

## Determination of Botanical Origin and Mineral Content of Propolis Samples from Balveren (Şırnak) Beekeepers Accommodation Areas

Mehmet FİDAN<sup>1</sup>, Süleyman Mesut PINAR<sup>2</sup>, Mehmet Emre EREZ<sup>3\*</sup>, Behçet İNAL<sup>4</sup>, Hüseyin EROĞLU<sup>2</sup>

<sup>1</sup>Siirt University, Faculty of Art and Science, Department of Biology, Siirt, TÜRKİYE

<sup>2</sup>Van Yüzüncü Yıl University, Faculty of Science, Department of Biology, Van, TÜRKİYE

<sup>3</sup>Van Yüzüncü Yıl University, Faculty of Science, Department of Molecular Biology and Genetics, Van, TÜRKİYE

<sup>4</sup>Siirt University, Agriculture Faculty, Department of Agricultural Biotechnology, Siirt, TÜRKİYE

ORCID ID: Mehmet FİDAN: <https://orcid.org/0000-0002-0255-9727>; Süleyman Mesut PINAR: <https://orcid.org/0000-0002-1774-7704>; Mehmet Emre EREZ: <https://orcid.org/0000-0002-4944-365X>; Behçet İNAL: <https://orcid.org/0000-0003-2215-2710>; Hüseyin EROĞLU: <https://orcid.org/0000-0001-9171-5607>

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**Abstract:** Researches on bee products have become popular in recent years. In fact, the content and component of bee products varies depending on many ecological and floristic factors and its nutritional and therapeutic properties are directly related to its content. Balveren (Şırnak province) beekeepers place their hives in locations with different geographical structure, floristic and topographic characteristics. This variability not only affects the quality of honey but also changes the properties of propolis. Studies on propolis, known as bee glue, have gained importance in recent years. As with other bee products, the propolis content also depends on the floristic characteristics of the region. In this study, propolis samples were collected from the regions where Balveren beekeepers stayed and their botanical origins, wax ratios, phenolic content, and mineral substance contents were analyzed. In the microscopic analysis, pollen grains belonging to 14 different families used by bees were determined. It was determined that the total phenolic and mineral contents of propolis vary completely depending on the location. With this study, the propolis properties of the hives in the region were tried to be revealed and it was aimed that this study would help the region's propolis to be used for technological and therapeutic purposes.

**Keywords:** Bee, pollen, total phenolic, wax, ICP OES.

### Balveren (Şırnak) Arıcılarının Konaklama Alanlarındaki Propolis Örneklerinin Botanik Kökeni ve Mineral İçeriğinin Belirlenmesi

Öz Arı ürünleri ile ilgili çalışmalar gün geçtikçe artmaktadır. Aslında arı ürünlerinin içeriği, bileşeni; ekolojik ve floristik birçok faktöre bağlı olarak değişkenlik göstermekte, besleyici ve tedavi edici özelliği ise içeriği ile doğrudan ilişkilidir. Balveren beldesi (Şırnak) arıcıları kovanlarını; coğrafik yapısı, floristik ve topografik özellikleri farklı lokasyonlara yerleştirmektedirler. Bu değişkenlik balın kalitesini etkilediği gibi propolis özelliklerini de değiştirmektedir. Kovan yapıstırıcısı olarak bilinen propolis ile ilgili çalışmalar son yıllarda önem kazanmaktadır. Diğer arı ürünlerinde olduğu gibi, propolis içeriği de bölgenin floristik özelliklere bağlıdır. Bu çalışmada Balveren arıcılarının konakladıkları bölgelerden propolis örnekleri toplanmış, botanik orijinleri, mum oranları, fenolik madde içerikleri ve mineral madde miktarları analiz edilmiştir. Yapılan mikroskopik analizlerde arıların kullandığı 14 farklı familyaya ait polen taneleri tespit edilmiştir. Fenolik madde ve mineral madde içeriklerinin ise tamamen propolis örneği alınan lokasyona bağlı olarak değişkenlik gösterdiği tespit edilmiştir. Bu çalışma ile bölgede bulunan kovanların propolis özellikleri ortaya konulmaya çalışılmış ve yapılan çalışmanın bölge propolislerinin teknolojik ve tedavi edici amaçlar ile kullanılmasına yardımcı olması hedeflenmiştir.

**Anahtar kelimeler:** Arı, Polen, toplam fenolik, balmumu, ICP OES.

#### 1. Introduction

Anatolia has a rich vegetation due to the factors such as its geographical structure, climatic characteristics, extreme microclimate area, and topographic structure. In addition, with its plant diversity/biological richness, it is a natural habitat for many living species (Davis, 1971). In this context, due to its natural structure and depending on its geographical diversity, beekeeping in Anatolia has been practiced by the people using traditional and modern techniques for many years (Üreten, 2011; Şenoğlu Fenerci, 2021).

Bees generally collect products for honey from different parts of the plants. These attractants are usually lipophilic substances found in flowers, leaves, leaf buds, mucus, gums, resins, and similar substances (Crane, 1999;

Bankova et al., 2014). Propolis is known as a resinous, fragrant mixture obtained by bees by mixing flowers, pollen, buds, and other plant products with their own salivary enzymes and metabolites (Anjum et al., 2019; Sforcin, 2016). In addition, propolis is a part of the protection mechanisms of the hives and is a mixture of plant resin and wax. This product enables bees to reduce disease and/or parasite effects due to its antimicrobial and antiseptic properties (Simone-Finstrom et al., 2017; Saelao et al., 2020). Propolis is also known as bee glue, which is used to mummify dead bees and to eliminate a potential source of microbial infection (Guzmán-Gutiérrez et al., 2018).

The content of propolis can originate from different plant species; thus, the type and amount of the content

\*Corresponding author: emreerez@hotmail.com

vary widely around the world. The specificity of the flora determines the chemical composition of propolis (Bankova et al., 2014). Volatile components give propolis a uniquely pleasant aromatic odor and contribute to its biological activity. Generally, raw propolis consists of 50% resin, 30% beeswax, 10% essential oils-balsams, 5% pollen, and 5% other organic compounds and minerals (Anjum et al., 2019). Propolis contains more than 500 components, including phenolic compounds (flavonoids, phenolic acids, and esters), fatty acids, sugars, minerals, and terpenoids (Kurek Gorecka et al., 2014; Kasote et al., 2017). There are many studies on the therapeutic properties of propolis as antibacterial (Sforzin et al., 2000), antifungal (Ota et al., 2001; Herrera et al., 2010), anti-inflammatory (Borrelli et al., 2002), anticancer (Sawicka et al., 2012), and antitumor (Oršolić & Bašić, 2003; Sobočanec et al., 2011; Bagatir et al., 2022).

Within the scope of this study, appropriate propolis sampling was made and samples were collected from beekeepers registered in Balveren Town (Şırnak province-Turkey). The collected samples were examined in two stages: microscopic and chemical analysis. The vegetative origins of the pollen samples were determined by examining the propolis samples with a light microscope. In addition, propolis surfaces were examined by electron microscopy. For chemical analyses, the varying total phenolic contents of propolis samples were revealed. In addition, the wax ratios of the samples were obtained by obtaining propolis extracts and the mineral content and ratios in the samples were determined by using the ICP-OES device. Thus; in this study, it was aimed to determine the propolis characteristics and chemical properties in the region and also the plant families used as a source material.

## 2. Material and Methods

### 2.1. Collection of Propolis Samples and Determination of Botanical Origins

Propolis samples were taken from the hives with the support of beekeepers who have been dealing with beekeeping for many years in and around Balveren. For this purpose, propolis samples were collected on 27-30 August 2021, usually in the morning (08:00-10:00) from 12 different locations (Table 1). Approximately 300 g propolis samples were collected from each locality, labeled, and brought to the laboratory environment and analysis processes were started.

Table 1. Location of propolis samples

Sample	Location
1.	Kaval valley (Hakkari province)
2.	Balveren (Şırnak province)
3.	Kaval valley (Hakkari)
4.	Beytuşşebap Gökce village (Şırnak province)
5.	Balveren (Şırnak province)
6.	Kaval valley (Hakkari province)
7.	Kaval valley (Hakkari province)
8.	Beytuşşebap Gökce village (Şırnak province)
9.	Kaval valley (Hakkari province)
10.	Beytuşşebap Dönmezler village (Şırnak province)
11.	Feraşın Valley (Şırnak province)
12.	Feraşın Valley (Şırnak)

### 2.2. Electron Microscopy Analysis

By forming extraction of propolis by ethanol, the colloidal mixture of propolis particles was dried in an oven at 40°C. The resulting dry extract was then suspended in ultrapure water to a concentration of about 1% w/v. The mixture was sonicated for 10 minutes to obtain a homogeneous suspension. Then, the dimensions and morphological properties of propolis particles were investigated using scanning electron microscopy (SEM) (Abdullah et al., 2019).

### 2.3. Preparation of Propolis Extracts

Pure propolis and wax (wax, fatty acid) were obtained by extraction for each propolis sample. The extraction method was carried out by modifying the methods of Cunha et al., 2004 and Negri et al., 2000. 3 g of each crude propolis sample was weighed and wrapped in filter paper, transferred to a 500 mL Soxhlet extractor (60°C). In the Soxhlet extractor, 750 mL of pure n-hexane (for wax extraction) and 750 mL of pure ethanol (for propolis extraction) were used. Extraction continued for 6 hours for each solvent. Filtering was done using Whatman filter papers. The resulting solution was evaporated on an evaporator to remove the solvent and the same procedure was repeated for each sample to yield pure propolis and wax samples. The initial weight and final weight of each sample were compared to determine the wax ratio.

### 2.4. Total Phenolic Substance Analysis

Folin-Ciocalteu (FCR) method was used for phenolic content. 1 mL of FCR reagent was added to the propolis extracts and incubated at room temperature for 3 minutes. Then, 1 mL of saturated Na<sub>2</sub>CO<sub>3</sub> (7%) was added. After 90 minutes of incubation at room temperature in the dark, absorbance was taken at 760 nm wavelength. It was prepared with solutions of different concentrations of Gallic acid (0.05-1 mg/mL) as a standard and the results were calculated as gallic acid equivalents (Su et al., 2007).

### 2.5. Elemental Analysis of Propolis Samples

For the elemental analysis of the propolis samples, 0.6-1.0 g of the samples were weighed and solubilized with the help of a microwave. For this, the weighed samples were transferred to pressure-resistant polytetrafluoroethylene (PTFE) containers and after adding HNO<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> (10.0/2.0) acid mixture, the digesting process was carried out in the Speedwave MWS-3 Berghof brand microwave oven under the conditions specified by Yüksel (2017). After the necessary procedures, elemental analysis was performed with Model Optima™ 7000 DV ICP-OES (Inductively Coupled Plasma Optical Emission Spectrometer) (Perkin Elmer, Inc., Shelton, CT, USA).

## 3. Results and Discussion

### 3.1. Detection of Botanical Origins

Microscopic images of propolis from different localities and pollen grains found in plants were compared with each other. It was found to be compatible with pollens belonging to the families of Asteraceae, Fabaceae, Hypericaceae, Salicaceae, Anacardiaceae, Lamiaceae, Apiaceae, Brassicaceae, Juglandaceae, Malvaceae, Asteraceae, Boraginaceae, Asparagaceae, Caryophyllaceae, and Euphorbiaceae (Fig. 1). It has also been stated in previous

studies that Asteraceae, Fabaceae, Lamiaceae are the families most visited by honey bees (Perveen & Qaiser 2003; Özhatay et al., 2012; Özaltan & Kocyigit, 2022).

Pollen grains belonging to 14 different families were detected in propolis samples. The pollen analysis study actually reveals the vegetation and diversity of the region. In pollen studies, identification is a very difficult and complex process on the basis of species identification, although it is clear on the basis of families. For this reason, only family-based determination was made in pollen samples obtained from propolis samples. Depending on the biodiversity and the region of origin of the natural substance, propolis has a different chemical composition.

### 3.2. Electron Microscope Images

Electron microscope images were obtained in order to determine whether there were differences in pattern and texture in the propolis samples. As a result of the electron microscopy analysis, no significant structure, pattern or texture difference could be detected in the images of the propolis samples (Fig. 2).

There were no unusual compounds found during visual or electron microscopic investigation. Rugged surfaces hidden by wax and extractive layers can be seen in every shot. The properties of Bulgarian propolis samples were found to be similar (Tylkowski et al., 2010).

### 3.3. Wax Ratios of Propolis Samples

It is known that balsam, active ingredient, and wax contents of propolis vary considerably. Researchers found high amounts of balsam that is found to be caused by high amounts of phenolic compounds and low amounts of wax. Indeed, Popova et al. (2017) stated that raw propolis contains between 40% and 60% balsam (Popova et al., 2007). It is also reported to be between 27.7% (Bonvehí & Gutierrez, 2011). It is reported that the amount of wax found in raw propolis in different Portuguese propolis samples varies between 4.8% and 16.0% (Dias et al., 2012). In a study conducted on ethanolic extracts of crude propolis samples collected from Brazil, China, and Uruguay, it was stated that while there was no wax in Uruguay propolis extract, the amount of wax for other regions varied between 2.40% and 30.60% (Bonvehí & Coll, 1994).

In our study, wax rates varied between 19% and 63% (Table 2). This situation is considered to be caused by the plant origin and phenolic and tannin substances in its content.

In the wax analysis, it was determined that the lowest rate was in the propolis of the Feraşin region and the highest wax rate was from the village of Beytüşşebap Dönmezler. It is considered that this situation occurs depending on the plant diversity.

### 3.4. Total Phenolic Substance Contents

As a result of the total phenolic analysis, it was detected that one of the active substances that gives the functional properties of propolis is phenolic compounds. The phenolic content of the collected samples varies considerably.

As a result of our study, the highest three values among the phenolic values belonging to 12 different regions were 4. Locality; 99.46 µg /mL, Locality 9; 111.3

µg /mL, and 7th Locality; 77.46 µg /mL. These values seem to be significant as they are quite high compared to other studies (Fig. 3). Ethanol, water and olive oil were used for extraction of Lithuania propolis samples and according to phenolic analysis; 12.7 /1.6 and 0.5 mg/mL GAE values were obtained respectively (Maden, 2013). Bonvehí & Gutierrez (2011) found that ethanol and propylene glycol extracts had total phenol content of 21 to 34 g/100 g and 20 to 30.3 g/100 g, respectively. It is estimated that the difference between the total phenolic compounds of propolis samples in 70 different places may be due to the geographical location and climatic characteristics.

The biological activities of propolis, such as its antioxidant and antibacterial properties, are dependent on its phenolic compounds. Numerous studies have shown that propolis type, origin, raw materials, and extraction techniques all affect changes in the chemical composition of propolis. It has been reported that the total amount of phenolic in Anatolian propolis ranges from 10.6-178 mg GAE/g and the amount of total phenolic increases as the amount of balsam increases (Keskin & Kolaylı, 2018). Aliyazicioğlu et al. (2013) reported that the total phenolic content for different Turkish propolis samples ranged from 115 to 210 mg GAE/g (Li et al., 2008). The total phenolic content of chestnut propolis was reported to range from 1.2 to 15.6 mg/g (Sarıkaya et al., 2009). It is clear that the total phenolic content of propolis samples obtained from different regions of Turkey varies in a wide range.

In the phenolic substance analysis, it was determined that there was variability in the samples taken from the Kaval valley. It was seen that the highest rates belonged to the Gökçe village of Beytüşşebap. This situation is considered to be caused by the source that the bees preferred for propolis.

### 3.5. Mineral Substance Contents

The presence of minerals, which are a natural part of terrestrial systems, can significantly affect the pharmacotherapeutic properties of derived products. It is important to know the essential mineral content of propolis that has a nutritional supplement or healing effect. The role of macroelements in development is well known. In addition, the mineral content in propolis gives more specific results regarding the condition of the region because pollen and propolis are much less processed by bees than beeswax and honey and more precisely reflect environmental contamination (Formicki et al., 2013). In this context, mineral substance values in our study vary considerably depending on the location. These variations occur especially in Ca, Mg, and Zn values (Table 3).

There is a close relationship between the level of heavy metals accumulated in the soil and plants and their content in bee products (Kabata-Pendias, 2011). Propolis is much more contaminated with toxic elements than polyfloral honey but both can be used as a bioindicator to assess the extent of environmental pollution by determining the level of accumulated toxic elements (Roman et al., 2011). Such elements, even in low concentrations, can cause many diseases and abnormalities in the functioning of the human organism.

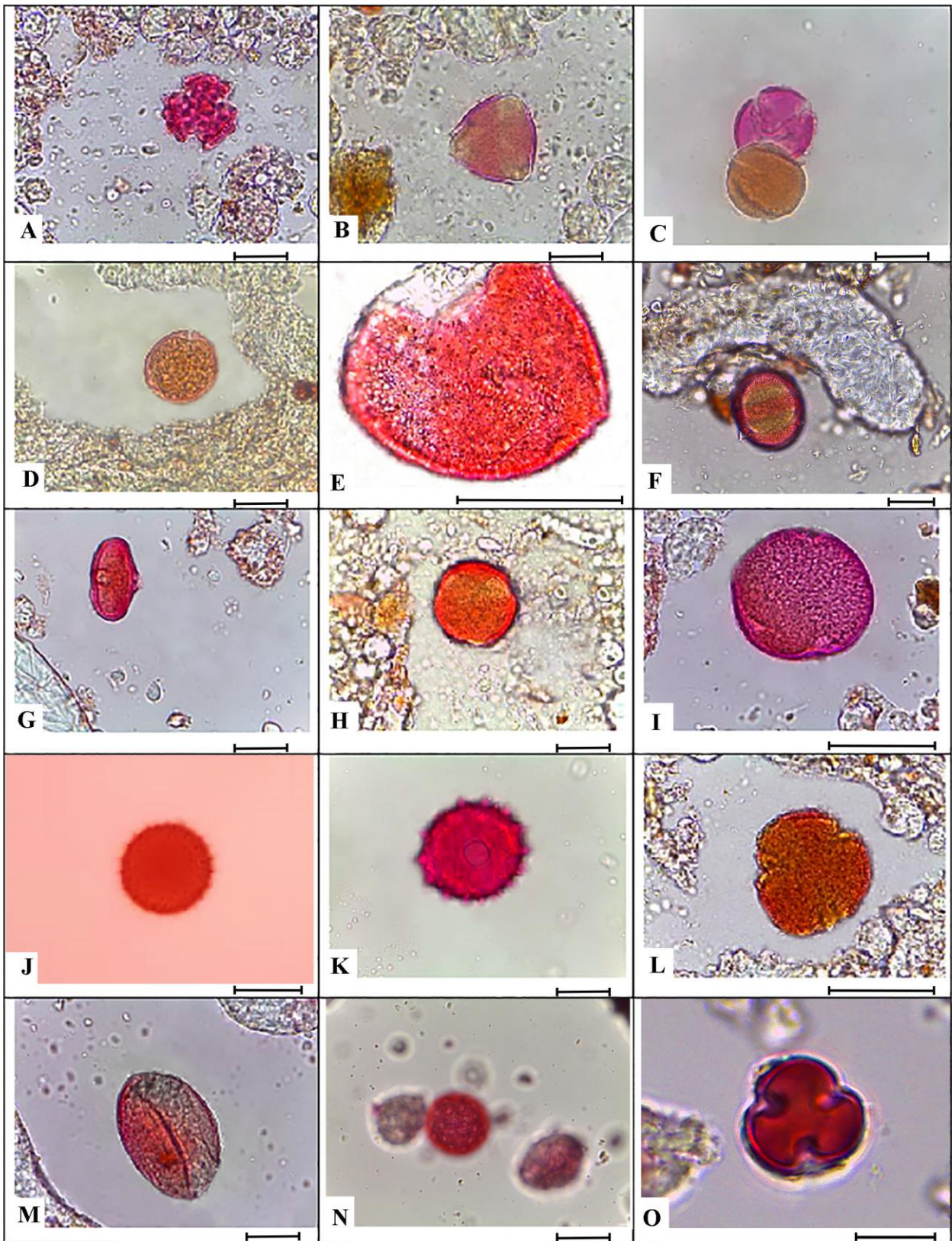


Figure 1. Pollen grains found in propolis samples (Bar : 20  $\mu$ m)

A- Asteraceae, B- Fabaceae, C- Hypericaceae, D- Salicaceae, E- Anacardiaceae, F- Lamiaceae, G- Apiaceae, H- Brassicaceae, I- Juglandaceae, J- Malvaceae, K- Asteraceae, L- Boraginaceae, M- Asparagaceae, N- Caryophyllaceae, O- Euphorbiaceae.

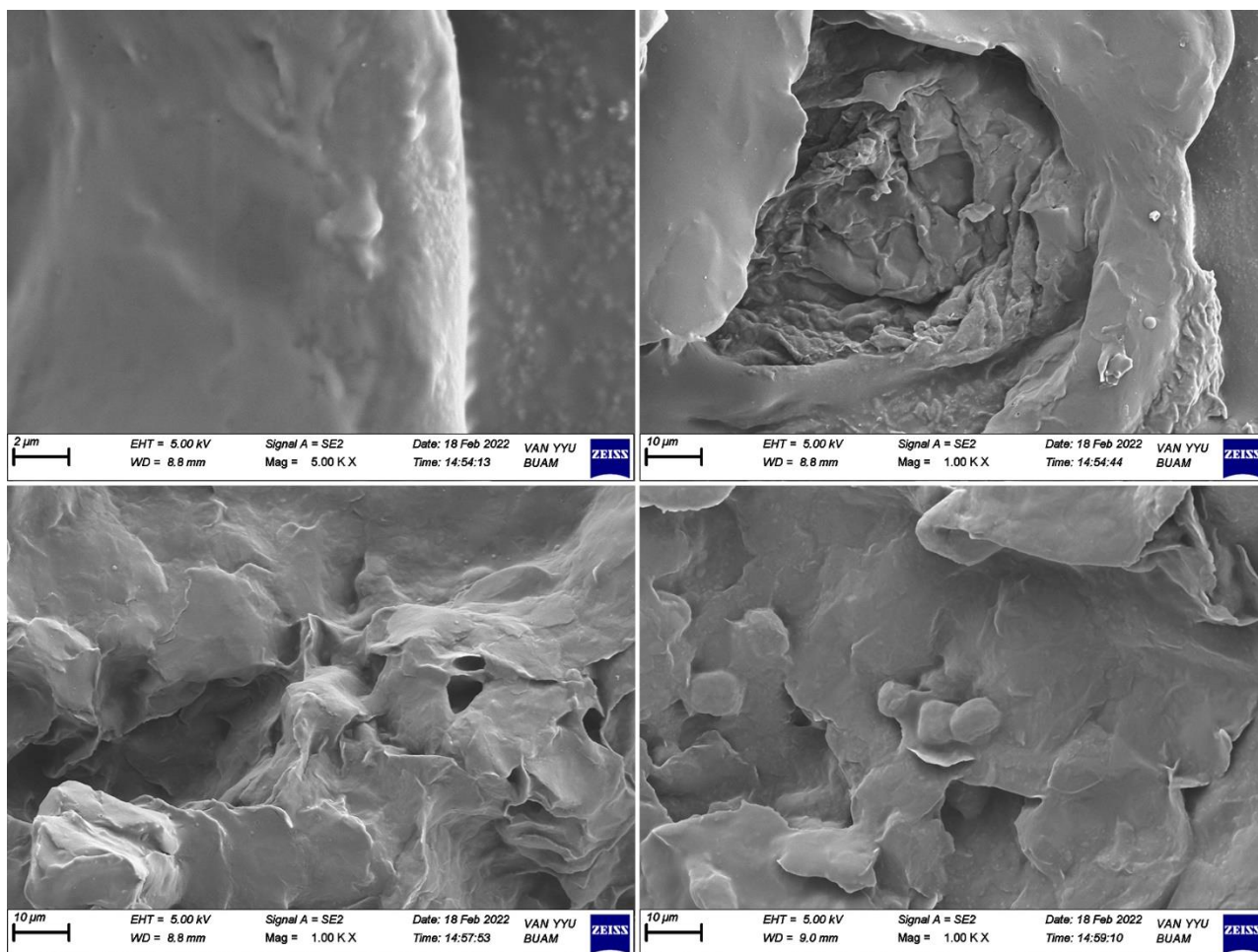


Figure 2. SEM images of propolis samples

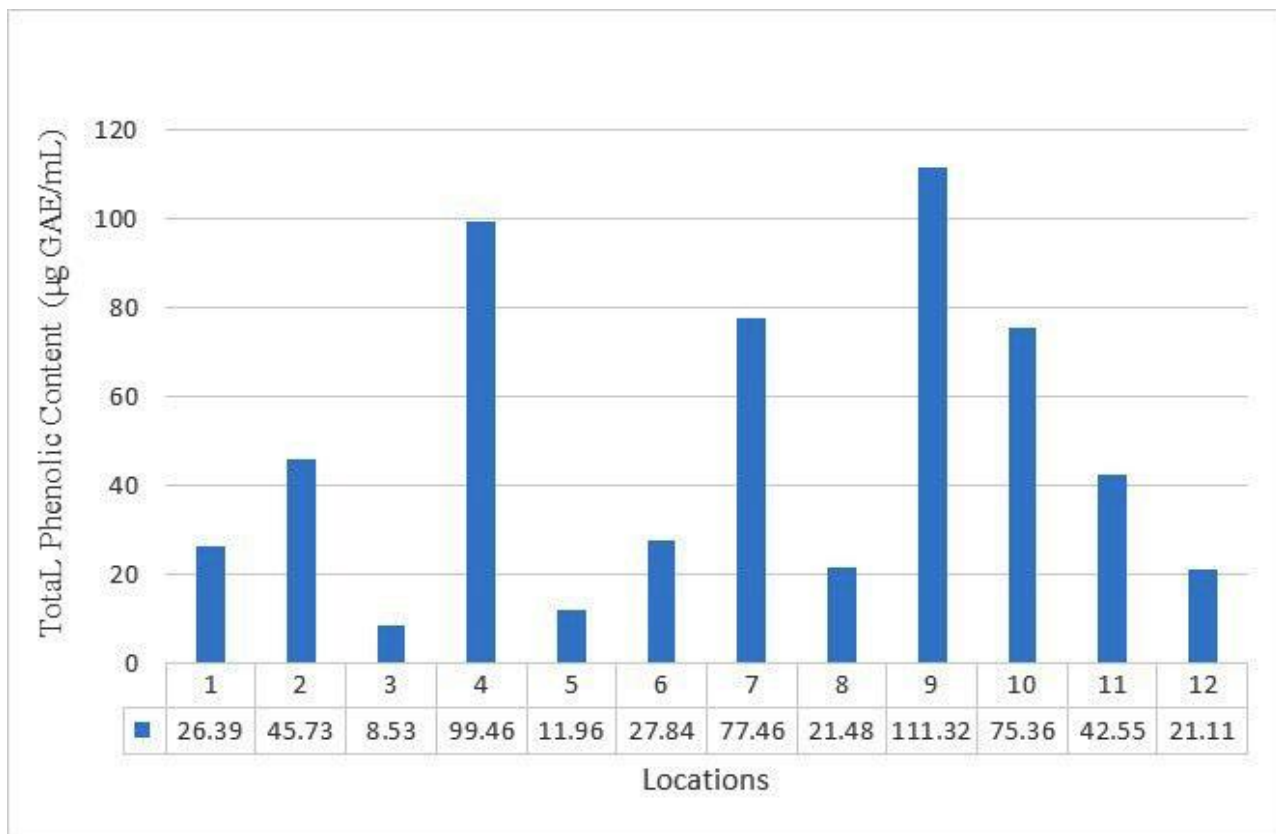


Figure 3. The total amount of phenolic substances (μg GAE/mL) contained in the propolis samples

Table 2 Analyzing the Wax content of propolis samples

Sample	Wax amount (g)	Pure propolis (g)	Wax (%)	Pure propolis (%)
1	5.76	7.57	43.21	56.80
2	6.46	4.98	56.50	43.50
3	8.57	5.95	59.06	40.94
4	6.88	4.09	62.73	37.27
5	5.54	5.25	51.37	48.63
6	4.06	2.37	63.18	36.82
7	7.59	7.76	49.47	50.53
8	6.08	4.54	57.27	42.73
9	6.94	6.97	49.92	50.08
10	10.28	8.79	53.94	46.06
11	4.23	16.66	20.25	79.75
12	3.87	16.49	19.04	80.96

Table 3. Mineral content of propolis samples ( $\mu\text{g}/\text{kg}$ )

Sample	Ca	Cu	Fe	Pb	Mg	Mn	Ni	P	Zn
1.	464.34	2.81	11.69	2.89	10.82	1.86	6.78	49.8	10.08
2.	398.88	1.903	7.12	5.25	52.87	2.10	4.61	59.3	25.76
3.	255.56	9.01	4.34	7.68	35.31	2.44	8.59	45.6	12.07
4.	377.12	5.44	29.23	7.64	19.99	2.63	1.38	26.5	29.11
5.	358.88	6.74	15.66	2.66	10.95	1.96	1.09	11.1	74.94
6.	786.81	1.739	49.24	1.55	78.22	3.39	2.56	32.4	12.50
7.	331.61	5.08	14.94	4.62	14.87	3.37	5.42	86.9	4.32
8.	168.35	1.47	21.68	5.66	25.38	2.19	1.49	13.6	9.88
9.	468.91	2.17	75.39	1.08	20.59	2.16	3.35	11.2	9.67
10.	707.36	3.20	14.49	1.57	13.02	1.96	1.99	32.6	10.64
11.	506.87	7.76	19.71	2.98	91.44	5.73	2.62	53.7	49.53
12.	486.32	2.49	14.76	2.43	48.72	4.39	3.44	39.3	85.04

Differences in the content of elements in individual mineral substances are also present in many other studies. Soil type and parameters, mobile metals, botanical origin of the samples, and weather conditions may cause differences in the mineral profile of the investigated propolis obtained from different locations. These facts can be used in some distant and different grouping of mineral substances in the same cluster or subset.

Finally, this study found that, especially due to Türkiye's rich biodiversity, the propolis sample taken from the Balveren beekeepers has an average level of phenolic and botanical origin and it can also be utilized as a natural source in the food and medicinal industries. Additionally, the identification of fully active components in Balveren propolis allows it to be regarded as a significant source of natural antioxidant chemicals.

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