread, corn flour, potato, vegetables, fruits) and 32 water samples were collected and analyzed for Pb, Cu, Cd and Zn using atomic absorption spectrometric method. The human health risk assessment through dietary exposure was evaluated by calculating the estimated daily intake of metals, the Target Hazard Quotients (THQ) and the Total Target Hazard Quotients (TTHQ) for normal daily consumption in adults. The average daily intakes of Pb, Cd and Zn were higher than the Tolerable Daily Intake (TDI) reference value for urban and rural residents, which showed that there was a potentially high risk of ingestion of food and water. The average daily intake of Cu with food and drinking water was lower than the corresponding TDI value, which indicated that Cu intake was not a potential hazard to the health of residents in the study areas. The individual target hazard quotients, calculated for food consumption in urban area decreased in the following order: Cd>Cu>Pb>Zn, and in rural area: Pb>Cd>Cu>Zn, having values higher than 1 for Pb and Cd in the rural area and approximately equal to 1 for Cd (urban) and Cu (rural), indicating non-carcinogenic adverse health effects. The TTHQs for the two areas (urban and rural) were 2.934 and 5.164, respectively, established cumulative adverse effects of metals. The results showed that vegetables, potato and bread were the main sources of metal intake from foodstuff for adults, but fruit, water and meat were secondary contributors.

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INTRODUCTION

Metals are ubiquitous in the environment [1], thus human exposure to metals through food cannot be avoided, as it leads to acute (short-term) or chronic (long-term) health effects [2]. Metals bioaccumulate in the food chain and could pose a health risk of consumers because of their toxicity, persistence in the environment and their lack of biodegradability [3]. Lead (Pb) and Cadmium (Cd) are nonessential elements and are involved in carcinogenesis, mutagenesis and teratogenesis processes and they may act as endocrine disruptors. Copper (Cu) and Zinc (Zn) are essential for maintaining the physiological functions and the biochemical processes of individuals, but their excess may exert negative effects in individuals [4].

Food consumption is the main exposure pathway to environmental contaminants [5]. The intake of metals through ingestion depends on food habits. Thus information on the intake of metals through food chain is important in assessing the risk to human health [6]. According to the FAO/WHO, different methods (total diet study, journal study) can be used to determine the amount ingested from a food

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contaminant, combining data on specific contaminants in food with individual food consumption data. Assessment of long-term exposure to metals by food consumption of the population in a studied area can be performed starting from monitoring food intake (by food categories) and concentration of metals determined by laboratory analysis, in the food collected from the respective areas. The food consumption by the inhabitants can be investigated using specific questionnaires.

For studies to estimate long-term exposure through food intake, the average consumption over a given period of time and its refer ence to reference values for the amounts of metal (mg/kg body weight) that may be consumed daily shall be taken into account, during a lifetime, without the manifestation of adverse effects on health [7].

In this study, long-term exposure to metals (Pb, Cu, Cd, Zn) was estimated by determining the average intake of metals by ingestion and comparing it with the reference values of Tolerable Daily Intake (TDI). Also, the non-carcinogenic health risk of the adult inhabitants in the investigated areas was assessed by calculating the Target Hazard Quotients (THQs) for individual metal and the Total Target Hazard Quotients (TTHQs) for combined metals due to dietary intake. THQ is the ratio of exposure to a reference dose. A THQ value below 1 indicates that the exposure level is lower than the reference dose, which means that daily exposure does not cause adverse health effects of the population during lifetime.

The investigated area is situated in a former industrial district, based on nonferrous extraction and metallurgic activities, resulting in heavy anthropogenic contamination of the environment with metals [8-10]. Also, the health risk of the inhabitants in investigated area by vegetable and milk consumtion was previously investigated [11,12].

The objective of this study was to assess the health risk by food and water ingestion, of resident adult population in two areas affected by metal pollution: 1. urban area of Baia Mare city and 2. rural area, composed of Sasar and Bozanta Mare villages, situated in the adjacent area of Baia Mare city and located near three tailings ponds and tailings dumps. In order to achieve this goal, the following steps were taken: 1. establishing the food ratio for each studied area, by completing questionnaires on the structure of the diet for three consecutive days, in order to assess the exposure to heavy metals; 2. analysis of food samples in order to determine the concentration of metals (Pb, Cd, Cu, Zn); 3. risk characterization and evaluation of the exposure of population groups in the Baia Mare area (urban and rural) to metals by ingesting food and drinking water by comparing with the values of TDI; by calculating the THQ and TTHQ quotients.

MATERIALS AND METHODS

Sampling

During 2019, a number of 230 foodstuffs were collected, representing the most consumed food in the investigated area. Also, 32 water samples were collected. In the urban area of Baia Mare city, 127 foodstuffs were sampled from grocery stores and food markets and 12 water samples from the water supply network (tap water). In the rural area, 103 foodstuffs were collected, mainly from the private households and gardens and also 20 water samples collected from their own wells or from the water network, depending on the case (part of the Sasar and Bozanta Mare villages are connected to the drinking water supply network of Baia Mare city). The number of samples collected from the investigated areas, urban and rural, by food category, is presented in table 1.

The samples were transported to the laboratory in polyethylene packages, with identification data, tightly closed to avoid their contamination.

Sample preparation and analysis

In order to get homogenized food samples, a domestic shredder was used, and only the the edible parts of food were considered. All chemicals were of analytical reagent grade. Ultra-pure deionized water type Milli-Q (Millipore, France) was used for preparation of standards and sample solutions. Concentrated 65% nitric acid, 30% hydrogen peroxide (H_2O_2) and metals stock standard solutions (1000 mg/l) were supplied by Merck, Germany.

Analytical determination

The analysis of Pb, Cu, Cd and Zn concentrations in food samples was performed after a microwave digestion treatment (Ethos Easy, Milestone, Italy) with nitric acid. One-half gm of each food sample was digested with 9 ml of nitric acid and 0.5 ml of 30% H_2O_2. Samples containing high levels of fats were pre-digested with concentrated nitric acid. The water samples were acidified with 65% nitric acid by adding 0.5 ml of acid to 100 ml of water, so that the pH of the sample was lower than 2, according to the standardized method [13].

Quantitative analysis of metals

The metal contents were measured using a graphite furnace atomic absorption spectrometer (PinAAcle 900T, Perkin-Elmer, USA). The Limits Of Quantitation (LOQs) of measured metals were calculated as five time of the standard deviation of a series of measurements of a solution against the blank absorbance.

Dietary survey and calculation of the dietary exposure

A dietary survey was conducted in order to calculate the

Table 1: Number of food samples by food category and water samples collected in rural and urban areas.

No.	Food category	No. of samples collected	
		urban area	rural area
1	Meat and organs	**15 samples:** 7 meat (3 pork, 2 beef, 2 poultry) and 8 organs (3 pork liver, 1 beef liver, 3 pork kidney, 1 beef kidney)	**10 samples:** meat (5 pork, 1 beef, 4 poultry) samples
2	Meat products	**15 samples:** 5 sausage, 5 salami, 5 baloney	**5 samples:** 4 sausage, 1 pork ham
3	Animal fat	**4 samples:** 2 pork lard, 2 bacon	**6 samples:** 2 pork lard, 4 bacon
4	Chicken eggs	**4 samples**	**6 samples**
5	Milk and cheese	**20 samples:** 4 milk, 8 matured cheese, 8 melted cheese samples	**8 samples:** 4 milk, 4 home made cow cheese
6	Bread	**10 samples:** 7 white, 2 black, 1 semi - white samples	-
7	Corn flour	**5 samples**	**3 samples**
8	Potatoes	**8 samples**	**8 samples**
9	Vegetables	**40 samples:** 15 leafy vegetables: 5 lettuce, 5 spinach, 5 cabbage samples and 25 fresh vegetables: 5 carrot, 4 pepper, 2 cucumber, 2 eggplant, 5 tomato, 5 green onion, 2 parsley samples	**47 samples:** 14 leafy vegetables (5 lettuce, 9 cabbage samples) and 33 fresh vegetables (8 carrot, 2 celery, 2 garlic, 6 tomato, 5 green onion, 10 parsley) samples
10	Fruits: apple	**6 samples**	**10 samples**
11	Water	**12 samples** from city network supply	**20 samples:** 12 wells and 8 from city network supply

daily food ingestion rate or the daily intake of each food category selected in this study, based on a 24-h recall in two non-consecutive days and a food frequency questionnaire, providing qualitative and quantitative information. The group selected for Urban area consisted of 40 subjects resident in the city of Baia Mare and was formed of 19 (47.5%) men and 21 (52.5% women), with a mean age of 59.25 ± 7.47 years. In rural area, the group consisted of 52 subjects, 18 (35%) men and 34 (65%) women, with a mean age of 65 ± 10.7 years. All consumed food categories were registered and converted to gram weights using a kitchen scale (KS1500, Zelmer, Poland). Each interviewed person was weighted with a personal scale (BS1200, Zelmer, Poland) alongside other individual identification parameters. The written informed consent was obtained from each individual. The average weight of the interviewed population was 70.6 kg and 69.0 kg for urban and rural areas, respectively, thus we assumed an average weight of 70 kg for both areas under investigation. The dietary intake of each food category was calculated by multiplying mean concentration of each chemical in food category and the consumption of that food category. The total dietary intake was obtained by summing the respective intakes of all food groups. Finally, the intake was also estimated according to the respective average body weight. In the calculation of the daily metal intake, the intake of drinking water was also considered. For calculations, when the concentration of a metal was under the respective Limit Of Quantification (LOQ), that value was assumed to be one-half of that limit (LOQ/2).

Health risk assessment

The health risk assessment is used to determine whether exposure to a chemical, at any dose, may increase the incidence of an adverse effect on human health [14]. The purpose of exposure assessment is to identify potential receptors, assess routes of exposure and quantify exposure. The estimation of the dose penetrated into the human body by contact with the contaminant is determined by calculating the average daily intake, which is the quantification of the contaminant ingested, inhaled or absorbed through the skin, per kilogram body weight, per day (mg/kg/day) [15]. In this study, only one route of exposure was selected, due to the specific use of the soil in the investigated area, as well as the nature of the determined metals: the ingestion of food by the resident population in the studied areas. Inhalation and dermal exposure have been neglected, according to other studies [16-18].

For the calculation of the average total daily food intake of each metal, the values of the average daily intake of each metal, in each food category (meat and organs, meat products, eggs, milk, cheese, animal fat, potato, vegetables, apple, bread, corn flour) and water were summed. In the calculation of daily metal intake, the water intake was added, and the average amount ingested being considered 2 l/day [19].

The daily intake of Cd, Pb, Cu and Zn for each food category was calculated by multiplying the average concentration of each metal analyzed (C) in each food category, with the average amount of food consumed by a person/day in that area (F_{IR}), according to equation (1) [17].

$$DI = F_{IR} C \quad (1)$$

Where;

DI - daily intake of metal by food and water consumption, (µg/person/day);

F_{IR} - daily food ingestion rate (from each food category), (g/person/day);

C - concentration of the metal in the food category, (mg/kg).

In this study, the health risk from simultaneous exposure to Pb, Cd, Cu, Zn was calculated by determining the Total Target Hazard Quotients (TTHQ), calculated by summing the individual target hazard quotients (of each metal), based on equation (2) [17,20].

Total THQ (TTHQ) = THQ Pb + THQ Cd + THQ Cu + THQ Zn (2)

RESULTS AND DISCUSSION

Concentrations of metals Pb, Cu, Cd, Zn in food and water samples

The LOQs for Pb, Cd, Cu and Zn determined by atomic absorption were 0.010 mg/kg, 0.005 mg/kg, 0.015 mg/kg, 0.010 mg/kg, respectively for food samples and 0.0025 mg/l, 0.005 mg/l, 0.020 mg/l, Zn 0.50 mg/l, respectively for water samples. The concentrations of metals Pb, Cu, Cd, Zn in each investigated food category collected in urban and rural areas are shown in tables 1-3, respectively.

The concentrations of metals varied largely, even within the same food category. In urban area, the determination of Pb, Cu, Cd, Zn in foods revealed the following:

- In case of fresh vegetables, 9 samples recorded higher concentrations than MAC for Pb, 4 samples for Cd, 9 samples for Cu and 3 samples for Zn. The average Pb concentration was above the MAC value (0.1 mg/kg), the averages of the concentrations of the other metals were below their corresponding MACs.

Table 2: Concentrations of metals Pb, Cu, Cd, Zn in each investigated food category, collected in urban area.

Food category	Pb, mg/kg mean ± SD[a)] range MAC[b)] / ([c)])	Cd, mg/kg mean ± SD range MAC[b)] / ([c)])	Cu, mg/kg mean ± SD range MAC[b)] / ([c)])	Zn, mg/kg mean ± SD range MAC[b)] / ([c)])
1. Meat and animal organs (*n* = 15)	0.138 ± 0.123 0.021 - 0.356	0.257 ± 0.476 <LOQ* - 1.837	1.703 ± 1.551 0.210 - 5.86	6.975 ± 3.803 2.911 - 18.66
From which:				
Meat (*n* = 7)	0.070 ± 0.098 0.021 - 0.289 0.1 / (6)	0.012 ± 0.017 <LOQ - 0.046 0.05 / (7)	0.578 ± 0.358 0.210 - 1.113 3.0 / (7)	5.135 ± 1.256 2.911 - 7.158 50 / (7)
Organs (*n* = 8)	0.197 ± 0.116 0.033 - 0.356	0.471 ± 0.584 0.010 - 1.837	2.687 ± 1.526 0.779 - 5.860	8.585 ± 4.608 4.797 - 18.66
From which:				
Liver (*n* = 4)	0.189 ± 0.119 0.033 - 0.322 0.5 / (4)	0.224 ± 0.177 0.010 - 0.378 0.5 / (4)	2.751 ± 2.212 0.779 - 5.860 3.0 / (3)	7.288 ± 2.096 5.243 - 9.708 50 / (4)
Kidney (*n* = 4)	0.206 ± 0.131 0.067 - 0.356 0.5 / (4)	0.717 ± 0.777 0.041 - 1.837 1.0 / (2)	2.624 ± 0.7297 1.859 - 3.587 3.0 / (3)	9.882 ± 6.376 4.797 - 18.66 50 / (4)
2. Meat products (*n* = 15)	0.703 ± 0.422 0.155 - 1.989 1.0 / (14)	0.027 ± 0.058 <LOQ - 0.226 0.1 / (14)	1.388 ± 1.755 0.481 - 7.652 5.0 / (14)	5.353 ± 11.11 1.128 - 45.36 50 / (15)
From which:				
Home - made and smoked sausages (*n* = 5)	0.651 ± 0.110 0.505 - 0.794	0.009 ± 0.009 <LOQ - 0.023	0.976 ± 0.235 0.755 - 1.365	2.723 ± 1.392 1.665 - 5.149
Salami (*n* = 5)	0.525 ± 0.269 0.155 - 0.864	0.019 ± 0.028 <LOQ - 0.067	0.806 ± 0.248 0.481 - 1.099	2.106 ± 0.526 1.642 - 3.012
Baloney (*n* = 5)	0.933 ± 0.655 0.318 - 1.989	0.053 ± 0.097 0.003 - 0.226	2.380 ± 2.967 0.694 - 7.652	11.23 ± 19.11 1.128 - 45.36
3. Animal fat (pork lard and pork bacon) (*n* = 4)	0.063 ± 0.028 0.035 - 0.102 1.0 / (4)	0.005 ± 0.002 0.003 - 0.008 0.1 / (4)	1.612 ± 0.501 1.021 - 2.044 - / (-)	1.528 ± 1.152 0.121 - 2.541 - / (-)
4. Eggs (*n* = 4)	0.031 ± 0.031 <LOQ - 0.0753 0.5 / (4)	0.010 ± 0.005 0.005 - 0.017 0.05 / (4)	3.098 ± 2.244 0.750 - 5.054 - / (-)	17.82 ± 8.691 10.88 - 30.12 - / (-)
5. Milk and cheese (*n* = 20)	0.398 ± 1.501 <LOQ - 6.764	0.004 ± 0.005 <LOQ - 0.0246	0.687 ± 0.846 0.013 - 3.682	5.197 ± 3.195 0.481 - 9.088
From which:				
Milk (*n* = 4)	0.013 ± 0.008 <LOQ - 0.0230 0.02 / (3)	0.004 ± 0.002 <LOQ - 0.006 0.01 / (4)	0.112 ± 0.085 0.013 - 0.217 0.5 / (4)	1.069 ± 0.4564 0.546 - 1.567 5.0 / (4)
Cheese (*n* = 16)	0.495 ± 1.675 <LOQ - 6.764 0.5 / (15)	0.004 ± 0.006 <LOQ - 0.025 0.05 / (16)	0.830 ± 0.892 0.013 - 3.682 2.5 / (15)	6.229 ± 2.684 0.481 - 9.088 25 / (16)
From which:				

Matured cheese (*n* = 8)	0.074 ± 0.112 <LOQ - 0.326	0.006 ± 0.008 <LOQ - 0.025	0.553 ± 0.571 0.013 - 1.590	7.114 ± 2.218 3.642 - 9.088
Melted cheese (*n* = 8)	0.9153 ± 2.365 <LOQ - 6.764	0.002 ± 0.001 <LOQ - 0.005	1.108 ± 1.097 0.096 - 3.682	5.345 ± 2.955 0.481 - 7.976
6. Bread (*n* = 10)	0.053 ± 0.023 0.026 - 0.084 1.0 / (10)	0.040 ± 0.041 0.004 - 0.116 0.2 / (10)	1.681 ± 0.511 0.895 - 2.611 5.0 / (10)	5.400 ± 2.157 0.128 - 8.002 15 / (10)
7. Corn flour (*n* = 5)	0.064 ± 0.023 0.046 - 0.099 1.0 / (5)	0.005 ± 0.001 0.004 - 0.006 0.2 / (5)	2.364 ± 0.290 1.984 - 2.658 5.0 / (5)	6.553 ± 1.234 4.513 - 7.823 15 / (5)
8. Potato (*n* = 8)	0.544 ± 0.837 0.025 - 2.473 0.1 / (3)	0.091 ± 0.132 0.003 - 0.3456 0.1 / (6)	3.035 ± 1.416 1.556 - 5.214 3 / (5)	7.004 ± 2.562 4.711 - 12.68 10 / (7)
9. Vegetables (*n* = 40)	0.286 ± 0.653 0.017 - 3.846	0.097 ± 0.091 0.004 - 0.368	3.168 ± 1.595 0.684 - 7.319	8.076 ± 3.344 3.785 - 15.13
From which:				
Fresh vegetables (*n* = 25)	0.110 ± 0.069 0.033 - 0.258 0.1 / (16)	0.076 ± 0.072 0.010 - 0.368 0.1 / (21)	2.952 ± 1.444 0.684 - 6.255 3.0 / (16)	7.141 ± 2.865 3.785 - 14.93 10.0 / (22)
Leafy vegetables (*n* = 15)	0.581 ± 1.015 0.017 - 3.846 0.3 / (7)	0.134 ± 0.109 0.004 - 0.346 0.2 / (10)	3.528 ± 1.815 0.839 - 7.319 - / (-)	9.635 ± 3.594 5.084 - 15.13 - / (-)
10. Fruits (apples) (*n* = 6)	0.307 ± 0.457 0.010 - 1.208 0.1 / (3)	0.021 ± 0.025 0.005 - 0.065 0.05 / (5)	1.677 ± 1.236 0.543 - 3.791 5.0 / (6)	2.204 ± 0.856 1.007 - 3.107 15 / (6)
	Pb, µg/l mean ± SD range MAC[d] / ([c])	**Cd, µg/l** mean ± SD range MAC[d] / ([c])	**Cu, mg/l** mean ± SD range MAC[d] / ([c])	**Zn, mg/l** mean ± SD range MAC[d] / ([c])
11. Water (*n* = 12)	4.28 ± 1.35 2.60 - 7.50 10 / (12)	0.46 ± 0.16 0.30 - 0.80 5.0 / (12)	0.088 ± 0.16 0.02 - 0.60 0.1 / (11)	1.04 ± 0.21 0.67 - 1.41 5.0 / (12)

a) Standard deviation
b) Maximum Admissible Concentration according to legislation, namely Hygiene Norms no. 975 of 1998, art. 93, Order no. 293/640/2001 - 1/2002, Order no. 1201/106 of 22 December 2003
c) Value in parentheses represents the number of samples below the MAC set by legislation
d) Maximum Admissible Concentration according to Law 458/2002 on drinking water quality (Official Gazette, Part I no. 552 of 29.07.2002)
*<LOQ: For statistical purposes, concentrations below the LOQ were considered as LOQ/2; thus, for Pb: LOQ/2 = 0.0045 mg/kg and for Cd: LOQ/2 = 0.0015 mg/kg

Table 3: The concentrations of metals Pb, Cu, Cd, Zn in each investigated food category collected in rural area.

Food category	**Pb, mg/kg** mean ± SD[a)] range MAC[b)] / ([c)])	**Cd, mg/kg** mean ± SD range MAC[b)] / ([c)])	**Cu, mg/kg** mean ± SD range MAC[b)] / ([c)])	**Zn, mg/kg** mean ± SD range MAC[b)] / ([c)])
1. Meat (*n* = 10)	0.104 ± 0.105 0.016 - 0.352 0.1 / (8)	0.044 ± 0.048 0.006 - 0.167 0.05 / (8)	1.291 ± 0.658 0.617 - 2.728 3.0 / (10)	15.45 ± 14.60 2.669 - 46.07 50 / (10)
2. Meat products (*n* = 5)	0.614 ± 0.522 0.074 - 1.438 1.0 / (4)	0.081 ± 0.072 0.031 - 0.207 0.1 / (4)	2.090 ± 1.415 0.913 - 4.306 5.0 / (5)	5.313 ± 1.975 3.128 - 8.121 50 / (5)
From which:				
Home made sausages (*n* = 4)	0.614 ± 0.522 0.074 - 1.438	0.081 ± 0.072 0.031 - 0.207	2.090 ± 1.415 0.913 - 4.306	5.313 ± 1.975 3.128 - 8.121
Pork ham (*n* = 1)	1.438	0.207	4.306	8.121
3. Animal fat (pork lard and pork bacon) (*n* = 6)	0.123 ± 0.078 0.046 - 0.264 1.0 / (6)	0.008 ± 0.004 0.003 - 0.015 0.1 / (6)	1.961 ± 0.925 0.884 - 3.557 - / (-)	2.009 ± 1.083 0.547 - 3.741 - / (-)
4. Eggs (*n* = 6)	0.162 ± 0.112 0.039 - 0.311 0.5 / (6)	0.047 ± 0.032 0.012 - 0.100 0.05 / (4)	2.400 ± 0.968 1.011 - 3.501 - / (-)	14.85 ± 5.475 7.315 - 22.04 - / (-)
5. Milk and cheese (*n* = 8)	0.225 ± 0.280 0.009 - 0.8140	0.028 ± 0.027 0.003 - 0.075	0.907 ± 0.804 0.047 - 2.237	3.551 ± 1.599 1.431 - 6.116
From which:				
Milk (*n* = 4)	0.041 ± 0.034 0.009 - 0.086 0.02 / (2)	0.010 ± 0.005 0.003 - 0.016 0.01 / (2)	0.366 ± 0.313 0.047 - 0.797 0.5 / (3)	2.641 ± 1.494 1.431 - 4.674 5.0 / (4)

Cow cheese (*n* = 4)	0.408 ± 0.304 0.097 - 0.814 0.5 / (3)	0.046 ± 0.029 0.009 - 0.075 0.05 / (2)	1.447 ± 0.7956 0.342 - 2.237 2.5 / (4)	4.462 ± 1.234 3.261 - 6.116 25 / (4)
6. Corn flour (*n* = 3)	0.607 ± 0.927 0.055 - 1.677 1.0 / (2)	0.147 ± 0.107 0.033 - 0.245 0.2 / (2)	2.349 ± 1.375 0.957 - 3.706 5.0 / (3)	4.293 ± 1.107 3.117 - 5.314 15 / (3)
7. Potato (*n* = 8)	0.242 ± 0.247 0.009 - 0.717 0.1 / (4)	0.161 ± 0.147 0.017 - 0.493 0.1 / (3)	2.757 ± 1.516 1.287 - 5.639 3.0 / (5)	9.211 ± 4.188 5.883 - 16.110 10 / (6)
8. Vegetables (*n* = 47)	2.080 ± 2.093 0.010 - 7.855	0.237 ± 0.2667 0.004 - 1.135	5.814 ± 5.546 0.517 - 35.62	24.93 ± 17.72 0.980 - 78.11
From which:				
Fresh vegetables (*n* = 33)	2.248 ± 2.365 0.010 - 7.855 0.1 / (4)	0.215 ± 0.254 0.004 - 0.967 0.1 / (15)	6.579 ± 6.260 0.517 - 35.62 3.0 / (7)	21.66 ± 18.48 0.980 - 78.11 10 / (11)
Leafy vegetables (*n* = 14)	1.684 ± 1.218 0.050 - 4.647 0.3 / (1)	0.287 ± 0.298 0.007 - 1.135 0.2 / (7)	4.010 ± 2.695 0.841 - 10.50 - / (-)	32.63 ± 13.40 12.08 - 69.11 - / (-)
9. Fruits (apples) (*n* = 10)	0.144 ± 0.174 0.009 - 0.571 0.1 / (6)	0.033 ± 0.018 0.012 - 0.073 0.05 / (9)	1.967 ± 0.7978 0.9110 - 3.590 5.0 / (10)	4.075 ± 2.878 0.7190 - 11.22 15 / (10)
	Pb, µg/l mean ± SD range MAC[d] / ([c])	**Cd, µg/l** mean ± SD range MAC[d] / ([c])	**Cu, mg/l** mean ± SD range MAC[d] / ([c])	**Zn, mg/l** mean ± SD range MAC[d] / ([c])
10. Water (*n* = 20)	6.30 ± 2.74 2.40 - 13.50 10 / (18)	1.80 ± 1.08 0.40 - 3.80 5.0 / (20)	0.055 ± 0.0254 0.020 - 0.130 0.1 / (19)	1.489 ± 0.6534 0.45 - 2.77 5.0 / (20)

a) Standard deviation
b) Maximum Admissible Concentration according to legislation, namely Hygiene Norms no. 975 of 1998, art. 93, Order no. 293/640/2001 - 1/2002, Order no. 1201/106 of 22 December 2003
c) Value in parentheses represents the number of samples below the MAC set by legislation
d) Maximum Admissible Concentration allowed according to Law 458/2002 on drinking water

- Regarding the leafy vegetables (lettuce, spinach, cabbage), 8 samples recorded higher concentrations than MAC for Pb and 5 samples for Cd; the Cu and Zn concentrations not being normalized. In the case of Pb, the mean concentration exceeded MAC approximately 2-fold.

- In case of apple category, 3 samples were identified as not corresponding for Pb and one sample for Cd. The average value of Pb is higher than MAC, and the average value of Cd falls within the limits set by legislation.

- In case of potatoes, concentrations of Pb and Cd were higher than their corresponding MAC in 5 samples and in 2 samples, respectively, and the average concentrations of investigated metals were below MACs.

- Regarding the edible organ samples, one liver sample recorded higher concentration for Cu, two kidney samples recorded higher concentrations for Cd and one sample for Cu, respectively. The averages concentrations of these metals were within the allowed values.

- In case of meat-derived products samples, a sample recorded higher concentrations for Pb, Cd and Cu, respectively. The averages concentrations were within the MAC values.

- In case of milk and dairy samples, one sample of milk and one sample of melted cheese recorded higher concentrations for Pb and one sample of melted cheese for Cu, but the averages of the Pb concentrations were below the MAC value.

- In the case of bread, corn flour, meat, eggs and animal fats, the metal concentrations were below their correspunding MACs.

- In case of drinking water, all the analyzed samples fall within the potability parameters imposed by the legislation, except one sample for Cu concentration, for the investigated metals.

The analysis of metal concentrations in food collected from rural area, in comparison with MAC, showed the following:

- In case of potatoes, 50%, 63%, 38% and 25% of samples exceeded the corresponding MACs for Pb, Cd, Cu and Zn, respectively. The average concentrations of Pb and Cd were higher than MACs.

- Regarding the fresh vegetables (onion, carrots, parsley, celery, garlic, tomatoes), 88%, 55%, 79% and 67% of samples exceeded the corresponding MACs for Pb, Cd, Cu and Zn, respectively. Average concentrations exceed MACs 22.5 times for Pb and 2.2 times for Cd, Cu and Zn.

- In case of leafy vegetables (lettuce, cabbage), 93% and 50% of samples exceeded MACs for Pb and Cd, respectively. Average concentrations exceed the corresponding MAC by 5.6 times for Pb and 2.2 times for Cd.
- The following results were obtained for apples: 4 samples out of 10 exceed the MAC for Pb, 1 sample exceeded the MAC for Cd, and Cu and Zn fall within the allowed values. The average concentration for Pb was higher than MAC, in contrast to the average values for Cd, Cu and Zn which were below their corresponding MACs.
- Corn flour samples collected from private households were within the allowed values in proportion of 67% for both Pb and Cd. For Cu and Zn, the samples recorded values below MACs.
- In meat samples (pork, beef, chicken) and meat products (homemade sausages, pork ham), 80% of the samples fall within the allowed values for Pb and Cd, and 100% of the samples fall within the allowed values for Cu and Zn. The average values are below the MACs for all analyzed metals.
- In the case of cow's milk samples, 2 samples exceeded MAC for Pb, 2 samples for Cd and 1 sample for Cu. The average Pb concentrations were above the MAC value, the other average values were within the accepted parameters.
- For cheese, 75% and 50% of the samples exceeded their corresponding MACs for Pb and Cd, respectively. The averages concentrations were within the values imposed by the MACs for every metal.
- For egg samples, Cd concentration was higher than MAC in 2 samples (67%), but the average concentration was within the MAC values. For Pb, the concentrations fall within the values allowed by the legislation, and Cu and Zn are not standardized.
- In the animal fat samples (lard and bacon) each metal concentration was below the MACs.
- In the case of drinking water, 90% of the samples fall within the limits of MACs for Pb and 100% for Cd, Cu and Zn. The average concentrations of metals were within the allowed values for MACs.

Risk assessment

This stage includes the following steps: 1. Assessment of the daily intake of metals by ingestion, 2. Exposure calculation for each of the investigated areas by comparison with TDI values, 3. Calculation of THQ for each metal, 4. Calculation of TTHQ.

In order to evaluate the exposure of the investigated population to the metals in the studied areas, the analysis of the daily intake on food categories was made, based on the processing the food frequency questionnaires.

Assessment of daily intake of metals through ingestion, in rural and urban areas

The calculated values of the total daily food intake for Pb, Cd, Cu and Zn in the urban area are shown in table 4, and for the rural area in table 5.

The mean Cu daily intake by participants in this study in urban area (2.25 mg/day) was slightly lower than 2.7 mg/day reported by Shokunbi, et al. [21] in foods analysed in the Nigerian Food Composition Table and 2.7 mg/day reported by Bowen [22] for Germans and was slightly higher than 2.1 mg/day reported by Rubio, et al. [23] for Spanish population.

For the studied adults population in urban area, the contribution of the food categories to the total daily intake of Pb, Cd, Cu, Zn was graphically represented in figure 1.

Due to the high concentrations of metals and the frequency of consumption, vegetables represent the food category that provides the highest intake of Pb, Cd, Cu, Zn by ingestion, for the inhabitants of the investigated urban area.

Figure 2 graphically represented the contribution of the most important food categories to the daily food intake of metals Pb, Cd, Cu, Zn of the adult inhabitants of the rural area. Due to the high concentrations of metals and the frequency of consumption, vegetables are the food category that provides the highest intake of Pb, Cd, Cu, Zn by ingestion of these foods.

By comparing the data obtained with those published in the literature, the daily dietary intake of metals in the studied areas was higher, for example, than in Great Britain (except Zn in urban areas, Baia Mare), as follows: dietary intake of Pb is 9.3, respectively 25 times higher in urban and rural areas; the daily food intake of Cd is 5, respectively 7 times higher in the two areas, the daily intake of Cu is 1.9, respectively 2.2 times higher in the two investigated areas, and the daily intake of Zn in the Baia Mare area represents 95% from that of the adult inhabitants of Great Britain, and in the studied rural area the contribution of Cu is 1.6 times higher than that of the inhabitants of Great Britain [24,25]. Also, the values obtained in this study are higher than those reported by Santos, et al. [26] regarding the daily intake of heavy metals by ingesting food by the adult population of Rio de Janeiro. For example, the average concentrations of Pb, Cd, Cu, and Zn in potatoes from the diet of the inhabitants of Baia Mare is 2.3; 17; 2.5 and 2.9 times higher, respectively, than the concentrations of these metals in the potatoes consumed by the inhabitants of Rio de Janeiro. The daily food intake for Pb and Cd for the studied areas is higher than the values given by Winter-Sorkina, et al. [27],

Table 4: Daily intake of Pb, Cd, Cu and Zn, in urban area.

Food category	Mean intake rate, g/day	Mean Pb conc., mg/kg	Mean Cd conc., mg/kg	Mean Cu conc., mg/kg	Mean Zn conc., mg/kg	Mean daily intake Pb, µg/ day	Mean daily intake Cd, µg/day	Mean daily intake Cu, µg/day	Mean daily intake Zn, µg/ day
Meat and organs	68.50 (35.52)*	0.138	0.257	1.703	6.975	9.453	17.61	116.66	477.79
Meat products	18.54 (17.81)	0.703	0.027	1.388	5.353	13.03	0.501	25.73	99.25
Eggs	15.25 (22.01)	0.031	0.010	3.098	17.82	0.473	0.153	47.25	271.76
Milk	139.2 (129.5)	0.013	0.004	0.112	1.069	1.810	0.557	15.59	148.81
Cheese	17.25 (20.64)	0.495	0.004	0.830	6.229	8.539	0.069	14.32	107.45
Animal fat	4.17 (8.76)	0.063	0.005	1.612	1.528	0.263	0.021	6.722	6.372
Potato	95.17 (70.71)	0.544	0.091	3.035	7.004	51.77	8.660	288.84	666.6
Vegetables	250.6 (79.43)	0.286	0.097	3.168	8.076	71.67	24.31	793.90	2023.8
Fruits	101.0 (73.46)	0.307	0.021	1.677	2.204	31.01	2.121	169.38	222.60
Bread	345.4 (69.49)	0.053	0.040	1.681	5.400	18.31	13.82	580.62	1865.2
Corn flour	5.00 (7.20)	0.064	0.005	2.364	6.553	0.320	0.025	11.820	32.765
Water	2000	$4.28\ 10^{-3}$	$0.46\ 10^{-3}$	0.088	1.04	8.560	0.920	176.00	2080.0
Total intake/day (µg/day)						**215.21**	**68.76**	**2246.8**	**8002.4**
Total intake/day (µg/kg bw/day)						**3.074**	**0.982**	**32.10**	**114.3**
RfD** (µg/day/kg bw)						**3.50**	**1**	**40**	**300**
PTDI***(µg/day/kg bw)						**3.57**	**1**	**500**	**1000**

*Standard deviation in parenthesis; **Oral reference dose; ***Provisional tolerable daily intake according to FAO/JECFA [17].

Table 5: Daily intake of Pb, Cd, Cu and Zn, in rural area.

Food category	Mean intake rate, g/day	Mean Pb conc., mg/kg	Mean Cd conc., mg/kg	Mean Cu conc., mg/kg	Mean Zn conc., mg/kg	Mean daily intake Pb, µg/ day	Mean daily intake Cd, µg/day	Mean daily intake Cu, µg/ day	Mean daily intake Zn, µg/ day
Meat and organs	70.28 (37.48)*	0.104	0.044	1.291	15.45	7.309	3.092	90.73	1085.8
Meat products	9.97 (7.17)	0.614	0.081	2.090	5.313	6.122	0.808	20.84	52.97
Eggs	33.43 (26.01)	0.162	0.047	2.4	14.85	5.416	1.571	80.23	496.44
Milk	88.46 (78.05)	0.041	0.010	0.366	2.641	3.627	0.885	32.38	233.62
Cheese	28.30 (28.83)	0.408	0.046	1.447	4.462	11.55	1.302	40.95	126.27
Animal fat	8.72 (9.84)	0.123	0.008	1.961	2.009	1.073	0.070	17.10	17.518
Potato	95.77 (58.79)	0.242	0.161	2.757	9.211	23.18	15.42	264.04	882.14
Vegetables	236.4 (103.6)	2.08	0.237	5.813	24.93	491.71	56.03	1374.2	5893.4
Fruits	59.29 (45.42)	0.144	0.033	1.967	4.075	8.539	1.957	116.62	241.61
Bread**	299.0 (121.3)	0.053	0.040	1.681	5.388	15.85	11.96	502.62	1611.0
Corn flour	17.24 (57.32)	0.607	0.147	2.349	4.293	10.46	2.534	40.50	74.01
Water	2000	$6.30\ 10^{-3}$	$1.80\ 10^{-3}$	0.055	1.49	12.600	3.600	110.00	2980.0
Total intake/day (µg/day)						**597.43**	**99.22**	**2690.2**	**13695**
Total intake/day (µg/day/kg bw)						**8.535**	**1.417**	**38.43**	**195.6**

*Standard deviation in parenthesis; **The two rural localities are supplied with bread by the same bakery units that supply Baia Mare, so we considered the average concentrations of metals the same as for urban

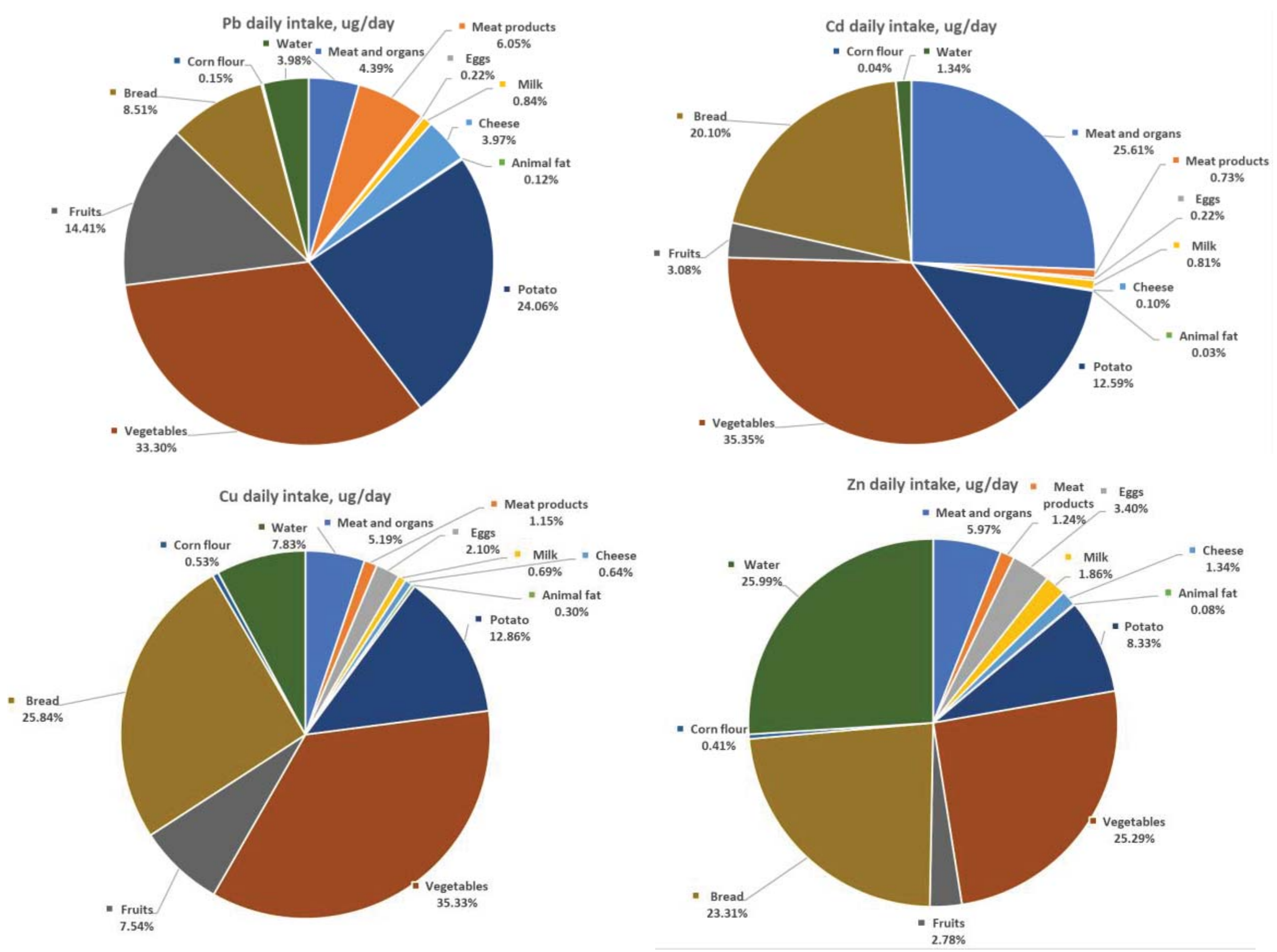

Figure 1 The contribution of the studied food categories to the daily food intake of Pb, Cd, Cu, Zn of the adult inhabitants of Baia Mare area.

for some European countries: Belgium, Denmark, Germany, Italy, Switzerland, the Netherlands. For example, the daily intake of Pb, respectively Cd by ingestion, calculated for the adult population in Switzerland is 25 µg/day, respectively 9 µg/day, namely 9 (24), respectively 8 (11) times lower than in Baia Mare (Sasar and Bozanta Mare).

Exposure assessment for both investigated areas by comparison with TDI values

The intake of each metal was calculated considering its concentration in food. To assess the health risk of daily intake of metals, these data were compared with Tolerable Daily Intake (TDI) values [5]. The FAO/WHO recommended values for TDI for Pb, Cd, Cu and Zn, with reference to the adult body, are given in table 6.

For urban residents, the daily intake of Pb, Cd, Cu, Zn through food and water revealed that: the mean total intake of Pb was 215.2 µg/person/day, representing 108% of TDI. Vegetables (including potatoes) contributed about 57% to the daily intake of Pb by ingestion. The average total intake of Cd was 68.75 µg/person/day, representing 97-121% of TDI. The contribution of Cd was mainly due to vegetables (35%), meat and organs (26%) and bread (20%). The average total intake of Cu was 2247 µg/person/day, representing 6.81% of TDI. The main food categories that contribute to the intake of Cu in the diet were vegetables (35%) and bread (26%). The average total intake of Zn was 8002 µg/person/day, representing 123% of TDI. Vegetables, drinking water and bread were the main contributors, with 25%, 26% and 23%, respectively, to the daily intake of ingested Zn.

For rural residents, the daily intake of Pb, Cd, Cu, Zn through food and water revealed that: the average total intake of Pb was 597.4 µg/person/day, representing 299% of TDI. Vegetables (including potatoes) contribute about 86% to the daily intake of Pb by ingestion. The average total intake of Cd was 99.22 µg/person/day, representing 140-174% of TDI. Vegetables (including potatoes) were the main foods that contributed to the daily intake of Cd, by 72%. The average total intake of Cu was 2690 µg/person/day, representing 8.2% of TDI. Vegetables and bread had an intake of 51%, respectively 19% to the daily ingested Cu. The average total intake of Zn was 13695 µg/person/day,

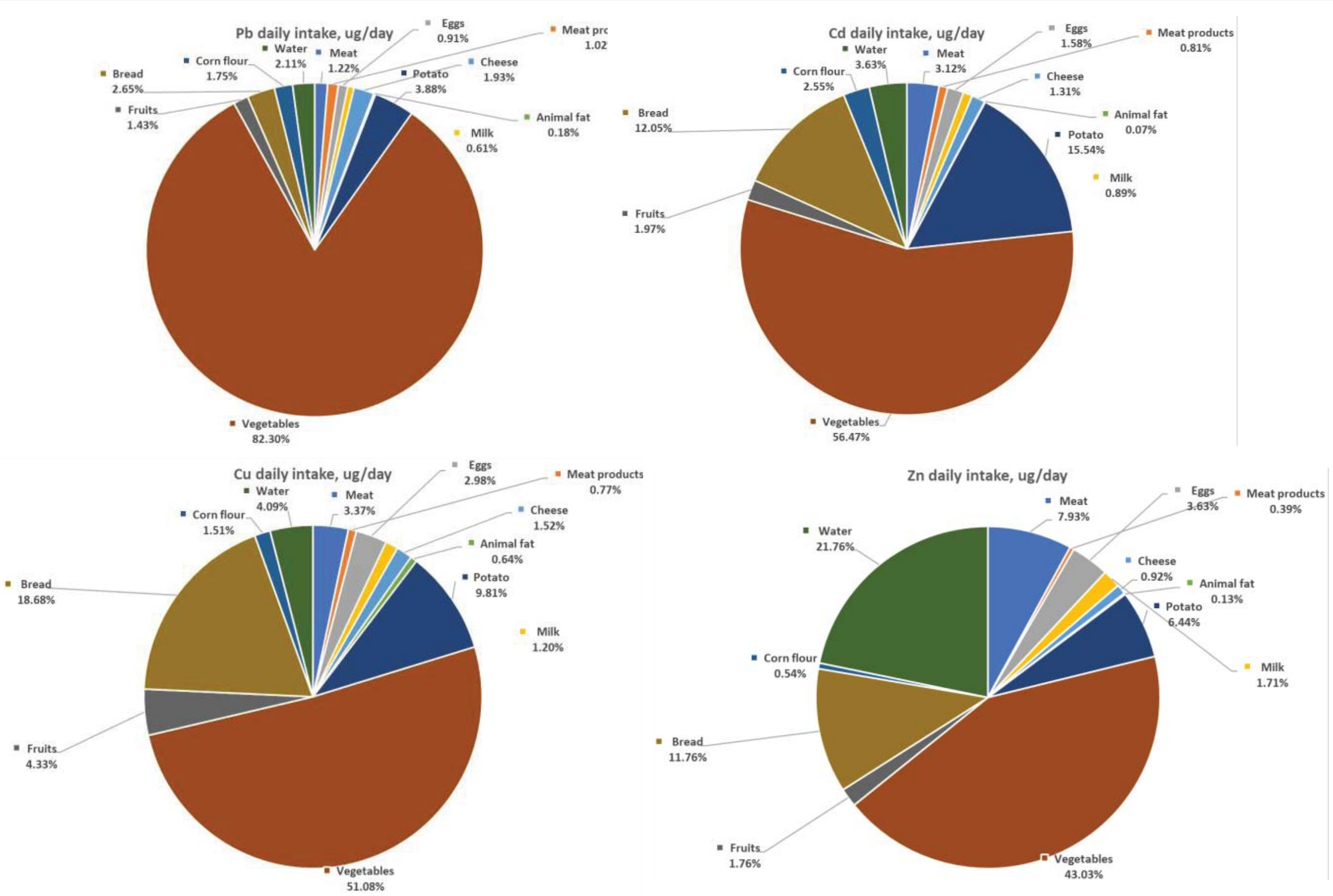

Figure 2 The contribution of the studied food categories to the daily food intake of Pb, Cd, Cu, Zn of the adult inhabitants of rural area.

Table 6: TDI values for Pb, Cd, Cu, Zn.

Metal	Pb	Cd	Cu	Zn
TDI, μg/day	200	57 - 71	33000	6500

representing 211% of TDI, indicating an excess of more than twice the allowable value of the TDI. Vegetables contributed with 43% to the daily intake of ingested Zn.

The average total intakes/person/day of Pb, Cd and Zn were higher than the Tolerable Daily Dose (TDI) for urban and rural residents, which indicated that there is a potentially high risk by ingestion of food and water. The average daily intake of Cu with food and drinking water was lower than the corresponding TDI value, which indicated that Cu intake was not a potential hazard to the health of residents in the study areas. The total intake/person/day of Pb, Cd, Cu and Zn in the investigated population group in the rural area was higher than in the investigated group in the urban area, by 2.8 times, 1.4 times, 1.2 times, and 1.7 times, respectively. The main food category that contributed to the intake of metals (Pb, Cd, Cu, Zn) in the studied areas, was represented by vegetables.

The value obtained for the daily intake of Pb by the ingestion of food and water for the inhabitants in the urban area (215.2 μg/day) was at the upper limit of the range of values reported in the literature (7-230 μg/day), and the value obtained for the daily intake of Pb for rural inhabitants (597.4 μg/day) was higher than these values reported in the literature [24,28-30]. The value obtained for the daily intake of Cd by the ingestion of food and water for the inhabitants in the urban area (68.75 μg/day) falls within the range of values reported in the literature: 4-84 μg/day [31], and the value obtained for the daily intake of Cd, by ingestion, for the inhabitants of the rural area (99.22 μg/day) was higher than the maximum limit of the interval. The values obtained for the daily intake of Cu by food and water intake for urban (2.25 mg/day) and rural (2.69 mg/day) were in the range of 1.2-4.8 mg/day reported for France and Germany [31,32]. The values obtained for the daily intake of Zn by the ingestion of food and water for the inhabitants of the urban area (8.00 mg/day) and of the rural area (13.69 mg/day) fall within the range of values reported in the literature: 7.0-18 mg/day [31,33]. The largest differences between the average daily intake of metals from rural to urban areas were found in vegetables (491.7 μg/day compared to 71.67 μg/day for Pb; 56.03 μg/day compared to 24.31 μg/day for Cd; 1374.2 μg/day compared to 793.9 μg/day for Cu and 5899 μg/day compared to 2024 μg/day for Zn, respectively). The explanation may lie in the fact that the rural area is directly exposed to winds blowing from nearby tailings dumps and tailings ponds and entraining suspended particles from their slopes. These suspended particles containing large amounts of metals are deposited on the leaves of vegetables, this being a way for metals to enter the plant, along with the absorption of metals from polluted soil. According to studies conducted in similar areas [34,35], 50% of the Pb content in vegetable leaves comes from the atmosphere. Thus, soil Pb can be a secondary source of exposure to plants, especially leafy vegetables.

Health multiple risk assessment by simultaneous exposure to more metals using THQ method

The non-carcinogenic risk assessment by food and water intake of the inhabitants of the studied areas was performed by calculating the Target Hazard Quotients (THQ) for each metal investigated (Pb, Cd, Cu and Zn, respectively), based on equation (3).

$$THQ = \frac{E_F \cdot E_D \cdot F_{1R} \cdot C}{R_f D_o \cdot W_{AB} \cdot T_A} \cdot 10^{-3} \quad (3)\ [36]$$

where;

E_F – exposure frequency (365 days/year);

E_D – exposure duration (70 years, estimated), equivalent to the average lifetime [48];

F_{IR} – food ingestion rate (g/person/day), estimated under community survey;

C – metal concentration in food (µg/g);

$R_f Do$ – reference oral dose (mg/kg/zi);

W_{AB} – average body weight for an adult (70 kg, estimated);

T_A – average duration of exposure to non-carcinogens (365 days/year x number of years of exposure, considered 70 years, in this study).

A THQ value greater than 1 indicates that the exposed population experiences a health risk, and a value below 1 indicates the relative absence of a health risk associated with the absorption of a metal through the consumption of contaminated food [34,36]. Oral reference dose values for studied metals are recommended by the United States Environmental Protection Agency (USEPA) [37] and are shown in table 7.

The individual Target Hazard Quotients (THQs) for the investigated metals and the TTHQ calculated for food intake in the two areas exposed to chronic metal pollution are shown in table 8.

The individual target hazard quotients, calculated for food consumption in urban area decreased in the following order: Cd>Cu>Pb>Zn, and in rural area: Pb>Cd>Cu>Zn, having values higher than 1 for Pb and Cd in the rural area and approximately equal to 1 for Cd (urban) and Cu (rural), indicating that the inhabitants of the investigated areas experience a potential health risk through food and water ingestion. Comparing the individual values of THQ, the potential health risk for the rural inhabitants was higher than for the urban area, for each investigated metal. These values suggested that the inhabitants under investigation are already experiencing adverse health effects due to food intake.

Comparing the THQ values obtained for Cd with the values reported for adults residing near the Zinc Processing Plant, Huludao, China (THQ Cd: 3.351-6.932), it is observed that the values are lower, but are comparable to the values calculated in the Hsingshan study (1.65), Lukang (1.73), Taishi (1.76) from Taiwan by oysters consumption [34,38]. The THQ Pb in rural area is comparable to that obtained for residents of Huludao, China (THQ Pb 2.085-2.447). The THQ Pb in the two studied areas were higher than those determined in the inhabitants of the Musi River, India, regarding the intake of vegetables grown on irrigated soil with contaminated waters (0.02-0.11) [39]. The values of THQ Cd (urban and rural), THQ Pb (urban) obtained in this study are lower, and THQ Cu (urban and rural) are higher than those determined by Zhuang, et al. [40] for the inhabitants near the Dabaoshan mine, China, which consumes rice (THQ Pb: 1.43-1.99; THQ Cd: 2.61-6.25; THQ Cu: 0.66-0.89; THQ Zn: 0.48-066). Also, the values obtained for Pb and Cd are comparable to those reported by Khan, et al. [41] for residents of Beijing, China who consume vegetables grown on irrigated soils with wastewater.

The graphical representation of TTHQ, depending on the THQ value for the studied metals, for the two areas under investigation is given in figure 3 and highlights the dominant

Table 7: Values of oral reference doses for Pb, Cd, Cu, Zn.

	Pb	Cd	Cu	Zn
R_fDo, µg/kg bw/day	4	1	40	300

Table 8: THQ for Pb, Cd, Cu, Zn and TTHQ.

Area	THQ Pb	THQ Cd	THQ Cu	THQ Zn	TTHQ
Baia Mare, urban	0.769	0.982	0.802	0.381	2.934
Sasar and Bozanta Mare, rural	2.134	1.417	0.961	0.652	5.164

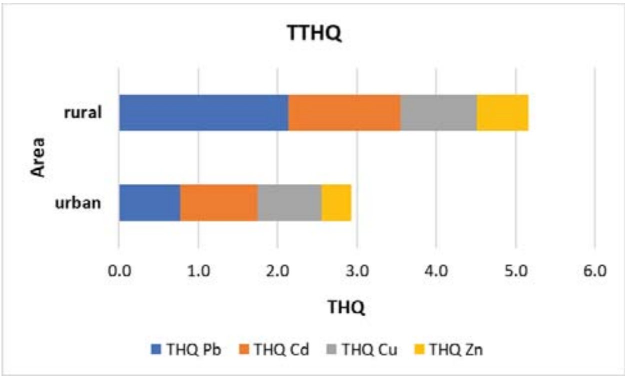

Figure 3 TTHQ associated with different food consumption, for both investigated areas, urban and rural.

contribution of Pb and Cd and the secondary contribution of Zn to the health risk of the inhabitants in the studied areas. The main contributors to the TTHQ value, in the two areas, were Pb and Cd (and Cu, in urban), being also the most toxic elements among those investigated.

The TTHQ quotients for the two areas (urban and rural, respectively) were 2.934 and 5.164, respectively, suggesting that the inhabitants under investigation are already experiencing adverse effects on human health due to food intake.

The results showed that the intake of food, especially vegetables was an important way of exposure of the population in areas with soil contaminated with metals. These results were consistent with those obtained in other studies [42,43]. The TTHQ value obtained for the urban area was lower, and for the rural area was comparable with those reported by Zheng, et al. [34], regarding the inhabitants near the Zinc Processing Plant, Huludao, China (5.79 - 9.90).

CONCLUSION

The present study was carried out in two areas (Urban: Baia Mare city, and rural: Sasar and Bozanta Mare villages situated in the proximity of Baia Mare city) and revealed high concentrations of metals in vegetables, as well as in other categories of food consumed by the inhabitants.

Food is an important route of exposure for metals, especially for the population consuming food/vegetables grown on contaminated soil. In the risk assessment process, the parameters specific to the investigated areas and the food intakes of metals were used. Specific exposure factors were obtained using dietary structure questionnaires. The results of the risk assessment indicated that the inhabitants of the investigated areas (urban and rural) experience a potential health risk through food intake. The most relevant route of exposure by ingestion is through the consumption of vegetables, especially in rural areas. One way to reduce the risk of exposure is by selecting vegetables for cultivation, especially those with lower metal concentrations, or lower bioconcentration factors. This study highlights the degradation of environmental factors in the studied areas and, implicitly, the potential danger on the health status of the exposed inhabitants. Authorities must control the phenomenon of pollution in the investigated areas, by limiting the spread of pollutants. In addition, soil remediation measures and the restriction of vegetable cultivation on contaminated soils are required.

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