Fluoride 2019 May 16. www.fluorideresearch.online/epub/files/038.pdf [Epub ahead of print]

THE INFLUENCE OF TEMPERATURE ON THE CONTENT OF FLUORIDE IN DIFFERENT TYPES OF HONEY

Katarzyna Janda,[a] Karolina Jakubczyk,[a] Daniel Styburski,[a]
Beata Bosiacka,[b] Izabela Gutowska,[a] Dariusz Chlubek[c]

Szczecin, Poland

ABSTRACT: Honey is the most popular bee product consumed by humans. It is known for its nutritional properties and health benefits, which include neuroprotective effects, the support of the circulatory system, and a beneficial influence on skin and respiratory system disorders. The aim of this study was to determine the influence of water temperature used for the preparation of honey solutions on fluoride (F) content. The study material included buckwheat honey, black locust honey, and two varieties of rapeseed honey. Honey solutions (10%) were prepared using distilled water with the following temperatures: 25°C, 70°C, 80°C, and 90°C. The content of F was between 0.019 and 0.384 mg/L. The buckwheat honey solutions had higher F concentrations than those prepared from black locust honey and rapeseed honey. The F content was significantly higher ($p<0.05$) in the buckwheat honey solution prepared with water at a temperature of 80ºC compared to the buckwheat honey solutions prepared at 25ºC, 70ºC, and 90ºC.

Keywords: Fluoride; Honey.

INTRODUCTION

Honey is the most popular bee product consumed by humans. It is known for its nutritional properties and health benefits. The market features many different types of honey, including nectar honey (flower), honeydew honey, and mixed (nectar-honeydew or honeydew-nectar). They are different not only in terms of sensory values, but also in how they influence human health. The most known varieties of honey include rapeseed honey, black locust honey, buckwheat honey, heather honey, lime honey, and polyflora honey. Honey is beneficial to health in many ways. It has a neuroprotective effect, it supports the functioning of the circulatory system,[1,2] and it helps with respiratory system disorders[3] and skin problems.[4] It is a product with complex chemical content, of which 80% is sugar. Most of the sugar, about 90%, includes glucose and fructose, which are easily assimilated monosaccharides. Oligosaccharides are present in honey in small amounts, but thanks to their prebiotic properties they have a positive influence on the human body.[5] The caloric value of honey in 100 g of product is between 320 and 330 kcal. Bee products are not only a rich source of natural nutrients, but they also contain biologically active compounds, including antioxidants.[6] Some of the substances that determine honey's antioxidant potential and play a significant role include: enzymes (e.g., catalase), vitamins (carotenoids, vitamin C, and vitamin E), organic acids, amino acids, and proteins, as well as numerous polyphenol compounds.[7]

Food products, including honey, are one of the sources of fluoride (F) in the human diet. However, not many studies have been carried out in this field.[8-10] The aim of the present study was to determine the influence of the temperature of water used to prepare honey solutions on F content.

[a]Department of Biochemistry and Human Nutrition, Pomeranian Medical University, Szczecin, Poland; [b]Department of Plant Taxonomy and Phytogeography, Faculty of Biology, University of Szczecin, Poland; [c]Department of Biochemistry and Medical Chemistry, Szczecin, Poland. For correspondence: D Chlubek; E-mail: dchlubek@pum.edu.pl

MATERIAL AND METHODS

The preparation of honey solutions: The study material included different varieties of honey (buckwheat honey, black locust honey, and two varieties of rapeseed honey) from the West Pomeranian Voivodeship (Poland).

Ten g of each honey were weighed and quantitatively transferred to a 100 mL measuring flask which was then filled with 25ºC, 70ºC, 80ºC, and 90ºC distilled water. The contents were stirred using a magnetic stirrer until a homogeneous solution was achieved. Subsequently, the solution was left to cool down to 25ºC. All infusions were prepared in triplicate.

Determining fluoride content: F concentrations in individual samples were measured by the potentiometric method with a fluoride ion-selective electrode (Orion 9409 BN, Thermo Scientific, USA). 1.0 mL of the sample was transferred to a plastic tube, and then 1.0 mL of TISAB II was added to this solution. After mixing, the potential difference of each sample was measured for 10 min; 5 min before the addition of the appropriate standard, and 5 min after the addition. According to the work of Łukomska et al.[11] F content in the samples was calculated based on the difference of potentials measured in each sample and the concentration of the added standard. The electrode was calibrated using standard solutions. The correctness of the analytical procedure was controlled by determining the concentration of F in NaF solutions with the known concentrations: 0.1, 1.0, 10.0 mg/L (Orion Company, USA). All assays were performed in triplicate.

Statistical analysis: In all the experiments, three samples were analysed and all the assays were carried out at least in triplicate. The statistical analysis was performed using Stat Soft Statistica 13.0 and Microsoft Excel 2010. The results are expressed as mean values and standard deviation (SD). One-way analysis of variance (ANOVA) and the Tukey post-hoc test were used. Differences were considered significant at $p \leq 0.05$.

RESULTS

In the studied honey solutions, the highest content of F was observed in buckwheat honey. Statistically, the highest content was found in the solution of this honey prepared with water at a temperature of 80ºC. Interestingly, the concentration of F in a solution prepared with 90ºC water decreased below the value labelled at 25ºC. Statistically, significant differences were observed between 80ºC and 25ºC ($p=0.03$), 80ºC and 70ºC ($p=0.009$), and 80ºC and 90ºC ($p=0.004$).

A similar tendency in relation to the content of the studied element was observed for black locust honey. However, for each of the temperatures used, the results were lower in comparison to buckwheat honey. In this case, the level of F in the solution was also the lowest at 90ºC, and statistically significant differences were observed only in relation to 80ºC and 90ºC ($p=0.008$).

The lowest content of F was observed in the two types of rapeseed honey. However, in relation to these two types, the highest level of F was labelled for solutions prepared using 90ºC water. For rapeseed honey I solutions, statistically significant differences were observed at 90ºC and 25ºC ($p=0.0006$), 90ºC and 70ºC ($p=0.0007$), whereas in relation to rapeseed honey II solutions, the differences were noted only at 25ºC and 90ºC ($p=0.01$) (Figure 1).

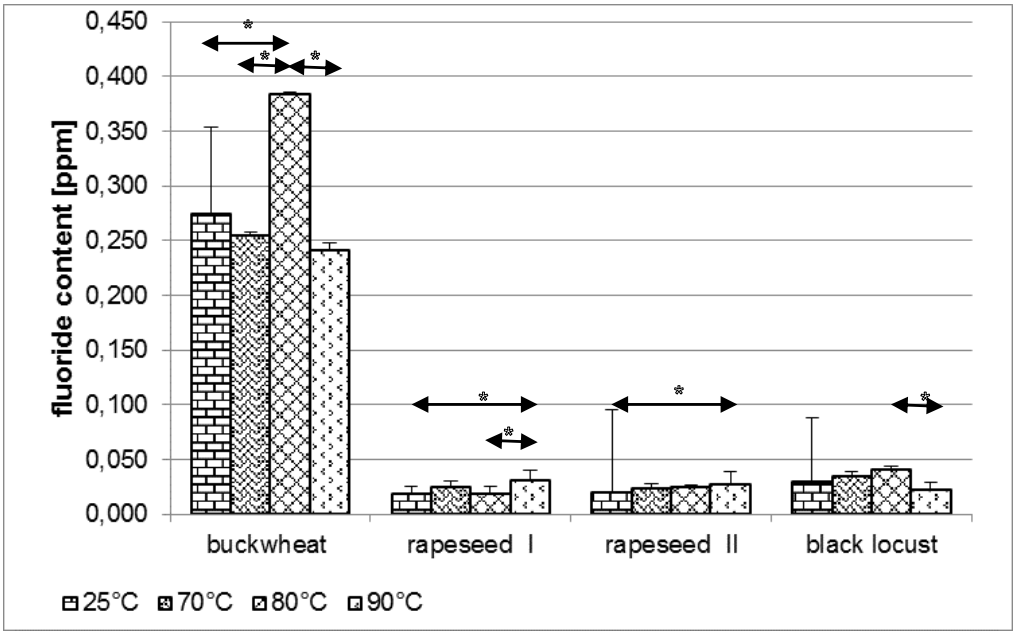

Figure 1. The content of fluoride in specific honey types depending on temperature. *p≤0.05.

The comparison of the acquired solutions also showed statistically significant differences in terms of the amounts of released F depending on temperature (Figure 2).

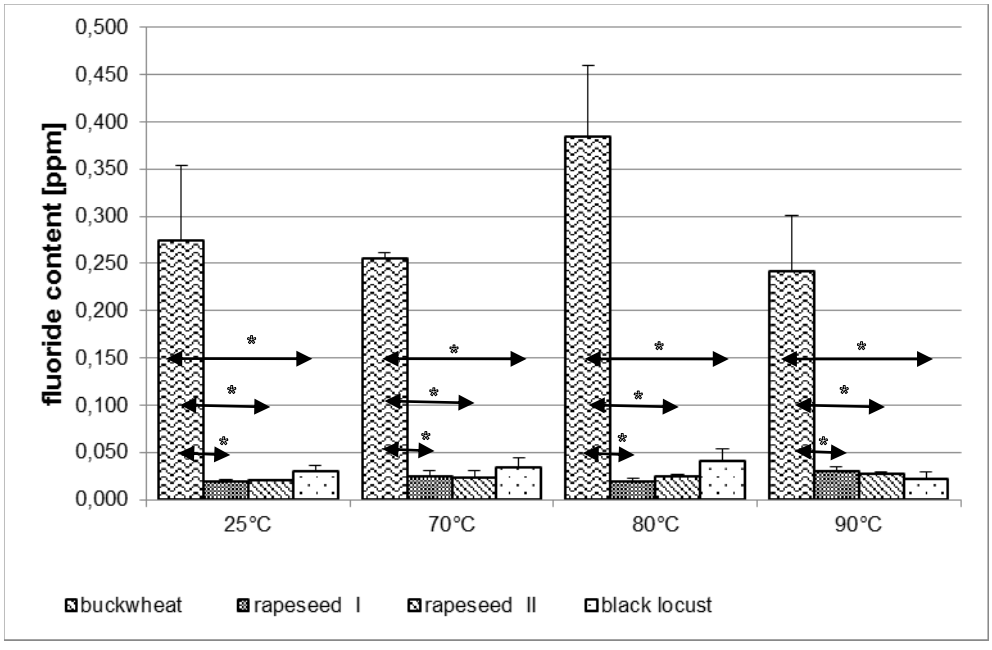

Figure 2. The content of fluoride in specific honey types depending on temperature of honey solutions. *p≤0.05.

The highest concentration of F was noted in the buckwheat honey solution at all studied temperatures, and it was significantly higher in comparison to the results achieved for black locust honey (for 25ºC p=0.0002; for 70ºC p=0.0002; for 80ºC p=0.0002; for 90ºC p=0.0002) and both types of rapeseed honey (for 25ºC p=0.0002 (rapeseed honey I and rapeseed honey II); for 70ºC p=0.0002 (I and II); for 80ºC p=0.0002 (I and II); for 90ºC p=0.0002 (I and II) which had the lowest values of F out of all of the studied material

DISCUSSION

Honey may be considered as a biological indicator of environmental quality and floral biodiversity. Its composition strongly depends on the territory in which it is produced and it is closely tied to the flora visited by bees for its production.[12] In many works, authors point out the influence of the origin of the resource on the physicochemical properties of honey. The differences were particularly visible in the enzymatic activity, ash content, hydroxymethylfurfural (HMF) content, pH, and electrical conductivity.

The mineral concentration in honey was found to be correlated with geographic and floral origins, geochemical soil composition, and local pollution because of the natural absorption of minerals by plants from the soil, water, and air.[13] The quality of honey depends on floral origin and chemical composition and it can be characterised according to its geographical source.[13] The major mineral elements present in honey are K, P, Mg, Al, Ca, Na, Fe, Mn, Cu, Zn, Cl, S, and Si. Some of the elements, such as I, Co, and Mo may occur naturally because of the geographical place, but honey may also be contaminated by heavy metals such as Pb, Cr, Hg, Sb, Ni, Cd, etc. due to contamination by environmental or anthropogenic sources.[13] Honey has a rather low mineral content (typically 0.1–0.2% in floral honey and 1% or higher in mellate honey) and it particularly varies depending on the pedoclimatic conditions, the botanical origin, and the technique of extraction. The dominant element in honey is potassium, followed by chlorine, sulphur, sodium, phosphorus, magnesium, silicon, iron, and copper.[13] Micro- and macroelements play an important role in human metabolism. Therefore, the analysis of this content in different nutrition products seems to be very important for public health studies. Honey is a product which has been intensely studied in recent years because of its health benefits for users. On the other hand, the determination of mineral content of honey seems to be needed not only due to the presence of necessary elements but also the content of heavy metals. Topical fluoride on teeth is seen to have a protective effect against dental caries but although F can be incorporated in teeth during their development up to the age of approximately 8 years[14] and can be incorporated in bone during both development and adult life,[15] fluoride is neither an essential trace element for humans nor necessary for the development of healthy teeth and bones.[16] F increases the synthesis of reactive oxygen species, inflammatory mediators, and impairs the action of enzymes.[17-19] Besides environmental contamination, the presence of F in honey is also associated with the fact that bees need water, in which F content differs to a large extent from region to region.[9] Meyer et al.[8] in the study of F influence on the health of bees noted that F concentrations in bees differ depending on the distance from the source of F emission. Additionally, the authors examined the honey samples and they reported that F content was always low, at the level of 1.4 mg/L. In another study,

Tong et al.[20] noted concentrations of 0.001 to 8.9 mg/L F in honey originating from New York and they concluded that the high amount was not hazardous. Grobler et al.[9] also noted a low amount of F (0.7 mg/L) in honey samples originating from the South Western Cape Province of South Africa. Additionally, they studied the effect of honey on teeth enamel condition and noted that the buffering capacity of honey is higher than that of saliva but lower than that of fruit juices suggesting that honey would be less harmful to enamel in comparison to fruit juice.[9] It is worth mentioning that honey did not show any signs of enamel demineralization after 3 hr of contact with the honey solution and this was probably caused by the significant presence of compounds including micro- (F) and macroelements (Ca, P). However, the study using artificial honey with the same mineral composition showed significant enamel demineralization after 30 min of contact,[9] which suggests that Ca, P, and F levels could be only partially responsible for the non-erosive action of honey and that something more in honey composition is responsible for its non-destructive function.[9]

Analysis of our results showed that buckwheat honey turned out to be the richest source of F. A portion of 10 g of this honey contains, depending on the temperature of water in which it was dissolved, from 0.0241 to 0.0384 mg F. The largest amount of this element goes into solution in water at a temperature of 80ºC. The other honeys deliver, in 10 g (quantity about 1 small spoon), from 0.0019 to 0.0035 mg F.

It would seem that honey as a raw material with a high content of simple sugars, may contribute to the demineralization of tooth enamel and the development of caries. But the results of studies of Tong et al.[20] showed that fructose and glucose had lower cariogenic properties than sucrose. Accordingly, it is possible that honey has less cariogenic activity than other sugars (e.g., fructose and sucrose).[21,22] However it should be remembered, that the lower cariogenic properties of honey probably depends on its many other properties, which all act together. The strong antimicrobial properties of honey[23] cause difficulties in the growth and development of bacteria that live in the oral cavity and plaque and are responsible for the formation of acids destroying the enamel. In addition, it has been proven that honey can enhance the remineralization of an initial enamel lesion[24] and *in vitro* tests have shown that pure honey, with a relatively low pH, does not have an erosive effect on human tooth enamel.[9]

CONCLUSIONS

The present study confirmed that the F content in water solutions of honey depend on both the type of honey and the temperature of water used to prepare the solutions. Although our research has shown that honey is not a rich source of F, honey may contain additional factors, such as antibacterial properties, which protect the teeth against enamel demineralization and the development of dental caries.

ACKNOWLEDGMENTS

This study was supported by the statutory budget of the Department of Biochemistry and Human Nutrition, Pomeranian Medical University in Szczecin, Poland.

REFERENCES

1 Khalil MI, Sulaiman SA. The potential role of honey and its polyphenols in preventing heart diseases: a review. AJTCAM 2010;7(4):315-21.

2 Rahman MM, Gan SH, Khalil MI. Neurological effects of honey: current and future prospects. Evid-Based Compl Alt Med 2014:958721.

3 Henatsch D, Wesseling F, Kross KW, Stokroos RJ. Honey and beehive products in otorhinolaryngology: a narrative review. Clin Otolaryngol 2016;41(5):519-31.

4 Burlando B, Cornara L. Honey in dermatology and skin care: a review. J Cosm Derm 2013;12(4):306-13.

5 Borawska J, Bednarski W, Gołębiewska J. Profile of honey saccharides and possibilities of applying Bifidobacterium to modify their composition and properties. Food Sci Technol 2011;3(76):29-39.

6 Bobiş O, Mărghitaş LA, Dezmirean D, Bonta V, Mihai CM. Beehive products: source of nutrients and natural biologically active compounds. J Agroaliment Proc Technol 2010;16(2):104-9.

7 Wilczyńska A. An attempt to use linear regression to determine the relationship between color and antioxidant properties of honey. Sci J Gdynia Maritime Univ 2013;80:13-9. [in Polish].

8 Meyer DF, Lunden JD, Weinstein LH. Evaluation of fluoride levels and effects on honey bees (*Apis mellifera* L.) (Hymenoptera: Apidae). Fluoride 1988;21(3):113-20.

9 Grobler SR, du Toit IJ, Basson NJ. The effect of honey on human tooth enamel in vitro observed by electron microscopy and microhardness measurements. Arch Oral Biol 1994;39(2):147-53.

10 Spano N, Guccini V, Ciulu M, Floris I, Nurchi VM, Panzanelli A, et al. Free fluoride determination in honey by ion-specific electrode potentiometry: Method assessment, validation and application to real unifloral samples. Arabian J Chem 2018;11(4):492-500.

11 Łukomska A, Jakubczyk K, Maciejewska D, Baranowska-Bosiacka I, Janda K, Goschorska M, et al. The fluoride content of Yerba mate depending on the country of origin and the conditions of the infusion. Biol Trace Elem Res 2015;167(2):320-5.

12 Ioannidou MD, Zachariadis GA, Anthemidis AN, Stratis JA. Direct determination of toxic trace metals in honey and sugars using inductively coupled plasma atomic emission spectrometry. Talanta 2005;15,5(1):92-7.

13 Dhahir SA, Hemed AH. Determination of heavy metals and trace element levels in honey samples from different regions of Iraq and compared with other kind. Am J App Chem 2015;3(3):83-92.

14 Gutowska I, Baranowska-Bosiacka I, Noceń I, Dudzińska W, Marchlewicz M, Wiszniewska B, et al. Changes in the concentration of elements in the teeth of rats with type 1 diabetes, in the peak stage of the disease with absolute insulin deficit. Biol Trace Elem Res 2011;139(3):332-40.

15 Kot K, Ciosek Ż, Łanocha-Arendarczyk N, Kosik-Bogacka D, Ziętek P, Karaczun M, et al. Fluoride concentration in cartilage, spongy bone, anterior cruciate ligament, meniscus, and infrapatellar fat pad of patients undergoing primary knee joint arthroplasty. Fluoride 2017;50(1):97-102.

16 Scientific Committee on Health and Environmental Risks (SCHER). Opinion of critical review of any new evidence on the hazard profile, health effects, and human exposure to fluoride and the fluoridating agents of drinking water. Brussels, Belgium: Directorate General for Health and Consumers, European Commission; 2011 May 16. pp. 2-4.

17 Łukomska A, Baranowska-Bosiacka I, Wolska J, Banaszczak M, Piotrowska K, Jakubczyk K, et al. Extract from nettle seeds (*Urtica dioica* L.) decreases the synthesis of lipoxin A4,

inhibiting the development of fluoride-induced inflammation in THP1 monocytes/macrophages. Fluoride 2017;50(4):412-24.

18 Dec K, Łukomska A, Maciejewska D, Jakubczyk K, Baranowska-Bosiacka I, Chlubek D, et al. The influence of fluorine on the disturbances of homeostasis in the central nervous system. Biol Trace Elem Res 2017;177(2):224-34.

19 Gutowska I, Baranowska-Bosiacka I, Goschorska M, Kolasa A, Łukomska A, Jakubczyk K, et al. Fluoride as a factor initiating and potentiating inflammation in monocytes/macrophages. Toxicol In Vitro 2015;29(7):1661-8.

20 Tong SC, Morse RA, Bache CA, Lisk DJ. Elemental analysis of honey as indicator of pollution. Forty-seven elements in honeys produced near highway, industrial and mining areas. Arch Environ Health 1975;30:329-32.

21 Arawwawala LDAM, Hewageegana HGSP. Health benefits and traditional uses of honey: Areview. J Apither 2017;2(1):9-14.

22 Ahmadi-Motamayel F, Rezaei-Soufi L, Kiani L, Alikhani MY, Poorolajal J, Moghadam M. Effects of honey, glucose, and fructose on the enamel demineralization depth. J Dent Sci 2013;8(2):147-50.

23 Aparna S, Srirangarajan S, Malgi V, Setlur KP, Shashidhar R, Setty S, et al. A comparative evaluation of the antibacterial efficacy of honey *in vitro* and antiplaque efficacy in a 4-day plaque regrowth model *in vivo*: preliminary results. J Periodontol 2012;83(9):1116-21.

24 Gocmen GB, Yanikoglu F, Tagtekin D, Stookey GK, Schemehorn BR, Hayran O. Effectiveness of some herbals on initial enamel caries lesion. Asian Pac J Trop Biomed 2016;6(10):846-50.