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MICROELEMENT COMPOSITION OF BASIC CONSUMPTION PRODUCTS IN THE TRANSCARPATHIAN REGION, UKRAINE

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ABSTRACT
Traditional food products (milk and vegetables) form the diet basis and are the main source of essential trace elements (microelements). The contents of trace elements in food products obtained from various landscape areas significantly differ. Transcarpathia has a pronounced tectonic and geological diversity. The young Carpathian Mountains face high tectonic and geological activity, which can affect the microelement composition of food products. A significant difference in the microelement composition of milk and vegetable mix was found for different landscape zones, for instance, food in lowland areas is richer in Fe, Cu, Zn, Mo, Co, P, Se, I, Br, F, Ca, and Mg while food in mountainous areas contains large amounts of As and Mn. Consistent patterns of microelement distribution in the food products from different landscape zones (lowland > foothill > mountainous) were estimated using the Spearman correlation coefficient. For milk samples: Fe: 0.91; Cu: 0.89; Mn: -0.94; Zn: 0.88; Mo: 0.89; Co: 0.94; Ca: 0.91; Mg: 0.91; P: 0.92; As: -0.94; Se: 0.82; I: 0.84; Br: 0.73; F: 0.82, for vegetable mix samples: Fe: 0.91; Cu: 0.85; Mn: -0.94; Zn: 0.79; Mo: 0.90; Co: 0.92; Ca: 0.94; Mg: 0.94; P: 0.80; As: -0.91; Se: 0.84; I: 0.91; Br: 0.73; F: 0.88. Pronounced inter-element correlation of microelements in food products is observed (the value of the Pearson correlation coefficient for all pairs of chemical elements is $r > 0.60$).

1. Introduction

Nutrition is a prerequisite for a healthy and full quality life. Each food item affects human health (Niva, 2007). Therefore, the quality and safety of food products requires special attention (Khan *et al.,* 2017; Sohail *et al.,* 2018; Barrett, 2010; Lam *et al.*, 2013; Röhr, 2005). Similarly, traditional food products should be carefully considered, since they are the basis of any diet (Guerrero *at el.,* 2010).

In most cases, nutritional value of food products is measured by the content of the main components (proteins, carbohydrates, fats, etc.) or biologically active substances (vitamins, polyphenols, etc.) (Chen *at el.,* 2017; Abuajah *at el.,* 2014; Rahaiee *at el.,* 2014; Kadnikova *et al.*, 2015; Sharma *et al.*, 2012). However, the micro- and macronutrient composition of food products is also an important quality criterion (Harmankaya *et al.*, 2012; Kizil and Turk, 2010; Özcan *et al.*, 2013; Rudawska and Leski, 2005; Simsek and Aykut, 2007; Škrbić and Onjia, 2007; Słupski *et al.*, 2005; Xiao *et al.*, 2016). New approaches, which include personalizing the choice of a diet, are an important component of rational and therapeutic nutrition and affect immunity and other crucial health components (Luo *at el.,* 2018; Kovalskys *at el.,* 2015; Shahidi, 2006). An important aspect of assessing food products' quality is their microelement composition, which can be used to measure the degree of ingestion of vital chemical elements into the human body when eating food, which affects human health (Bilandžić *at el.,* 2015; Marles, 2017; Martínez-Ballesta *et al.*, 2010). The content of microelements (essential trace minerals) in food products depends on their content in soils and rocks. Therefore, the geology and geochemistry of the studied areas can significantly affect the contents of essential trace elements in foods.

Based on the reference diet of Ukrainian citizens, milk and dairy products (equivalent to milk) account for about 42% (1,022 kg per day) of the diet, vegetables – about 26% (potatoes – 0.359 kg per day, other vegetables – 0.279 kg per day) of the diet (MHPU, 1997). These products, which are also traditional, form the basis of a person's diet, including residents of the Transcarpathian region. Since most residents of the Transcarpathian region consume household-produced dairy products and vegetables, the latter's microelement composition depends on how they were cultivated and obtained. The Transcarpathian region is characterized by significant landscape diversity (height difference varies from 100 m to 2100 m) including geochemical zones and terrain features. Therefore, traditional food products (milk and vegetables) obtained in different landscape zones can differ much in terms of microelement composition. This affects the entry of vital trace elements into the human body and, therefore, health. No such studies for traditional food products of the Transcarpathian region have been conducted before.

This paper presents data on the microelement composition (based on 16 elements, including Hg and Pb safety indicators) of traditional food products (whole cow's milk and vegetable mix) of the Transcarpathian region taking into account the area's landscape and geochemical diversity. The contents of microelements: Fe, Cu, Mn, Zn, Mo, Co, As, Se, I, Br and F, macroelements: P, Ca and Mg, and safety indicators: Hg and Pb were studied. The choice of Hg and Pb as a safety indicator is based on their high toxicity to humans, even in small amounts (Boudebbouz *at el.,* 2021). Based on the generalization of the results of analyses, major features of the microelement composition of traditional food products have been identified, which may have a decisive effect on the health of the region's population. A similar study was carried out for raw cow milk in Slovakia (Pšenková *at el.,* 2020).

2. Materials and methods

2.1. Study area

The research study was conducted for all landscape zones of the Transcarpathian region in 2018. It covers a mountainous area (3 districts: Rakhiv, Mizhhirya, and Volovets) located on the outer flysch areas (the Krosno Zone) and inner covers and rock zones; a foothill area (6 districts: Svaliava, Irshava, Khust, Tiachiv, Perechyn, and Velykyi Bereznyi) located on different territories and covering the Volcanic Carpathians, the Marmara massif, outer flysch areas, and partially inner covers and rock zones; and a lowland area (4 regions: Berehove, Vynohradiv, Mukachevo, and Uzhhorod) located within the Transcarpathian deflection. Tectonic and geochemical peculiarities of the studied area are considered in the study (Sukharev *et al.*, 2020).

On Figure 1, shows a fragment of tectonic map of the Ukrainian Carpathians (Transcarpathian region) with research areas.

On Figure 2, shows a fragment of a geologic map of the Ukrainian Carpathians (Transcarpathian region) (Sukharev *et al.*, 2020).

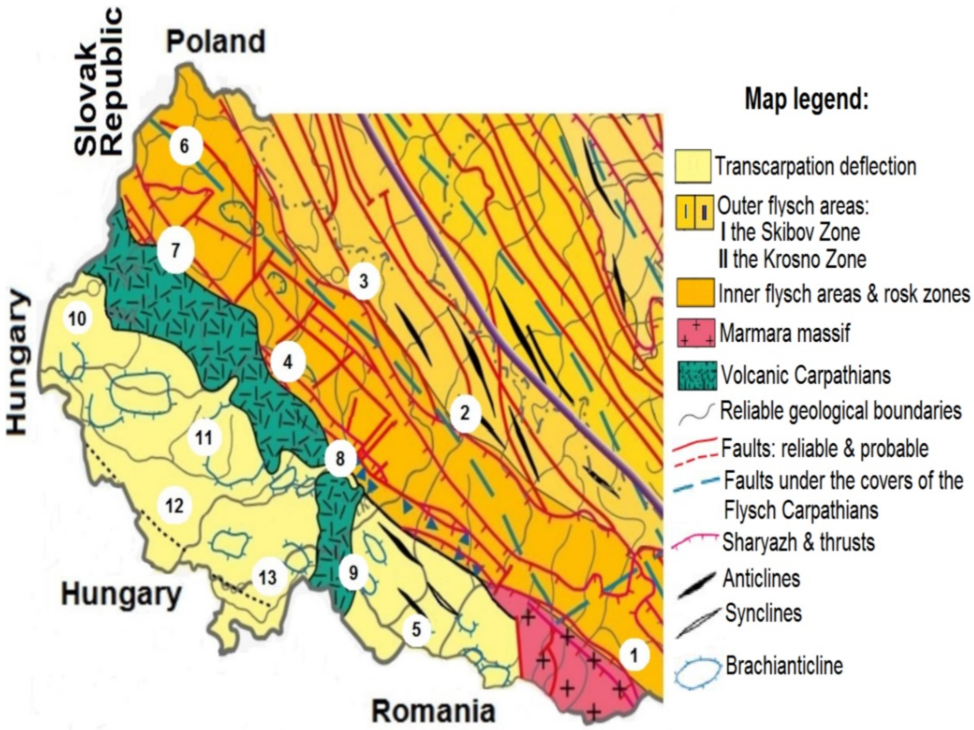

Figure 1. Fragment of tectonic map of Ukrainian Carpathians (Transcarpathian region). Research areas (district):
1 – Rakhiv; 2 – Mizhhirya; 3 – Volovets; 4 – Svaliava; 5 – Tiachiv; 6 – Velykyi Bereznyi; 7 – Perechyn; 8 – Irshava; 9 – Khust; 10 – Uzhhorod; 11 – Mukachevo; 12 – Berehove; 13 – Vynogradiv

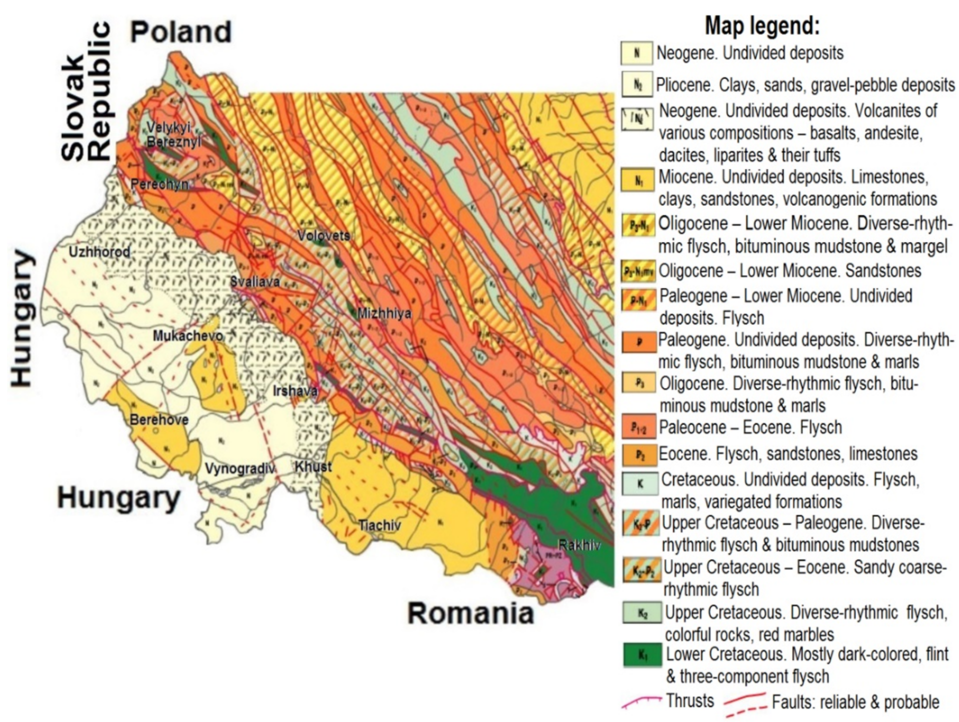

Figure 2. Fragment of geologic map of Ukrainian Carpathians (Transcarpathian region)

2.2. Sampling and sample preparation

Samples of the whole cow's milk were obtained by mixing 10 different milk samples from different households in respective districts taking into account the areas' landscape and terrain features.

Vegetable mix samples (potato : white cabbage : beet : onion = 3:1:1:0.5) were prepared for each district respectively, from different households, averaging 10 samples after grinding. Samples were homogenized. The ratio of vegetable mix components was selected based on consumer needs and considering the reference diet of Ukrainian citizens. Vegetables were grinded (homogenized) using a blender with polymer blades.

When sampling milk and vegetables, we took into account the number of villages in each study district and the ratio of the area of different landscapes in them. The samples were collected in the way that they would represent the features of the territories.

Dry ashing (determination of Fe, Cu, Zn, Mn, Mo, Co, Pb, Ca, and Mg) and wet ashing (determination of P, As, Se, I, Br, F, and Hg, for iodine with KOH) techniques were used for sample preparation. These sample preparation methods are traditional (Lars, 2000; Lars and Joakim, 2000; Nielsen, 2017; Sahrawat *et al.*, 2006). For dry ashing, a muffle furnace was used (Protech, PT-1400M, Italia). The acids (and KOH) used for wet ashing were and preparation of solutions (after dry ashing) of analytical grade.

For example, the preparation of milk samples with dry ashing was carried out as follows. Twenty-five mL of each sample was put in ceramic crucibles and dried in 450°C by heater. After that, crucibles containing the samples were put in the oven at 450°C for 4 hours until the sample turned to ash. In the next step, 5.0 mL of nitric acid (1.0 $mol \cdot L^{-1}$) was added to the vessel containing the sample and heated to dissolve of ash. After cooling the solution, the volume was increased to 50 mL with nitric acid 0.1 $mol \cdot L^{-1}$.

2.3. Analytical methods and instruments

All the reagents used in this research were of analytical grade. In the research study, double-distilled water was used. Standard procedures were used to determine microelements in food products (Nielsen, 2017). Standard solutions for making calibration curves (determination of As, Ca, Co, Cu, Fe, Pb, Mg, Mn, Mo, Se, Zn) were obtained by diluting commercial multi-element standard stock solution (SPEX QC-21, USA). Thus, the total content of trace elements in food products was determined.

Atomic absorption spectroscopy was used to determine Cu, Zn, Fe, Pb, Mn, Mo, Co (electrothermal technique: graphite furnace, chemical modifier $Pd(NO_3)_2$), Hg (cold vapor technique), and As (hydride generation technique). Experimental conditions (wavelength, nm): Cu – 324.8, Zn – 213.9, Fe – 248.3, Mn – 279.5, Mo – 313.3, Co – 240.7; Pb – 283.3; Hg – 253.7; As – 193.7. AAS vario® 6 (Analytik Jena AG, Germany), a hydrogen generator (Varian VGA-76, USA), and ultrapure $Pd(NO_3)_2$ (Suprapur®, Merck, Germany) were involved in the study.

Flame atomic emission spectroscopy was used to determine Ca and Mg (wavelength, nm: Ca – 434, Mg – 385; FPA-2-01, LLC LabTime Ltd., Russia). The following methods were used to determine specific elements: F^- and Br^- – potentiometry (SevenCompact S220, Mettler Toledo, USA); iodine – inverse voltammetry (Ecotest-VA-iodine, Russia); Se – spectrofluorimetry (λ_{abs} = 378 nm, λ_{em} = 520 nm, Hitachi F-7000, Hitachi Ltd., Japan); and P – spectrophotometry (Shimadzu UV-1800, Shimadzu Co., Japan).

2.4. Statistics and Mapping

Standard statistical methods were used; in particular, the Pearson and Spearman correlation coefficients were calculated using the SPSS Statistics (IBM) and OriginPro (OriginLab Corporation) programs. The ArcGIS 10.2.1 program was used to map the territory of the Transcarpathian region based on the results of microelements' determination in the food products.

The results of trace elements' determination in milk samples are presented in Tables 1-3.

3. Results and discussions

Table 1. The results of determination of some trace elements (metals) in the milk from the Transcarpathian region (n = 6; P = 0.95)

Milk samples‡ (district)	**Trace element content, mg·L⁻¹**					
	Fe	**Cu**	**Zn**	**Mn**	**Mo**	**Co**
Mountain area						
Rakhiv	2.17±0.11	0.13±0.01	2.34±0.12	0.21±0.02	0.031±0.002	0.021±0.002
Mizhhirya	1.96±0.10	0.18±0.01	2.61±0.13	0.19±0.02	0.029±0.002	0.027±0.003
Volovets	2.34±0.12	0.22±0.02	2.53±0.13	0.22±0.02	0.036±0.002	0.025±0.003
Average content	*2.16±0.20*	*0.18±0.05*	*2.49±0.15*	*0.21±0.02*	*0.032±0.004*	*0.024±0.003*
Foothill area						
Svaliava	2.51±0.12	0.31±0.02	3.19±0.19	0.15±0.01	0.052±0.004	0.052±0.005
Tiachiv	2.72±0.13	0.29±0.02	3.08±0.20	0.18±0.01	0.042±0.003	0.056±0.005
Velykyi Bereznyi	2.66±0.12	0.25±0.02	2.88±0.18	0.14±0.01	0.047±0.004	0.044±0.004
Perechyn	2.63±0.13	0.27±0.02	3.27±0.21	0.17±0.01	0.044±0.003	0.048±0.005
Irshava	2.84±0.13	0.33±0.02	3.92±0.22	0.13±0.01	0.051±0.004	0.051±0.005
Khust	2.81±0.14	0.34±0.02	3.60±0.19	0.11±0.01	0.051±0.005	0.066±0.006
Average content	*2.70±0.19*	*0.30±0.05*	*3.32±0.60*	*0.15±0.04*	*0.048±0.006*	*0.053±0.013*
Lowland area						
Uzhhorod	2.89±0.14	0.31±0.02	3.79±0.20	0.09±0.01	0.061±0.005	0.073±0.007
Mukachevo	2.97±0.15	0.38±0.03	4.02±0.22	0.12±0.01	0.054±0.004	0.089±0.007
Berehove	3.09±0.15	0.40±0.03	4.29±0.23	0.10±0.01	0.055±0.004	0.091±0.007
Vynohradiv	3.02±0.14	0.36±0.02	4.11±0.19	0.08±0.01	0.058±0.005	0.087±0.007
Average content	*3.00±0.11*	*0.36±0.05*	*4.05±0.26*	*0.10±0.02*	*0.057±0.004*	*0.085±0.012*

Note: ‡ – milk density is 1.027 kg·L^{-1}; Safety indicators: Hg content from < 0.5 to 1.03 μg·L^{-1}; Pb – 5.7-13.1 μg·L^{-1}. The lowest Hg and Pb content in milk samples from mountainous areas. Permissible content of Hg and Pb in milk ≤ 10 μg·kg^{-1} and ≤ 20 μg·kg^{-1} respectively (EC, 2015; FAO/WHO, 2011).

Table 2. The results of determination of some trace elements (non-metals) in the milk from the Transcarpathian region (n = 6; P = 0.95)

Milk samples (district)	**Trace element content, μg·L⁻¹**				
	As	**Se**	**I**	**Br**	**F**
Mountain area					
Rakhiv	32.2±3.8	21.4±2.1	50.9±4.8	344±47	48.1±6.6
Mizhhirya	28.3±3.4	19.3±2.0	47.7±4.7	406±55	56.9±6.8
Volovets	27.1±3.3	24.8±2.9	63.0±5.5	368±46	38.5±56
Average content	*29.2±3.0*	*21.8±3.0*	*53.9±9.1*	*373±33*	*47.8±9.3*
Foothill area					
Svaliava	19.9±2.7	52.5±5.7	115±11	492±61	59.4±6.7
Tiachiv	21.3±2.9	43.9±5.1	127±11	890±89	83.9±9.6
Velykyi Bereznyi	25.8±2.9	52.1±5.7	96±9	423±52	63.1±7.7
Perechyn	22.1±2.5	49.3±5.5	112±10	549±63	74.8±8.8
Irshava	23.6±2.6	50.5±5.5	118±10	641±68	73.2±8.7

Khust	17.4±2.2	54.8±5.6	122±11	677±70	68.7±7.5
Average content	*21.7±4.3*	*50.5±6.6*	*115±19*	*612±278*	*70.5±13.3*
Lowland area					
Uzhhorod	12.8±1.7	51.4±5.7	115±10	624±67	77.5±8.8
Mukachevo	14.1±1.8	52.3±5.7	137±12	589±65	91.2±9.6
Berehove	18.7±2.0	61.7±6.6	153±12	753±78	118.7±12.9
Vynohradiv	15.2±1.9	58.8±6.5	144±12	805±82	104.3±11.1
Average content	*15.2±3.5*	*56.1±5.6*	*137±22*	*693±112*	*97.9±20.8*

Table 3. The results of macroelements determination in the milk from the Transcarpathian region (n = 6; P = 0.95)

Milk samples (district)	Macroelement content, mg·L^{-1}			Ca/Mg
	P	Ca	Mg	
Mountain area				
Rakhiv	97±5	1985±109	173±10	11.5
Mizhhirya	111±5	2077±108	189±11	11.0
Volovets	103±5	2104±110	182±11	11.6
Average content	*104±7*	*2055±70*	*181±8*	*11.4±4*
Foothill area				
Svaliava	154±7	2197±116	221±12	9.9
Tiachiv	151±7	2136±103	213±12	10.0
Velykyi Bereznyi	129±6	2098±109	218±12	9.6
Perechyn	136±7	2174±115	202±11	10.8
Irshava	163±7	2263±106	243±13	9.3
Khust	168±8	2389±107	267±14	9.0
Average content	*150±21*	*2210±179*	*227±40*	*9.8±1.0*
Lowland area				
Uzhhorod	177±8	2410±113	279±15	8.6
Mukachevo	179±8	2373±111	231±13	10.3
Berehove	163±8	2488±114	253±14	9.8
Vynohradiv	182±8	2421±116	298±16	8.1
Average content	*175±12*	*2423±65*	*265±34*	*9.2±1.1*

The results of trace elements' determination in milk samples are presented in Tables 1-3.

The studied milk samples are characterized by rich microelement composition while safety indicators (content of Hg and Pb) confirm their safety. Our data are consistent with the results of determining the content of heavy metals in cow's milk from the mountainous area of Romania (Cadar *at el.,* 2015). The level of essential trace elements in milk samples significantly varies (mg·L^{-1}): Fe: 1.96-3.09; Cu: 0.13-0.40; Zn: 2.34-4.29; Mn: 0.08-0.22; Mo: 0.029-0.061; Co: 0.021-0.091; P: 97-182; As: 0.013-0.032; Se: 0.019-0.062; I: 0.048-0.153; Br: 0.34-0.81; F: 0.039-0.119. The content of the majority of trace elements (Fe, Cu, Zn, Mo, Co, P, Se, I, Br, F) in milk samples from the mountainous area is significantly lower than in milk samples from the foothill and lowland areas. The exception is As and Mn, the content of which is higher in milk from the mountainous area than in milk from the foothill and lowland areas. For example, Figure 3 shows a map of the Transcarpathian region according to the distribution of arsenic content in milk, with the highest content being observed in the milk sample from Rakhiv district (it has the highest average height above sea level). This is obviously associated with the relatively high As content in this region's soils as evidenced by the presence of arsenide mineral waters in the Rakhiv district.

Microelement distribution in milk samples from different landscape zones (lowland area > foothill area > mountainous area) was estimated using the Spearman coefficient: Fe: 0.91; Cu: 0.89; Mn: -0.94; Zn: 0.88; Mo: 0.89; Co: 0.94; Ca: 0.91; Mg: 0.91; P: 0.92; As: -0.94; Se: 0.82; I: 0.84; Br: 0.73; F: 0.82. Thus, consumption of milk from the lowland landscape zone of the Transcarpathian region provides a more complete supply of microelements to the human body. The authors (Falandysz *et al.*, 2008; Falandysz *et al.,* 2012) also established similar patterns of content of some trace elements in mushrooms of the mountains and lowland areas.

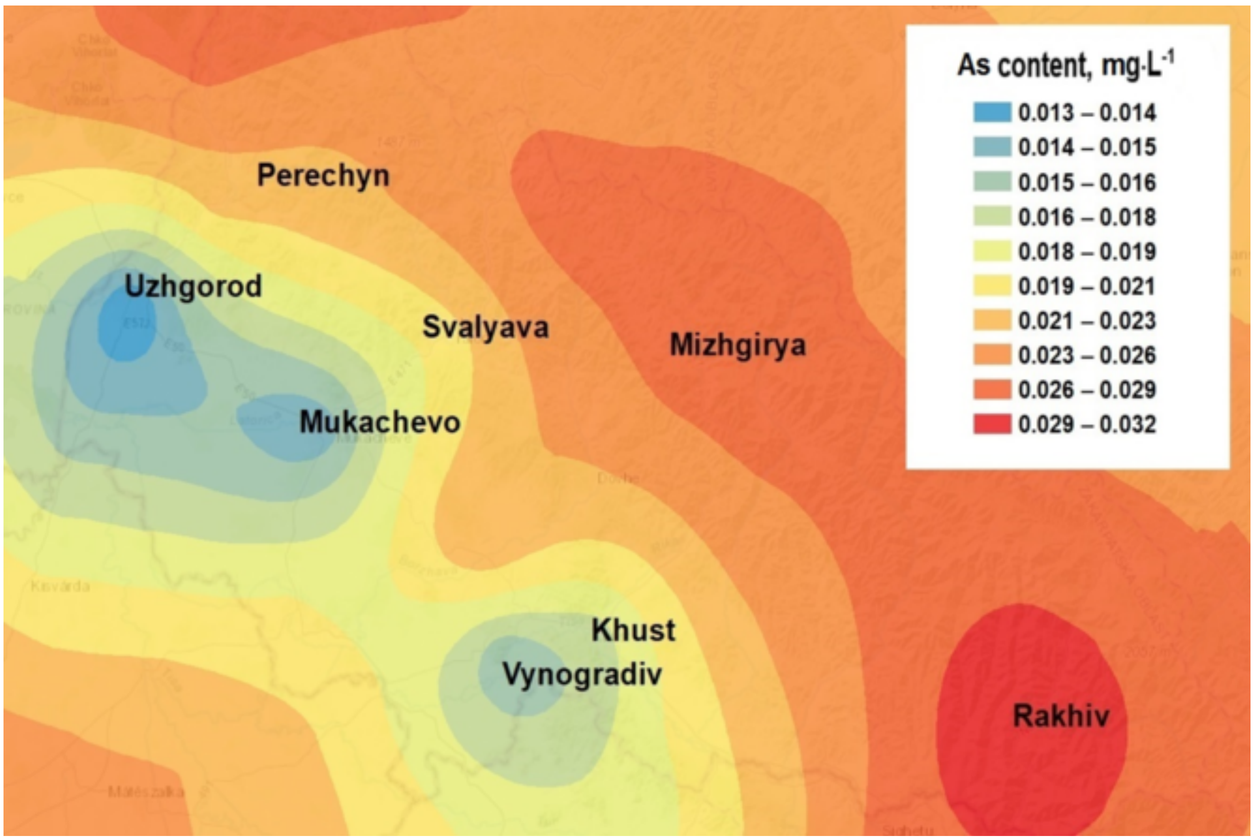

Figure 3. Map of arsenic distribution in milk samples from the Transcarpathian region

A similar consistent pattern is observed for the macroelement content in milk samples from different landscape zones. For example, Ca content in milk samples varies at the rate of 1985-2488 mg·L^{-1}, Mg – 173-298 mg·L^{-1}, and their ratio (Ca/Mg) varies between 8.1 and 11.5. Though Ca and Mg content in milk samples from the mountainous area is lower than in foothill and lowland areas, the ratio of Ca/Mg content in milk samples from the mountainous area is higher (Fig. 4).

Importantly, the content of different trace elements in milk samples correlates with each other, and the values of the Pearson coefficient of inter-element correlation are high (r > 0.60) as can be seen in Table. 4. A particularly pronounced correlation is observed for such pairs of microelements as (values of the Pearson coefficient): Fe:Cu – 0.92; Fe:Zn – 0.92; Fe:Mo – 0.90; Fe:Co – 0.91; Fe:Se – 0.91; Fe:I – 0.95; Cu:Zn – 0.95; Cu:Co – 0.93; Cu:P – 0.92; Cu:Se – 0.90; Cu:I – 0.95; Zn:Co – 0.94; Zn:P – 0.93; Zn:I – 0.91; Mo:P – 0.92;

Mo:Se – 0.91; Co:P – 0.92; Co:I – 0.92; Co:F – 0.90; P:I – 0.90; Se:I – 0.94. An inverse pattern regarding other trace elements is observed for Mn and As (Table 4). This may indicate a relative stability of the chemical composition of milk even from different landscape zones. In order to illustrate the revealed inter-element patterns of microelement content in milk samples, Figure 5 presents the respective correlation diagram. The authors (Pilarczyk *at el.,* 2013) also showed significant very high or high positive inter-element correlations in milk.

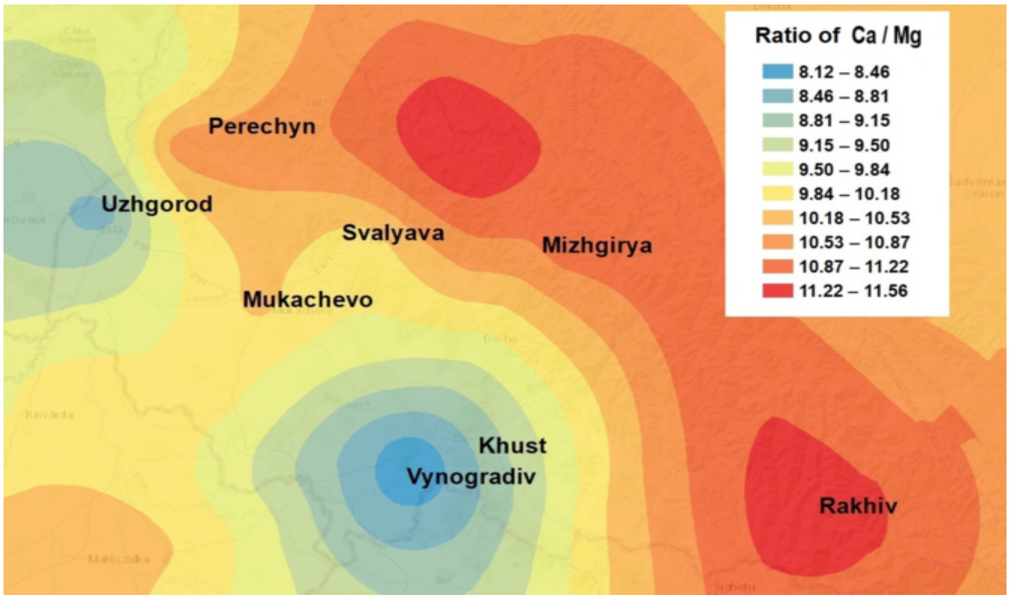

Figure 4. The map of the Transcarpathian region by the ratio of Ca/Mg in milk

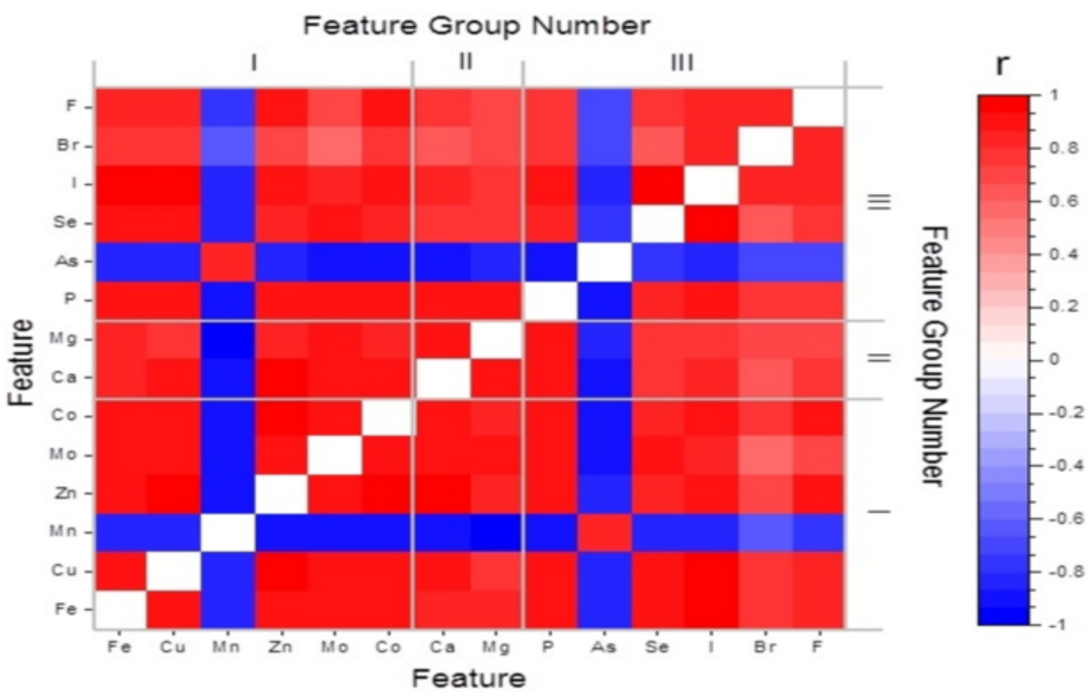

Figure 5. Correlation diagram of inter-element correlation of microelement content in milk samples (Pearson's correlation coefficient): I – trace elements metals; II – macro elements; III – trace elements non-metals

Table 4. The values of the Pearson coefficient for consistent patterns of inter-element composition of milk from the Transcarpathian region (σ < 0.05)

-	Fe	Cu	Mn	Zn	Mo	Co	Ca	Mg	P	As	Se	I	Br	F
Fe	X	0.92	0.85	0.92	0.90	0.91	0.86	0.82	0.88	-0.81	0.91	0.95	0.76	0.82
Cu	0.92	X	-0.83	0.95	0.88	0.93	0.91	0.79	0.92	-0.85	0.90	0.95	0.74	0.81
Mn	-0.85	-0.83	X	-0.90	-0.92	-0.89	-0.91	-0.96	-0.91	0.83	-0.86	-0.80	-0.61	-0.75
Zn	0.92	0.95	-0.90	X	0.89	0.94	0.94	0.85	0.93	-0.83	0.86	0.91	0.73	0.87
Mo	0.90	0.88	-0.92	0.89	X	0.87	0.87	0.89	0.92	-0.88	0.91	0.86	0.60	0.68
Co	0.91	0.93	-0.89	0.94	0.87	X	0.93	0.83	0.92	-0.90	0.84	0.92	0.74	0.90
Ca	0.86	0.91	-0.91	0.94	0.87	0.93	X	0.89	0.89	-0.88	0.79	0.83	0.67	0.79
Mg	0.82	0.79	-0.96	0.85	0.89	0.83	0.89	X	0.89	-0.83	0.79	0.77	0.69	0.69
P	0.88	0.92	-0.91	0.93	0.92	0.92	0.89	0.89	X	-0.93	0.86	0.90	0.76	0.75
As	-0.81	-0.85	0.83	-0.83	-0.88	-0.90	-0.88	-0.83	-0.93	X	-0.77	-0.82	-0.67	-0.68
Se	0.91	0.90	-0.86	0.86	0.91	0.84	0.79	0.79	0.86	-0.77	X	0.94	0.67	0.74
I	0.95	0.95	-0.80	0.91	0.86	0.92	0.83	0.77	0.90	-0.82	0.94	X	0.83	0.87
Br	0.76	0.74	-0.61	0.73	0.60	0.74	0.67	0.69	0.76	-0.67	0.67	0.83	X	0.80
F	0.82	0.81	-0.75	0.87	0.68	0.90	0.79	0.69	0.75	-0.68	0.74	0.87	0.80	X

Table 5. The results of determination of trace elements (metals) in vegetable mix from the Transcarpathian region (n = 6; P = 0.95)

Samples of vegetable mix (district)	**Trace element content, mg·kg^{-1}**					
	Fe	**Cu**	**Zn**	**Mn**	**Mo**	**Co**
Mountain area						
Rakhiv	3.92±0.29	0.62±0.05	2.55±0.17	3.18±0.29	0.053±0.004	0.018±0.002
Mizhhirya	4.14±0.28	0.73±0.06	2.74±0.18	2.93±0.27	0.062±0.005	0.025±0.002
Volovets	4.39±0.31	0.71±0.06	2.61±0.17	3.32±0.29	0.069±0.005	0.021±0.002
Average content	*4.15±0.24*	*0.69±0.07*	*2.63±0.11*	*3.14±0.21*	*0.061±0.008*	*0.021±0.004*
Foothill area						
Svaliava	4.77±0.33	0.86±0.07	3.66±0.23	2.18±0.21	0.082±0.006	0.032±0.003
Tiachiv	5.45±0.35	0.71±0.06	3.18±0.20	2.74±0.25	0.069±0.005	0.035±0.003
Velykyi Bereznyi	5.22±0.34	0.77±0.06	2.93±0.18	2.07±0.20	0.066±0.005	0.028±0.003
Perechyn	5.09±0.34	0.81±0.07	3.47±0.22	2.54±0.24	0.071±0.006	0.029±0.003
Irshava	6.61±0.42	0.92±0.07	3.69±0.22	1.91±0.19	0.077±0.006	0.037±0.004
Khust	5.88±0.36	0.94±0.07	3.28±0.21	1.52±0.15	0.087±0.006	0.042±0.004
Average content	*5.50±1.11*	*0.84±0.13*	*3.37±0.44*	*2.16±0.64*	*0.075±0.012*	*0.034±0.008*
Lowland area						
Uzhhorod	6.92±0.41	0.83±0.07	3.57±0.22	1.28±0.13	0.117±0.008	0.037±0.003
Mukachevo	6.69±0.39	1.07±0.08	3.96±0.23	1.67±0.17	0.084±0.006	0.051±0.005
Berehove	7.81±0.44	1.21±0.09	4.69±0.27	1.19±0.13	0.093±0.007	0.053±0.005
Vynohradiv	7.63±0.43	1.32±0.10	4.51±0.26	1.34±0.14	0.108±0.007	0.048±0.005
Average content	*7.26±0.57*	*1.11±0.28*	*4.18±0.61*	*1.37±0.30*	*0.101±0.017*	*0.047±0.010*

Note: Safety indicators: Hg content from < 0.5 to 3.1 μg·kg^{-1}; Pb – 3.4-21.4 μg·kg^{-1}. The lowest Hg and Pb content in vegetable mix samples from mountainous areas. Permissible content of Hg and Pb in vegetable ≤ 10 μg·kg^{-1} and ≤ 100 μg·kg^{-1} respectively (EC, 2015; FAO/WHO, 2011).

Table 6. The results of determination of some trace elements (non-metals) in vegetable mix from the Transcarpathian region (n = 6; P = 0.95)

Samples of vegetable mix (district)	Trace element content, μg·kg^{-1}				
	As	Se	I	Br	F
Mountain area					
Rakhiv	92.9±9.9	3.13±0.47	7.07±0.81	102±16	101±19
Mizhhirya	88.1±9.5	4.19±0.66	6.24±0.70	125±19	121±21
Volovets	78.7±8.7	5.38±0.85	7.85±0.83	113±17	92±18
Average content	*86.6±7.9*	*4.23±1.15*	*7.05±0.81*	*113±12*	*105±16*
Foothill area					
Svaliava	59.3±7.5	7.04±0.95	23.7±2.1	153±26	132±24
Tiachiv	61.7±7.7	7.98±0.99	33.0±2.8	258±36	169±25
Velykyi Bereznyi	77.0±8.6	5.07±0.75	14.4±1.4	140±25	137±24
Perechyn	66.2±8.1	6.32±0.86	18.5±1.8	161±26	153±25
Irshava	70.5±8.5	8.05±0.98	26.9±2.5	192±29	144±24
Khust	51.4±7.5	7.11±0.87	38.3±3.6	204±32	148±24
Average content	*64.4±13.0*	*6.93±1.86*	*25.8±12.5*	*185±73*	*147±22*
Lowland area					
Uzhhorod	33.4±4.6	8.86±0.97	43.1±3.9	181±28	172±26
Mukachevo	60.9±7.6	8.90±1.01	30.9±2.8	165±27	181±27
Berehove	54.2±7.5	10.4±1.03	53.8±4.7	231±33	228±29
Vynohradiv	47.1±6.6	11.1±1.04	51.0±4.5	246±35	217±29
Average content	*48.9±15.5*	*9.82±1.28*	*44.7±13.8*	*206±41*	*200±28*

Table 7. The results of macroelements determination in vegetable mix from the Transcarpathian region (n = 6; P = 0.95)

Samples of vegetable mix (district)	Macroelement content, mg·kg^{-1}			Ca/Mg
	P	Ca	Mg	
Mountain area				
Rakhiv	334±17	118±6	151±8	0.78
Mizhhirya	359±18	144±8	142±8	1.01
Volovets	349±17	157±8	179±9	0.88
Average content	*347±13*	*140±22*	*157±22*	*0.89±0.12*
Foothill area				
Svaliava	401±19	192±10	218±11	0.88
Tiachiv	398±19	176±9	213±11	0.83
Velykyi Bereznyi	355±18	168±9	204±11	0.82
Perechyn	380±19	184±9	185±9	0.99
Irshava	413±19	201±10	231±11	0.87
Khust	393±19	219±11	255±12	0.86
Average content	*390±35*	*190±29*	*218±37*	*0.88±0.11*
Lowland area				
Uzhhorod	409±19	241±12	273±13	0.88
Mukachevo	422±20	203±11	266±13	0.76
Berehove	431±21	253±12	311±14	0.81
Vynohradiv	438±20	232±11	307±14	0.76
Average content	*425±16*	*232±29*	*289±23*	*0.80±0.08*

In general, milk can be considered a microelement-rich food product. Based on the daily microelement intake, it is possible to calculate to what extent consumption of traditional food products satisfies human need for micronutrients (Goldhaber, 2003; Prashanth *et al.*, 2015; Santos *et al.*, 2004). A person's need for essential trace elements varies between 18-50 mg per day (Mertz, 1981). The degree of digestibility of microelements from food should also be considered. For example, when consuming milk in generally accepted amounts, a resident of the Transcarpathian region satisfies the need for As: in mountainous areas – by 116% (calculation according to the maximum recommended amount), in foothill areas – by 100%, in lowland areas – by 100% (calculation according to the minimum recommended amount). The recommended amount for arsenic is 15-25 mg per day (Uthus and Seaborn, 1996). For iodine (the recommended amount is 150 mg per day), a different situation is observed. For instance, a resident of the mountainous area of the Transcarpathian region consuming milk in generally accepted quantities satisfies the need for iodine by only 36%, while a resident of lowlands – by 91.5%. This should be considered when developing diets.

The results of microelements' determination in vegetable mix samples are presented in Tables 5-7. Chemical composition of the vegetable mix (Tables 5-7) significantly differs from the microelement composition of milk (Tables 1-3).

Vegetable mix is richer in Fe, Cu, Mn, Mo, P, As, and F than milk, but milk contains larger amounts of Co, Se, I, Br, and Ca. In particular, the mean contents of iron in vegetable mix is 5.64 $mg \cdot kg^{-1}$ versus 2.62 $mg \cdot L^{-1}$ in milk; copper – 0.88 $mg \cdot kg^{-1}$ versus 0.28 $mg \cdot L^{-1}$ in milk; manganese – 2.22 $mg \cdot kg^{-1}$ versus 0.15 $mg \cdot L^{-1}$ in milk; molybdenum – 0.079 $mg \cdot kg^{-1}$ versus 0.046 $mg \cdot L^{-1}$ in milk; phosphorus – 387 $mg \cdot kg^{-1}$ versus 143 $mg \cdot L^{-1}$ in milk, and fluorine – 151 $\mu g \cdot kg^{-1}$ versus 72.1 $\mu g \cdot L^{-1}$ in milk. Herewith, milk contains significantly larger amounts of cobalt (0.054 $mg \cdot L^{-1}$ versus 0.034 $mg \cdot kg^{-1}$ in vegetable mix); selenium (42.8 $\mu g \cdot L^{-1}$ versus 6.99 $\mu g \cdot kg^{-1}$ in vegetable mix); iodine (102 $\mu g \cdot L^{-1}$ versus 25.9 $\mu g \cdot kg^{-1}$ in vegetable mix); bromine (559 $\mu g \cdot L^{-1}$ versus 168 $\mu g \cdot kg^{-1}$ in vegetable mix), and calcium (2229 $mg \cdot L^{-1}$ versus 187 $mg \cdot kg^{-1}$ in vegetable mix). Given that milk and vegetables constitute the basis of the human diet (about 68%), milk can be considered the main source of Ca, I, Se, and Co while vegetables are the main source of Fe, Cu, P, Mn, Mo, As, and F.

The chemical composition of vegetable mix also demonstrates relative stability. Like in milk, microelement content of vegetable mix samples from different landscape zones is significantly different. A vegetable mix from lowlands tends to be richer in trace elements (except for As and Mn) than a vegetable mix from foothills and mountains, which was estimated using the Spearman coefficient with the following values (lowland area > foothill area > mountainous area): Fe: 0.91; Cu: 0.85; Mn: -0.94; Zn: 0.79; Mo: 0.90; Co: 0.92; Ca: 0.94; Mg: 0.94; P: 0.80; As: -0.91; Se: 0.84; I: 0.91; Br: 0.73; F: 0.88. Thus, vegetables from the lowlands of the Transcarpathian region are a more complete source of trace elements for people.

Unlike milk, for which Ca/Mg ratio is 8.1–11.5, for vegetable mix Ca and Mg content is comparable and Ca/Mg ratio varies from 0.76 to 1.01 (Figure 6). For vegetable mix, as well as for milk samples, pronounced correlations of inter-element composition are also observed, which was estimated using the Pearson coefficient (Table 8). The most pronounced are the correlations for such pairs of chemical elements (values of the Pearson coefficient): Fe:Zn – 0.90; Fe:Co – 0.91; Fe:Ca – 0.92; Fe:Mg – 0.96; Fe:P – 0.90; Fe:Se – 0.93; Fe:I – 0.92; Fe:F – 0.91; Cu:Zn – 0.93; Zn:Co – 0.90; Zn:P – 0.94; Zn:Se – 0.91; Zn:F – 0.91; Mo:Ca – 0.90; Co:Mg – 0.92; Co:P – 0.92; Co:Se – 0.90; Co:F – 0.90; Ca:Mg – 0.94; Ca:Se – 0.90; Ca:I – 0.93; Mg:P – 0.90; Mg:Se – 0.94; Mg:I – 0.96; P:Se – 0.97; P:I – 0.90; Se:I – 0.93; Se:F

– 0.90; I:F – 0.91. For clarity of the revealed correlations of inter-element composition of vegetable mix, Figure 7 presents the respective correlation diagram.

Table 8. The values of Pearson coefficient for consistent patterns of inter-element composition of vegetable mix from the Transcarpathian region (σ < 0.05)

-	Fe	Cu	Mn	Zn	Mo	Co	Ca	Mg	P	As	Se	I	Br	F
Fe	X	0.87	-0.90	0.90	0.82	0.91	0.92	0.96	0.90	-0.77	0.93	0.92	0.75	0.91
Cu	0.87	X	-0.80	0.93	0.70	0.89	0.80	0.88	0.85	-0.60	0.85	0.81	0.63	0.85
Mn	-0.90	-0.80	X	-0.82	-0.85	-0.87	-0.93	-0.92	-0.83	0.82	-0.81	-0.87	-0.62	-0.81
Zn	0.90	0.93	-0.82	X	0.73	0.90	0.86	0.89	0.94	-0.70	0.91	0.87	0.70	0.91
Mo	0.82	0.70	-0.85	0.73	X	0.71	0.90	0.87	0.79	-0.94	0.84	0.85	0.57	0.71
Co	0.91	0.89	-0.87	0.90	0.71	X	0.87	0.92	0.92	-0.72	0.90	0.89	0.75	0.90
Ca	0.92	0.80	-0.93	0.86	0.90	0.87	X	0.94	0.89	-0.91	0.90	0.93	0.72	0.83
Mg	0.96	0.88	-0.92	0.89	0.87	0.92	0.94	X	0.90	-0.85	0.94	0.96	0.74	0.88
P	0.90	0.85	-0.83	0.94	0.79	0.92	0.89	0.90	X	-0.79	0.97	0.90	0.80	0.88
As	-0.77	-0.60	0.82	-0.70	-0.94	-0.72	-0.91	-0.85	-0.79	X	-0.84	-0.88	-0.69	-0.72
Se	0.93	0.85	-0.81	0.91	0.84	0.90	0.90	0.94	0.97	-0.84	X	0.93	0.83	0.90
I	0.92	0.81	-0.87	0.87	0.85	0.89	0.93	0.96	0.90	-0.88	0.93	X	0.86	0.91
Br	0.75	0.63	-0.62	0.70	0.57	0.75	0.72	0.74	0.80	-0.69	0.83	0.86	X	0.82
F	0.91	0.85	-0.81	0.91	0.71	0.90	0.83	0.88	0.88	-0.72	0.90	0.91	0.82	X

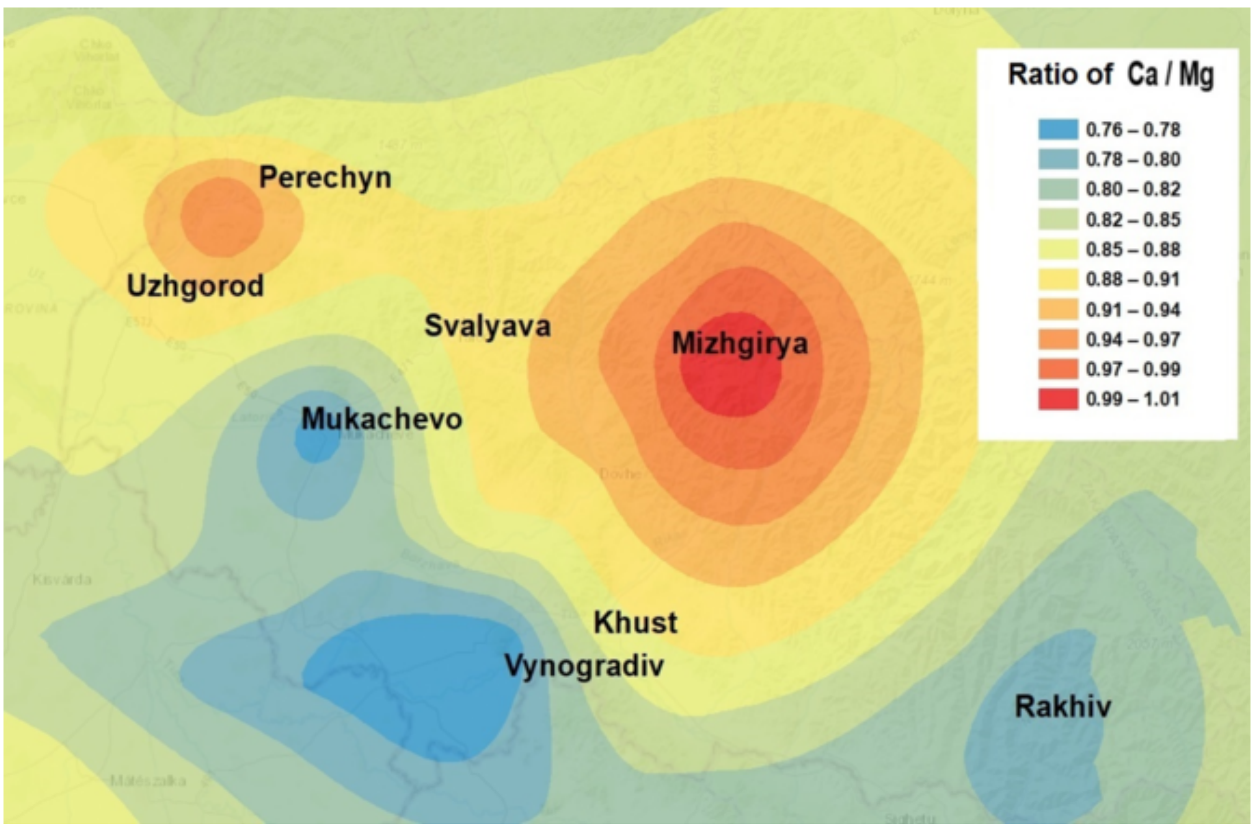

Figure 6. The map of the Transcarpathian region by the ratio of Ca/Mg in vegetable mix

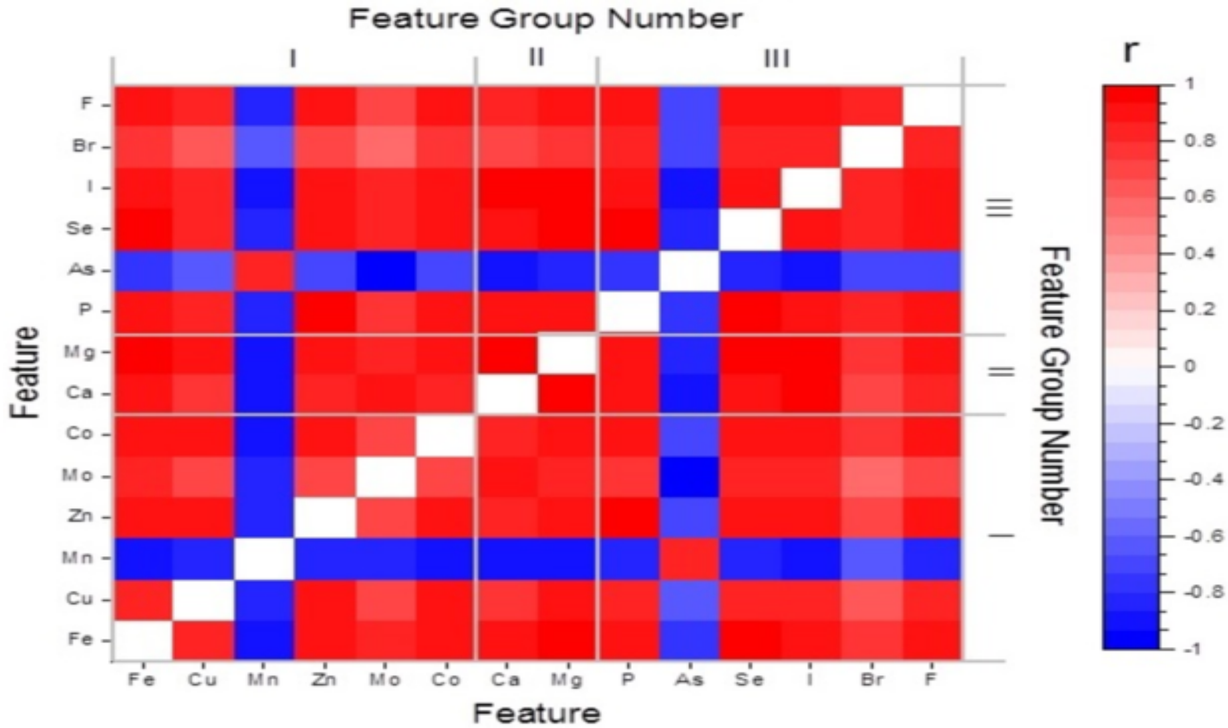

Figure 7. Correlation diagram of inter-element correlation of microelement content in vegetable mix samples (Pearson's correlation coefficient): I – trace elements metals; II – macro elements; III – trace elements non-metals

The content of toxic elements in vegetable mix is low (Hg to 0.0031 mg·kg^{-1}; Pb to 0.0214 mg·kg^{-1}) but is higher than in milk samples.

In general, milk and vegetables from the Transcarpathian region are rich in microelements (with the exception of I, F, and Se) and some macroelements (Ca and Mg), and they contain a low amount of toxic components (Hg and Pb). Therefore, traditional food products play an important role in providing the human body with trace elements and maintaining population health.

4. Conclusions

The microelement composition (based on 16 chemical elements) of traditional food products (milk and vegetable mix) from the Transcarpathian region was compared and it was proven that milk can be considered the main source of Ca, I, Se, and Co for human body while vegetables are the main source of Fe, Cu, P, Mn, Mo, As, and F. A significant difference was found in the microelement composition of milk and vegetables from different landscape zones; food products from the lowland area are richer in Fe, Cu, Zn, Mo, Co, P, Se, I, Br, F, Ca, and Mg while food products from the mountainous area have large amounts of As and Mn. This is obviously connected with the geochemical features of these areas. Consistent patterns of trace element distribution in food products from different landscape zones: lowland area > foothill area > mountainous area. It was found that the food products from the Transcarpathian region demonstrate relative stability of chemical composition. There is pronounced inter-element correlation of trace element content in food products, the value of Pearson coefficient for all pairs of chemical elements is $r > 0.60$. Results generalization suggests that traditional food products from the Transcarpathian region, taking into account their share in the human diet, almost completely satisfy daily human need for essential microelements (except for I, Se, and F).

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