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Element content of Yemeni honeys as a long-time marker to ascertain honey botanical origin and quality

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A B S T R A C T

This work was aimed at studying variations in the element content of five famous Yemeni honeys of different floral origin. For this purpose, samples of honey were collected over three years from four mono-floral and one multi-floral honey. Element levels were determined by ICP-AES. Results show a highly reproducible level of element for each honey type and a significant change in element content in honey as a function of the floral source. In addition to demonstrating the impact of the floral origin on Yemeni-honey mineral content, our compiled values will be highly useful to ascertain honey quality once Yemen will return to less-trouble times.

1. Introduction

Honey is defined by the Official Journal of the European Communities as “the natural sweet substance produced by *Apis mellifera* bees from the nectar of plants or from secretions of living parts of plants or excretions of plant-sucking insects on the living parts of plants, which the bees collect, transform by combining with specific substances of their own, deposit, dehydrate, store and leave in honeycombs to ripen and mature” (European Community, 2002). Honey is predominantly used as food source but its inclusion in multiple traditional medicine preparations is frequent since it is produced in almost every country (Eteraf-Oskouei & Najafi, 2013). Honey primarily contains sugar and water. It is also composed of more than 200 substances including variable amounts of minerals (Ball, 2007; Missio da Silva, Gauche, Gonzaga, Costa, & Fett, 2016). At least 54 chemical elements divided into 3 groups have been identified in honey (Solayman et al., 2016). The three groups are: the major (or macro) elements such as Na, K, Ca, Mg, or P; the minor (or trace) elements such as Cu, Pb, Zn, Mn, or Cd; and heavy metals. Distribution of these elements in honey is generally used as a quality factor (Conti, 2000).

Honey types of distinct floral origin are considered premium products and are therefore often traded at a higher price than honeys originating from mixed or unspecified botanical source (Donarski, Jones, Harrison, Driffield, & Charlton, 2010). Mono-floral Yemeni honey is one of the most famous types of honey; it is also one of the most expensive honeys in the world. The average price per kilogram of the Sidr honey, that bees make from *Ziziphus spina-christi L.* flowers is up to US$ 200. Other famous mono-floral Yemeni honey types are Sumar, Sal, and Salam honeys that are bee-made from the *Acacia tortilis*, *Euphorbia cactus*, and *Acacia ehrenbergiana* flowers, respectively.

Dishonest geographical origin indication is fraudulent and punishable according to trading laws. Several methods have been recently developed to detect this kind of fraud (Arvanitoyannis, Chalhoub, Gotsiou, Lydakis-Simantiris, & Kefalas, 2005; Roshan et al., 2013). Confusion between honey types of identical geographic origin but from different floral origin should also be avoided for financial reasons and consumer/producer protection. Finally, even though geographical origin and honey type are

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respected, variation in honey quality over time is unsatisfactory and discordant with consumer trust and elevated prices.

We report herein the mineral analysis of four mono-floral and one multi-floral expensive Yemeni honeys in order to investigate the effects of the floral origin on the element content of honey. Our study, performed over a three-year period, was initially simply aimed at characterizing the elements of the most famous types of mono-floral Yemeni honeys. Because of the troubled times that arose in Yemen at the end of our study, our results will also be useful to finely evaluate the possibly deep environmental impact of war on honey quality as well as to ascertain the return of Yemeni honey quality once peace is back.

2. Materials and methods

2.1. Sample collection

Honey samples were collected directly from the hives with the assistance of the beekeepers, at the end of the flowering season for mono-floral honeys to ensure the sample authenticity. All samples were certified as genuine by General Authority for Standardization and Metrology (Sana'a, Yemen).

Monofloral honey samples were collected in the governorates of Hadramawt, Hodeidah, and Ibb (Fig. 1) in 2014, 2015, and 2016 before the resumption of fighting. Large difference in blooming time between the honey-yielding species (Adgaba et al., 2017) unambiguously ascertained honey monofloral nature. Hadramawt governorate afforded two kinds of monofloral honeys: Sidr honey made from *Ziziphus spina-christi* flowers and collected in November around the city of Wadi-Dawan, and Sumar honey bee-made from *Acacia tortilis* flowers, and collected in April. Salam honey made from *Acacia ehrenbergiana* flowers was collected in April in the village of Mount Ras (governorate of Hodeidah). Sal honey bee-made from *Euphorbia cactus* Ehrenb. flowers was collected around Dhi As Sufal, in the governorate of Ibb in March.

Multi-floral (Maraiy Mahwetey) honey originated from the governorate of Ta'izz; it was collected in June. For each type of honey, twelve samples were collected from as many hives. Consequently, this study was based on the analysis of 180 samples, 144 mono-floral samples and 36 of multi-floral origin. All samples were stored in the dark at 4 °C prior to analysis.

2.2. Chemicals

Distilled deionized water 18.2 MX cm resistivity, obtained from a Milli-Q Millipore system (Bedford, MA, USA), was used to prepare all aqueous solutions. Nitric acid 69.5% (Fluka-Traceselect™, Buchs, Switzerland) and hydrogen peroxide 30% (Aldrich, Milan, Italy) were reagent-grade solvents.

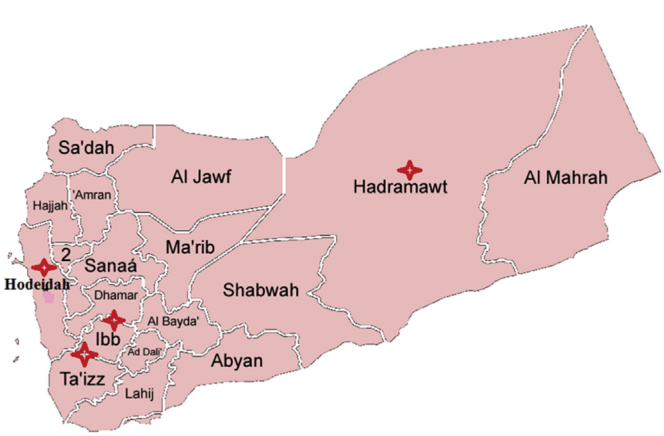

Fig. 1. Map of Yemen indicating the collection sites.

2.3. Sample preparation

Calibration solutions were prepared from standard solutions of 1000 mg/L of each element (P, Mn, Na, Pb, Cr, Cu, Zn, Fe, K, Mg, Ca) supplied by Perkin Elmer (Waltham, Mass., USA) or Aldrich (Milan, Italy). Glass- and plastic-ware was soaked overnight in 10% v/v nitric acid and rinsed three times with deionized water before use.

For mineral analysis, 0.5 g of honey was accurately weighed in vessel tubes containing 5 mL of concentrated HNO_3 and 2 mL H_2O_2 (30%). The tubes were sealed with a Teflon cap and digested in a microwave oven digester (Milestone Ethos Sel, Sorisole, Italy). Samples were digested at 100 °C for 5 min, bought up to 140 °C at 8 °C/min, then held at 140 °C for 5 min, allowed to cool. After cooling, 1 mL of 1% aq. HNO_3 was added and the final volume adjusted to 25 mL with deionized water. Measurements were performed by inductively coupled plasma - atomic emission spectrometry (ICP-AES). All analyses were performed three times.

2.4. Apparatus

ICP-AES spectrometer (JobinYvon, Ultima 2) with axial viewed plasma was used for determination the mineral contents in each honey sample. The operating conditions were set as follows: power 1.15 kW; plasma flow gas 12 l/min; auxiliary gas flow 1.5 l/min; nebulizer gas flow 0.2 l/min. Wavelengths used for the quantification were: chromium 205.560 nm, zinc 213.857 nm, copper 324.752 nm, iron 238.204 nm, potassium 766.490 nm, magnesium 279.077 nm, manganese 257.610 nm, sodium 588.995 nm, lead 220.353 nm, iron 238.204 nm, and calcium 317.933 nm.

2.5. Statistical analysis

Collected data were statistically analyzed from the mean values of three replicates per honey sample. Means were compared using the least significant difference (LSD) test at $P < 0.05$ (Steel & Torrie, 1982, pp. 137–171). Statistical analysis was performed using the Statgraphics Plus package.

3. Results and discussion

In ordinary conditions, honey minor-component composition is directly influenced by local climatic, geologic and environmental conditions (Islam, Khalil, Islam, & Gan, 2014). In addition, the variety of available nectar-secreting plants at a given time is another important factor influencing honey composition (Ball, 2007; Bilandzic et al., 2014; Czipa, Andrási, & Kovács, 2015; Rashed and Soltan, 2004). Beekeeper, farming practices, storage conditions can also contribute to modify honey composition (White, Riethop, Subers, & Kushini, 1962, p. 124). In times of war, additional uncontrollable parameters can inevitably lead to a change in honey composition and particularly in element honey content. The amount of time necessary to return to initial honey composition is impossible to predict.

In parallel, great differences in elemental composition with respect to honey floral origin have been frequently reported (Devillers et al., 2002; Fernandez-Torres et al., 2005; Golob, Dobersek, Kump, & Necemer, 2005; Lachman et al., 2007; Nayik & Nanda, 2016; Oroian, Amariei, Leahu, & Gutt, 2015; Pisani, Protano, & Riccobono, 2008). Variation in element content in honey has been even primarily attributed to botanical origin rather than any other cause (Bogdanov, Haldimann, Luginbühl, & Gallmann, 2007).