

DETERMINATION OF HONEY GEOGRAPHIC ORIGIN ACCORDING TO ITS ELEMENTAL COMPOSITION BY THE METHOD OF X-RAY FLUORESCENCE

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Abstract

The aim of the research was to determine features of the elemental composition of polyfloral honey from the Odessa region (Ukraine) by the method of x-ray fluorescence for using these features in the geographic marking of the region of honey origin. A choice of honey from the Odessa region is explained by its relatively good ecology, optimal maritime climate and enough motley grass resources for gathering nectar by bees. At the same time the Odessa region occupies the fourth place among regions – honey producers in Ukraine with a right to export honey beyond the country with specific weight 10 % from the total export of this product.

11 samples of fresh honey and 14 samples of honey, stored during one year were studied as to the content of 22 mineral elements. The elemental analysis of honey was realized on the energy-dispersive x-ray fluorescent spectrometer ElvaX Light SDD. Each sample was studied for 5 times. The obtained results were statistically processed by the standard methodology. The determination distinctness of mass shares of metals is no worse than 0,1 %. Limits of finding admixtures of heavy metals in the light matrix are no worse than 1 ppm. The studies were realized during 2016–2017.

It has been established, that lyophilization of honey doesn't essentially influence determination parameters of the elemental composition of honey by the method of x-ray fluorescence. The analysis of energy-dispersive spectrums of mineral elements determined that marker elements for honey from the Odessa region are Cl, K, Ca, that prevail among other studied mineral elements. There has been established the typical intensity of a signal of energy-dispersive spectrums for Cl, K, Ca of honey from the Odessa region that correspond to the following values: for fresh honey Cl from 27075 to 29429; K from 47 296 to 41 546; Ca from 7572 to 6928; for honey, stored during one year: Cl from 40383 to 37044; K from 43 589 to 42 591; Ca from 15495 to 10006. These parameters

may serve as markers for honey from the Odessa region. At the same time the obtained results may be used for controlling the quality of natural honey by its element composition for identifying a geographic place of origin.

Keywords: polyfloral honey, elemental analysis, energy-dispersion, X-ray fluorescent spectrometer, Cl, K, Ca.

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1. Introduction

Bee honey is a product with unique food and healing properties, so its consumption in the world continuously grows. At the same time honey production is one of the most important branches of agriculture of the USA, some EU countries, Africa, Asia and Ukraine [1]. This natural product is very popular among persons of elder age and among children because of its consumption properties. It is well-known, that due to the essential amount of mineral substances, vitamins, simple sugars, organic acids, antioxidants and enzymes, honey is considered as a good product with nutritive, healing and cosmetic properties [2].

Different types of honey contain near 300 diverse components, 100 of them are always present in each type. Main components of honey are carbohydrates, water, proteins, free amino acids, enzymes, vitamins, mineral substances. It is commonly known, that the honey composition is influenced by the region, where it was produced. That is why honey from certain regions may have better properties, comparing with other honey types, so higher demand and price [3–5].

The resolution of the European Parliament presents a list of products, subjected to the highest risk of fraud in the food sphere [6]. Honey is included in this list. One of fraud types is declaration of the botanical or geographic origin of it. European consumers worry about a possibility of falsification, especially if talk about more expensive types of honey. At producing honey with indicating a place of its origin, there may take place its falsification by mixing it with one from different regions [7, 8].

Beekkeepers of many countries have patented specific markers of honey that allow to determine its origin place and to confirm its specific signs. It allows to honey protect from a certain place from such falsifications as mixing with other honey types [9].

A traditional determination of geographic and/or botanical origin of honey is a study of pollen of bee plants [10]. But this method is rather laborious and insufficiently reliable, especially for polyfloral honey, because pollen of bee plants may be added in honey artificially. In this connection scientists offer different marker combinations for identifying a geographic origin of honey: content of carbohydrates, proteins, furfural hydroxyl oxymethyl, and also definitions of the complex of physical and chemical quality parameters of honey [10, 11].

Most scientists think that the mineral content of honey may be a marker sign of its origin that is confirmed by the following data of scientific publications. Mineral substances of honey are presented by 37 macro- and microelements that have importance for its food value. The ratio of correspondent elements in honey depends on soil, where bee plants grow and from the environment. The content of mineral substances in honey varies from 0,006 to 3,45 %. Light floral honey types contain near 0,2–0,3 % of these elements, dark floral from 0,5 to 0,6 %. Honey includes 13 elements, most spread in the earth's crust (B, Si, Al, Fe, Cu, Na, K, Mg, H, Ca, P, S, Mn). It is universally recognized, that all honey types contain calcium, magnesium, manganese, phosphorus, silicon, cooper and nickel. Especially much K, P, Ca are in honey. At the same time Tl, Ag, As, Ba, Bi, Cd, Co, Cr, Ge, Hg, Ni, Pd, Sb, Sn, V, Zn, W were found in honey from different countries [10–13].

Chinese researchers determined using mass-spectrometry with the inductively-connected plasma (ICP-MS) that most spread minerals in honey from different regions of China are K, Ca, Na and Mg. So they state that the mineral composition of honey depends on its region of origin [10]. The same method revealed that most spread macroelements in honey from the Southern and Eastern districts of Turkey were K, Na and Ca, and most spread microelements – Zn and Cu [13]. Using another method (spectral x-ray fluorescent spectroscopy), it was determined, that K and Ca are observed most often in honey samples from four regions of the Rio-de-Janeiro state (Brazil) [11].

The study of scientists from different countries demonstrated that the quality and quantity ratio of mineral substance differs in honey of different regions that may be a reliable marker of the geographic origin of honey.

The problem of Ukraine is that honey is exported in big parties, but packed and marked in Europe, so it is written on a label not “Ukrainian honey”, but “honey, produced in EU countries”. Consumers are ready to pay more for specific attractive features of honey, signed on a mark, for example, indication of an origin place that is ecologically safe. At the process of marking honey, its name may be added by information about its origin place. Ukrainian honey may have indications of a place of geographic origin on a mark, for example “Ukrainian honey” or an origin region – “Odessa honey”, if producers confirm its specific markers and methods of their identification. But unfortunately there is no such marking for Ukrainian honey for today. It is connected with a fact that honey is a multi-component product, and it is difficult to determine specific features of its composition, inherent to a certain location.

There are no studies in Ukraine about regional peculiarities of the content of micro- and macroelements and their ratio in honey as marker parameters for determining its geographic origin that is an important marketing factor for determining its consumption price, including one at the world market. The topicality of the research theme is determined also by the fact that together with a function of individualization of unique honey as a commodity, produced at the territory of a certain geographic object, geographic marking of an origin place plays an important role of legal protection of its unique properties and also informs a consumer about their uniqueness.

The aim of this research was to determine peculiarities in the elemental composition of natural polyfloral bee honey from the Odessa region (Ukraine) that differs from honey from other regions. The method of x-ray fluorescence was used for it. Fixed features of the elemental composition of Odessa honey may be used for identifying this region of honey origin at international trade.

2. Materials and methods

Samples of polyfloral honey from the Odessa region (Ukraine) were used in the study for determining its elemental composition by the method of x-ray fluorescence. This method advantageously differs by its speed and distinctness comparing with other ones, and at the same time it allows to obtain results by most chemical elements. The elemental analysis of honey was realized on the energy-dispersive x-ray fluorescent spectrometer ElvaX Light SDD (LTD „ELVATECH”, Ukraine).

There were studied 11 samples of fresh honey and 14 ones of honey, stored during one year. This storage term was chosen, because it is universally recognized, that honey keeps its properties during a year at optimal storage conditions, and after year some of them may worsen, for example, organoleptic, bactericidal ones and enzyme composition. The Odessa region was chosen for the study, because this region of Ukraine with a little number of industrial enterprises – polluters of the external environment, and most its territory is occupied by resort zones. The region has a warm, soft, maritime climate and many territories with gardens of fruit trees and meadows with motley grass that are valuable resources of bee plants. For example, according to the ecological rating of Ukrainian regions, made by the Ukrainian organization “Green fund”, emissions of polluting substances in the atmosphere in 2017 in the Odessa region were 0,9 t/km², whereas in such regions as Kharkiv – 1,4 t/ km², Lugansk – 2,8 t/ km², Cherkasy – 2,3 t/km², Lviv – 5,0 t/km² [14]. In the Odessa region there are many cultured and wild plants that provide honey, unique by its taste qualities. The most popular honey sort of this region is meadow one, gathered by bees from motley grass. The Odessa region occupies the fourth place among regions – honey producers in Ukraine with a right to export honey beyond the country with specific weight 10 % from the total export of this product.

2. 1. Experimental procedures

At first there was carried out the comparative study of the elemental composition of honey from the Odessa region by x-ray fluorescent spectrometer in native (natural polyfloral honey) and lyophilized samples after their sublimation. Honey samples were placed on the support of the en-

ergy-dispersive x-ray fluorescent spectrometer. The elemental composition of “light” elements and “heavy” ones was determined according to the regimes, cited below.

The studies were conducted as follows: a batch of a honey sample with mass 10 g was studied using the energy-dispersive x-ray fluorescent spectrometer with an x-ray tube and rhodium anode. At studying the first group of “light” elements (from Na to Sc), the tension on the tube was 10 kW without a filter of primary x-ray radiation. For the analysis of the group of “heavy” elements (from Ti) there was used the tension of the x-ray tube as 40 kW and the aluminium filter of primary x-ray radiation with width 800 mcm. The current force was chosen automatically for achieving load level 50000 impulses/s. Each sample was studied 5 times. The obtained results were statistically processed using Statistica software (version 8) for determining parametric indicators, namely determining mean values (M) and error of mean (m) [15]. The determination distinctness of mass shares of metals is no worse than 0,1 %. Limits of finding admixtures of heavy metals in the light matrix are no worse than 1 ppm.

3. Results

The obtained results are presented in **Table 1**. The data of **Table 1** demonstrate that lyophilization almost didn't influence the intensity of a signal from the chemical elements of the sample. It has been also established, that the most intensity of a signal in the energy-dispersive spectrum of the elemental composition of honey was in such elements as Cl, K, Ca.

Table 1

Elemental composition of honey samples of the Odessa region before and after lyophil drying

No. by order	Atom number of element	Element	Signal intensity	
			Before sublimation	After sublimation
1	12	Mg	1087±11	641±6
2	13	Al	1054±12	982±8
3	14	Si	1773±9	2722±13
4	15	P	2648±7	4740±17
5	17	Cl	77568±21	82409±21
6	19	K	76392±14	75472±14
7	20	Ca	30505±11	41335±11
8	23	V	–	140±8
9	24	Cr	1631±13	1590±13
10	25	Mn	363±5	473±5
11	26	Fe	10498±15	11048±13
12	27	Co	412±7	511±11
13	28	Ni	6125±18	6370±10
14	29	Cu	2428±12	2817±9
15	30	Zn	2859±13	1183±17
16	35	Br	1681±11	1367±11
17	37	Rb	1838±14	1918±14
18	38	Sr	–	2514±12
19	39	Y	2139±17	3066±9
20	48	Cd	1111±15	1202±8
21	50	Sn	1646±11	1191±9
22	51	Sb	1293±8	–

The results of the study of polyfloral honey samples, stored during 1 year, are presented below (**Table 2**).

Table 2

Elemental composition of 14 samples of polyfloral honey from the Odessa region, produced in 2016 (after storage during 1 year)

Elements	Samples													
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
	Signal intensity													
Mg		235			623	460	391			260	234	383	350	224
Al	559	441	391	312	634	474	570	360	365	490	388	632	482	350
Si	956	600	516	1554	625	342	649	1054	414	1044	512	906	540	363
P	1138	1527	1500	1336	1325	1284	1535	1115		1863	1040	1714	1433	995
Cl	41505	46665	42285	32124	43218	35316	39930	38764	39375	48218	43290	47185	41413	25869
K	35995	54696	52396	42029	48304	31599	36556	39266	50328	49241	37460	55895	47189	29287
Ca	17458	15853	18102	11044	14746	11821	12911	18646	26543	20477	13212	17602	17153	1366
V	136	155	–	–	–	–	113	–	–	–	106	–	–	–
Cr	639	814	542	728	719	418	381	467	622	484	436	564	675	394
Mn	194	273	305	354	228	144	–	–	158	–	–	317	–	159
Fe	3554	5049	2953	4597	4446	2825	2926	2604	4016	2882	2917	3589	4120	2871
Co	260	199	–	314	304	77	–	–	181	106	178	74	–	166
Ni	2392	2984	2254	2398	2380	1810	2112	2076	2497	1907	1991	2038	2321	2086
Cu	837	–	715	–	997	703	726	681	970	642	582	861	1140	600
Zn	–	356	436	389	–	212	466	257	610	267	266	–	305	–
Ge	–	–	421	–	–	–	–	–	–	–	–	–	–	–
As	–	–	–	703	–	–	557	–	–	–	–	–	–	–
Se	–	–	–	–	–	–	–	–	815	–	–	–	–	–
Br	899	677	–	–	1671	763	1082	–	485	–	1355	–	1078	–
Rb	–	1483	–	–	513	–	943	839	2104	–	784	–	1442	–
Sr	–	–	–	–	–	–	–	–	846	–	946	–	–	808
Y	–	1137	–	1086	1264	–	1084	1180	545		1338	920	612	1235
Cd	–	–	474	–	347	–	–	641	–	–	1061	583	–	–
Sn	–	–	864	–	–	–	–	–	–	970	1011	–	1135	–
Sb	–	–	–	–	–	–	–	969	–	–	–	1095	982	1455
I	–	1588	1475	–	815	622	–	–	1309	1673	–	744	–	–
Ba	–	–	2218	–	–	–	–	–	–	–	–	–	–	–
Hg	–	–	–	–	–	–	–	–	748	–	–	–	–	–

Table 2 presents the intensity of a signal of elements, found in the honey samples, selected in 2016 and studied after storage during 1 year.

28 elements were found in the studied samples. 8 elements of 28 constantly were present in all honey samples: Al, Si, Cl, K, Ca, Cr, Fe, Ni.

At that the most signal intensity was typical for such elements as Cl, K, Ca. The rest of elements occurred in not all honey samples, but most often the following: P (13 samples); Cu (12 samples); Zn, Y (10 samples) Mn (9 samples), Br (8 samples), the other elements were present in the insufficient number of samples. The data of table 2 also testify that the elements Cl, K, Ca are

constantly present in honey of the Odessa region and generate strongest signal of x-ray radiation comparing with the other elements.

Then samples of fresh polyfloral honey were studied using the energy-dispersive x-ray fluorescent spectrometer as it was described above. The research results are presented in **Table 3**.

Table 3

Elemental composition of 11 sample of fresh polyfloral honey from the Odessa region, produced in 2017

Elements	Samples										
	1	2	3	4	5	6	7	8	9	10	11
	Signal intensity										
Mg	179	226	237	–	301	104	440	224	9	–	122
Al	479	582	347	519	437	408	422	481	508	497	544
Si	455	527	418	572	544	467	228	370	497	458	349
P	1184	–	1024	–	979	855	930	1508	–	1393	915
Cl	23952	37274	26092	31224	25038	37860	23398	37049	38135	16014	27685
K	26189	76037	28857	51351	25999	32186	18554	45529	73060	23513	55727
Ca	4470	10752	4191	6089	2753	12390	3823	9030	12419	4075	6215
V	151	–	–	–	–	–	–	–	–	–	89
Cr	514	610	696	824	462	729	585	421	542	558	708
Mn	197	–	–	–	129	–	189	294	211	303	139
Fe	3839	4011	4255	5019	2976	4396	3524	3292	3699	3359	4261
Co	103	243	161	313	–	99	141	169	375	193	145
Ni	–	2146	2474	–	2147	2255	1830	2269	2608	2237	2413
Cu	755	1027	1149	1103	801	950	832	753	987	807	915
Zn	10718	2285	486	885	223	639	334	394	–	12096	–
Ga	–	377	–	–	–	–	–	–	–	–	–
Ge	–	620	–	–	–	–	246	–	–	–	–
As	–	–	–	–	–	442	–	–	–	–	442
Se	696	–	–	–	–	–	–	–	–	–	–
Br	–	764	–	–	–	1013	–	–	–	–	–
Rb	–	1093	–	922	–	1114	–	–	766	–	1323
Sr	1098	741	–	–	–	–	774	–	946	–	–
Y	–	1208	–	–	–	800	1239	–	1176	–	1272
Cd	–	–	380	–	–	–	–	806	–	–	638
Sn	–	1690	–	–	–	–	961	–	–	–	–
Sb	–	–	–	–	–	860	–	–	–	–	1164
I	–	–	1116	–	–	773	–	1450	–	–	–
Ba	–	–	–	–	1868	–	–	–	–	2173	–
W	–	–	–	–	–	–	517	–	–	–	–

As it can be seen on table 3, the following 8 elements are constantly present in all 11 honey samples: Al, Si, Cl, K, Ca, Cr, Fe, Cu. The following elements are present in honey samples inconsistently: Co (in 10 samples) Mg, Zn and Ni (in 9 samples) P (in 8 samples) Mn (in 7 samples).

Thus, in honey, stored during 1 year and in fresh one there was registered the constant presence of such elements as: Si, Cl, K, Ca, Cr, Fe. Whereas Ni was present in all honey samples, stored during 1 year, and Cu was revealed in all samples of fresh honey. The obtained results were compared with literary data, where peculiarities of the elemental composition of honey from different countries

were indicated [10–13]. For example, such elements as K, Ca, Na and Mg prevailed in Chinese honey, in separate regions of Turkey – K, Na and Ca, in Brazil - K and Ca. The Odessa region is typical by the content of Cl ions in air that influenced the content of this element in honey. The ratio of correspondent elements depends on soil, where bee plants grow and from the external environment, so it may be a marker parameter. The content of these elements is closely connected with honey authenticity and may serve a marker parameter, typical for polyfloral honey of the Odessa region.

Thus, if honey will be supplied to the international market from the Odessa region of Ukraine as a cleaner one, it must contain the greatest amount of three cited elements, determined by the most intensity of the radiological signal. If honey will be supplied from a more ecologically polluted region, for example, Kharkiv one – it undoubtedly has much Cl and the other marker elements. At the same time bee plants of the Odessa region are more biologically active (enzymes, vitamins), because this region has more number of sunny days and softer climate. So, honey has more healing biologically active substances.

4. Conclusions

As a result of this study, it has been established that honey samples from the Odessa region constantly contain the following elements: Al, Si, Cl, K, Ca, Cr, Fe, Cu (in fresh honey) and Si, Cl, K, Ca, Cr, Fe, Ni (for honey samples, stored during 1 year).

The most intensity of x-ray signals was observed for such elements as Cl, K, Ca. Honey, stored during 1 year had more intensity of spectral lines of Cl, K, Ca comparing with fresh honey, that is explained by its dehydration at storage and relatively higher density. At the same time it has been determined, that K is the first element by the intensity level of x-ray signal, Cl – on the second place, then Ca.

Thus, at studying honey from the Odessa region by the energy-dispersive x-ray fluorescent spectrometer, it has been established, that Cl, K, Ca are the most spread elements. These three elements generated the following values of x-ray signal: for fresh honey: Cl – 27075–29429; K – 47296–41546; Ca – 7572–6928; for honey, stored during 1 year: Cl – 40383–37044; K – 43589–42591; Ca – 15495–10006.

The presented results allow to define the geographic location of honey, and the described method may be used by specialists of laboratories that determine quality parameters of food products as an expressed method of the honey quality evaluation.

An advantage of this study is its easiness and speed of conduction; its results are evaluated by the intensity of x-ray signal in spectrograms, obtained by the energy-dispersive x-ray fluorescent spectrometer. The most signals in honey of the Odessa region must correspond to such three elements as Cl, K, Ca. The obtained results will be useful for honey producers in the concrete Ukrainian region – namely in Odessa one. Using this method for determining marker parameters of the elemental composition of honey, fixed by us, it is possible to confirm the authenticity of honey from the Odessa region and to mark it correspondingly. In this connection Ukraine will receive the new brand product and a great possibility to realize it by more profitable prices at the international market. The results of these studies may serve as an idea for analogous studies and searchers for markers of honey by its elemental composition in other regions of Ukraine, for example, Transcarpathian one, where honey is also eco-safe and healing, and also for different regions of other countries.

Despite the fact that the offered method may be used in practice, it must be improved for the future to obtain quantity parameters of Cl, K, Ca in honey for more concretization of obtained data. The determination of Cl, K, Ca in honey by such indirect parameters as an x-ray signal intensity by the energy-dispersive x-ray fluorescent spectrometer must be added by the method of recalculation and obtaining of quantity parameters that need additional studies. It is a field of further studies.

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