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Original article

# Potential Solutions to Combat the Antibiotic and Pesticide Problem in Honey

Ekim Mor [a] & Seyhun Yurdugül [a, *]

[a] Department of Biology, Abant Izzet Baysal University, Bolu, Turkey.

**Abstract**

The presence of pesticides and antibiotics are regarded as a great problem in honey since honey should be one of the purest food in the world. This is mostly due to the uncontrollable and intensive use of pesticides in agriculture and preference of antibiotics in apiculture for getting rid of bacterial infections. This review emphasizes the importance of these substances as well as the health-related problems in honey and proposing different novel food processing techniques to combat this problem without disrupting the physicochemical properties of honey. Among these novel technologies, cold plasma, ultrasonication, and high hydrostatic pressure are either reported or recommended to fight against unwanted organisms causing quality loss in honey. Therefore using these technologies were reported to be environmentally friendly, as since they are included in non-thermal methods, avoiding temperature abuse and they are cost-independent. Cold plasma technology is a recommended method to get rid of not only viable bacteria but also it is found to be effective against spores.

**Keywords:** Antibiotics, pesticides, honey, cold plasma, ultrasonication, high hydrostatic pressure.

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* **Corresponding author:**

Seyhun Yurdugül, Department of Biology, Abant Izzet Baysal University, 14280, Bolu, Turkey.
Email: yurdugul_s@ibu.edu.tr

## INTRODUCTION

Antibiotics and pesticides in natural foodstuff such as honey are considered as a serious problem. Mainly, European Union (EU) regulations implied that honey should not contain inorganic or organic compounds other than its composition, even the preservative agents, as well as addictive substances are not allowed (Derebaşı et al., 2014). Interestingly, a research carried out in Bangladesh pointed out that the presence of toxic heavy metal elements was proven in honey even in low amounts of Co, Pb, and Cd, pointing out that these metals can also transmit from different environments to honey as well (Paul et al., 2017).

Pesticides are reported to be present in nectar and pollen, and they are regarded to have importance for the bee health, as well as harmful for human health due to the toxicity characteristic of the chemical and the length and magnitude of exposure. If a human being is exposed to them, the harmful effects of pesticides are related with mild skin irritation, defects, tumors, genetic changes, blood and nerve disorders, endocrine disruption, and even coma or death on the human body. The farmers and their relatives have the highest exposure potential to them, next children are the other vulnerable group due to their weight and age. The other important thing to be considered is their ability of biomagnification, bioaccumulation, and bioconcentration in the organism even after years. Although the main aim of pesticide use is to kill or either controlling of unwanted organisms, pests, weeds etc., one of the main reason that the producers' use pesticides is the extensive protection of fruits and vegetables, as well as increasing the yield and farm revenues (URL 1)

Bee can produce honey and other related products naturally, which was used as a general nutritional and medicinal purpose product e.g. treatments of a sore throat, cough, earaches, infected leg ulcers, measles, eye disorders, and gastric ulcers. Honey, as well as propolis and polen, are rich in simple sugars, minerals and enzymatic and non-enzymatic antioxidants (Tillotson et al., 2006).

Likewise, as the other foodstuff, honey is subjected to various types of the illicit substitution of one substance for another and contamination. Mainly, in the markets of the underdeveloped countries, there are too many jars of honey without a brand, label, as well as they are subjected to adulteration. These substances are generally pesticides, herbicides, antibiotics, or heavy metals. Their ingestion may cause important health problems. Labeling of honey should associate qualified assays, confirming its safety. Health authorities have to implement enforcement by law that would stop any misuse of honey production, storage and analysis to keep its safety worldwide (Waili et.al, 2012).

The aim and scope of this review are to imply the removal of the pesticides and antibiotics in honey by proposing the use of novel technologies, especially the cold plasma to avoid the harmful effects of them.

**Antibiotics and their presence in honey**

Antibiotic residues are reported to be present in honey at an intensive amount since they are used in apiculture for getting rid of bacterial infections. Antibiotic use is preferred in large-scale honey production (URL 2). A common example is oxytetracycline, involved in the treatment of American (AFB) and European foulbrood diseases (EFB) caused by Paenibacillus (Bacillus) larvae and *Streptococcus pluton*, respectively. Different antibiotics such as enrofloxacin, streptomycin, monensin, lincomycin, erythromycin etc. are reported to be present in beekeeping affairs. Antibiotic usage in beekeeping is only permitted in various European countries. Therefore, no maximum residue limits (MRLs) are reported for antibiotics in honey according to the European Community regulations (Mutinelli, 2003), which means that honey containing antibiotic residues are not permitted to be sold (Johnson and Jadon, 2010).

The frequent utilization of antibiotics leads its accumulation of their residues in honey and it is considered as a problem in the food industry because of underqualified product manufacturing and decreased interest in the market. These residues may probably display toxicity on the human body since they are not easily detoxified by the liver. By the investigation of these residues in bee products, it leads to evaluate the risk of these foods in living organisms and gives information on the dose of these antibiotics applied to the field crops around the hives. These residues can cause allergic symptoms in hypersensitive people, hemopoietic system problems, or lead to disorders in an indirect way, such as induction of resistant bacterial strains. Other than these, ß-lactam group of antibiotics, even at a low dose, lead to cutaneous eruptions, dermatitis, gastrointestinal problems and sometimes anaphylactic shock. These residues may lead to microbiological hazards, carcinogenicity, reproductive problems, and teratogenicity in the future (Tillotson et al., 2006).

Antibiotic residues are determined in honey due to their frequent use in apiculture, in order to get rid of bacterial problems. They arise commonly from the environmental problems, as well as inadequate beekeeping applications. Even though they are limited, some international studies are focused on these residual analyses in honey. In a study conducted by Al-Waili et al., 2012, high levels of oxytetracycline and chloramphenicol residues were detected in honey. Moreover, another frequently used antibiotic is tetracycline, some other antibiotics, including lincomycin, erythromycin, streptomycin, monensin, and enofloxacin are commonly present in the market. In a study conducted in Southern Marmara region of Turkey, 50 different honey samples were investigated for their erythromycin content. Four samples were found to include erythromycin remnants between the 50-1776 $ng \cdot kg^{-1}$ interval (Johnson and Jadon, 2010). Erythromycin was included into bee cake and it was given to the bees in a definite hive in order to analyze the transmission of erythromycin remnants to the honey structure; and it was found to be approximately 28 $ng \cdot kg^{-1}$ (Al-Waili et al., 2012).

In another assay, the degradation kinetics were monitored by a linear model and it was found that the oxytetracycline degradation was observed in chestnut honey for a period of 116 days. In floral honey, the remnants of chloramphenicol and sulphathiazole were kept for 661 and 581 days, respectively, and in meadow, honey streptomycin was found to stay for 321 days (Gacic et al., 2015). According to Wen et al., 2017, the microbial degradation of antibiotics by Escherichia spp. and Candida spp. showing the highest efficiency to get rid of doxycycline 92.52% and 91.63%, respectively. Even though tetracycline (TC) is said to be persistent and toxic, it is widely preferred. Wang et al. (2017) proposed a novel model for the biodegradation of tetracycline in an anaerobic bacterial fuel cell, including a mixed substrate of tetracycline-glucose. In a week, an approximate degradation of 79.1% tetracycline was obtained in the microbial fuel cell. Candida spp. SMN04, a novel yeast culture, obtained from the pharmaceutical wastewaters, was reported to be able to use cefdinir as a carbon source for its growth in a mineral containing the medium.

In apiculture, the main commonly preferred antibiotics for the cure of bacteriological disorders are chloramphenicol, oxytetracycline, streptomycin, and sulphathiazole. A study was designed to detect the destruction kinetics of these antibiotic residues present in various types of honey kept in the dark at 25 °C. Seventy-four honey samples with pollen and favorable physicochemical properties provided from the Croatian market and quantitative assay of antibiotic remnants were analyzed by ELISA and HPLC analysis. Antibiotic remnants were not detected in the different honey samples analyzed, including chestnut, meadow, floral, acacia and honeydew. Subsequently, the antibiotics oxytetracycline, chloramphenicol sulphathiazole, and streptomycin were added as 10, 200, 200, 0.5 in mg/kg to each of the tested honey samples. Intensive amounts of antibiotics have been put in the honey because of the mixed antibiotic mass fraction and the different chemical structure of each antibiotic. The dynamics of disruption of the analyzed antibiotics were shown to change depending on the type of honey to which the antibiotic was supplemented at 25 °C, during a six-month storage in the dark. Although the half-life of oxytetracycline in the floral, meadow, acacia and honeydew honey kept in the dark at 25 ° C was 15, 16, 17 and 19 days, in the chestnut honey the dissociation lacked even after 60 days of storage, respectively. The half-life of chloramphenicol and sulphathiazole was shown to be more than six months in all of the investigated samples. The quickest dissociation was observed in oxytetracycline spiked into the acacia honey, following streptomycin and sulphathiazole addition, while the most retarded dissociation was observed when chloramphenicol was spiked into the floral one. In agreement with the data presented using a linear model of dissociation kinetics, the oxytetracycline dissociation was 116 days in chestnut honey, the longest of the others. Streptomycin is protected in meadow honey for 321 days and also sulphathiazole and chloramphenicol are protected as the longest in floral honey for 661 and 581 days, respectively (Gacic et al., 2015). Yet, in some fifteen kind of honeys obtained from Switzerland, out of some 350 samples, the finding of sulfanilamide residues could not be explained by such apicultural application. Bees sometimes gather nectar from meadows treated with the herbicide asulam. Such honey is contaminated by asulam and its distortion product sulfanylamide. This is the first instance that the use of a herbicide leads the existence of remnants of an antimicrobial active substance from sulfonamides in food. The relation of this result depends on the fact that the preference of the herbicide asulam might lead to untolerated remnant levels of sulfanilamide in a product for human consumption (Kaufman and Kaenzig, 2004).

**The pesticide problem**

Chemical assay indicates that honey bees (*Apis mellifera*) and hive products include different pesticides derived from various sources. The most plentiful pesticides are acaricides applied by beekeepers to control *Varroa destructor* (Johnson et al., 2013). Other than Varroa, black queen cell virus and deformed wing virus are the other organisms that divert the beekeepers to use these pesticides (Traver et al., 2018). A widely used, organophosphate-based pesticide, acephate which is preferred to eliminate the piercing and/or sucking insects in field crops was reported to give indirect harm to bee (Yao et al., 2018). Fungicides may be incorporated when treated to flowering crops located next to the hive. Acaricides, antimicrobial drugs, and fungicides don't display an intensive toxicity to bees independently, but when they are served in combined form their toxic characteristics may be increased interactively. The studies conducted related to mortality ratio in adult worker bees not only displayed an interaction among acaricides, but also between acaricides and antibiotics and acaricides and fungicides. The tau-fluvalinate was reported to display higher toxic characteristics when combined with other acaricides and most other compounds tested (15 of 17) while the characteristics of amitraz were almost the same (1 of 15). The prochloraz, a fungicide which can inhibit the biosynthesis of sterols increased the toxic property of the tau-fluvalinate, coumaphos, and fenpyroximate acaricide types, by inhibiting the detoxicative cytochrome P450 monooxygenase action. Four other sterol biosynthesis inhibiting fungicides ascended the toxic properties of tau-fluvalinate in a dose-related action, therefore, possible evidence of P450 increase was seen at the lowest fungicide concentration. Sublethal amitraz pre-exposure elevated the toxic potential of the three P450 removed acaricides, but toxic characteristic of amitraz was not varied by sublethal exposure to the same three acaricides. The genetic characteristics of the bees involved were changed related to two-fold difference of tau-fluvalinate (Johnson et al., 2013).

It is assumed that an interaction with acaricides in bees resembles the drug interactions in different organisms in that P450 related toxin removal display a crucial function. Similar to drugs in human, the interaction of pesticides displays a complexity proven by non-transition, year based change and inducing the enzymes of toxin removal in bees (Johnson et al., 2013).

In a study conducted by Kasiotis et al. in 2018, a GC-MS/MS method in order to detect and quantify phenolics in monoterpene nature, as well as thymol and carvacrol in the honey bee, including a simple protocol by extracting in ethyl acetate, is reported. It was subsequently applied to samples of honeybee after death cases were reported to analyze the quantity of the afore-mentioned compounds between the period of 2015-2017. In parallel, other commonly consumed acaricides, such as amitaz, tau-fluvalinate, and coumaphos were also investigated by an LC-ESI-MS/MS multi-residue method related to the modification of QuEChERS technique. Degraded amitraz; DMF and DMPF and coumaphos oxon products were also analyzed. The most important acaricides found were coumaphos, thymol, metabolites DMF and DMPF, and in a small quantity tau-fluvalinate, with concentrations for compounds changing from the low ng/g scale up to approximately 60,000 ng/g bee body weight. The greatest concentrations were expressed for coumaphos and thymol. Preliminary risk assessment using hazard quotient (HQ) as the criterion, showed negligible risk from acaricides as individual components of bees. However, the possible synergism among acaricides or acaricides and other pollutant substances must be taken into consideration.

**Novel technologies to combat these harmful substances**

Biological warfare is proposed in the field crop production as an alternative to combat easily with the problems by the frequent use of antibiotics and pesticides such as *B. thurngiensis* toxins etc (Batabjal and Nijkamp, 2014). But honey should be treated with novel food processing technologies to avoid antibiotics and pesticide problem, although limited applications were reported on some of them such as cold plasma on food. Plasma technology is one of the novel methods proposed to get rid of pesticides and antibiotics in the meantime. This technique finds its application through offering various potential trials in food and biomedical industry. The potential applications of plasma technology in the food industry are based on leading to increasing in food quality through the decontamination and improvement of food, the degradation of toxins and modifying the surface of packaging materials. Cold plasma is reported to be an effective method for pathogen elimination and spoilage, therefore, up to now, no adverse effects were reported on food quality. It can be another alternative for significant inactivation of mycotoxins and pesticides since this technique was tested on the agricultural produce. The inactivation mechanism of cold plasma at the molecular level is under investigation, and further research should be carried out to evaluate its influence on reactive gas chemistry, toxicology, ecology as well as the economy (Pankaj and Keener, 2017). Another study was carried out by Heo et al. (2014) providing that the atmospheric air plasma was remotely used to inactivate microorganisms as well as to remove pesticides in enclosed appliances such as an airtight box and commercial refrigerator. At a level of 99% saturation in 120 seconds were achieved during the sterilization of bacteria and the elimination of pesticides in a 292 liter containing refrigerator via 99% elimination in a very short time, indicating that this technique may be preferable during fresh keeping of agricultural products and elimination of unwanted substances on them (Heo et al., 2014).

According to Sarangapani et al. (2017) cold plasma is reported to be a potential biological decontamination method to avoid microbiological and chemical risks related to food commodities including fruits and vegetables. The effect of cold plasma exposure in order to degrade pesticides, including imidacloprid and boscalid on blueberries subsequently to sustain important quality criteria of them during post exposure was investigated by a high-voltage dielectric barrier discharge plasma reactor. The efficiency of this method after 80 kV-five minutes of cold plasma exposure was 80.18% in boscalid and 75.62% in imidacloprid exposure. The total phenolics and flavonoid content were found to rise significantly for one-minute exposure at all applied voltages. On the other hand, vitamin C levels were significantly decreased at high doses of plasma. No significant change in physical properties like color was observed, but some acceptable changes were recorded in the firmness of the fruits. The quality retention of blueberries was provided by using cold plasma (Sarangapani et al., 2017). Another study was carried out on apple and spinach samples exposed to omethoate by generating low-temperature plasma through a dielectric barrier corona discharge and it was found that, after twenty minutes of treatment, the number of omethoate remnants in apple and spinach was (94.55 +/- 0.01)% and (95.55 +/- 0.01)%, respectively. In lower than twenty minutes of exposure, the amount of moisture, ascorbic acid and beta-carotene were not significantly changed by low-temperature plasma. The detailed investigation revealed that low-temperature plasma may degrade unsaturated double bond structure in omethoate by producing phosphate radicals, causing omethoate degradation. Consequently, it is reported that no quality change was observed by using suitable doses during low-temperature plasma treatment and efficient degradation of omethoate residues in fruits and vegetables were obtained (Shi et al., 2018).

The effect of cold plasma on bacteria is another important criteria to be discussed. *Clostridium difficile* is a spore former microorganism, causing colitis and antibiotic-related diarrhea, even in the developed countries. The spores of *C. difficile* are harsh and their viability may be prolonged even for months, causing high mortality rate healthcare-linked infections. Cold plasma treatment of *C. difficile* spores was analyzed by a study conducted by Connor et al. (2017) including the analysis of the criteria affecting the sporicidal efficacy, such as oxygen percentage in helium carrier gas, as well as the effect of cold plasma on spores from different strains belonging to five evolutionary clades. The strains from different clades demonstrated varying resistance to the cold plasma such as the strain R20291, the global ribotype 027 type present in most epidemics, as the most resistant. After the exposure to plasma, near to three log reduction was effectively obtained in spore counts of all strains (Connor et al., 2017). This study can be a model for eliminating *C. botulinum* spores present in honey which causes infant botulism in babies less than one-year-old. Although this technique does not find its way to be applied to honey, we would like to propose cold plasma to test in honey to evaluate its effect, especially on physicochemical properties.

Other than cold plasma, another technique, ultrasound may be helpful in getting rid of the antibiotics, solely or in combination. A study performed by Serna-Galvis et al., 2016 aimed to work on the ultrasonic removal of a penicillin-derived antibiotic (oxacillin) in pharmaceutical wastewater. This water was treated with high-frequency ultrasound, mixed with the antibiotic in combination with mannitol or calcium carbonate. The high-frequency ultrasonic treatment performed more than 120 minutes easily cleaned oxacillin and its activity was destroyed (Serna-Galvis et al., 2016). In another study, the disruption kinetics of the tetracycline in aqueous solution was analyzed by ultrasonication together with sulfate. A preliminary study was carried out using the only peroxydisulfate, ultrasound and ultrasound activated peroxydisulfate. The results showed that the tetracycline disruption rate increased with the increase of initial peroxydisulfate concentration, temperature, and ultrasonic power, but decreased with the increase of the tetracycline level at the beginning implies that, ultrasound activated peroxydisulfate was found to be an alternative for tetracycline disruption in aqueous solutions (Nasseri et al., 2017).

In the industry, when honey was processed by heat around 50° C leads to a liquified texture, by delaying crystal formation and decreasing the crystal counts as well as the viscosity. Heat processing of honey generates 5-hydroxymethylfurfural (HMF), a toxic substance decreasing the quality of honey. The combined treatment of high hydrostatic pressure (HHP) between 220-330 MPa interval and ultrasonic processing at 24 kHz was tested on different honey samples in a study by Onur et al.(2018). Depending upon short time-processing, the maintenance of quality attributes and being convenient, ultrasound treatment is preferred to thermal processing. The HMF level was said to be lowered by using HHP processing, as well as liquefied honey crystals were obtained (Onur et al., 2018). In another study, HHP treatment of 600 MPa in 0, 2, 5, 8, 12 and 15 minutes of a honey brand in Mexico resulted in reduction of microorganisms, bioactivity retention, including antioxidative activity, ascorbic acid, total phenolics and carotenoids as well as diastase activity, 5-hydroxymethylfurfural, fructose and glucose concentration and rheological properties. The viable microbial population is reduced by HHP treatment even for fifteen minutes below the detection limits. In addition, the antioxidative activity and total phenolics were increased by 30% and 6%, respectively at two minutes' treatment time. The ascorbic acid, fructose, glucose, and maltose concentration during HHP treatment were found to be the same as the treatment period reported above (Leyva-Daniel et al., 2017).

## CONCLUSION

Honey is one of the common food items which should be safe, regarding children, as well as adults since the existence of antibiotics and pesticides emphasize a great problem. The current situation implies that efforts should be made in order to cope with this problem without disrupting the physicochemical property of honey. We would like to point out that since it is regarded as a low-cost method, cold plasma technology can be advised to assist the degradation of these harmful substances in honey regarding different food applications. In addition, the use of other novel food processing technologies, such as ultrasound, and/or high hydrostatic pressure together or independently may be a good alternative to degrade the harmful substances in honey, since they show not only cost-independent but also environmental friendly characteristics.

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