

Toxicological Assessment of Algerian Honeys: Heavy Metal Contamination as an Indicator of Environmental and Public Health Risks [†]

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[†] Presented at the 29th International Electronic Conference on Synthetic Organic Chemistry (ECSOC-29); Available online: <https://sciforum.net/event/ecsoc-29>.

Abstract

Honey, beyond its nutritional and therapeutic value, can act as a natural bioindicator of environmental quality. In this study, fourteen unifloral and multifloral honeys from northern Algeria were analysed for toxic metals using atomic absorption spectrometry and inductively coupled plasma mass spectrometry. Concentrations ranged as follows: Cu (0.133–1.975 µg/kg), Fe (1.11–28.04 mg/kg), Zn (4.40–26.30 mg/kg), Cd (1.119–39.521 µg/kg), and Pb (11.515–77.216 µg/kg). Some samples, particularly for Cd and Pb, exceeded international safety limits, indicating potential health risks. The results confirm the importance of routine monitoring and highlight honey's relevance in food safety and environmental surveillance.

Keywords: honey; heavy metals; AAS; ICP-MS; contamination; food safety

Academic Editor(s): Name

Published: date

Citation: Tani, N.K.; Allali, H.; Puścion-Jakubik, A.; Aissaoui, N.; Socha, K. Toxicological Assessment of Algerian Honeys: Heavy Metal Contamination as an Indicator of Environmental and Public Health Risks. *Chem. Proc.* **2025**, volume number, x. <https://doi.org/10.3390/xxxxx>

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1. Introduction

Honey is a natural product highly valued for its nutritional richness, therapeutic potential, and cultural significance. Beyond its role as food, it serves as an effective environmental indicator because its composition reflects both botanical sources and ecological conditions surrounding the hive [1]. Bees forage over wide areas, during which they not only collect nectar but also gather particles of pollutants and trace metals, which may later be transferred into honey [2]. Consequently, honey provides an integrated view of environmental quality and potential consumer exposure.

Heavy metals are particularly concerning due to their persistence and bioaccumulative nature. Essential elements such as Cu, Zn, and Fe play physiological roles at trace levels, whereas Cd and Pb are toxic even in small amounts [3]. Exposure to these contaminants has been associated with renal and hepatic dysfunction, neurodegenerative disorders including Alzheimer's and Parkinson's diseases, and increased carcinogenic risk [4,5].

The release of these metals into ecosystems is amplified by anthropogenic activities such as industry, traffic, and agriculture, raising the likelihood of their entry into the food chain.

Several investigations have demonstrated the utility of honey as a biomonitor of environmental contamination, as its mineral and heavy metal composition reflects both soil and vegetation in the surrounding habitat [6]. Furthermore, such analyses provide insight into honey's nutritional and functional qualities, since minerals contribute to bioactivity [7]. Despite Algeria's favourable climate, diverse flora, and strong beekeeping tradition, systematic studies on heavy metal contamination in Algerian honeys remain limited [8].

The present study therefore investigates toxic elements—Cu, Fe, Zn, Cd, and Pb—in fourteen unifloral and multifloral honeys from northern Algeria. Its purpose is to evaluate potential health risks associated with consumption and to examine honey's role as a natural tool for environmental surveillance.

2. Materials and Methods

2.1. Honey Samples

Fourteen honey samples (MH1–MH14), representing both unifloral and multifloral varieties, were collected during the 2022 production season from beekeepers operating in several regions of Algeria, including Tlemcen, Sidi Bel Abbes, Mostaganem, Mascara, Tiaret, El-Bayadh, Naâma (Mechria), Laghouat (Aflou), Chlef, Aïn Defla, Medea, and Tebessa (Figure 1). Their floral sources included *Nasturtium officinale*, *Thymus ciliatus* subsp. *coloratus*, *Scolymus hispanicus*, *Eucalyptus globulus*, *Citrus sinensis*, *Euphorbia guyoniana*, *Ziziphus lotus*, *Petroselinum crispum*, *Daucus carota*, and *Rosmarinus officinalis* (Table 1). All samples were coded, stored in sterilised glass containers, and maintained at 4 °C until further analysis.



Figure 1. Sampling map of Algerian honey collection sites.

Table 1. Botanical origin and geographical source of honey samples.

Region	Sample	Scientific Name	Season/Year of Harvest
Tlemcen	MH 1	<i>Nasturtium officinale</i> R.Br.	Spring 2022
	MH 2	<i>Thymus ciliatus</i> subsp <i>coloratus</i>	Spring 2022
Sidi Bel Abbes	MH 3	<i>Scolymus hispanicus</i> L.	Spring 2022
Mostaganem	MH 4	<i>Eucalyptus globulus</i> Labill.	Summer 2022
	MH 5	<i>Citrus sinensis</i> L.	Spring 2022
Mascara	MH 6	<i>Citrus sinensis</i> L.	Spring 2022

Tiaret	MH 7	<i>Eucalyptus globulus</i> Labill.	Summer 2022
El-Bayadh	MH 8	<i>Euphorbia guyoniana</i> Boiss. & Reut.	Summer 2022
Naâma (Mechria)	MH 9	<i>Ziziphus lotus</i> L.	Summer 2022
Laghouat (Aflou)	MH 10	<i>Euphorbia guyoniana</i> Boiss. & Reut.	Spring 2022
Chlef	MH 11	<i>Petroselinum crispum</i> (Mill.) Fuss.	Spring 2022
Aïn Defla	MH 12	<i>Eucalyptus globulus</i> Labill.	Summer 2022
Medea	MH 13	<i>Daucus carota</i> L.	Spring 2022
Tebessa	MH 14	<i>Rosmarinus officinalis</i> L.	Spring 2022

2.2. Reagents and Solutions

All chemicals were of analytical grade. Ultrapure concentrated nitric acid (69%) and certified standard solutions of cadmium [$\text{Cd}(\text{NO}_3)_2$ in 0.5 mol/L HNO_3 , 1000 mg/L] and lead [$\text{Pb}(\text{NO}_3)_2$ in 0.5 mol/L HNO_3 , 1000 mg/L] were obtained from Merck (Darmstadt, Germany). Calibration solutions for Fe, Cu, and Zn were prepared by serial dilution of 1 g/L stock standards with ultrapure water generated using a Millipore Simplicity UV purification system.

2.3. Sample Homogenisation and Digestion

Each honey sample was first homogenised with a T 18 digital Ultra-Turrax (IKA, Staufen, Germany) to achieve a uniform texture. Portions of 200–300 mg were accurately weighed into PTFE vessels, mixed with 4 mL of spectrally pure HNO_3 , and subjected to closed-vessel microwave digestion (Speedwave, Berghof, Germany). The digestion process followed a controlled temperature–pressure ramp, progressively increasing from 170 °C at 20 atm to 190 °C at 30 atm and finally to 210 °C at 40 atm, with corresponding microwave powers of 80%, 90%, and 90%, respectively, each maintained for 10 min. After digestion, the system was cooled to 50 °C under 40 atm for 18 min with zero microwave power to stabilise the solutions. Mineralised samples were then quantitatively transferred to polypropylene tubes, diluted with ultrapure water, and stored at –20 °C pending analysis.

2.4. Determination of Mineral Elements

Iron was quantified by flame atomic absorption spectrometry (AAS) at 248.3 nm with Zeeman background correction. Samples were diluted to fit within a calibration range up to 5 mg/L, and the detection limit was 0.19 mg/kg. Copper and zinc were determined using a Hitachi Z-2000 spectrometer, employing electrothermal atomisation for Cu (324.8 nm) and flame AAS for Zn (213.9 nm), with detection limits of 0.53 µg/L and 0.013 mg/L, respectively.

2.5. Trace Metal Determination by ICP-MS

Cadmium and lead concentrations were measured by inductively coupled plasma mass spectrometry (ICP-MS, NexION 300D, PerkinElmer, USA) under standard operating conditions. Kinetic energy discrimination and collision cell technology were applied to minimise polyatomic interferences. Method detection limits, determined from ten replicate blanks ($3 \times \text{SD}$), were 0.017 µg/kg for Cd and 0.16 µg/kg for Pb.

All measurements were performed in triplicate, and results were expressed on a wet weight basis.

2.6. Data Processing and Statistical Analysis

Results are expressed as mean $\pm$ standard deviation from triplicate measurements. Statistical analyses were performed to compare metal concentrations among honeys of

different botanical and geographical origins. Pearson's correlation coefficient was applied to examine relationships between elements, with statistical significance set at $p < 0.05$.

3. Results and Discussion

3.1. Elemental Content of Algerian Honey Samples

The analysis of fourteen Algerian honey samples (Table 1) revealed a wide range of concentrations for five key elements. The measured ranges were: Iron (Fe) (1.11–28.04 mg/kg), Copper (Cu) (0.133–1.975 $\mu\text{g/kg}$), Zinc (Zn) (4.40–26.30 mg/kg), Cadmium (Cd) (1.119–39.521 $\mu\text{g/kg}$), and Lead (Pb) (11.515–77.216 $\mu\text{g/kg}$). These concentrations varied considerably across the samples, reflecting their diverse floral and geographical origins, a finding consistent with other studies [8,9]. The distribution of these elemental concentrations is graphically represented in the histogram in Figure 2.

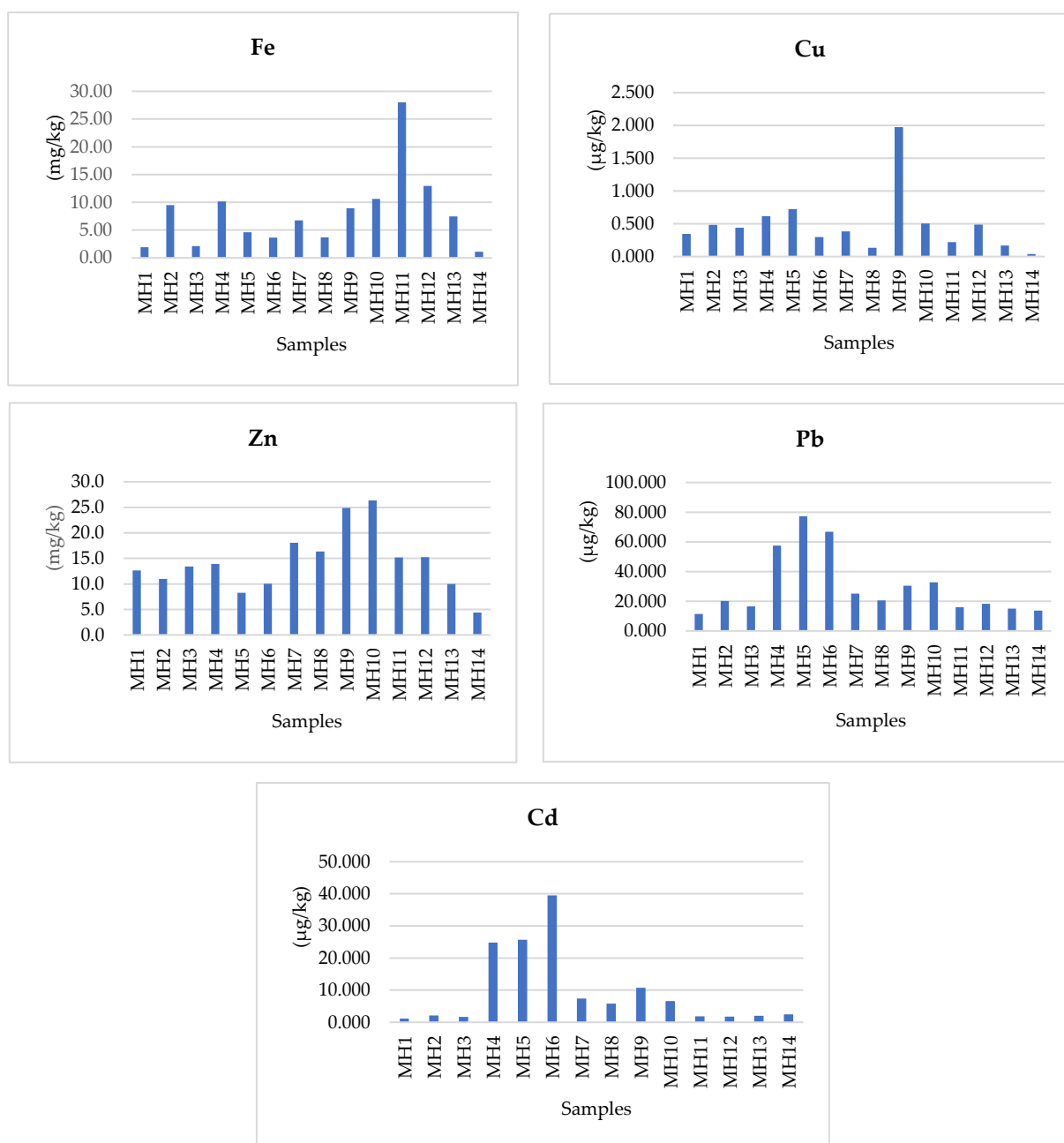


Figure 2. Distribution of mineral concentrations across studied honey samples.

3.2. Essential Micronutrients: Fe, Cu, and Zn

The nutritional and therapeutic properties of honey are closely linked to its mineral content. Findings on the concentrations of essential micronutrients, such as Fe, Cu, and Zn, highlight both the benefits and potential concerns associated with honey consumption.

The iron content of the Algerian honeys analysed ranged from 1.11 mg/kg in rosemary honey (MH14; Tébéssa) to 28.04 mg/kg in parsley honey (MH11; Chlef) (Figure 2). This range places them within or above the concentrations reported for honeys from other Mediterranean countries, such as Morocco (1.46–13.95 mg/kg) [10], Tunisia (0.83–3.54 mg/kg) [11], and Portugal (0.18–2.68 mg/kg) [12]. A comparison with a previous Algerian study (8.48–59.60 mg/kg) [8] shows that the concentrations observed in the present samples are largely of the same order of magnitude. This higher iron content could be attributed to the specific Algerian floral sources and soil composition. The notably high Fe concentration in parsley honey (MH11) is consistent with the fact that this plant is naturally rich in iron.

From a food safety perspective, the Provisional Tolerable Weekly Intake (PTWI) for iron is set at 5.6 mg/kg b.w. by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) [13]. While the majority of the honeys examined contained iron levels below this guideline, the high concentrations observed in some samples, such as parsley honey (MH11), suggest a potential dietary risk, particularly for vulnerable groups like children, if consumed in large quantities.

Copper concentrations ranged from 0.037 ± 0.043 µg/kg in rosemary honey (MH14) from Tébéssa to 1.975 ± 2.375 µg/kg in jujube tree honey (MH9) from Naâma. These values are notably lower than those reported for honeys from Morocco (≤ 0.1 mg/kg) [10] and Tunisia (0.12–0.34 mg/kg) [11], but fall within the lower end of the wide ranges found in Portugal (0.00–5.35 mg/kg) [12] and France (0.06–1.71 mg/kg) [14] (Table 2). The significant variation in copper content observed in the samples highlights the influence of both botanical origin and geographical location. The highest concentration was found in the jujube tree honey (MH9) from the semi-arid region of Naâma, a value considerably higher than that of the rosemary honey (MH14) from the more temperate region of Tébéssa. This suggests that the composition of the soil and the local environment directly impact the accumulation of copper in the honey.

Regarding food safety, while copper is an essential element, an excessive intake can be toxic. The Provisional Tolerable Weekly Intake (PTWI) for Cu is set at 3.5 mg/kg b.w. [15]. It is reassuring that the highest copper concentration recorded in this study is well below this established threshold. This confirms that the consumption of these honeys does not pose a toxicological risk from a copper standpoint. Elevated copper concentrations in honey are generally attributed to environmental factors and human activities, such as the use of fungicides, fertilisers, and pesticides in agricultural areas [16].

Zinc concentrations in the analysed honeys were found to be exceptionally high, ranging from 4.40 mg/kg to 26.30 mg/kg. These values exceed those reported for honeys from Morocco (≤ 0.1 –0.69 mg/kg) [10], Tunisia (0.42–2.06 mg/kg) [11], Portugal (0.03–3.29 mg/kg) [12], and Spain (2.34–3.47 mg/kg) [17]. A significant variation in zinc content was observed between different botanical origins. The highest level, at 26.30 mg/kg, was detected in spurge honey (MH10) from the region of Laghouat, a concentration notably higher than the minimum recorded value of 4.40 mg/kg in a sample of rosemary honey (MH14) from Tébéssa. As an essential micronutrient, zinc is vital for human health, playing a key role in immune function, wound healing, and numerous metabolic processes. Furthermore, a significant positive correlation was found between the concentrations of Fe and Zn ($p < 0.05$). This finding indicates that these two elements are likely co-absorbed by the plants from the soil, suggesting a common geological or geochemical origin for their presence in the honeys [18].

3.3. Toxic Elements: Pb and Cd and Their Health Impacts

The study also focused on potentially toxic elements, namely lead and cadmium, which are well-known for their negative impacts on living organisms and the environment [19]. The concentrations of these toxic elements decreased in the order of Pb > Cd.

The concentration of lead in the honeys analysed ranged from 11.515 µg/kg in watercress honey (MH1; Tlemcen) to 77.216 µg/kg in orange tree honey (MH5; Mostaganem). A comparison with international data (Table 2) reveals that these concentrations are notably higher than those reported for honeys from Tunisia (0.01–0.05 mg/kg) [11], but are comparable to or slightly lower than those from Morocco (≤ 0.1 mg/kg) [10]. They remain well below the wide ranges reported for France (0.28–1.08 mg/kg) [14] and Italy (9–209 mg/kg) [20].

The highest lead levels were concentrated in honeys from specific geographical origins. The maximum concentration, at 77.216 ± 14.313 µg/kg, was found in orange tree honey (MH5) from Mostaganem. Other significant values were also recorded in jujube tree honey (MH9) from Naâma (30.451 ± 10.345 µg/kg), spurge honey (MH10) from Laghouat (32.749 ± 0.045 µg/kg), and orange tree honey (MH6) from Mascara (66.912 ± 33.272 µg/kg). These elevated levels suggest a potential link to external contamination from sources such as traffic or industrial emissions, particularly as these honeys were collected from major agricultural and potentially polluted areas.

With regard to food safety, the Provisional Tolerable Weekly Intake (PTWI) for Pb is set at 25 µg/kg b.w. for adults [21]. It is a cause for concern that several samples, including MH5, MH6, MH9, and MH10, exhibited concentrations exceeding this established threshold. These findings indicate a potential dietary risk, with the elevated concentrations being of particular concern for children due to their lower body mass and higher susceptibility.

Analysis of the honeys revealed cadmium concentrations varying between 1.119 ± 0.580 µg/kg in watercress honey (MH1; Tlemcen) and a maximum of 39.521 ± 23.609 µg/kg in orange tree honey (MH6; Mascara). These values are significantly higher than those reported for honey from Turkey (<1 µg/kg) [6] and France (0.08–0.25 µg/kg) [14] (Table 2). Certain samples showed remarkably high levels, signalling a potential food safety concern. Besides the highest value in orange tree honey (MH6), other samples with elevated concentrations included eucalyptus honey (MH4) from Mostaganem (24.837 ± 19.339 µg/kg) and jujube tree honey (MH9) from Naâma (10.733 ± 5.462 µg/kg). The presence of such high levels is particularly worrying as exposure to cadmium is associated with a long biological half-life and the element is classified as a Group 1 carcinogen [22].

A highly significant positive correlation was observed between the concentrations of Cd and Pb ($p < 0.05$), strongly suggesting a common origin of contamination for both elements, likely stemming from local anthropogenic activities such as industrial pollution or agricultural runoff.

Table 2. Literature-based concentrations of trace elements in Mediterranean honeys for comparative assessment.

Element	Present Study	Algeria [8]	Morocco [10] N = 29	Tunisia [11] N = 6	Portugal [12] N = 16	France [14] N = 86	Spain [17] N = 140	Italy [20] N = 40	Turkey [6] N = 71
Fe	1.11–28.04	8.48–59.60	1.46–13.95	0.83–3.54	0.18–2.68	0.56–86.76	2.26–4.70	<1–4.4	<1–7254.62 µg/kg
Cu	0.133–1.975 µg/kg	1.66–9.62	≤ 0.1	0.12–0.34	0.00–5.35	0.06–1.71	0.74–1.88	0.06–5.4	<1–929 µg/kg
Zn	4.40–26.30	0.22–13.90	≤ 0.1 –0.69	0.42–2.06	0.03–3.29	0.17–6.42	2.34–3.47	<0.5–8.9	<1–237 µg/kg

Pb	11.515–77.216 µg/kg	0.54–132.73 µg/kg	≤0.1	0.01–0.05	-	0.28–1.08	46.32–31.50 µg/kg	9–209	<1 µg/kg
Cd	1.119–39.521 µg/kg	0.24–8.14 µg/kg	-	-	-	0.08–0.25	4.21–4.56 µg/kg	1.3–4.2	<1 µg/kg

From each report the min-max range is mentioned. All values are in mg/kg, unless stated otherwise. N: the number of honey samples.

3.4. Honey as a Bioindicator of Environmental Quality

Honey's composition is a direct reflection of its surrounding environment, making it an excellent natural bioindicator for monitoring environmental quality and pollution. The significant variations in elemental content among the samples, even from the same botanical source but different regions (e.g., spurge honey from El-Bayadh and Laghouat, or eucalyptus honey from Mostaganem and Tiaret), underscore the link between honey composition and local human pressures. The unexpectedly high levels of Cd and Pb in certain samples point to potential environmental pollution sources, likely stemming from vehicle emissions, industrial activity, or agricultural runoff [19]. Using honey as a tracer for the spatial analysis of these pollutants can provide a low-cost and effective tool for ecological assessment in Algeria.

4. Conclusions

The analysis of Algerian honeys revealed that their mineral and heavy metal content varies considerably depending on their botanical and geographical origin. High levels of essential elements, such as iron and zinc, indicate a superior nutritional quality and promising characteristics for these honeys. However, the presence of lead and cadmium at concentrations exceeding safety standards in some samples poses a potential health risk and confirms the role of honey as an environmental sentinel against pollution. The significant positive correlation between cadmium and lead also suggests a common source of anthropogenic contamination. Consequently, rigorous monitoring of these products is essential to not only guarantee consumer safety but also to secure a premium market position, promoting both public health and sustainable apiculture.

Author Contributions: Conceptualization, H.A.; methodology, H.A. and A.P.-J.; software, N.K.T.; validation, N.K.T., N.A. and A.P.-J.; formal analysis, N.K.T.; investigation, N.K.T., N.A. and A.P.-J.; resources, H.A. and K.S.; data curation, N.K.T.; writing—original draft preparation, H.A.; writing—review and editing, H.A.; visualization, H.A.; supervision, H.A. and A.P.-J.; project administration, H.A.; funding acquisition, H.A. and K.S. All authors have read and agreed to the published version of the manuscript.

Funding: Funding for this study was provided by the PRFU project B00L01UN130120220004 (Ministry of Higher Education and Scientific Research, Algeria; Abou Bekr Belkaïd University, Tlemcen). The authors declare no additional funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: All relevant data are contained within the article.

Acknowledgments: The authors gratefully acknowledge the institutional support provided by the Ministry of Higher Education and Scientific Research and Abou Bekr Belkaïd University, Tlemcen. Special thanks are extended to Professor Katarzyna Socha of the Department of Bromatology, Medical University of Białystok, for her invaluable collaboration and assistance in the realization of the mineral analyses. Her contribution was instrumental to the successful completion of this work.

Conflicts of Interest: The authors declare no conflict of interest.

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