



# Honey varieties vs metal and pesticide content – literature review and own research

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## Abstract

**Introduction and objective.** Although monofloral honeys are regarded as more valuable than multifloral types, they lack a clear uniform definition in European countries concerning the proportions of predominant pollen types. In addition, honey contains various secondary plant metabolites, enzymes and co-enzymes, which provide health-promoting properties; however, it can also accumulate heavy metals and pesticide residues.

**Review methods.** A literature review was performed using the databases PubMed, Google Scholar concerning the content of metals in the soil, flower and bee pollen in varietal honey. Literature was collected on the influence of pesticides contained in honey on their impact on the human health. Own research selected three varieties of Polish monofloral honey (linden, black locust, rapeseed), which were analyzed using a spectrometer to determine the concentration Ca, Cu, Fe, K, Mg, Mn, Na, Zn.

**Brief description of the state of knowledge.** Literature data indicate that a polluted or treated environment can contribute to the accumulation of *inter alia* heavy metals and pesticides in pollen, honey, beeswax, and the honeybee itself. Such contamination is influenced by various environmental factors, e.g. contaminants from the flower can be passed to the bee through contact with contaminated pollen and incorporated in honey. However, in the monofloral honeys analysed in this study, there were combinations of health-promoting elements that exert synergistic effects.

**Summary.** The results obtained provide new qualitative and quantitative data on the composition and potential contamination of varietal honeys over the past 10 years, a period characterised by legislative changes aimed at reducing pesticide and metal contamination of terrestrial ecosystems.

## Key words

food safety, metal and pesticide contamination, monofloral honey

## INTRODUCTION

**Rules for classification of varietal honeys.** The pollen composition of honey is determined by its botanical and geographical origin [1], and in some countries, each variety is named based on its predominant pollen component. In Poland, the variety of honey and its quality parameters are defined by the Regulation of the Ministry of Agriculture and Rural Development of 3 October 2003 on detailed requirements concerning the commercial quality of honey (*Journal of Laws* 2003, No. 181, item 1773), and the Polish Standard for the Properties of Honey (PN-88/A-77626).

The European Commission provides a legal definition for the composition of blossom and honeydew honeys, but not for monofloral honeys, even though the latter is often sold at higher prices than the multifloral varieties [2]. Hence, European countries lack uniform regulations specifying the percentage of predominant pollen in monofloral honeys; rather, each country has its own list of monofloral honeys, and specifies the percentage of pollen contained therein [3] (Tab. 1).

**Table 1.** Required percentage of total pollen count (upper limit) for individual monofloral honeys according to national legislation in the selected country

Name of pollen by legal type [3]	HR	DE	EL	IT	PL	RO	RS
	[%]						
<i>Brassica napus</i>	60	80			45		
<i>Robinia pseudoacacia</i>	20				30	30	20
<i>Tilia</i> spp.	20	20			20	30	25
<i>Arbutus unedo</i>	10						
<i>Calluna vulgaris</i>	20	45			45		20
<i>Castanea sativa</i>	85	90	87				85
<i>Citrus</i> spp.	10	20	3	10			
<i>Erica</i> spp.		45	45				
<i>Eucalyptus</i> spp.		85					
<i>Gossypium</i> spp.			3				
<i>Helianthus</i> spp.		50	20				40
<i>Lavendula</i> spp.	10						
<i>Medicago sativa</i>							30
<i>Phacelia tanacetifolia</i>	60						
<i>Rosmarinus officinalis</i>							20
<i>Salvia officinalis</i>	15						
<i>Satureja montana</i>	20						
<i>Taraxacum officinale</i>							20
<i>Thymus</i> spp.			18	15			
<i>Trifolium</i> spp.		70					
General monofloral	45	45	45				

Croatia (HR), Germany (DE), Greece (EL), Italy (IT), Poland (PL), Romania (RO), Serbia (RS)

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### Health-promoting properties of honeys with a dominant presence of specific plant pollens.

In addition to pollen, monofloral honey is rich in various secondary compounds with positive effects on health [4]. For example, honeys are believed to have antibiotic effects and to support ulcer healing and kidney function (*Brassica* spp.), to promote digestion (*Robinia* spp.), improve muscle tone (*Salix* spp.), ease blood circulation and slow atherogenesis (*Aesculus* spp., *Prunus* spp., *Salix* spp.), and support hepatoprotection and liver function (*Aesculus* spp.). They are also believed to possess calming and sedative properties (*Robinia* spp., *Tilia* spp.) and act as diuretics (*Prunus* spp.).

**Main contaminants in honey that may affect quality.** Being a local product, the composition of bee honey is influenced by local pollution by heavy metals [5] and pesticides [6]. Such contamination may cause health problems related to organ damage and an increased risk of cancer in the consumer.

Many metals have important functions in the human body, and their presence in small amounts in honey can have many benefits. However, excessive amounts, which may be present in honey from polluted areas [7], can cause hypersensitivity or even allergy. Metals can also exert an adjuvant effect on the immune system of a person sensitised to pollen allergens, resulting in enhanced immune reactivity [8].

It was assumed that:

- 1) honeys from cultivated plants are more contaminated with pesticides and metals than those from uncultivated plants;
- 2) pesticide and metal contamination of species or monofloral honeys can cause hypersensitivity, allergy, and possibly trigger or aggravate pathological conditions.

## OBJECTIVE

The aim of the study was to determine the following, on the basis of the literature review and our own research:

- 1) the state of contamination of varietal honey and monofloral honey (as described by beekeepers) with pesticides and metals;
- 2) the effects of consuming contaminated honeys on hypersensitivity and allergies, their toxic effects and potential to aggravate existing conditions;
- 3) sources of pesticide and metal contamination in pollen.

The research will result in:

- 1) sensitising consumers to information about the country of origin of honey;
- 2) drawing attention to the type of honey resulting from its pollen characteristics;
- 3) warning of rare but possible health problems for people with food allergies.

## REVIEW METHODS

**Literature review.** The review included scientific articles and relevant legislation, with most attention paid to recent literature. The nomenclature was taken from studies on the presence of pesticides and metals in honeys, although such naming is generally not regulated by legal standards. Information was collected on the content of metals in soil and bee pollen, and in various types of varietal honeys.

The following terms were combined in the search engine:

- pesticides / veterinary drugs / neonicotinoids / coumaphos / chlorfenvinphos in pollen, bee bread, honey, beeswax, honey bees; residues in varietal honeys (acacia, buckwheat, goldenrod, linden, pine, rapeseed, raspberry, rosemary, sunflower); sources of residues, hypersensitivity, pollen, honey, allergy;
- metals / heavy metals / macroelements / microelements content (Zn, Cu, Sr, Al, As, Ni, Cr, Cd, Pb, Hg, Rb, Ti) in monofloral honeys (anise, astragalus, basil, bean, buckwheat, carob, cedrus, chasteberry, chestnut, citrus, clover, cotton, cynoglossum, dandelion, dorycnium, eucalyptus, euphorbia, phacelia, ferula, fir, goldenrod, hawthorn, heather, indigobush, ivy, Jerusalem thorn, lavender, linden, lucerne, mandarin, mint, oak, onosma, orange, parsley, pine, potentilla, rapeseed, raspberry, rhododendron, rosemary, sage, sainfoin, savory, spruce, strawberry tree, sulla, sunflower, thyme, vetch, willow, yellow-thistle);
- metals / heavy metals / macroelements / microelements content (Mn, Zn, Cu, Fe, Ca, Mg, K, Na, Pb, Al, Cd, Ni, Cr) in soil with name of plants (*Betula*, *Brassica*, *Facelia*, *Fagopyrum*, *Melilotus*, *Prunus*, *Robinia*, *Tilia*);
- metals / heavy metals / macroelements / microelements content (Mn, Zn, Cu, Fe, Ca, Mg, K, Na, Pb, Cd, Cr, Al, Ni, As) in bee pollen from *Facelia*, *Malus*, *Salix*, *Taraxacum*.

### Own research – marking grains of pollens and metals in monofloral honeys.

Three varieties of honey were selected for research: black locust (*Robinia pseudoacacia* L.), linden (*Tilia* spp.) and rapeseed (*Brassica napus* L.), which came from areas in Poland with very little industrialization.

The percentage of pollen grains of individual species of plants present in honey was determined on the basis of honey pollen analysis. The honey sample weighing 10 g was dissolved in 20 ml of distilled water at 50 °C (in water bath). After centrifuging for 10 minutes at 3,000 rpm, removing the liquid from the sediment and adding water to 20 ml, the sample was centrifuged again at 3,000 rpm. With 0.1 ml of sediment, a 0.5 cm layer of water was left, and with 0.3 ml – 1 cm of water. For microscopic examination, 0.1 ml of sediment was taken.

Samples were analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, ICAP 7400 Duo, Thermo Scientific) to determine the concentration of: Ca (wavelengths 315.887 nm), Cu (224.700 nm), Fe (238.204 nm), K (766.490 nm), Mg (280.270 nm), Mn (257.610 nm), Na (589.592 nm), Zn (202.548 nm). At least 0.5 g of the samples were mineralized with 2 mL of 65% HNO<sub>3</sub> (Suprapur, Merck) and 1 mL of non-stabilized 30% H<sub>2</sub>O<sub>2</sub> solution (Suprapur, Merck), using microwave digestion system MARS 5, CEM. Samples of reference material (NIST SRM 1486 Bone Meal) were prepared in the same manner as samples.

**Statistical analysis – own research.** The research results were subjected to statistical analysis and presented in the form of descriptive characteristics. A one-way ANOVA was performed to determine the effect of 'honey variety' on the concentration of the tested metals. Pearson's linear correlations were established for the relationship between the numbers of pollen and the concentrations of these metals, regardless of the honey variety. In order to indicate the correlation between the total number of pollen and

concentration of metals, the colour gradation principle was adopted according to the coefficient of determination  $R^2$ .

## DESCRIPTION OF THE STATE OF KNOWLEDGE

**Pesticides and veterinary drugs in honey – sources of residues.** Many of the crops visited by bees are treated with pesticides and other agrochemicals. Many beekeepers also use mite repellents against varroa; these preparations are deposited in pollen, bee bread, honey, beeswax and the honey bees themselves [6]. Calatayud-Vernich et al. [9] report higher levels of chlorpyrifos and acetamiprid in apiaries located in agricultural areas, with the most contaminated product being wax.

Amulen et al. [10] indicate a direct link between the presence of certain pesticide residues in bee products and the visited crops. David et al. [11] found oilseed rape pollen to be contaminated with spiroxamine, carbendazim, neonicotinoids, thiamethoxam and clothianidin, the demethylation-inhibitor (DMI) fungicides difenoconazole and trifloxystrobin, and to a lesser extent, boscalid, pyraclostrobin and fluoxastrobin. Most of these substances, at lower concentrations, were found in the pollen of hand-picked field flowers at the edge of cultivated fields. Cappellari et al. [12] report that in semi-natural areas, pollen contamination by pesticides was minimised only at the beginning of the season, and the presence of perennial plants also increased the risk of pollen contamination by pesticide residues.

In general, the level of pesticide contamination in honey was much less than in bee pollen or bee bread, and its consumption does not generally pose a risk to humans. However, Shi et al. [13] report that neonicotinoids accumulate in honey, with higher concentrations present compared to pollen or soil.

Végh et al. [6] report that secondary contamination by pesticides and veterinary medicines can occur in the hive, and suggest that coumaphos and chlorfenvinphos may accumulate in beeswax to an extent that poses a potential health risk to honey consumers. The concentration of coumaphos in beeswax is influenced by *inter alia* the use of contaminated wax pads and possible contamination of beeswax during treatment. Also, it can be influenced by the distance between the coumaphos strip and the wax sampling location, frequency of application, and the use of coumaphos in bee colonies in the area, and other species that may possibly be contact with bees; it is also affected by the proximity of hives receiving varroa treatment to untreated hives. To avoid contamination of bee products and larvae with coumaphos, it is recommended to replace it with acaricides [14].

**Pesticides and veterinary drugs – degree of contamination in honeys.** El-Nahhal [15] assessed the presence of hazardous insecticide, acaricide, fungicide and herbicide residues in honeys from different countries and classified them into the first three toxicity classes (Ia, Ib, II and III) according to guidelines of the World Health Organization (WHO) [16]. Based on the relative average index (Rel Ave), indicating the mean concentration of contaminant residues, Polish honeys were placed in Class III for insecticides, alongside Mexico, Ethiopia and France ( $0.51 < \text{Rel Ave} < 0.80$ ), for acaricides alongside Slovenia, and for fungicides alongside Italy. No data on herbicide contamination was noted from Poland.

Considering art. 32 Regulation (WE) No. 396/2005 [17], the European Food Safety Authority (EFSA) is required to submit an annual report evaluating the levels of pesticide residues in foods on the European market. According to the 2021 data, only 119 of 1,035 honey samples (11.5%) contained detectable pesticide residues below the maximum residue limit. Overall, 28 different pesticides were detected, with the most common being thiacloprid and acetamiprid [18].

**Pesticides and veterinary drugs – residues in varietal honeys.** Although the pesticide residue content in honey is well documented [6, 15, 18], only a few countries have performed such analyses on varietal honeys [19]. In Polish honeys, studies of neonicotinoids indicated the presence of thiamethoxam in goldenrod (254.97 ng/g), rapeseed (368.26 ng/g), buckwheat (494.47 ng/g) and raspberry (652.42 ng/g), and thiacloprid in linden (348.19 ng/g) [20].

Scripcă and Amariei [19] reported that the maximum limits set by European legislation for endosulfan were exceeded in mint and rapeseed honeys, with the respective levels reaching 0.42 and 5.14 ng/g. Neonicotinoids were found in 27% of analysed honey samples. Chloramphenicol (0.2 ng/g to 0.8 ng/g) was also identified, but only in rapeseed honey. However, taking into account the lowest estimated daily intake (EDI), pine, multiflower, sunflower, acacia, lime and rapeseed honeys did not pose a threat to humans [21].

Ligor et al. [20] found that the levels of neonicotinoids (thiamethoxam, clothianidin, imidacloprid, acetamiprid and thiacloprid) did not exceed the limit of quantification (LOQ) in honey dominated by goldenrod, phacelia, lime, rosemary or buckwheat pollen. No neonicotinoids were found in sunflower honey, in two samples of rapeseed honey or two samples of acacia honey from Poland and Romania.

**Hypersensitivity to pollen in honey that may cause allergy.** Pollen analysis indicates that honey can often contain anemophilous plant pollen, which can cause allergy symptoms [22]. Food allergies to honey are most often caused by *Asteraceae* pollen, but also by grass and tree pollen allergens. Patients with pollinosis or food allergy, and those allergic to bee venom, may experience symptoms when consuming honey which has been powdered with pollen from wind-pollinated plants during the pollen season, resulting in double exposure [23, 24].

**Legal regulations concerning the assessment of pesticide and veterinary drug residues in food, including honey.** It is important to monitor honey for contaminants to ensure the health of the population, particularly children and the elderly. It is assumed that pesticides placed on the market should not exert harmful effects on humans [25].

In 2020, the European Food Safety Authority published two pilot studies [26, 27] on the risks posed by various pesticide residues in food. The studies examined the possibility of acute effects on the nervous system and the possibility of chronic thyroid problems, such as hypothyroidism, hyperplasia and tumours, in four populations of adults, three populations of children (3–9 years) and three populations of toddlers (1–3 years). It was found that cumulative exposure did not exceed the regulatory threshold for pesticides on the nervous system for most populations, with some exceptions for children and toddlers. Cumulative assessment groups (CAG) were established for erythrocyte acetylcholinesterase inhibition in

brain tissue and/or erythrocytes; such exposure was mainly associated with chlorpyrifos, triazophos and omethoate [26]. The regulatory threshold for chronic thyroid problems was not exceeded in any population group [27]. Coumaphos does not pose a health risk to the consumer based on a small daily intake of honey, and has not been used to protect crops since 2009 [28].

In organic beekeeping, the use of allopathic, chemically-synthesised medicinal products for prophylactic purposes is prohibited. In addition, the plant sources of pollen must be at least 3 km from any source of pollution or production outside agriculture [29]. Panseri et al. [30] propose a strategy to select unpolluted areas for organic honey production.

**Pesticides and veterinary drugs – preventing contamination of honeys by their residues.** Bees should be provided with potentially uncontaminated resources to ensure honey quality [12]. In the case of varietal honeys derived from arable crops, beekeepers should allow time for pesticide levels to fall after spraying before introducing bees to an agricultural area.

Honeybees are protected as livestock by legislation and the norms of beekeeping [31]. Therefore, beekeepers are obliged to comply with various regulations regarding their activities, the import, transit and sale of animals, and the use of veterinary medicinal products and feeds.

The quality of resources for honey bees can be improved by replacing chemical methods of insect and weed control with physical and biological methods, as well as with integrated pest management techniques, and by reducing pesticide use within the honey bees' flight range (i.e. crop and grassland areas). Contamination has been found to be significantly reduced by selecting advanced pesticide spraying techniques for specific plant species, eliminating pesticides with systemic effects and restricting honey bee movement for at least 48–72 hours after pesticide application [15]. Within the hive, there may be movement of pesticides, e.g. coumaphos, from the contaminated wax to the honey [6]. Reducing the level of contamination in the wax can reduce the potential damage to bee colonies and products [14].

**Metals in honey – sources of residues.** Honey is regarded as a natural product and should contain an elemental composition that promotes health. However, the quality of pollen or nectar in the honey is influenced by the state of the environment (quality of air, water, soils) and the use of agricultural techniques, such as fertilisers and plant protection products [20].

Honey may contain metals from industrial activities, coal burning and municipal waste; it can also accumulate from road transport via exhaust, car tyres or brake pads, and from street surfaces [32]. Metal contamination is also influenced by the degree of soil contamination, climatic conditions and air quality; the presence of contaminants on the plant (flower), contact of the bee with contaminated pollen, and the conditions for honey production in the hive.

Literature data indicate the presence of low concentrations of heavy metals in pollen (Tab. 2). Heavy metals accumulate in agricultural soils through prior application of fertilizers and pesticides (As, Cd, Cu, Pb), the current introduction of mineral multi-component fertilizers or sewage sludge, and from the presence of dust in the atmosphere from point sources, especially near mining activities. In addition to these traditional sources, metal-containing nanomaterials enter

**Table 2.** Metal content in bee pollen [mean, mg/kg]

Elements	Pollen			
	<i>Facelia</i> spp. [34]	<i>Malus</i> spp. [35]	<i>Salix</i> spp. [36]	<i>Taraxacum</i> spp. [35]
Al	2.42		53.67	
Ca	1067.00		921.00	
Cd	0.05	0.04	0.10	0.08
Cr	< 0.20	2.10	0.47	5.33
Cu	9.03	1.99	5.78	4.90
Fe	56.21	18.94	100.39	40.75
K	6239.00		3616.00	
Mg	553.00		747.00	
Mn	21.90		31.94	
Na	18.02		27.51	
Ni		0.20	1.51	0.66
Pb	0.10	0.69		1.82
Zn	55.01	22.72	57.50	52.92

the soil as active ingredients or preservatives in biosolids or pesticides and fertilisers [33].

The metal content of the soil in the plot dominated by the different species of melliferous plants varies greatly (Tab. 3). Praus et al. [37] report that the levels of lead ( $0.046\text{--}0.140\ \mu\text{g g}^{-1}$ ) and nickel ( $0.12\text{--}4.30\ \mu\text{g g}^{-1}$ ) in bees demonstrated a significant linear correlation with (bio)available Pb ( $0.012\text{--}0.254\ \mu\text{g g}^{-1}$ ) and pseudo-comprehensive Ni ( $17.1\text{--}36.4\ \mu\text{g g}^{-1}$ ) in soil. However, no significant Spearman's correlation was noted between the different heavy metals analysed in soil and *Brassica napus* inflorescences, except between Pb and Fe, and between Pb and Cu [38].

Higher levels of environmental pollution are associated with greater mercury accumulation in the soil, which is then taken up with water and minerals by plants, and then passed to honey via the bees. The mean mercury content determined in honeys from Poland did not exceed the permissible standards (max. value in honey is  $10\ \mu\text{g/kg}$ , UE Directive 2018/73) [51].

**Metals in honey – contamination of honeys.** Although the quality of honey is influenced by its region of origin, a greater influence is held by the chemical composition of the soil. Pietrelli et al. [52] report that the concentrations of metals in bee forage are proportional to their concentration in the soil.

Metals are taken up by plants, and then by the bee with pollen and nectar. In a polluted environment, the presence of heavy metals can deteriorate the quality of the honey. Honeys from apiaries located close to main traffic routes will be characterised by higher elemental concentrations [53]. Tomczyk et al. [54] report higher concentrations of heavy metals in bees than in their honey. Atanasov [38] reports that pollen contains higher levels of *inter alia* lead, cadmium and copper, compared to bee bread, and propose that this is due to it being a generative cell.

**Metals in honey – residues in varietal honeys.** The levels of toxic metals in varietal honeys are generally low, with the exception of some individual types, e.g. buckwheat (Ti), lime (Hg), acacia (Co and Al) [5, 55, 56, 57] (Tab. 4). The honeys with names assigned by beekeepers demonstrate

**Table 3.** Metal content in soil on the surface dominated by individual plant species [range mg/kg]

Element	Species							
	<i>Betula</i> spp. [39]	<i>Facelia</i> spp. [40]	<i>Fagopyrum</i> spp. [41]	<i>Melilotus</i> spp. [42, 43, 44]	<i>Prunus</i> spp. [45]	<i>Brassica</i> spp. [38, 46, 47]	<i>Robinia</i> spp. [48, 49]	<i>Tilia</i> spp. [50]
Ca	731–17050	-	-	4620–176600	-	-	3613–5396	1010–71370
Cd	0.24–31.40	-	0.10–0.43	0.02–9.50	-	0.24–31.40	0.20–3.91	0.01–1.92
Cu	1.20–8.00	-	-	0.25–9.87	97.17–102.56	0.02–0.42	9.33–50.29	0.01–35.86
Fe	72.00–191.00	-	-	0.48–17.00	-	0.20–3.67	21355–30842	2.03–625.80
K	45–101	9	-	52–4890	413–445	42–55.18	86–1331	20–4050
Mg	46–113	3	-	110–4250	-	7	54–5543	340–15710
Mn	3.7–20.0	-	-	8.8–321.6	-	-	485.4–765.9	2.3–454.1
Na	25–220	-	-	10–800	-	-	471–795	10–201
Ni	-	-	-	0.35–18.82	21.52–23.41	-	27–50.83	-
Pb	24.00–48.00	-	5.49–20.98	0.35–264.40	22.00–24.00	0.11–0.70	12.57–157.99	-
Zn	7.80–31.00	-	20.00–80.00	1.10–375.80	73.34–75.45	0.03–1430.70	77.60–543.97	1.36–286.70

similar values for heavy metal concentrations, although high concentrations were noted in chestnut (for Ti) and in fir (for Rb) [57, 58].

Metals essential to physiological function, such as Zn or Cu, are also present in honey; however, excessive amounts can lead to harmful effects. High levels of Cu and Al were found in acacia honey, Zn was in rapeseed honey and Rb in lime honey (Tab. 4).

**Metals in honey – regulating the assessment of residue levels.** The quality requirements of honey are specified in national and international standards. The most important of these are the World Standard, approved in 2001 by the Codex Alimentarius Commission (Codex Alimentarius: Draft revised standard for honey 2001) and EU Directive 2001/110 (Council Directive 2001/110/EC relating to honey, 2002). The requirements for Polish honey are specified in

**Table 4.** Metal content in monofloral honeys and honeys named by beekeepers

Name of honey	Metals [range mg/kg]													References
	Zn	Cu	Sr	Al	As	Ni	Cr	Cd	Pb	Co	Hg	Rb	Ti	
<b>by legal type</b>														
rapeseed	0.32– 6.96	0.05– 2.76	0.03– 0.07	0.30– 4.56	LOD– 0.01	0.02– 0.90	LOD– 0.03	LOD– 0.09	LOD– 0.60	0.01– 0.07	0.02– 0.21	0.15	LOD– 0.21	5, 19, 54, 59–71
linden	0.14– 6.93	0.14– 1.56	0.05– 0.30	0.14– 3.41	LOD– 0.02	LOD– 0.72	LOD– 1.21	LOD– 0.72	LOD– 0.86	LOD– 0.07	0.01– 1.25	0.15– 8.60	0.00– 0.23	5, 56–61, 63, 64, 66–82
acacia	0.30– 8.58	LOD– 5.16	0.06– 0.26	0.08– 11.90	LOD– 0.01	LOD– 1.82	LOD– 0.72	LOD– 0.03	LOD– 1.15	LOD– 0.50	0.01– 0.89	0.38– 0.80	0.00– 0.12	19, 49, 53, 55–63, 65–70, 73, 75–81, 83–88
buckwheat	0.48– 9.30	0.23– 4.87	0.04– 0.12	0.20– 0.36		LOD– 0.43	0.03	LOD– 0.24	0.01– 1.78	LOD		0.80	0.38	5, 19, 61, 63, 64, 71, 78, 82, 89
chestnut	0.34– 5.74	0.05– 1.00	0.00– 1.32	0.21– 3.47	0.00– 0.87	0.00– 0.74	0.00– 0.84	0.00– 0.84	0.00– 0.01	0.01– 0.60	0.01– 0.50	0.00– 0.29	8.99– 23.00	55–58, 65, 68, 72, 73, 75, 79, 84, 85, 88, 90–92
citrus	0.32– 2.02	0.03– 0.50		1.87	0.10	0.08– 0.65	0.02– 0.92	0.00– 0.01	0.01– 0.04	0.50	0.01			55, 56, 79, 90
clover	0.38– 0.54	0.05– 0.32	0.04	0.22	0.01	0.02– 1.20	LOD– 0.02	0.02– 0.09	0.08– 0.43	LOD– 0.00		1.60	0.02– 0.09	63, 64, 93
dandelion	0.59– 2.97	0.25– 9.67		1.82– 2.84		0.13– 0.42		0.01– 0.09	0.01– 0.09				0.06– 0.15	5, 19, 54, 71
eucalyptus	0.09– 0.62	0.03– 0.21		5.19	0.10	0.13– 0.54	0.01– 0.70	0.00– 0.01	0.01– 0.09	0.50	0.00			55, 56, 79, 90
facelia	0.34– 3.01	0.00– 0.62	0.04	0.40– 3.02	LOD– 0.07	LOD– 0.04	LOD– 0.05	LOD	LOD– 0.40	LOD		0.48		59, 64, 66, 68
heather	0.52– 3.80	0.04– 0.80	0.04– 0.30	0.43– 2.01	0.00– 1.66	LOD– 5.59	LOD– 18.80	0.00– 0.98	0.00– 8.72	LOD– 0.07		0.81– 7.70	0.12	63, 64, 72, 80, 85, 94, 95
lavender	0.80– 1.69	0.18– 0.24	0.15– 0.21	0.06	0.01– 0.08	0.04– 0.07	0.01– 0.13	0.01– 0.28	0.02– 0.84	LOD– 0.02		0.62– 0.77		58, 72, 84, 85
lucerne	0.30	0.15						0.02	0.22					63
rosemary	0.10			2.30	0.29	0.08– 2.23	0.69	0.07	0.76					58, 94
sunflower	0.39– 6.88	LOD– 4.63	0.03– 0.35	0.17– 13.56	LOD– 0.15	LOD– 0.18	LOD– 0.11	LOD– 0.13	LOD– 0.44	0.01– 0.37	0.00– 0.73	1.10	0.00	19, 59, 60, 63, 65–69, 73, 76–79, 91, 96, 97

**Table 4.** Metal content in monofloral honeys and honeys named by beekeepers (continuation)

Name of honey	Metals [range mg/kg]												References	
	Zn	Cu	Sr	Al	As	Ni	Cr	Cd	Pb	Co	Hg	Rb		Ti
thyme	0.56– 1.15	0.11– 0.43		0.61– 2.82	0.00– 0.69	LOD– 0.85	0.01– 0.04	LOD– 0.00	LOD– 0.31	LOD– 0.01	0.05– 0.38		0.02	65, 79, 98–100
<b>by beekeepers description</b>														
anise	0.84	0.29				0.53	0.02	0.00	0.08					79
astragalus	0.75– 2.37	0.02– 0.36	0.17– 0.32			0.03– 0.41	0.00	0.00– 0.04	0.00– 0.04	0.02– 0.03		0.39– 0.41		72, 79, 85
basil	2.00	0.29												60
bean	0.31	0.11				0.21	0.00	LOD– 0.02	0.00– 0.20					71, 79
carob	1.01	0.42	0.29			0.05	0.02	0.01	0.01	0.02		1.46		72
cedrus	0.10	0.04				0.08	0.00	0.00	0.00					79
chasteberry	0.05– 0.44	0.01– 0.20	0.12	LOD	LOD	0.02– 0.07	0.00– 0.10	0.00– 0.02	0.00– 2.79	LOD				79, 84
cotton	0.53– 2.37	0.03– 2.25				0.16		0.00	0.02					79, 97
cynoglossum	0.39	0.10			0.01	0.44	0.01	0.01	0.35	0.00				93
dorycnium	0.68	0.23			0.01	0.62	0.01	0.01	0.08	0.00				93
euphorbia	0.50	0.03				0.15	0.01	0.00	0.00					79
ferula	0.62	0.17				0.25	0.00	0.00	0.00					79
fir	1.04– 1.46	0.77– 2.10	0.05	3.28– 27.35	0.00– 0.39	0.01– 0.56	0.01– 0.02	LOD– 0.02	0.01– 0.13	LOD– 0.04	0.06– 1.38	16.88	0.10	58, 65, 99, 100
goldenrod	1.05– 1.38	0.15– 0.16		0.59– 0.78		0.26– 0.47		0.01– 0.04	0.04– 0.24				0.23– 0.27	5, 54, 71
hawthorn	0.96	0.07												59
indigobush	1.15	0.25		1.09	0.00	0.03	0.01	0.00	0.01	0.01	0.36			65
ivy	1.13	0.16		1.16	0.00	0.02	0.02	0.00	0.00	0.00	0.45			65
jerusalem thora	0.76– 2.94	0.19– 0.86	0.15– 0.63	0.18– 4.04	0.00– 0.10	0.02– 0.90	0.01– 0.04	0.00– 0.36	0.00– 1.67	LOD– 0.02	0.66			65, 84
mandarin	1.33	0.14		1.84	0.00	0.01	0.01	0.00	0.01	0.00				95
mint	1.17– 2.67	0.15– 0.65		0.92	0.00	0.19– 1.02	0.02– 0.04	0.00– 0.08	0.01– 0.33	0.01	0.44			19, 65, 79
oak	LOD– 2.11	0.06– 2.19	0.34	0.91– 2.89	0.00	0.06– 0.77	0.01– 0.03	0.00– 0.01		0.01– 0.02	0.94	4.33		65, 72, 85, 101
onosma	0.77	0.24			0.19	0.06	0.01	0.02	0.29	0.00				93
orange	0.49– 1.75	0.02– 0.56		0.59– 7.15	0.58	0.02– 0.24	LOD– 0.01	LOD	0.04– 0.24	LOD	0.19			55, 100
parsley	0.08	0.02				0.09	0.00	0.00	0.00					79
pine	0.52– 9.30	0.05– 1.21	0.08	0.40– 3.21	0.53– 0.69	0.12– 0.83	0.01	LOD– 0.21	LOD– 0.28	LOD– 0.08	0.03	2.65	0.53– 0.69	63, 72, 79, 97–100
potentilla	0.53	0.49			0.63	0.34	0.01	0.04	0.44	0.00				93
raspberry	1.24– 3.65	0.35– 3.09				0.01		0.00– 0.02	0.00– 0.27					19, 61, 71
rhododendron	0.10– 1.00	0.08– 0.43	0.14	1.20	0.01	0.05– 0.18	0.01– 0.09	0.00	0.00– 0.60	0.01		3.30– 4.20		57, 72, 79
sage	0.68– 3.60	0.26– 1.00		0.35– 1.65	0.00– 0.19	0.01– 0.99	0.00– 0.59	0.00– 0.01	0.00– 0.54		0.33			58, 65, 73, 88, 94, 95
sainfoin	0.57	0.28				0.36	0.01	0.00	0.00					79
savory				1546.70	4.02	453.00	3.75	12.80	7.25					94
spruce	2.61	1.185	0.13	8.97		0.09		0.02		0.04		6.71	0.02	58
strawberry tree	0.24– 9.12	0.05– 0.25		0.90– 1.64	0.00– 0.00	0.01– 0.12	0.00– 0.10	0.00– 0.00	0.00– 0.07	0.00– 0.01	0.36– 0.94			65, 79, 95
sulla	0.33– 0.46	0.04		0.76		0.03	0.02		0.01					55, 90
willow	0.43	0.27	0.04	0.27		0.02	LOD		0.00	0.02		0.80	LOD	64
vetch	0.39– 0.63	0.02– 0.18		4.39			0.00	0.00– 0.77	0.04					59, 63
yellow-thistle	0.81	0.32				0.47	0.00	0.00	0.00					79

LOD – limits of detection

the Polish Standard for bee honey: PN-88/A-77626. The commercial quality of honey is specified in the Regulation of the Minister of Agriculture and Rural Development, dated 29 May 2015.

The permissible amounts of additives and other foreign substances in foodstuffs or stimulants, as well as contaminants, are specified by the Regulation of the Minister of Health, dated 27 December 2000 (*Journal of Laws* 2001, No. 9, item 72). According to this regulation, the permissible content of mercury is 0.01 mg/kg fresh weight.

The Joint FAO/WHO Expert Committee (JEC) on Additives determined the permissible doses of heavy metals for ingestion by adults per week (PTWI – Provisional Tolerable Weekly Intake), i.e. doses that do not yield toxic effects: Pb = 0.025 mg/kg b.w., Hg = 0.0016 mg/kg b.w., and Cd = 0.007 mg/kg b.w. These values include all possible routes by which heavy metals can enter the human body, i.e. through food, inhaled air or through the skin. According to PN-88/A-77626:1988, which was withdrawn in 2014, the highest permissible concentration of the following heavy metals in bee honey were Pb 1.08 mg/kg, Zn 3.4 – 47.4 mg/kg, Cd 0.02 mg/kg, and Hg 0.011 mg/kg.

Currently, Commission Regulation (EC) No. 1881/2006 of 19 December 2006, setting maximum levels for certain contaminants in foodstuffs, is binding for member states. The regulation covers, *inter alia* mycotoxins (aflatoxins, ochratoxin A, *Fusarium* toxins, patulin), metals (lead, cadmium, mercury, inorganic tin), monochloropropane-1,2-diol (3-MCPD), dioxins and dioxin-like PCBs, as well as polycyclic aromatic hydrocarbons (PAHs), melamine, and erucic acid. However, while it specifies the limit for cadmium at 0.20 mg/kg fresh weight for fresh herbs, and lead 0.30 mg/kg for leafy vegetables, there is no information applicable to honey.

**Metals in honey – preventing contamination.** For the consumer it is important that honey has health-promoting effects, and that harmful substances are not present. It is

therefore important to buy honey from a reputable source with a veterinary number on the package. The responsibility for nectar selection, and honey collection and packaging lies with the beekeeper [102].

While no clear regulations exist regarding the heavy metal content of honey, it is important to systematically assess the environmental contamination of bee forage. Such measures will increase the health-promoting value of the honeys and ensure consumer safety, especially those sensitive to allergens [103].

However, as honey consumption is typically quite low, i.e. 0.60 kg of honey per person per year, the levels of toxic and beneficial metals in honey may not significantly influence consumer health. The concentrations of metals in honeys of different botanical origin are recorded at levels that do not pose a threat to the health of a potential consumer in relation to the applicable standards [104].

## OWN RESEARCH

**Presence of metals in varietal honeys.** Own findings (Tab. 5) indicate higher values for magnesium and manganese in rapeseed honeys than in others. Their presence may be due to the use of fertilisers needed for oilseed rapeseed cultivation.

The findings also indicate correlations between total pollen counts and the contents of Mn, Cu and Na (Tab. 6). Positive linear correlations were detected for the following systems: Cu with Mn, Fe with Mn, Mg with Mn, Na with Mn, Cu with Mg, Mg with Na, Mg with Ca.

The proportion of pollen in a honey influences its appearance: e.g. rapeseed and robinia honey are light yellow, and lime honey – golden yellow. This will influence the attractiveness of the honey to the consumer. Most pollen grains have similar sizes and shapes: *Robinia* spp. measure 34.0 µm and are rounded, *Tilia* spp. measure 35.8 µm and are also rounded, while *Brassica* spp. measure 33.6 µm and are either rounded or prolate in shape (Fig. 1).

**Table 5.** Characteristics of monofloral honeys

Mh	Variable	Tpn	Td	Mn	Zn	Cu	Fe	Mg	Ca	K	Na
Acacia	mean	185	14	0.435	3.01	0.270	2.24	13.2	54.3	1004	26.7
	min.	25	7	0.105	1.97	0.047	1.01	6.8	29.0	402	3.0
	max.	496	21	1.059	5.06	0.636	3.14	21.7	96.4	1626	59.4
	SD	211	6	0.446	1.45	0.265	1.03	7.5	31.4	679	24.1
	CV [%]	114	42	103	48	98	46	57	58	68	90
Linden	mean	178	16	0.237	3.78	0.167	1.86	16.4	78.4	1652	12.7
	min.	34	5	0.130	2.14	0.097	1.23	13.8	51.0	1274	8.4
	max.	380	22	0.416	5.92	0.330	3.37	20.7	140.2	1844	23.7
	SD	117	6	0.094	1.42	0.076	0.77	2.9	30.2	186	5.9
	CV [%]	66	38	40	37	46	41	17	38	11	66
Rapeseed	mean	295	13	1.198	3.42	0.305	3.06	18.0	69.5	816	17.0
	min.	38	4	0.426	2.10	0.217	2.03	14.5	49.7	326	14.6
	max	400	17	3.283	5.05	0.443	3.55	23.2	97.2	1706	21.8
	SD	172	6	1.393	1.26	0.10	0.69	3.8	20.0	636	3.3
	CV [%]	59	47	116	37	33	23	21	29	78	19

min – minimum values; max – maximum values; SD – standard deviation; CV – coefficient of variation; Mh – monofloral honeys; Tpn – total pollen number; Td – number of pollen taxons in a sample of varietal honey

**Table 6.** Linear correlation between the honey parameters tested:  $r$  ( $p < 0.0500$ ),  $N = 15$ ) and graphical representation of the determination index  $R^2$  (colored fields)

Variable	Tpn	Td	Mn	Zn	Cu	Fe	Mg	Ca	K	Na
Tpn	1.0000									
Td	<b>0.6742</b>	1.0000								
Mn	<b>0.6207</b>	0.2509	1.0000							
Zn	0.1773	0.2069	0.1269	1.0000						
Cu	<b>0.6157</b>	0.2367	<b>0.8266</b>	0.0865	1.0000					
Fe	0.2368	-0.0799	<b>0.7315</b>	-0.1699	0.4970	1.0000				
Mg	0.4811	0.1590	<b>0.5791</b>	0.1893	<b>0.6565</b>	0.4563	1.0000			
Ca	0.2440	0.3072	0.4611	0.3867	0.2114	0.5126	<b>0.6068</b>	1.0000		
K	0.1082	0.3348	0.1553	0.0416	0.0961	-0.0319	0.4452	0.2942	1.0000	
Na	<b>0.5431</b>	0.3939	<b>0.7622</b>	0.1526	<b>0.8132</b>	0.2904	0.3411	0.2163	0.0655	1.0000

Tpn – Total pollen number; Td – number of pollen taxa in a sample of varietal honey.

Positive correlation coefficient (red) and negative correlation coefficient (blue)

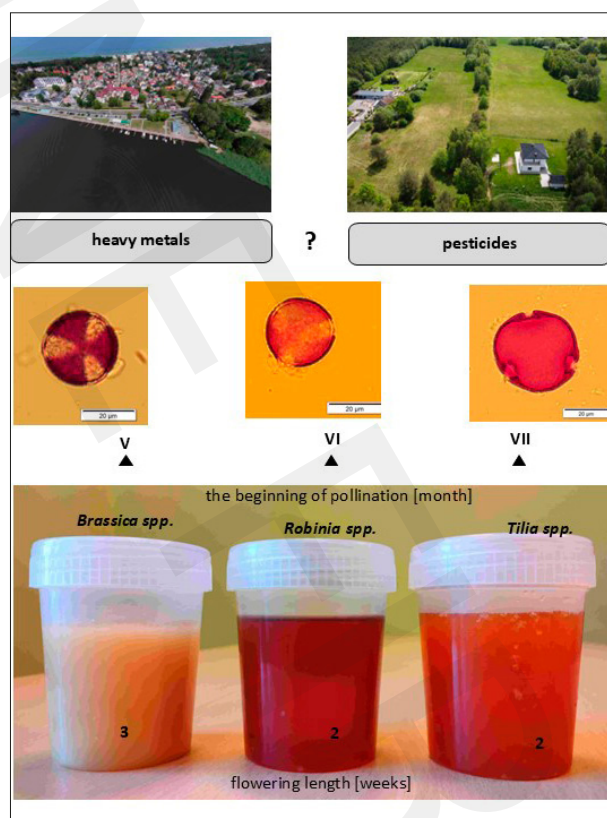
Values of the determination index ( $R^2$ ) for a positive correlation:



Determination index ( $R^2$ ) for a negative correlation:



Notation in bold - statistically significant correlation



**Figure 1.** Graphical abstract

## SUMMARY

### Information on the origin of a honey and its characteristics.

In addition to its nutritional value, honey is known to influence cell metabolism, kill gram-positive and gram-negative bacteria, and supports liver function. The health-promoting effects of varietal honeys are also emphasised by Traditional Medicine.

However, the consumption of honey has also been associated with negative effects; for example, some are believed to have allergenic properties. In addition, it has previously been recommended that pregnant women consume only pasteurised honey to counteract the effects of possible contamination with *Clostridium botulinum* spores, which are a risk factor for the development of botulism in newborn. Despite this, studies indicate that women with a normally-functioning intestinal flora do not need to avoid honey during pregnancy [105]. Furthermore, honey is not recommended for children under one year of age, as their digestive system would not be able to cope with any bacteria that may be contained in the honey.

Consumers are well aware that regulations concerning honeybee protection against pesticide residues in honey differ between regions. As such, indicating the country of origin of the honey makes it easier to make an informed choice when shopping. In Poland, the Regulation of the Minister of Agriculture and Rural Development of 13 October 2023, amending the Regulation on the labelling of particular types of foodstuffs, came into force on 18 April 2023 [106]. the



regulation requires that the names of all the countries of origin for a blend of honeys are included on the label.

El-Nahhal [15] reports that Polish honeys demonstrate the same level of contamination as those of other economically-developed countries. Even so, the maximum residue limits (MRLs) for pesticides in honey (e.g. coumaphos or atrazine) are based on national regulations, and the lack of standardisation creates problems in international trade and marketing [107].

As organically-sourced honey is often more expensive, it is important for its quality to be confirmed at a higher level than the beekeeper. As it is also important to assess the specific and cumulative human exposure to contaminants in the diet [30], organic honeys should also be subject to regular testing to identify residual contaminants in both the environment and food.

Own findings indicate that the presence of metals in honey is negligible, which may be attributed to their gradual removal during the various stages of production. The profile of metals differed between the studied honeys; generally, the highest concentrations were shown for potassium; however, statistically significant linear correlations were noted between pollen count and certain elements. In Poland, it is also beneficial that honey is required by legislation to be labelled with its country of origin, because the issue of metal contamination is widely known.

## CONCLUSIONS

- 1) Honey demonstrated varying levels of pesticide and metal residues depending on the quality of the natural environment.
- 2) Honeys from crops are more contaminated with pesticides and metals than those from uncultivated plants. The accumulation of a given metal in honey is influenced by the biochemical characteristics of bees, such as biological barrier function and the bioavailability of trace elements from ingested pollen or nectar.
- 3) Food allergy symptoms can arise in response to the pollen in monofloral honeys, as well as the presence of wind-distributed pollen, such as birch.
- 4) The pesticide coumaphos can accumulate in beeswax and thus be transferred to honey which, even with large consumption of contaminated honey, does not have a harmful effect on the health of adults.
- 5) When purchasing honey, its safety can be guaranteed by using reliable sources and providing information on the country of origin. However, due to its allergenic properties, it is recommended to also indicate the dominant pollen type present in the variety.
- 6) The most common, statistically correlated combinations of health-promoting elements accompanying manganese include copper, iron, magnesium and sodium.

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