



Potentially toxic trace elements in bee bread, propolis, beeswax and royal jelly – A review of the literature and dietary risk assessment

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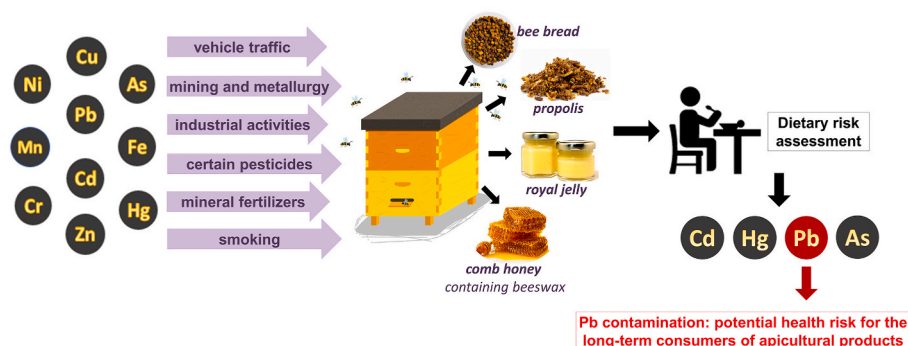
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HIGHLIGHTS

- Propolis and bee bread are better indicators of pollution than royal jelly.
- The food safety risk posed by As, Cd and Hg in bee products is low for long-term consumers.
- The accumulation of lead in apicultural products may be a food safety concern.
- Beehives should be placed in ecologically clean environments to ensure food safety.

GRAPHICAL ABSTRACT



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ABSTRACT

Scientific evidence suggests that apicultural products accumulate pollutants present in the hive environment, thus, they can be used as bioindicators. However, our understanding on the food safety implications of the presence of potentially toxic trace elements in these products remains incomplete. In our study, available data on the trace metal content of bee bread, propolis, beeswax and royal jelly, as well as their possible sources are reviewed. Furthermore, dietary risk assessments were conducted for elements that do not have any biological role in humans by comparing the estimated exposures with official reference values. In the case of elements with genotoxic carcinogen potential, the margin of exposure (MoE) approach was applied. The observed concentration ranges vary over a wide range for Fe (0.94–2125.20 mg/kg), Zn (<LOQ – 2790.00 mg/kg), Cu (<LOQ – 40.93 mg/kg), Mn (<LOQ – 204.80 mg/kg), Ni (<LOQ – 75.90 mg/kg), Cr (<LOQ – 56.28 mg/kg), Pb (<LOQ – 160.10 mg/kg), As (<LOQ – 8.47 mg/kg), Cd (<LOQ – 76.69 mg/kg) and Hg (<LOQ – 1.7 mg/kg) in beehive products from different geographical origins. These variances can be attributed to the diversity of soil types, climatic conditions, floral sources, beekeeping practices and anthropogenic activities in the environment. Available data suggest that Pb can be present in apicultural products at concentrations exceeding a thousand µg/kg, which poses a significant food safety threat to long-term consumers.

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1. Introduction

The rapid development of industrial and agricultural activities, mining and urbanization has led to a significant increase of pollutant emissions (Khaneghah et al., 2020; Sharma et al., 2023). Heavy metals are extensively studied pollutants that persistently accumulate in the environment due to their stability and non-degradability (El-Kady and Abdel-Wahhab, 2018). In scientific literature, the term “heavy metal” commonly refers to metals and metalloids associated with environmental pollution, toxicity, and adverse effects on living organisms. According to their definition, heavy metals are “naturally occurring metals having an atomic number greater than 20 and an elemental density greater than 5 g/cm³”. However, there is a concern about the general use of this term because it usually refers not only to metals but also to metalloids. Therefore, researchers suggest using alternative terms, such as “potentially toxic trace elements” (Ali and Khan, 2017; Pourret et al., 2021).

The contamination of foods by trace metals and metalloids is considered a major food safety issue due to their ubiquitous presence and long-term environmental persistence (Matin et al., 2016; Rana et al., 2018; Sharma et al., 2023). Several metals (e.g. Fe, Zn, Cu, Mn and Se) are essential for the physiological and biochemical processes in living organisms, however, health problems may arise if they are ingested in excessive amounts (Khaneghah et al., 2020). The toxicity of trace elements depends on various factors including the dose, exposure conditions, bioavailability and affected biological species. Different chemical species of metals and metalloids may also differ considerably in terms of toxicity. For example, inorganic arsenic is more toxic than organic forms (Nordberg and Nordberg, 2016). Lead, arsenic, cadmium and mercury are particularly important in terms of human toxicity, because even small amounts of these elements can be highly toxic, and they do not have any confirmed biological role (Bartkowiak, 2022; Khaneghah et al., 2020; Nordberg and Nordberg, 2016).

A large number of studies confirm that apicultural products accumulate pollutants present in the hive environment, including substances from agricultural and industrial activities, as well as beekeeping practices. As a result, bees and their products can be used as bioindicators (Aljedani, 2020; Bogdanov, 2006; Cunnigham et al., 2022; Golubkina et al., 2016; Nowak and Nowak, 2021; Sharma et al., 2023; Zafeiraki et al., 2022). Honeybees (*Apis mellifera*) typically forage within a 2 km distance from the hive, but in cases of limited food sources, they can fly up to 10 km (Garbuzov et al., 2015). Consequently, beehive products may become contaminated with pollutants from a large area. Since apicultural products are consumed by humans as nutraceuticals or food supplements, their contamination with trace elements may have food safety implications (Nowak and Nowak, 2021; Sharma et al., 2023; Tutun et al., 2022; Végh et al., 2021). The potentially toxic trace element content of honey and bee pollen, as well as the dietary risk posed by them, have been comprehensively reviewed in recent studies (Fakhri et al., 2019; Végh et al., 2021). Therefore, our study is limited to publications that address the trace element content of other edible bee products.

2. Methodology

The review followed the process proposed by Impellizzeri and Bizzini (2012). The first step involved outlining the main questions, including the followings: “To what extent do potentially toxic trace elements accumulate in bee bread, propolis, beeswax and royal jelly?”; “How does the source environment affect the trace element accumulation in these products?”; “What kind of techniques are generally used for the determination?”. Studies were identified by conducting searches on google scholar using combinations of the following keywords: „toxic element” OR „heavy metal” OR „trace metal” OR „trace element” OR „PTE” OR „inorganic contaminant” AND „apicultural product” OR „bee bread” OR „propolis” OR „beeswax” OR „honeycomb” OR „royal jelly”.

Additional relevant studies were found by reviewing the references of the selected articles. During the initial screening, duplicate studies, irrelevant studies, studies lacking data on As, Cd, Hg or Pb concentrations, studies related only to method validation, and studies published before January 1, 2000, were excluded. Remaining articles were grouped by product type for easier handling. A total of 60 studies were included in our review. Data were extracted regarding the concentration ranges of Fe, Zn, Cu, Mn, Ni, Cr, Pb, As, Cd, and Hg in the discussed apicultural products, as well as the characteristics of the source area and the determination technique used.

Furthermore, chronic dietary risk assessments were conducted on the content of Pb, As, Cd and Hg in apicultural products, since these elements are highly toxic even in small concentrations and do not have any identified biological function in humans. Daily exposures were estimated by considering the mean concentration values reported in the reviewed studies and the recommended maximum daily intake values. Mean concentrations were calculated considering all samples (values below the quantification limit were considered as zero). For bee bread, propolis and royal jelly, daily dosages of 10 g, 3 g, and 2 g, respectively, were considered based on the recommendations of a widely known commercial distributor of beekeeping products (Apiland, 2023). Beeswax can be consumed in the form of comb honey, which consists of pure honeycomb cells filled with honey. Based on a report by the European Food Safety Authority (EFSA, 2020), it is estimated that beeswax intake may average 4 g per day in certain populations, thus, this value was used in our risk assessment. Average body weights of 70 kg for men and 60 kg for women were assumed during the calculations (Rubio et al., 2017). For Cd and Hg, the exposure values were compared to the tolerable intake values established by FAO/WHO (2019). Since Pb and As possess genotoxic and carcinogenic potential, the margin of exposure (MoE) approach was applied during the risk assessment of these elements considering the benchmark dose lower confidence limit (BMDL) values reported by the European Food Safety Authority (EFSA, 2009; EFSA, 2010).

3. Results and discussion

3.1. Trace metals in bee bread, propolis, beeswax and royal jelly – key findings

In Table 1, scientific data on the concentration ranges of ten trace metals (Fe, Zn, Cu, Mn, Ni, Cr, Pb, As, Cd and Hg) in bee bread, propolis, beeswax and royal jelly are summarized. Among these products, propolis has been the subject of extensive research, whereas a limited number of studies have addressed the trace metal content of royal jelly. The samples were originated from various regions worldwide, but the available information is incomplete, particularly regarding products from North America, Africa, and Australia. The most commonly used instrumental techniques for determining inorganic contaminants in beehive products include atomic absorption spectrometry (AAS), inductively coupled plasma optical emission spectrometry (ICP OES), and inductively coupled plasma mass spectrometry (ICP-MS). Besides, flame atomic absorption spectrometry (FAAS), electrothermal atomic absorption spectrometry (ETAAS) (mostly graphite furnace atomic absorption spectrophotometry) and cold vapor atomic fluorescence spectrometry (CV AFS) have also been employed in some studies.

Elements were tested in the reviewed studies (n = 60) with the following frequencies: Pb (87%), Cd (82%), Zn (77%), Cu (68%), Fe (63%), Mn (60%), Cr (60%), Ni (55%), As (48%) and Hg (38%). Available data indicates that the median concentrations of trace elements detected in apicultural products follow this order: Fe, Zn, Mn, Cu, Ni, Pb, Cr, Cd, As and Hg. Essential elements are typically present in multiple concentrations in these products compared to the latter. However, lead, arsenic, cadmium, and mercury were detected in exceptionally high concentrations in products originating from polluted areas, such as urban, industrial, and mining sites (Gajger et al., 2019;

Table 1
Concentration of potentially toxic trace elements in bee bread, propolis, beeswax and royal jelly.

Samples (n ^o)	Country of origin	Source area	Technique	Concentration range ($\mu\text{g}/\text{kg}$)										Reference
				Fe	Zn	Cu	Mn	Ni	Cr	Pb	As	Cd	Hg	
bee bread (3)	Portugal	no data	AAS	64000–404000	n.a. ^b	10000–13000	76000–90000	n.a.	n.a.	<LOQ	n.a.	<LOQ - 50	n.a.	Aylanc et al. (2023)
bee bread (23)	Albania	no data	ICP OES	22000–565000	14000–87000	2200–16800	4900–65000	3200–27000	240–4100	110–320	n.a.	30–140	n.a.	Pavlova et al. (2021)
bee bread (17)	Bulgaria	no data	ICP OES	58000–298000	29000–56000	5400–15500	15000–58000	410–8200	120–900	80–250	n.a.	40–90	n.a.	Pavlova et al. (2021)
bee bread (5)	Turkey	no data	ICP-MS	84060–317020	52550–73960	10440–20020	17400–97090	1840–4080	720–1740	60–690	20–260	5310–67060	<LOQ	Mayda et al. (2020)
bee bread (1)	Russia	no data	AAS	n.a.	14600	4300	n.a.	n.a.	n.a.	250	<LOQ	70	<LOQ	Murashova et al., 2019
bee bread (8)	Lithuania	no data	ICP-MS	22920–77640	10780–43700	4400–19420	6720–39380	n.a.	187–849	81–468	n.a.	12–61	n.a.	Adaskeviciūtė et al., 2019
bee bread (1)	Morocco	no data	ICP OES	273000	33100	7000	26000	260	n.a.	70	n.a.	54	n.a.	Bakour et al. (2019)
bee bread (1)	Egypt	unpolluted	AAS	9958	1614	265	n.a.	n.a.	n.a.	1094	n.a.	146	n.a.	Omran et al. (2019)
bee bread (1)	Egypt	polluted	AAS	14952	2580	342	n.a.	n.a.	n.a.	1338	n.a.	182	n.a.	Omran et al. (2019)
bee bread (12)	Serbia	no data	ICP-MS	43070–57520	27310–46940	5–11	19510–204800	1–4	107–184	56–183	21–43	32–137	<LOQ	Ciric et al. (2019)
bee bread (68)	Bulgaria	no data	AAS	28490–142900	8540–19430	6720–13440	6950–35430	380–1060	n.a.	650–1470	n.a.	70–270	n.a.	Zhelyazkova (2018)
bee bread (1)	Moldavia	unpolluted	ICP-MS	92700	48200	8400	21370	880	110	210	40	60	<LOQ	Golubkina et al. (2016)
bee bread (1)	Russia	industrial	ICP-MS	37600	15500	2000	12640	200	110	340	30	110	<LOQ	Golubkina et al. (2016)
bee bread (12)	Egypt	no data	F AAS	51598–184916	18230–130214	3056–21428	1066–4646	n.a.	<LOQ	162–4052	<LOQ	91–1870	n.a.	Esmael et al. (2016)
bee bread (75)	Poland	industrial	ICP OES	n.a.	n.a.	n.a.	n.a.	n.a.	38–251	25–198	12–892	1–80	n.a.	Roman et al. (2016)
bee bread (252)	Poland	no data	Automatic analyser	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	<1 - 8	Madras-Majewska and Jasiński (2005)
propolis (6)	Serbia	no data	ICP OES	109000–376000	195569	1700–5000	6500–16300	590–1230	800–5600	3000–11700	40–340	36–362	n.a.	Ristivojevic et al. (2023)
propolis (7)	Turkey	no data	x-ray FLU	117000–627000	<LOQ - 40700	n.a.	3450–14100	<LOQ - 200	n.a.	<LOQ - 350	n.a.	n.a.	n.a.	Mutlu et al. (2023)
propolis (9)	Poland	mainly rural	ICP OES	107500–300300	900–54200	1300–4500	3700–18200	5000–7000	6400–8100	300–5200	n.a.	<LOQ - 100	n.a.	Milek et al. (2022)
propolis (12)	Turkey	no data	ICP OES	4340–75390	4320–85040	1470–9010	1860–5730	1090–8590	n.a.	1080–7680	n.a.	n.a.	n.a.	Fidan et al. (2022)
propolis (8)	Pakistan	no data	AAS	355170–1331460	257–472	n.a.	40860–77440	1250–64230	2–15	4580–10500	n.a.	10–412	n.a.	Akbar et al. (2022)
propolis (30)	Turkey	no data	ICP OES	69000–568000	7980–102000	610–6080	1610–28000	480–75900	300–4710	580–4380	<LOQ - 1360	n.a.	<LOQ - 180	Tutun et al. (2022)
propolis (1)	Romania	industrial	F AAS	n.a.	4195	3203	2184	1146	2344	651	n.a.	80	n.a.	Mititelu et al. (2022)
propolis (1)	Romania	agricultural	F AAS	n.a.	947	402	1026	876	1868	160	n.a.	16	n.a.	Mititelu et al. (2022)
propolis (20)	Italy	urban	ICP-MS, CV AFS	n.a.	17000–120000	n.a.	n.a.	<LOQ - 200	260–1340	100–560	<LOQ - 230	3–119	3–16	Conti et al. (2022)
propolis (252)	Hungary	no data	ICP OES, ICP-MS	36800–1450000	5340–2790000	573–26900	887–21100	90–28800	91–38400	n.a.	n.a.	6–1480	n.a.	Soós et al. (2021)
propolis (6)	Poland	agricultural	ICP OES, ICP-MS	76000–160000	11000–19000	980–3000	5000–9700	250–860	250–720	440–1000	50–110	30–80	n.a.	Matuszewska et al. (2021)
propolis (6)	Italy	no data	CV AFS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	<LOQ - 16	Astolfi et al. (2021)

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Table 1 (continued)

Samples (n ^o)	Country of origin	Source area	Technique	Concentration range ($\mu\text{g}/\text{kg}$)										Reference
				Fe	Zn	Cu	Mn	Ni	Cr	Pb	As	Cd	Hg	
propolis (3)	Borneo	suburban, unpolluted	ICP OES	19840–69640	<LOQ - 2970	10850–24250	6980–106030	102–220	884–2740	195–1000	726–4450	<LOQ - 664	n.a.	Abdullah et al. (2020)
propolis (1)	Turkey	no data	ICP-MS	1425820	74950	4290	35660	2590	3080	n.a.	n.a.	1530	<LOQ	Ecem Bayram, 2020
propolis (2)	Brazil	no data	ICP-MS	102660–347750	25010–33210	5110–9010	2818–6069	3110–3510	160–800	n.a.	n.a.	650–660	<LOQ	Ecem Bayram, 2020
propolis (1)	China	no data	ICP-MS	287010	8000	1470	2500	2180	560	n.a.	n.a.	1070	<LOQ	Ecem Bayram, 2020
propolis (1)	Ethiopia	no data	ICP-MS	861970	14100	3680	52520	1360	2260	n.a.	n.a.	1970	<LOQ	Ecem Bayram, 2020
propolis (19)	Brazil	no data	F AAS, GF AAS	n.a.	n.a.	570–11600	n.a.	n.a.	n.a.	<LOQ - 720	<LOQ - 8470	< LOQ - 30	n.a.	Hodel et al. (2020)
propolis (1)	Russia	no data	AAS	n.a.	59100	5000	n.a.	n.a.	n.a.	2260	<LOQ	780	<LOQ	Murashova et al., 2019
propolis (5)	Lithuania	no data	ICP-MS	234200–304500	31900–102100	2360–14310	15000–25100	n.a.	4350–12130	3490–9490	n.a.	12–41	n.a.	Adaskevičiūtė et al., 2019
propolis (1)	Poland	no data	ICP-MS	245000	52400	8530	28800	n.a.	4320	4600	n.a.	72	n.a.	Adaskevičiūtė et al., 2019
propolis (10)	North Macedonia	lowland	AAS	n.a.	21–38	n.a.	n.a.	n.a.	n.a.	33–49	n.a.	27–38	n.a.	Naco et al. (2017)
propolis (10)	North Macedonia	mountain	AAS	n.a.	13–31	n.a.	n.a.	n.a.	n.a.	22–42	n.a.	11–18	n.a.	Naco et al. (2017)
propolis (40)	North Macedonia	rural	AAS	n.a.	20–31	18–29	n.a.	n.a.	12–38	31–42	n.a.	13–30	n.a.	Bogdanova Popov et al. (2017)
propolis (6)	Turkey	no data	ICP OES	1740–3090	14060–35670	2620–4600	4970–16760	4240–37080	1130–3660	840–2490	n.a.	30–130	n.a.	Şahinler et al. (2017)
propolis (10)	Serbia	no data	ICP OES	116000–284000	19200–241000	2220–8700	3980–14360	500–1590	710–9900	2000–9700	<LOQ	69–310	<LOQ	Tosic et al. (2017)
propolis (1)	Moldavia	unpolluted	ICP-MS	134000	32100	1540	6070	410	460	2080	60	50	7	Golubkina et al. (2016)
propolis (2)	Russia	industrial	ICP-MS	45300–106000	6800–8700	640–1040	2970–3000	210–280	170–450	1520–3180	30–60	20–20	<LOQ	Golubkina et al. (2016)
propolis (1)	Mongolia	mining	ICP-MS	386000	52600	2700	10160	540	4250	16070	170	70	<LOQ	Golubkina et al. (2016)
propolis (6)	Greece	polluted	CV AAS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	<LOQ	Maragou et al. (2016)
propolis (5)	Turkey	polluted	AAS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	46–961	19–578	1695–76691	<LOQ	Matin et al. (2016)
propolis (1)	Russia	near a highway	AAS	17740	5890	13700	n.a.	n.a.	n.a.	123630	n.a.	182	n.a.	Eskov et al. (2015)
propolis (39)	Spain	no data	ICP OES, ICP-MS	46100–874000	11100–460700	<LOQ - 33400	n.a.	500–29900	800–48900	< LOQ - 74000	n.a.	n.a.	n.a.	González-Martín et al. (2015)
propolis (52)	Chile	no data	ICP OES, ICP-MS	181800–1538000	5500–105000	<LOQ - 6200	n.a.	< LOQ - 9700	1400–5500	< LOQ - 8000	n.a.	n.a.	n.a.	González-Martín et al. (2015)
propolis (42)	Brazil	no data	F AAS, GF AAS	n.a.	<LOQ - 50000	<LOQ	<LOQ - 170000	n.a.	<LOQ - 19300	<LOQ - 160100	n.a.	<LOQ - 640	n.a.	Finger et al. (2014)
propolis (8)	Poland	industrial	F AAS	28000–101000	17700–71500	n.a.	n.a.	1990–9810	n.a.	890–2940	n.a.	13–54	n.a.	Formicki et al. (2013)
propolis (4)	Argentina	no data	ICP OES	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	1890–9940	n.a.	n.a.	n.a.	Pierini et al. (2013)
propolis (13)	Spain	no data	ICP OES	312000–1270000	163000–1364000	2080–4720	6420–27100	640–3650	330–2730	70–3750	<LOQ - 130	38–110	2–14	Serra Bonvehí and Orantes Bermejo (2013)
propolis (32)	mainly China	no data	ICP OES	310400–2125200	44400–386400	<LOQ - 14950	3670–88230	<LOQ - 3110	<LOQ - 11580	1670–55370	<LOQ - 920	240–1190	n.a.	Gong et al. (2012)
propolis (18)	Korea	no data	ICP-MS	n.a.	229–672	<LOQ - 130	n.a.	<LOQ - 67	<LOQ	<LOQ - 538	<LOQ	<LOQ - 18	<LOQ	Woo et al. (2012)
propolis (96)	Argentina	no data	NAA	400000–1945000	11000–105000	n.a.	n.a.	n.a.	600–3750	n.a.	20–600	n.a.	n.a.	Cantarelli et al. (2011)
propolis (20)	Poland	industrial	ICP OES	n.a.	16880–99680	1730–9570	n.a.	n.a.	n.a.	560–9940	87–1238	69–802	n.a.	Roman et al. (2011)
propolis (1)	Croatia	no data	AAS	1014430	234170	20220	21020	n.a.	n.a.	2818	448	<LOQ	18	Cvek et al. (2008)
propolis (1)	Romania	no data	ICP OES	n.a.	n.a.	3630	n.a.	n.a.	n.a.	60	n.a.	250	n.a.	Dobrinás et al. (2006)
propolis (3)	Italy	city center	AAS	n.a.	n.a.	n.a.	n.a.	n.a.	6–7	4–4	n.a.	5–7	n.a.	Conti and Botré (2001)
propolis (12)	Italy	suburbia	AAS	n.a.	n.a.	n.a.	n.a.	n.a.	2–4	1–3	n.a.	1–2	n.a.	Conti and Botré (2001)

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Table 1 (continued)

Samples (n ^o)	Country of origin	Source area	Technique	Concentration range ($\mu\text{g}/\text{kg}$)										Reference
				Fe	Zn	Cu	Mn	Ni	Cr	Pb	As	Cd	Hg	
beeswax (1)	South Korea	no data	ICP-MS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	11	2	n.a.	1	Kim et al. (2022)
beeswax (7)	Slovakia	industrial	ICP-MS	8277–160775	4037–118606	<LOQ - 12967	182–41904	94–1919	82–982	59–3193	<LOQ - 61	<LOQ - 60	12–90	Zafeiraki et al. (2022)
beeswax (8)	Pakistan	no data	AAS	1971–18054	1265–8299	<LOQ - 324	27–491	<LOQ - 322	5–256	<LOQ - 928	n.a.	<LOQ - 95	n.a.	Ullah et al. (2022)
beeswax (163)	Italy	city	ICP-MS, CV AFS	n.a.	3000–729000	n.a.	n.a.	<LOQ - 1700	20–1210	40–6510	30–140	30–289	1–11	Conti et al. (2022)
beeswax (6)	Italy	no data	CV AFS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	<LOQ - 13	Astolfi et al. (2021)
beeswax (1)	Vietnam	rural	ICP-MS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	644	19	46	<LOQ	Ngat et al. (2020)
beeswax (1)	Vietnam	semi-rural	ICP-MS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	525	80	<LOQ	<LOQ	Ngat et al. (2020)
beeswax (1)	Russia	no data	AAS	n.a.	780	570	n.a.	n.a.	n.a.	180	<LOQ	7	<LOQ	Murashova et al., 2019
beeswax (1)	Saudi Arabia	near traffic	AAS	5972	1283	1034	365	474	1768	114	n.a.	59	n.a.	Aljedani (2020)
beeswax (1)	Saudi Arabia	urban	AAS	9698	1542	1139	501	598	2016	137	n.a.	60	n.a.	Aljedani (2020)
beeswax (1)	Saudi Arabia	industrial	AAS	18516	6272	1913	1311	678	2307	215	n.a.	75	n.a.	Aljedani (2020)
beeswax (1)	Saudi Arabia	unpolluted	AAS	11195	776	95	1059	<LOQ	3	<LOQ	n.a.	<LOQ	n.a.	Aljedani (2020)
beeswax (1)	Italy	no data	ICP OES, ICP-MS	310	n.a.	<LOQ	n.a.	n.a.	20	n.a.	<LOQ	<LOQ	n.a.	Astolfi et al. (2020)
beeswax (32)	Israel	no data	ICP OES	n.a.	<LOQ - 34400	n.a.	370–1960	<LOQ	264–597	83–499	<LOQ	<LOQ	<LOQ - 62	Bommuraj et al. (2019)
unfiltered wax (1)	Spain	no data	ICP-MS	20700	10300	<LOQ	510	<LOQ	<LOQ	3500	15	20	1700	Navarro-Hortal et al. (2019)
filtered wax (1)	Spain	no data	ICP-MS	600	1300	<LOQ	140	<LOQ	<LOQ	2900	48	11	100	Navarro-Hortal et al. (2019)
beeswax (7)	Croatia	no data	GF AAS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	20–31	1–49	0–95	Kosanović et al. (2019)
beeswax (1)	Egypt	unpolluted	AAS	15466	2052	391	n.a.	n.a.	n.a.	1750	n.a.	160	n.a.	Omran et al. (2019)
beeswax (1)	Egypt	polluted	AAS	16696	4606	620	n.a.	n.a.	n.a.	1388	n.a.	194	n.a.	Omran et al. (2019)
light beeswax (1)	Croatia	near traffic	x-ray fluorescence	285900	262900	12800	30570	14840	53930	4060	n.a.	n.a.	n.a.	Gajger et al. (2019)
dark beeswax (1)	Croatia	near traffic	x-ray fluorescence	134100	190800	40930	21570	17830	56280	5200	n.a.	n.a.	n.a.	Gajger et al. (2019)
light beeswax (1)	Croatia	rural	x-ray fluorescence	218000	137700	28630	22530	13200	48900	1230	n.a.	n.a.	n.a.	Gajger et al. (2019)
dark beeswax (1)	Croatia	rural	x-ray fluorescence	56470	93800	21970	16630	12170	41030	2770	n.a.	n.a.	n.a.	Gajger et al. (2019)
light beeswax (1)	Croatia	industrial	x-ray fluorescence	221500	166800	30830	32870	12170	54270	2500	n.a.	n.a.	n.a.	Gajger et al. (2019)

(continued on next page)

Table 1 (continued)

Samples (n ^a)	Country of origin	Source area	Technique	Concentration range ($\mu\text{g}/\text{kg}$)										Reference
				Fe	Zn	Cu	Mn	Ni	Cr	Pb	As	Cd	Hg	
dark beeswax (1)	Croatia	industrial	x-ray fluorescence	123700	105700	23200	18830	12800	47770	5430	n.a.	n.a.	n.a.	Gajger et al. (2019)
beeswax (1)	Russia	near a highway	AAS	3140	47520	12350	n.a.	n.a.	n.a.	736	n.a.	150	n.a.	Eskov et al. (2015)
beeswax (8)	Poland	no data	F AAS	108000–334000	19100–81200	n.a.	n.a.	1890–7350	n.a.	150–3130	n.a.	5–99	n.a.	Formicki et al. (2013)
beeswax (1)	France	no data	ICP-MS	n.a.	1520	n.a.	16100	n.a.	n.a.	<LOQ	12	6	n.a.	Saunier et al. (2013)
beeswax (3)	Italy	city center	AAS	n.a.	n.a.	n.a.	n.a.	n.a.	63–94	180–206	n.a.	45–52	n.a.	Conti and Botré (2001)
beeswax (12)	Italy	suburbia	AAS	n.a.	n.a.	n.a.	n.a.	n.a.	32–76	57–146	n.a.	<LOQ - 29	n.a.	Conti and Botré (2001)
royal jelly (1)	Poland	agricultural	ICP OES, ICP-MS	3900	21000	4200	730	250	20	70	10	2	n.a.	Matuszewska et al. (2021)
royal jelly (5)	unknown	no data	ICP-MS	7240–9200	14950–19510	2810–3670	520–1220	<LOQ - 40	20–180	<LOQ	<LOQ	<LOQ	<LOQ	Ecem Bayram et al. (2021)
royal jelly (9)	Turkey	no data	ICP-MS	940–23370	6360–66440	430–12780	100–1710	<LOQ - 720	20–40	<LOQ - 10	<LOQ	<LOQ - 10	<LOQ	Ecem Bayram et al. (2021)
royal jelly (6)	Italy	no data	CV AFS	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	<LOQ	Astolfi et al. (2021)
royal jelly (1)	Italy	no data	ICP OES, ICP-MS	9230	n.a.	4260	n.a.	n.a.	34	n.a.	<LOQ	<LOQ	n.a.	Astolfi et al. (2020)
royal jelly (1)	Russia	no data	AAS	n.a.	6380	3100	n.a.	n.a.	n.a.	2800	20	<LOQ	<LOQ	Murashova et al., 2019
royal jelly (5)	Lithuania	no data	ICP-MS	6800–9330	18300–19700	7510–9810	<LOQ	n.a.	210–270	205–452	n.a.	1–3	n.a.	Adaškeviciūtė et al., 2019
royal jelly (1)	Germany	no data	ICP-MS	12410	24100	11100	<LOQ	n.a.	280	418	n.a.	2	n.a.	Adaškeviciūtė et al., 2019
royal jelly (30)	Bulgaria	no data	ET AAS	12000–21000	19000–29000	4000–4900	340–1690	35–94	200–2300	20–980	11–74	1–6	n.a.	Balkanska et al. (2017)
royal jelly (1)	France	no data	ICP-MS	n.a.	906	n.a.	304	n.a.	n.a.	168	5	7	n.a.	Saunier et al. (2013)
royal jelly (6)	France	no data	ICP-MS	9100–22100	19400–24800	4100–8100	700–4350	140–1700	330–2970	17–287	n.a.	2–19	<LOQ - 36	Stocker et al. (2005)
royal jelly (1)	China	no data	ICP-MS	11000	24400	4000	780	150	340	51	n.a.	1	1	Stocker et al. (2005)

^a number of observations.^b n.a.: not analysed.

Golubkina et al., 2016; Matin et al., 2016; Navarro-Hortal et al., 2019; Roman et al., 2011). The presented studies do not provide information on the chemical species of the trace elements, although these properties affect toxicity (Nordberg and Nordberg, 2016).

3.2. Factors influencing the content of potentially toxic trace elements in apicultural products

Relatively large variations can be observed regarding the concentrations of elements reported by different studies, suggesting that the metal and metalloid contents of bee products are significantly influenced by external factors. Several studies confirm that the element composition of apicultural products is dependent on their geographical origin (Aljedani, 2020; Ecem Bayram, 2020; Golubkina et al., 2016; Ngat et al., 2020; Omran et al., 2019). These differences can be attributed to various factors such as soil types, climatic conditions, floral sources, beekeeping practices and anthropogenic activities of the location sites (Cvek et al., 2008; Pohl et al., 2020; Bogdanova Popov et al., 2017). Pollen and propolis undergo less transformations by bees compared to wax and honey, thus, they may reflect environmental contamination more precisely (Eskov et al., 2015; Formicki et al., 2013). The concentrations of inorganic contaminants in royal jelly samples are relatively constant and low, possibly because the body of nurse bees filter these elements during secretion. Probably similar processes take place as reported by recent studies demonstrating that honeybees have an ability to filter metals from nectar when converting it into honey (Borsuk et al., 2021; Tomczyk et al., 2023). This may explain why the potentially toxic trace element content of royal jelly is not significantly influenced by the geographical and botanical origins (Balkanska et al., 2017; Stocker et al., 2005). The contamination of hive products with trace elements is a result of interrelated environmental processes that occur with varying intensity, making the identification of the origin of trace elements in bee products a very complex task (Murashova et al., 2019).

The concentration and mobility of metals in the soil are affected by soil properties, such as granulometric composition, organic matter content, pH, oxidation-reduction potential and the activity of microorganisms (Fijałkowski et al., 2012). The transfer of metals and metalloids from soil to plants is controlled by additional factors related to plant physiology, including plant type, rate and type of root secretions, root surface area and transpiration (Hodel et al., 2020). The above-listed factors may influence the element content of nectar, pollen, and resin; however, these possible effects should be the subject of further research. Climatic conditions, such as temperature, humidity, rainfall, and light may also have an impact on the metal deposition of different plant tissues (Shahid et al., 2017). According to Pohl et al. (2020), high wind activity and low pluviometric precipitation in the dry season may lead to an increased air pollution, resulting in an enhanced metal accumulation in floral sources of apicultural products. Findings of our previous study (Véghe et al., 2021) also indicated that dry weather favours the metal accumulation in bee pollen.

Plants specifically select, translocate, and accumulate metals and metalloids from the soil; thus, the element composition of hive products is greatly influenced by their botanical origin (Adaškevičiūtė et al., 2019; Gong et al., 2012; Pohl et al., 2020; Shahid et al., 2017; Tomczyk et al., 2023; Véghe et al., 2021). Pohl et al. (2020) demonstrated that the effect of the geographical origin on the element composition of bee pollen can only be quantified when comparing monofloral pollen sources from the same botanical origin. In such cases, observed differences can be attributed solely to variations between location sites. This approach should also be followed in research dealing with the metal contamination of bee bread and propolis. The plant source of apicultural products is not specified in most studies, which suggest that multifloral samples were used by researchers. In this regards, further studies are needed to investigate the effect of anthropogenic factors on the metal contamination of hive products of identified botanical origins.

Processing methods may have a significant impact on the trace metal content of apicultural products. Previous research has shown that the concentration of trace metals is considerably lower in the ethanolic extract of propolis compared to raw propolis (Cvek et al., 2008; Orsi et al., 2018). Since propolis is usually consumed in the form of alcoholic tinctures (Tosic et al., 2017), the risk posed by potentially toxic trace elements may be reduced during processing. In addition, the harvesting method also influences the concentration of lead in propolis as it was demonstrated by Sales et al. (2006). According to this study, harvest methods of plastic meshes result in significantly lower Pb contamination of the final product compared to the traditional method (separating wedges with a spatula). It is a common beekeeping practice throughout the world to recycle old, dark honeycombs for the production of new foundations. As a consequence, trace metals and other contaminants may accumulate in beeswax over several decades (Gajger et al., 2019; Orantes-Bermejo et al., 2010). The study of Gajger et al. (2019) revealed that the concentrations of inorganic contaminants are significantly higher in dark, old combs compared to light, freshly constructed combs. Navarro-Hortal et al. (2019) demonstrated that a certain filtration process (briefly a treatment with activated carbon and diatomaceous earth) is applicable to address this problem.

The majority of studies focus on the effects of anthropogenic activities on the content of inorganic contaminants in apicultural products, as they can serve as indicators of environmental pollution in the vicinity of beekeeping sites (Tutun et al., 2022). Trace elements can be transferred to honeybees and bee products from various environmental sources in the vicinity of beehives including soil, vegetation, air, and water (Astolfi et al., 2020). The co-occurrence of certain metals is a common phenomenon, suggesting that these metals may originate from the same pollution source (Esmael et al., 2016; Finger et al., 2014; Nowak and Nowak, 2021). For example, dust fallout may contain remarkable amount of Cd, Pb and Fe (Formicki et al., 2013). Pb primarily enters the air through vehicle traffic (Aljedani, 2020), but its transfer from soil to plant tissues is not significant (Pavlova et al., 2021; Tomczyk et al., 2023). On the other hand, Cd is a highly mobile element, and easily assimilated by plants due to the available form of Cd²⁺ ion. (Bartkowiak, 2022; Bogdanov, 2006). The concentration of Cd in agricultural regions may be significant due to its presence in pesticides and mineral fertilizers (Esmael et al., 2016; Tosic et al., 2017). Cd may also enter bee products through air pollution from industrial activities such as incinerators (Bogdanov, 2006).

3.3. Food safety aspects of trace elements in bee bread, propolis, beeswax and royal jelly

3.3.1. Toxicological importance of Cd, Hg, Pb and As

The Cd, Hg, Pb and As contamination of apicultural products are in the focus of the following sections, because these elements are highly toxic even in small concentrations and do not have any identified biological function in humans. Considering their toxicological importance, chronic dietary risk assessments were carried out regarding the presence of these elements in apicultural products.

Cadmium is naturally present in the environment, but also generated as a result of agricultural and industrial activities, such as copper and nickel smelting, fossil fuel combustion as well as the production of phosphate fertilizers, PVC products and colour pigments (Gnechi et al., 2020). Exposure to cadmium occurs primarily through the ingestion of contaminated food and water, but also through inhalation and tobacco use (Taha et al., 2018). The International Agency for Research on Cancer (IARC, 2023) has categorized cadmium as carcinogenic to humans (Group 1). Exposure to this metal may be related to various types of cancer, including the liver, kidneys, breast, lung and prostate (Gnechi et al., 2020). Itai-itai disease is caused by severe Cd poisoning, which appeared as a result of human activities related to mining industries in Japan. The main symptoms of this disease are osteoporosis, bone deformities and renal dysfunction (El-Kady and Abdel-Wahhab, 2018).

Mercury enters the environment as a result of natural and anthropogenic activities (Astolfi et al., 2021). In the environment, Hg occurs in elemental (Hg^0), inorganic (HgS , HgCl_2) and organic (mostly methylmercury, MeHg) forms. Inorganic mercury can be transformed to MeHg via methylation by microorganisms in aquatic environments. Humans are exposed to mercury mainly by consuming aquatic animals containing bioaccumulated MeHg, especially in populations living near oceans, lakes, and rivers (Díez, 2009). Possible symptoms of severe MeHg poisoning include loss of peripheral vision, lack of coordination of movements, muscle weakness as well as impairment of speech, hearing, or walking (EPA, 2023). This disease is known as Minamata disorder (El-Kady and Abdel-Wahhab, 2018). IARC has categorized methylmercury compounds as possibly carcinogenic to humans (Group 2 B), while mercury and inorganic mercury compounds are not classifiable as to their carcinogenicity (Group 3) (IARC, 2023).

Lead is an abundant trace element in the environment, which is present mainly in the oxidation state of +2. About only 5% of the Pb occurring in the world originates from natural sources, e.g. volcanic exhalations. Anthropogenic sources of Pb emissions include fossil fuel combustion, road transport, manufacturing, mining, smelting and printing (Holecý and Mousavi, 2012). This metal is severely toxic to humans and affects almost every organs (Rana et al., 2018). The most affected target is the central nervous system, but the digestive, respiratory and reproduction system, as well as the normal DNA transcription process are also disrupted by excessive lead exposure (Wani et al., 2015). Inorganic Pb was categorized by IARC (2023) as probably carcinogenic to humans (Group 2 A). In adults, lead deposits mainly in the bones, but children can store less Pb in these tissues. As a consequence, the concentration of Pb in their blood increases, which can lead to the development of various symptoms (El-Kady and Abdel-Wahhab, 2018; Malavika et al., 2021).

Arsenic is a metalloid that exhibits characteristics of both metals and non-metals. It is ubiquitous in the environment in different states of oxidation and can exist in several organic and inorganic forms (Palma-Lara et al., 2020). Arsenic can reach the environment from volcanic activities, but its main sources are smelting and mining. Besides, consuming coal and fossil fuels, as well as the use of certain pesticides also contribute to arsenic contamination (El-Kady and Abdel-Wahhab, 2018). Humans are exposed to arsenic mainly by contaminated drinking water. Approximately 70–90% of inorganic arsenic is absorbed by the human gastrointestinal tract and distributed through the blood to different organs (Palma-Lara et al., 2020). The International Agency for Research on Cancer (IARC, 2023) has categorized arsenic and inorganic arsenic compounds as carcinogenic to humans (Group 1). The most affected organ of arsenic accumulation is the liver, but high exposure may also increase the risk of tumors in the kidneys, lungs and bladder (Palma-Lara et al., 2020).

3.3.2. Dietary risk assessment of Cd and Hg detected in apicultural products

Cadmium and mercury are non-genotoxic carcinogenic metals, for which tolerable intake values, such as TDI (tolerable daily intake), PTWI (provisional tolerable weekly intake) and PTMI (provisional tolerable monthly intake) are established. These values are estimates of the amount of a chemical that can be taken daily, weekly, or monthly per unit body weight over a lifetime without appreciable health risk (WHO, 2021). For the exposure assessment, estimated daily intake values were determined considering the mean concentrations reported in the reviewed studies, the maximum daily dosages recommended by Apiland (2023) and an assumed average body weight of 70 kg (for men) of 60 kg (for women) (Rubio et al., 2017). In the case of beeswax, a daily dosage of 4 g was taken into account, based on the technical report of the European Food Safety Authority (EFSA, 2020). Daily exposures were estimated by using the following equation: *Estimated daily exposure*

$$\left(\frac{\text{ng}}{\text{kgbw}}\right)_{\text{day}} = \text{Mean concentration} \left(\frac{\mu\text{g}}{\text{kg}}\right) \times \text{Max. recommended daily dosage (g)} \div \text{Average body weight (kg)} \times 10$$

Calculated exposures were compared to tolerable intake values reported by FAO-WHO (2019) and expressed as percent contribution by the following equation:

$$\text{Contribution (\%)} = \text{Estimated daily exposure} \left(\frac{\text{ng}}{\text{kgbw}}\right)_{\text{day}} \div \text{Tolerable daily exposure} \left(\frac{\text{ng}}{\text{kgbw}}\right)_{\text{day}} \times 100$$

Contributions above 10% were considered a high risk, because apicultural products are not major elements of a general diet, and their contribution to the daily energy intake is typically below 10%.

Results of the risk assessments for cadmium and mercury are presented in Table 2. Samples intentionally harvested from polluted areas (urban, industrial, mining site etc.) are marked with an asterisk (*). Our results suggest that the Cd and Hg content of the four apicultural products generally do not pose a chronic risk to the consumers, not even in case of products from sites exposed to pollution. Nevertheless, contribution values exceeded 10% in a few cases. Matin et al. (2016) detected very high (19388 and 76691 mg/kg) Cd concentrations in propolis samples originated from a Turkish village located near an industrial district and a highway. Among others, petrochemical industry, iron and steel factories, refineries, gas turbines and natural gas combined cycle power plants were operating in the industrial zone. Cd was also present in exceptionally high amounts in Turkish bee bread samples (Mayda et al., 2020). The long-term daily consumption of these products appears to pose a high risk to the consumers as they contribute to the PTMI established for Cd by more than 100%. This observation suggests that it is important to locate beehives in ecologically clean areas not only to protect the colony, but also in order to minimize the food safety risks associated with the consumption of apicultural products. Navarro-Hortal et al. (2019) detected a very high mean concentration of mercury (1700 $\mu\text{g}/\text{kg}$) in unfiltered beeswax samples, but after filtration, a 94% reduction was observed for this element. The long-term daily consumption of the unfiltered beeswax would contribute to the PTWI established for Hg by approximately 17%. Summing it all up, the contamination of the discussed bee products with Cd and Hg poses a low food safety risk. Nevertheless, beekeepers should prioritize placing their hives in a clean environment.

3.3.3. Dietary risk assessment of Pb and As detected in apicultural products

For elements with genotoxic carcinogenic potential, no tolerable intake values can be established. Instead, the use of the Margin of Exposure (MoE) approach is recommended. MoE is the quotient of a reference value (usually benchmark dose) and the estimated human exposure. It is a dimensionless number, the acceptability of which is determined by its magnitude. This approach is not applicable to assess the extent of a risk, but the relative magnitude of MoE values can provide useful information. For genotoxic carcinogenic substances, applying the “as low as reasonably achievable” (ALARA) approach is recommended irrespective of the calculated MoE (Cunningham et al., 2011).

In Table 3, the chronic risk assessments for lead and arsenic detected in beehive products are summarized. Products harvested from polluted areas are marked with an asterisk (*). Chronic exposures were calculated in the same way as for non-genotoxic carcinogenic elements. Assumed maximum daily intakes were estimated by taking into account the minimum margin of exposures sufficient to ensure that there is no appreciable human risk. Based on previous studies, a MoE value of 1 for arsenic (Menon et al., 2020) and 10 for lead (EFSA, 2010) are applicable for food safety risk assessments. Benchmark dose lower confidence limits were established by the European Food Safety Authority (EFSA, 2009; EFSA, 2010), of which the lowest values were taken into account during the estimation to present the worst-case scenario. Contributions

Table 2
Risk assessment of non-genotoxic carcinogenic elements in apicultural products.

Element	Product	Mean conc. ($\mu\text{g}/\text{kg}$)	Reference	Estimated daily exposure ^a (ng/kgbw/day)		PTMI/PTWI ^b	Tolerable daily intake ^c (ng/kgbw/day)	Contribution ^d (%)	
				Men	Women			Men	Women
Cadmium	bee bread	4	Aylanc et al. (2023)	0.57	0.67	PTMI = 25 $\mu\text{g}/\text{kgbw}$	833	0.07	0.08
	bee bread	60	Pavlova et al. (2021)	8.57	10.00			1.03	1.20
	bee bread	70	Pavlova et al. (2021)	10.00	11.67			1.20	1.40
	bee bread	27248	Mayda et al. (2020)	3893.00	4541.83			467.30	545.24
	bee bread	70	Murashova et al., 2019	10.00	11.67			1.20	1.40
	bee bread	28	Adaškevičiūtė et al., 2019	4.00	4.67			0.48	0.56
	bee bread	54	Bakour et al. (2019)	7.71	9.00			0.93	1.08
	bee bread	146	Omran et al. (2019)	20.86	24.34			2.50	2.92
	bee bread ^a	182	Omran et al. (2019)	26.00	30.33			3.12	3.64
	bee bread	71	Ciric et al. (2019)	10.14	11.83			1.22	1.42
	bee bread	136	Zhelyazkova (2018)	19.43	22.67			2.33	2.72
	bee bread	60	Golubkina et al. (2016)	8.57	10.00			1.03	1.20
	bee bread ^a	110	Golubkina et al. (2016)	15.71	18.33			1.89	2.20
	bee bread	510	Esmael et al. (2016)	72.86	85.00			8.75	10.20
	bee bread ^a	19	Roman et al. (2016)	2.71	3.16			0.33	0.38
	propolis	177	Ristivojevic et al. (2023)	7.59	8.86			0.91	1.06
	propolis	100	Mitek et al. (2022)	4.29	5.01			0.51	0.60
	propolis	177	Akbar et al. (2022)	7.59	8.86			0.91	1.06
	propolis ^a	80	Mititelu et al. (2022)	3.43	4.00			0.41	0.48
	propolis	16	Mititelu et al. (2022)	0.69	0.81			0.08	0.10
	propolis ^a	31	Conti et al. (2022)	1.33	1.55			0.16	0.19
	propolis	64	Soós et al. (2021)	2.74	3.20			0.33	0.38
	propolis	40	Matuszewska et al. (2021)	1.71	2.00			0.21	0.24
	propolis	421	Abdullah et al. (2020)	18.04	21.05			2.17	2.53
	propolis	1176	Ecem Bayram, 2020	50.39	58.80			6.05	7.06
	propolis	2	Hodel et al. (2020)	0.08	0.09			0.01	0.01
	propolis	780	Murashova et al., 2019	33.43	39.00			4.01	4.68
	propolis	31	Adaškevičiūtė et al., 2019	1.33	1.55			0.16	0.19
	propolis	72	Adaškevičiūtė et al., 2019	3.09	3.61			0.37	0.43
	propolis	22	Naco et al. (2017)	0.94	1.10			0.11	0.13
	propolis	22	Bogdanova Popov et al. (2017)	0.94	1.10			0.11	0.13
	propolis	70	Şahinler et al. (2017)	3.00	3.50			0.36	0.42
	propolis	162	Tosic et al. (2017)	6.94	8.10			0.83	0.97
	propolis	50	Golubkina et al. (2016)	2.14	2.50			0.26	0.30
	propolis ^a	20	Golubkina et al. (2016)	0.86	1.00			0.10	0.12
	propolis ^a	70	Golubkina et al. (2016)	3.00	3.50			0.36	0.42
	propolis ^a	20577	Matin et al. (2016)	881.87	1028.85			105.87	123.51
	propolis	182	Eskov et al. (2015)	7.80	9.10			0.94	1.09
	propolis	130	Finger et al. (2014)	5.57	6.50			0.67	0.78
	propolis ^a	28	Formicki et al. (2013)	1.20	1.40			0.14	0.17
	propolis	72	Serra Bonvehí and Orantes Bermejo (2013)	3.09	3.61			0.37	0.43
	propolis	600	Gong et al. (2012)	25.71	30.00			3.09	3.60
	propolis	1	Woo et al. (2012)	0.04	0.05			0.00	0.01
	propolis ^a	194	Roman et al. (2011)	8.31	9.70			1.00	1.16
	propolis	<LOQ	Cvek et al. (2008)	low	low			low	low
	propolis	250	Dobrinas et al. (2006)	10.71	12.50			1.29	1.50
	propolis ^a	6	Conti and Botré (2001)	0.26	0.30			0.03	0.04
	propolis	2	Conti and Botré (2001)	0.09	0.11			0.01	0.01
	wax ^a	13	Zafeiraki et al. (2022)	0.74	0.86			0.09	0.10
	wax	8	Ullah et al. (2022)	0.46	0.54			0.05	0.06
wax ^a	18	Conti et al. (2022)	1.03	1.20	0.12	0.14			
wax	46	Ngat et al. (2020)	2.63	3.07	0.32	0.37			
wax ^a	<LOQ	Ngat et al. (2020)	low	low	low	low			
wax	7	Murashova et al., 2019	0.40	0.47	0.05	0.06			
wax ^a	59	Aljedani (2020)	3.37	3.93	0.40	0.47			
wax ^a	60	Aljedani (2020)	3.43	4.00	0.41	0.48			
wax ^a	75	Aljedani (2020)	4.29	5.01	0.51	0.60			
wax	<LOQ	Aljedani (2020)	low	low	low	low			
wax	<LOQ	Astolfi et al. (2020)	low	low	low	low			
wax	<LOQ	Bommuraj et al. (2019)	low	low	low	low			
wax	20	Navarro-Hortal et al. (2019)	1.14	1.33	0.14	0.16			
wax	11	Navarro-Hortal et al. (2019)	0.63	0.74	0.08	0.09			
wax	12	Kosanović et al. (2019)	0.69	0.81	0.08	0.10			
wax	160	Omran et al. (2019)	9.14	10.66	1.10	1.28			
wax ^a	194	Omran et al. (2019)	11.09	12.94	1.33	1.55			
wax	150	Eskov et al. (2015)	8.57	10.01	1.03	1.20			
wax	44	Formicki et al. (2013)	2.51	2.93	0.30	0.35			

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Table 2 (continued)

Element	Product	Mean conc. ($\mu\text{g}/\text{kg}$)	Reference	Estimated daily exposure ^a (ng/kgbw/day)		PTMI/PTWI ^b	Tolerable daily intake ^c (ng/kgbw/day)	Contribution ^d (%)	
				Men	Women			Men	Women
Mercury	wax	6	Saunier et al. (2013)	0.34	0.40	PTWI = 4 $\mu\text{g}/\text{kgbw}$	571	0.04	0.05
	wax ^a	48	Conti and Botré (2001)	2.74	3.20			0.33	0.38
	wax	<18	Conti and Botré (2001)	low	low			low	low
	royal jelly	2	Matuszewska et al. (2021)	0.06	0.07			0.01	0.01
	royal jelly	<LOQ	Ecem Bayram et al. (2021)	low	low			low	low
	royal jelly	<LOQ	Astolfi et al. (2020)	low	low			low	low
	royal jelly	<LOQ	Murashova et al., 2019	low	low			low	low
	royal jelly	2	Adaškevičiūtė et al., 2019	0.06	0.07			0.01	0.01
	royal jelly	2	Adaškevičiūtė et al., 2019	0.06	0.07			0.01	0.01
	royal jelly	3	Balkanska et al. (2017)	0.09	0.11			0.01	0.01
	royal jelly	7	Saunier et al. (2013)	0.20	0.23			0.02	0.03
	royal jelly	11	Stocker et al. (2005)	0.31	0.36			0.04	0.04
	royal jelly	1	Stocker et al. (2005)	0.03	0.04			0.00	0.00
	bee bread	<LOQ	Mayda et al. (2020)	low	low			low	low
	bee bread	<LOQ	Murashova et al., 2019	low	low			low	low
	bee bread	<1	Ciric et al. (2019)	low	low			low	low
	bee bread	<LOQ	Golubkina et al. (2016)	low	low			low	low
	bee bread ^a	<LOQ	Golubkina et al. (2016)	low	low			low	low
	bee bread	1	Madras-Majewska and Jasiński (2005)	0.14	0.16			0.03	0.03
	propolis	50	Tutun et al. (2022)	2.14	2.50			0.38	0.44
	propolis ^a	9	Conti et al. (2022)	0.39	0.46			0.07	0.08
	propolis	4	Astolfi et al. (2021)	0.17	0.20			0.03	0.04
	propolis	<LOQ	Ecem Bayram, 2020	low	low			low	low
	propolis	<LOQ	Murashova et al., 2019	low	low			low	low
	propolis	<LOQ	Tosic et al. (2017)	low	low			low	low
	propolis	7	Golubkina et al. (2016)	0.30	0.35			0.05	0.06
	propolis ^a	<LOQ	Golubkina et al. (2016)	low	low			low	low
	propolis ^a	<LOQ	Golubkina et al. (2016)	low	low			low	low
	propolis ^a	<LOQ	Maragou et al. (2016)	low	low			low	low
	propolis ^a	<LOQ	Matin et al. (2016)	low	low			low	low
	propolis	8	Serra Bonvehí and Orantes Bermejo (2013)	0.34	0.40			0.06	0.07
	propolis	<LOQ	Woo et al. (2012)	low	low			low	low
	propolis	18	Cvek et al. (2008)	0.77	0.90			0.14	0.16
	wax	1	Kim et al. (2022)	0.06	0.07			0.01	0.01
	wax ^a	30	Zafeiraki et al. (2022)	1.71	2.00			0.30	0.35
	wax ^a	5	Conti et al. (2022)	0.29	0.34			0.05	0.06
	wax	6	Astolfi et al. (2021)	0.34	0.40			0.06	0.07
	wax	<LOQ	Ngat et al. (2020)	low	low			low	low
	wax ^a	<LOQ	Ngat et al. (2020)	low	low			low	low
	wax	<LOQ	Murashova et al., 2019	low	low			low	low
wax	62	Bommuraj et al. (2019)	3.54	4.13	0.62	0.72			
wax	18	Kosanović et al. (2019)	1.03	1.20	0.18	0.21			
wax	1700	Navarro-Hortal et al. (2019)	97.14	113.33	17.01	19.85			
wax	100	Navarro-Hortal et al. (2019)	5.71	6.66	1.00	1.17			
royal jelly	<LOQ	Ecem Bayram et al. (2021)	low	low	low	low			
royal jelly	<LOQ	Astolfi et al. (2021)	low	low	low	low			
royal jelly	<LOQ	Murashova et al., 2019	low	low	low	low			
royal jelly	21	Stocker et al. (2005)	0.60	0.70	0.11	0.12			
royal jelly	1	Stocker et al. (2005)	0.03	0.04	0.01	0.01			

^aCollected from polluted areas (urban, industrial, mining site etc.).

^a Determined by mean concentrations reported in scientific studies. Daily dosages of 10, 3, 4 and 2 g/day for bee bread, propolis, beeswax and royal jelly (Apiland, 2023; EFSA, 2020), as well as an assumed average body weight of 70 kg (men) or 60 kg (women) were considered (Rubio et al., 2017).

^b Provisional Tolerable Monthly Intake/Provisional Tolerable Weekly Intake reported by FAO/WHO (2019).

^c Determined by calculation.

^d Percent contribution of the estimated daily intake to the tolerable daily intake.

of the estimated exposures to the assumed maximum daily intake values were expressed as a percentage. As no information is available on the chemical form of arsenic in beehive products, calculations were conducted for inorganic arsenic, which also shows the worst-case scenario.

Results of the risk assessment suggest that the arsenic contamination of the products can be characterized by a low food safety risk, because the contribution of the estimated daily exposure values to the assumed maximum daily intake was generally below 10%. On the other hand, contribution values regarding the lead content of apicultural products varied between <1 and >10.000. Exceptionally high lead content

(123.630 mg/kg) was detected by Eskov and co-workers in a propolis sample harvested near the Moscow–Nizhny Novgorod highway. Nevertheless, several samples not intentionally harvested in polluted areas also exhibited concerning levels of lead contamination. Based on the data obtained, the lead content of the discussed hive products appears to be as high to pose a potential risk to their long-term consumers. The maximum level of lead concentration in certain foods are regulated at European level (Commission Regulation (EU) 2023/915). For food supplements, a maximum concentration level of 3000 $\mu\text{g}/\text{kg}$ is set, which is exceeded by a relatively large proportion of the samples tested in the

Table 3
Risk assessment of genotoxic carcinogenic elements in apicultural products.

Element	Product	Mean conc. (µg/kg)	Reference	Estimated daily intake ^a (ng/kgbw/day)		BMDL ₀₁ BMDL ₁₀ ^b (µg/kgbw/day)	Assumed maximum daily intake ^c (ng/kgbw/day)	Contribution ^d (%)	
				Men	Women			Men	Women
Lead	bee bread	40	Aylanc et al., 2023	5.71	6.66	BMDL ₀₁ : 0.5	50	11.43	13.32
	bee bread	130	Pavlova et al. (2021)	18.57	21.67			37.14	43.33
	bee bread	190	Pavlova et al. (2021)	27.14	31.66			54.29	63.33
	bee bread	308	Mayda et al. (2020)	44.00	51.33			88.00	102.67
	bee bread	250	Murashova et al., 2019	35.71	41.66			71.43	83.32
	bee bread	230	Adaškevičiūtė et al., 2019	32.86	38.34			65.71	76.67
	bee bread	70	Bakour et al. (2019)	10.00	11.67			20.00	23.33
	bee bread	1094	Omran et al. (2019)	156.29	182.34			312.57	364.68
	bee bread ^a	1338	Omran et al. (2019)	191.14	223.00			382.29	445.99
	bee bread	113	Ciric et al. (2019)	16.14	18.83			32.29	37.66
	bee bread	988	Zhelyazkova (2018)	141.14	164.66			282.29	329.33
	bee bread	210	Golubkina et al. (2016)	30.00	35.00			60.00	70.00
	bee bread ^a	340	Golubkina et al. (2016)	48.57	56.67			97.14	113.33
	bee bread	1240	Esmael et al. (2016)	177.14	206.66			354.29	413.33
	bee bread ^a	93	Roman et al. (2016)	13.29	15.51			26.57	31.01
	propolis	7849	Ristivojevic et al. (2023)	336.39	392.46			672.77	784.91
	propolis	1000	Milek et al. (2022)	42.86	50.00			85.71	100.01
	propolis	3834	Fidan et al. (2022)	164.31	191.70			328.63	383.39
	propolis	7313	Akbar et al. (2022)	313.41	365.65			626.83	731.29
	propolis	1200	Tutun et al. (2022)	51.43	60.00			102.86	120.00
	propolis ^a	651	Mititelu et al. (2022)	27.90	32.55			55.80	65.10
	propolis	160	Mititelu et al. (2022)	6.86	8.00			13.71	16.01
	propolis	150	Mutlu et al., 2023	6.43	7.50			12.86	15.00
	propolis ^a	350	Conti et al. (2022)	15.00	17.50			30.00	35.00
	propolis	660	Matuszewska et al. (2021)	28.29	33.01			56.57	66.01
	propolis	648	Abdullah et al., 2020	27.77	32.40			55.54	64.80
	propolis	218	Hodel et al. (2020)	9.34	10.90			18.69	21.79
	propolis	2260	Murashova et al., 2019	96.86	113.00			193.71	226.01
	propolis	6274	Adaškevičiūtė et al., 2019	268.89	313.71			537.77	627.41
	propolis	4600	Adaškevičiūtė et al., 2019	197.14	230.00			394.29	459.99
	propolis	37	Naco et al. (2017)	1.59	1.86			3.17	3.71
	propolis	37	Bogdanova Popov et al. (2017)	1.59	1.86			3.17	3.71
	propolis	1530	Şahinler et al. (2017)	65.57	76.50			131.14	153.00
	propolis	5205	Tosic et al. (2017)	223.07	260.25			446.14	520.50
	propolis	2080	Golubkina et al. (2016)	89.14	104.00			178.29	207.99
	propolis ^a	2350	Golubkina et al. (2016)	100.71	117.50			201.43	234.99
	propolis ^a	16070	Golubkina et al. (2016)	688.71	803.50			1377.43	1606.99
	propolis ^a	357	Matin et al. (2016)	15.30	17.85			30.60	35.70
	propolis	123630	Eskov et al. (2015)	5298.43	6181.50			10596.86	12363.00
	propolis	10044	González-Martín et al. (2015)	430.46	502.20			860.91	1004.41
	propolis	2600	González-Martín et al. (2015)	111.43	130.00			222.86	260.00
	propolis	9850	Finger et al. (2014)	422.14	492.50			844.29	984.99
	propolis ^a	1676	Formicki et al. (2013)	71.83	83.80			143.66	167.60
	propolis	5615	Pierini et al. (2013)	240.64	280.75			481.29	561.49
	propolis	1470	Serra Bonvehí and Orantes Bermejo (2013)	63.00	73.50			126.00	147.00
	propolis	19920	Gong et al. (2012)	853.71	996.00			1707.43	1991.99
	propolis	104	Woo et al. (2012)	4.46	5.20			8.92	10.40
	propolis ^a	5740	Roman et al. (2011)	246.00	287.00			492.00	574.00
	propolis	2818	Cvek et al. (2008)	120.77	140.90			241.54	281.80
	propolis	60	Dobrinas et al. (2006)	2.57	3.00			5.14	6.00
propolis ^a	4	Conti and Botré (2001)	0.17	0.20	0.34	0.40			
propolis	2	Conti and Botré (2001)	0.09	0.11	0.17	0.21			
wax	11	Kim et al. (2022)	0.63	0.74	1.26	1.47			
wax ^a	691	Zafeiraki et al. (2022)	39.49	46.07	78.97	92.14			
wax	115	Ullah et al. (2022)	6.57	7.67	13.14	15.33			
wax ^a	560	Conti et al. (2022)	32.00	37.33	64.00	74.67			
wax	644	Ngat et al. (2020)	36.80	42.93	73.60	85.87			
wax ^a	525	Ngat et al. (2020)	30.00	35.00	60.00	70.00			
wax	180	Murashova et al., 2019	10.29	12.01	20.57	24.01			
wax ^a	114	Aljedani (2020)	6.51	7.60	13.03	15.19			
wax ^a	137	Aljedani (2020)	7.83	9.14	15.66	18.27			
wax ^a	215	Aljedani (2020)	12.29	14.34	24.57	28.68			
wax	<LOQ	Aljedani (2020)	low	low	low	low			
wax	255	Bommuraj et al. (2019)	14.57	17.00	29.14	34.00			
wax	3500	Navarro-Hortal et al. (2019)	200.00	233.33	400.00	466.67			
wax	2900	Navarro-Hortal et al. (2019)	165.71	193.33	331.43	386.66			

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Table 3 (continued)

Element	Product	Mean conc. ($\mu\text{g}/\text{kg}$)	Reference	Estimated daily intake ^a (ng/kgbw/ day)		BMDL ₀₁ BMDL ₁₀ ^b ($\mu\text{g}/\text{kgbw}/\text{day}$)	Assumed maximum daily intake ^c (ng/kgbw/day)	Contribution ^d (%)	
				Men	Women			Men	Women
	wax	1750	Omran et al. (2019)	100.00	116.67			200.00	233.33
	wax ^a	1388	Omran et al. (2019)	79.31	92.53			158.63	185.06
	wax ^a	4060	Gajger et al. (2019)	232.00	270.67			464.00	541.33
	wax ^a	5200	Gajger et al. (2019)	297.14	346.66			594.29	693.33
	wax	1230	Gajger et al. (2019)	70.29	82.01			140.57	164.01
	wax	2770	Gajger et al. (2019)	158.29	184.67			316.57	369.34
	wax ^a	2500	Gajger et al. (2019)	142.86	166.67			285.71	333.34
	wax ^a	5430	Gajger et al. (2019)	310.29	362.01			620.57	724.01
	wax	736	Eskov et al. (2015)	42.06	49.08			84.12	98.16
	wax	1914	Formicki et al. (2013)	109.37	127.60			218.74	255.20
	wax	<LOQ	Saunier et al. (2013)	low	low			low	low
	wax ^a	193	Conti and Botré (2001)	11.03	12.87			22.06	25.74
	wax	103	Conti and Botré (2001)	5.89	6.87			11.77	13.74
	royal	70	Matuszewska et al. (2021)	2.00	2.33			4.00	4.67
	jelly								
	royal	<LOQ	Ecem Bayram et al. (2021)	low	low			low	low
	jelly								
	royal	2800	Murashova et al., 2019	80.00	93.33			160.00	186.67
	jelly								
	royal	319	Adaškevičiūtė et al., 2019	9.11	10.63			18.23	21.26
	jelly								
	royal	418	Adaškevičiūtė et al., 2019	11.94	13.93			23.89	27.86
	jelly								
	royal	150	Balkanska et al. (2017)	4.29	5.01			8.57	10.01
	jelly								
	royal	168	Saunier et al. (2013)	4.80	5.60			9.60	11.20
	jelly								
	royal	99	Stocker et al. (2005)	2.83	3.30			5.66	6.60
	jelly								
	royal	51	Stocker et al. (2005)	1.46	1.70			2.91	3.41
	jelly								
Arsenic	bee bread	138	Mayda et al. (2020)	19.71	23.00	BMDL ₀₁ : 0.3	300	6.57	7.67
	bee bread	<LOQ	Murashova et al., 2019	low	low			low	low
	bee bread	32	Ciric et al. (2019)	4.57	5.33			1.52	1.78
	bee bread	40	Golubkina et al. (2016)	5.71	6.66			1.90	2.22
	bee	30	Golubkina et al. (2016)	4.29	5.01			1.43	1.67
	bread ^a								
	bee bread	<LOQ	Esmael et al. (2016)	low	low			low	low
	bee	325	Roman et al. (2016)	46.43	54.17			15.48	18.06
	bread ^a								
	propolis	141	Ristivojevic et al. (2023)	6.04	7.05			2.01	2.35
	propolis	680	Tutun et al. (2022)	29.14	34.00			9.71	11.33
	propolis ^a	80	Conti et al. (2022)	3.43	4.00			1.14	1.33
	propolis	70	Matuszewska et al. (2021)	3.00	3.50			1.00	1.17
	propolis	3205	Abdullah et al. (2020)	137.36	160.25			45.79	53.42
	propolis	720	Hodel et al. (2020)	30.86	36.00			10.29	12.00
	propolis	<LOQ	Murashova et al., 2019	low	low			low	low
	propolis	<LOQ	Tosic et al. (2017)	low	low			low	low
	propolis	60	Golubkina et al. (2016)	2.57	3.00			0.86	1.00
	propolis ^a	45	Golubkina et al. (2016)	1.93	2.25			0.64	0.75
	propolis ^a	170	Golubkina et al. (2016)	7.29	8.51			2.43	2.84
	propolis ^a	163	Matin et al. (2016)	6.99	8.16			2.33	2.72
	propolis	91	Serra Bonvehí and Orantes Bermejo (2013)	3.90	4.55			1.30	1.52
	propolis	<LOQ	Gong et al. (2012)	low	low			low	low
	propolis	<LOQ	Woo et al. (2012)	low	low			low	low
	propolis	223	Cantarelli et al. (2011)	9.56	11.15			3.19	3.72
	propolis ^a	657	Roman et al. (2011)	28.16	32.85			9.39	10.95
	propolis	448	Cvek et al. (2008)	19.20	22.40			6.40	7.47
	wax	2	Kim et al. (2022)	0.11	0.13			0.04	0.04
	wax ^a	23	Zafeiraki et al. (2022)	1.31	1.53			0.44	0.51
	wax ^a	<30	Conti et al. (2022)	low	low			low	low
	wax	19	Ngat et al. (2020)	1.09	1.27			0.36	0.42
	wax ^a	80	Ngat et al. (2020)	4.57	5.33			1.52	1.78
	wax	<LOQ	Murashova et al., 2019	low	low			low	low
	wax	<LOQ	Astolfi et al. (2020)	low	low			low	low
	wax	<LOQ	Bommuraj et al. (2019)	low	low			low	low
	wax	22	Kosanović et al. (2019)	1.26	1.47			0.42	0.49
	wax	15	Navarro-Hortal et al. (2019)	0.86	1.00			0.29	0.33
	wax	48	Navarro-Hortal et al. (2019)	2.74	3.20			0.91	1.07
	wax	12	Saunier et al. (2013)	0.69	0.81			0.23	0.27

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Table 3 (continued)

Element	Product	Mean conc. ($\mu\text{g}/\text{kg}$)	Reference	Estimated daily intake ^a (ng/kgbw/day)		BMDL ₀₁ BMDL ₁₀ ^b ($\mu\text{g}/\text{kgbw}/\text{day}$)	Assumed maximum daily intake ^c (ng/kgbw/day)	Contribution ^d (%)	
				Men	Women			Men	Women
	royal jelly	10	Matuszewska et al. (2021)	0.29	0.34			0.10	0.11
	royal jelly	<LOQ	Ecem Bayram et al. (2021)	low	low			low	low
	royal jelly	<LOQ	Astolfi et al. (2020)	low	low			low	low
	royal jelly	20	Murashova et al., 2019	0.57	0.67			0.19	0.22
	royal jelly	25	Balkanska et al. (2017)	0.71	0.83			0.24	0.28
	royal jelly	5	Saunier et al. (2013)	0.14	0.16			0.05	0.05

^aCollected from polluted areas (urban, industrial, mining site etc.).

^a Determined by mean concentrations reported in scientific studies. Daily dosages of 10, 3, 4 and 2 g/day for bee bread, propolis, beeswax and royal jelly (Apiland, 2023; EFSA, 2020), as well as an assumed average body weight of 70 kg (men) or 60 kg (women) were considered (Rubio et al., 2017).

^b Benchmark dose lower confidence limit (BMDL) values established by EFSA (EFSA, 2009; EFSA, 2010).

^c Calculated by taking into a MoE value of 1 for arsenic (Menon et al., 2020) and 10 for lead (EFSA, 2010).

^d Percent contribution to the assumed maximum daily intake.

reviewed studies, especially in the case of propolis and beeswax. Based on these results, these products may not be recommended for children as they can store less Pb in their bones and are more susceptible to the excessive exposure to this element (El-Kady and Abdel-Wahhab, 2018). Our observations suggest that specific regulations need to be established at international level to reduce the food safety risk posed by the presence of inorganic contaminants in beehive products other than honey.

4. Conclusions and future perspectives

Due to the rapid pace of urbanization, land-use changes, and industrialization, the occurrence of inorganic contaminants in foodstuffs has highly increased in the last decades. The contamination of apicultural products with trace metals and metalloids is ubiquitous and becoming an important subject of research. Bees and their products serve as bioindicators because they reflect the degree of pollution in the hive environment. In our study, the possible sources of contamination of bee bread, propolis, beeswax and royal jelly with potentially toxic trace elements are discussed. Available data on the concentration of trace metals in these products are summarized in our review. Besides, chronic dietary risk assessments are conducted on the cadmium, mercury, lead, and arsenic exposure arising from the consumption of the discussed products.

A great variability can be observed between the results of different studies regarding the potentially toxic trace element concentrations in hive products because it is highly influenced by external factors, such as soil types, climatic conditions, processing, and the level of pollution in the hive environment. As the element composition of bee bread and propolis is also affected by their botanical origin, it is advisable to use monofloral samples of selected plants in future research. According to our observations, the trace element composition of propolis is widely researched, whereas royal jelly has received limited attention regarding its trace element content. The Pb and Cd content of apicultural products were investigated by a large proportion of the presented studies, however, further research is needed on the As and Hg contamination. Essential trace elements are generally present in multiple concentrations compared to Cd, Hg, Pb and As, although, the latter may also accumulate to large extent in products originated from areas exposed to environmental pollution.

Results of the risk assessments indicate that the lead contamination of hive products pose a significant health risk to their long-term consumers, especially to children. Consequently, specific international

regulations need to be established to minimize food safety risks posed by the trace metal content of apicultural products. Based on available data, it is advisable for beekeepers to locate beehives in ecologically clean areas in order to protect consumers of apicultural products. Nevertheless, additional investigations need to be carried out to evaluate the trace metal content of beehive products originated from organically managed apiaries. Further studies are needed on the chemical species of the trace elements present in apicultural products, because it has a significant impact on their toxicity. Research should also be conducted on the possible decontamination processes that may be applicable to reduce the concentration of inorganic contaminants in beehive products.

Author contribution

Rita Végh: Conceptualization, Methodology, Investigation, Writing-Original Draft, Review & Editing.; Mariann Csóka: Supervision, Writing-Original Draft, Review & Editing.; Zsuzsanna Mednyánszky: Writing-Original Draft, Review & Editing.; László Sipos: Conceptualization, Methodology, Supervision, Writing-Original Draft, Review & Editing, Funding acquisition.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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