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Review

Honey Quality and Microplastic Migration from Food Packaging: A Potential Threat for Consumer Health?

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Abstract: In ancient Greece, people said that "honey is the Food of the Gods", and they were right. They believed that honey fell from the sky, with the morning dew, on the flowers and leaves, and from this point, the bees collected it. Honey is one of the most nutritious food products, which can be found in most homes. A lot of honey products are stored in different types of packaging materials, including plastics. Plastic packaging has been studied for the migration of plasticizers, chemical compounds, and MPs and NPs in foodstuffs. Most of them have been achieved through food simulations, while some studies managed to detect and isolate MPs/NPs. Recent studies presented evidence for the presence of MPs/NPs in honey products but not directly connected to food packaging or to the different types of honey and their properties (viscosity, pH value, and moisture content) or their storing conditions (temperature, humidity, light, and time). Spectroscopic and analytical techniques like Raman, FTIR, HPLC, and GC-MS are in the foreground for MP/NP detection and identification, but a universal way of isolation, detection, characterization, and quantification has not yet been found. This leaves an open field for more work to be done to clarify the factors affecting the migration of plastic packaging material in honey.

Keywords: microplastics; nanoplastics; honey; food packaging; microplastics migration; ATR; Raman; spectroscopy

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1. Introduction

Plastics deposition in the environment has been proven harmful to the ecosystem and human health [1,2]. MPs have been detected in seawater and drinking water [3–5], marine life [6,7], beverages like German beer [8], and in foodstuffs such as honey [9,10]. Many studies collected in a recent review paper have been published concerning the detection of possible food contaminants in fresh foods from their plastic packaging [11]. The use of plastics as food packaging materials is widespread due to their strong, durable, lightweight, versatile, and cost-effective nature. Polymeric materials such as LDPE, HDPE, PET, and PP have been widely used in food preservation and transportation [11]. However, many plastics used in food packaging, as mentioned above, wear out over time due to environmental conditions; that causes a problem. In recent years, many researchers all over the word have studied the migration of MPs from food packaging [12] into different foodstuffs such as cheese [13] and meat [14].

Honey is a highly appreciated natural food due to its essential properties for human health [15]. It consists mainly of sugars (predominantly the monosaccharides fructose and glucose, small amounts of the disaccharide sucrose and other oligosaccharides, and higher sugars) [16]; enzymes; vitamins; minerals; organic acids; essential oils; esters; pollens;

and proteins [17]. It has been famous for its therapeutic properties for centuries [18]. As reported recently, a combination of lime or green tea with honey had anti-inflammatory and antimicrobial effects during the COVID-19 period [16]. The presence of phenolic compounds, flavonoids, carotenoid derivatives, and organic acids transfuses this anti-inflammatory and antioxidant characteristic of honey. Vitamins and minerals are likewise meaningful, variated upon geographical origins of honey [19]. Furthermore, honey has been used in medicine for epidermic wounds as an antimicrobial and healing agent, due to some characteristics in combination, such as osmolarity, low water content and pH, high viscosity, high sugar, phenolic acids, and flavonoid levels. Finally, the presence and action of glucose oxidase on the glucose of honey produces H_2O_2, which has antibacterial potential [20,21].

Honey, as a viscous nonhomogeneous colloid substance, has a lot of different properties. The multiple different types of honey are based on their floral origin, presenting varying properties such as pH, water content, viscosity, sugar content, and color parameters. When honey is stored, all these properties are affected by the following main factors: temperature, moisture content, light, and time of storage. Long periods of honey storage influence the color, the optical density, the refractive index of honey [22], and, also, the humidity of honey, which is a very important parameter in honey quality, delineating the stability against fermentation and granulation [23]. The moisture content of honey, in its turn, strongly influences the flavor, preservation, viscosity, specific weight, crystallization, and flavor, contributing to the development of fermenting microorganisms [24]. All the aforementioned parameters, especially in long periods of storage, cause different polymer degradation and could potentially affect plastic migration from plastic packaging.

The analysis of the honey quality as MP migration has also been studied using spectroscopic and analytical techniques. Honey quality has been mainly studied using the following spectroscopic techniques: fluorescence spectroscopy [25], Raman and FTIR spectroscopy [26–31], and photoacoustic spectroscopy [32]. More recently, spectroscopic techniques have been used in combination with multivariate statistical analysis and other chemometric methods (PCA and PLS) [33] or through the usage of artificial neural networks for rapid quantification approaches [34]. Last, but not least, analytical methods such as HPLC–based method GC–MS and X–ray diffraction, are very helpful and used for microstructure analysis, respectively [35]. The spectroscopic determination of MPs/NPs in honey is mainly carried out by Raman and FTIR spectroscopy [9,10] and with the aid of the dispersive liquid–liquid microextraction–gas chromatography–mass spectroscopy method (DLLME–GC–MS) [36].

The main objective of this review is to trigger thinking concerning the existence of MPs as a global and realistic problem with potential risks to human health [37]. There is a necessity to develop widely applied methods for the identification of MPs/NPs in honey and, also, in other foods, with a focus on the sustainability of food quality and to avoid possible health risks. To safeguard human health, research should aim towards excellent food quality, which includes checking food products for a MP/NP presence.

2. Packaging Materials and Their Degradation Parameters

Packaging materials can be classified into multiple types. The most commonly used types for food packaging are glass, metals, and plastic. Each packaging material has its unique advantages and disadvantages related to endurance, usability, and cost. In the following sections, we present in detail the different types, characteristics, environmental friendliness, and risk in human health.

2.1. Plastic (Polymeric) Packaging

Plastics are used in many types of food packaging and containers for a variety of reasons: they help protect foods from damage, provide food safety, and extend the freshness of food. Plastic packaging comes in a wide range of materials, each of which offers specific features regarding its appearance, robust temperature range, compatibility for specific food usage, lifetime barrier properties, and environmental footprint. It can be used for hot and

cold food products and for microwave reheating. Due to plastic's good barrier properties against water, carbon dioxide, oxygen, and nitrogen, products in plastic packaging retain their flavor, aroma, and nutritional value and are protected from external contamination. Lightweight plastic packaging also contributes to lighter truck loads, which helps to reduce the energy used for transportation, lowers shipping costs, and reduces the amount of waste generated [11,38]. Furthermore, design freedom, durability, and cost–effectiveness are some of the most important advantages of plastic usage for food packaging [39].

2.1.1. Polymeric Food Packaging Materials

The most common plastics used by the food packaging industry are these seven: PET, HDPE, LDPE, PC, PVDC, PP, and PS [40,41]. Regarding the PVC usage for food packaging, health concerns have risen [42,43], mainly due to the migration of plasticizer findings [44]. However, its usage in the EU is not prohibited, and it complies with the applicable harmonized regulations [45]. Ortho-phthalates, referred to as phthalates, expose human health to risks [46], even though researchers are trying to design efficient PVC plasticizers with reduced migration rates [47]. In particular, concerning honey products, the plastic materials used for honey packaging are PET, LDPE, and HDPE where honey bottles are concerned [48]. Generally, copolymer PET12/LDPE is the most-used clear package for honey products, as it can meet the fundamental requirements for a honey sachet package at the lowest cost [49]. Lastly, in pilot honey packaging studies that were performed in our laboratory using Raman spectroscopy, we confirmed that PVC is one of the main honey packaging materials. Furthermore, LDPE and PP were also found to be used as plastic packaging for honey sachets, as well as PET and PS for honey bottles and boxes.

2.1.2. Polymer Degradation Parameters

Polymer degradation, referring to the alteration of the physical properties of a polymer, is linked to changes in its chemical composition, which are caused by oxidation or other external factors such as temperature, light, and moisture. Additionally, when food is stored or heated inside a plastic package, factors like temperature and UV radiation could possibly affect the chemical stability of the polymer packaging [50,51]. Concurrently, liquid foodstuffs can possibly affect the inner side of the package, since their properties such as acidity (pH), the water content, and viscosity are also affected by temperature, sunlight (UV radiation), or by a forced heating effect. In a recent study where polymer degradation was conducted, plastic items were exposed for a period of one week to UV light, UV light and water (stirred at room temperature), only water (30 °C), and only heat (40 °C) [52]. Prolonged UV exposure caused increased mechanical degradation in polyolefins such as PE and PP due to changes in the polymer structure. Specifically, prolonged exposure of PE and PP to UV light at wavelengths 300 and 370 nm, respectively, caused polymer degradation [53]. Through Isotopic signature analysis (13C/12C) for PP candy containers, it was determined that UV degradation was equal to degradation from hot water with light and more than the degradation caused only by heat at 40 °C [52]. Generally, polymer science suggests that polymers, especially nylon and PET, degrade at temperatures above 95 °C, which means that the molecular structure of the polymer itself undergoes disturbances [54].

2.2. Glass Packaging

Glass packaging is the most reliable, safer, and healthier food packaging material, especially for honeys [55]. It has a lot of advantages, such as 100% recyclability, almost zero chemical interactions with the food, and durability. These advantages prevent contamination and help long-term preservation, keeping the aroma and flavor almost unchanged [56,57].

2.3. Metal Packaging

Metal packaging such as "tinned iron canisters" [58] were firstly made in England in 1699 and were first used for roasted beef preservation in 1760. Due to the high recyclability and its magnetic properties, metal packaging is used even nowadays in the food industry, especially for packaging cheese spread, mustard cream, butter, honey, condensed milk, mayonnaise, tomato ketchup, jams, jellies [58], and many others. The most common types of metals used in food packaging is aluminum, stainless steel, and free steel [58].

There are some safety concerns about metal packaging due to the migration of substances, such as bisphenol A, lead, cadmium, mercury, aluminum, iron, nickel, bulging of the cans, tin dissolution, blackening, and corrosion, causing health problems [58]. For these reasons, metal surfaces are coated with protective lacquers to prevent metal–food interactions and the migration of metal components to the food [58].

3. MPs/NPs Migration from Plastic Packaging into Food

3.1. Generic

Plasticizers and microplastics have been identified in food, having migrated from food packaging materials [46,59]. Human health is at risk, a proven fact by scientific studies that presented that plastics can induce carcinogenesis in humans [2]. Plasticizers such as DEHP, DBP, DEHA, and their metabolites have migrated, especially from fast food packaging materials to foods that, even at low concentrations, can cause endocrine-disrupting effects [60]. For this reason, researchers are focused on studying the use of plasticizers with biodegradable films for ecofriendly food packaging applications [61] and alternative plasticizers safer for human health [62]; however, MP/NP migration is an important problem that still remains a strong research question.

3.2. Important Research Works for MPs/NPs and Plasticizers Migration into Food

Nowadays, unfortunately, almost everything in nature is polluted by MPs/NPs [63], especially aquatic ecosystems [64,65]. There is MP/NP contamination in the food chain, especially in water, drinks, and beverages [66,67]. Recently, scientific research proved that even plastic cutting boards are a source of MPs in meat [68].

3.2.1. Food Simulations

Since the mid–1990s, most research studies have used food simulations to study MP/NP migration from plastic packaging into food. "A food simulant is a chemical with characteristics that imitate food, which is used to model migration of (FCMs) for regulatory testing purposes" [69]. Distilled water is frequently used as a simulant for aqueous foods (pH > 4.5), acetic acid in water for acidic foods (pH $\leq$ 4.5), and 95% ethanol in water for fatty foods; the migration rate is usually measured by bringing into contact the plastic films under investigation with the simulant [70]. Additionally, olive and corn oil were used as simulants for fatty foods [71]. The migration testing of plasticizers [71] but, also, plastic monomers from plastic packaging began in 1989 [72]. Generally, the one side of plastic (plastic films [73]) is exposed to oil to study the migration, and the detection is achieved with GC or/and HPLC [74] and HPLC–UV [75] methods in a microwave or heating conditions of the oil (from 40 to 150 °C) [76]; plastics used are special food tapers or roasting bags [75]. In one relevant experiment, after 10 days heating at 44 °C, PP had an overall migration in olive oil of less than 10 mg/dm^2. The total migration in water, 10% ethanol, and 3% acetic acid simulants was lower than that in olive oil [77]. Generally, it was observed that, above 44 °C, there is migration from food packaging [73]. Recently, similar studies were conducted to observe the migration of a styrene monomer and polystyrene fragments in food and drink simulants from PS packaging, where PS samples were cut into small pieces 2 to 3 cm in size and, after that, were immersed in 100 mL of the simulant, as is presented in the following Table 1 [69,71].

Table 1. Food simulants used for migration testing [69,71].

Simulants	Contact Foods
10% Ethanol/distilled water	Aqueous foods (pH > 4.5)
3% Acetic acid	Acidic food (pH < 4.5)
50% aq. Ethanol	Diary food products
95% aq. Ethanol, olive, and corn oil	High fat content foods

3.2.2. Real Food Samples

Since 1995, migrant determination from nylon food packaging was studied [78]. Most of the research works were oriented toward the migration of chemical substances with food simulations. Despite this fact, there are some studies for MP migration from plastic packaging into real food samples. In 2003, scientists tested the migration of ethylene terephthalate oligomers from roasting bags into olive oil, where the HPLC–UV method could quantify ethylene terephthalate oligomers in olive oil [75]. Furthermore, dairy products with different fat contents packed in PS containers were studied according to the storage period and temperature. The observations were that, by increasing the temperature and extending the storage time, the migration rate of styrene was greater, and the fat content intensified the migration rate even more [79]. Recently, our lab documented PE migration from food packaging in edam, kefalotyri, and parmesan cheese samples using Raman and infrared (ATR/FTIR) spectroscopy [13]. The experimental procedure is described in Figure 1.

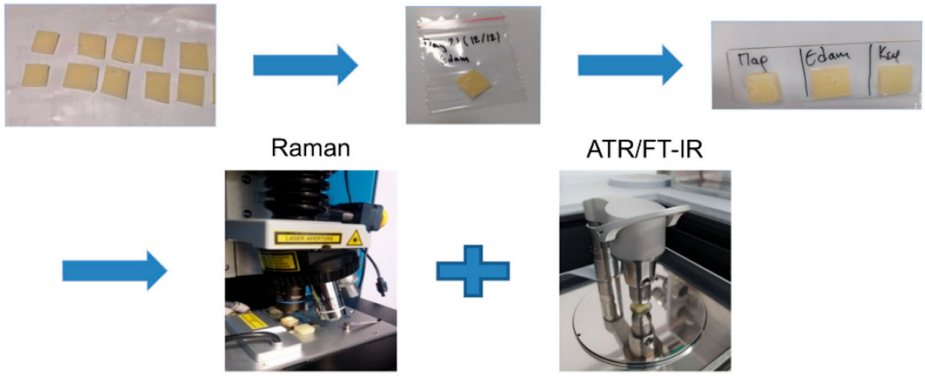

Figure 1. Sample preparation process [13].

Additionally, MP migration from food packaging was also studied in meat samples. Specifically, the surface of chicken meat was studied for PS MPs due to the use of food trays made of XPS in meat product placement and transportation. Researchers found that MP particles were trapped between the meat food tray containers and the sealing film; an analysis proved that the MP XPS per kg of packaged meat ranges from 4.0 to 18.7 [80].

Other important works, with the input of SEM and FTIR, demonstrated that MP migration occurs during the normal use of PE rice cooking bags and ice cube bags, as well as of nylon teabags. The MPs released from teabag samples were 1.13 ± 0.07 mg of nylon from each teabag; the effect of temperature is strong in the release of MPs [81]. Finally, the first study of its kind on the MP contamination of soft drinks, cold tea, and energy drinks was in 2022 [82], which, in combination with the research work on the investigation of MPs/NPs in packaged beverages [83] intended for human consumption, are very triggering for future research and environmental considerations about human food and drink consumption, since most studies have focused on bottled water, beer, milk, and refreshments display possible human risks [2,59].

4. Honey Properties and Postharvest Handling

Honey, as a viscous nonhomogeneous colloid substance, has a lot of different properties that change under the effects of heat and time storage. Different types of honey, based on floral origin, have their properties differentiated, such as pH, water content, viscosity, sugar content, and color parameters. All these factors could potentially affect the plastic migration from plastic packaging.

4.1. Types of Honey

There is a plethora of honey types around the world [84]. Their differences are based on their floral, geographical origin, mode of production, and/or presentation [85,86]. Usually, it is labeled after the botanical source it is produced from, like Acacia, Linden, Floral, Buckwheat, Heather, Rape, Honeydew, and Nectar honeydew [87]. Honey can be discriminated according to origin or postharvest manipulation, as seen in Table 2.

Table 2. Honey type discrimination methods.

Honey Discrimination	Examples of Honey Types
Geographical origin	Linden (Europe), Orange blossom (Spain and Mexico), Clover (New Zealand and Canada), Sage (California), Eucalyptus (Australia), Tupelo (Georgia/Florida), Dandelion (Eurasia), Wildflower (multiple flower sources or varieties), Manuka (New Zealand [88], Treatment of wounds [89]), Acacia (North America and Europe), Buckwheat (United States and parts of Canada), Sourwood (Southeast and Midwestern United States), Floral (Greek honey [90])
Flower origin	Multi-flower Monofloral honeys: Acacia, litchi, orange, coffee lavender, blueberry, Eucalyptus, Rosemary, Forest [91]
Mode of production and/or presentation	Comb honey, chunk honey or cut comb in honey, drained honey, extracted honey, pressed honey, filtered honey

Finally, a lot of studies have been conducted the last two decades concerning the examination of honey properties from different honey types from all over the word. Asian honeys such as red pine honey from Turkey [92], Malaysian honeys [93], Iranian honeys [94,95], European honeys such as Polish honeys [96], Greek honeys [97], Belgium honeys [98], American honeys such as Yatei honey from Argentina [99], Apis mellifera L. honey from Central Brazil [24], African Moroccan honeys [100], and bitter and sweet honey from the Mediterranean coast of Algeria [101] have been examined for their special properties, both physical and chemical. The list is enormously large and unending.

4.2. Honey Properties

Each type of honey has special properties due to their unique physicochemical parameters, such as electrical conductivity; water content; total acidity; total sugar; sucrose; viscosity; pH; and color parameters such as L*, α*, and b* [87]. Additionally, different honeys have different phenolic and flavonoid contents, which has a high correlation with their antioxidant activity [86,93] and with their antimicrobial activity [22,97]. Factors such as low pH, hydrogen peroxide, and high osmolarity contribute to the high antibacterial activity of honey [97].

4.2.1. Viscosity/Crystallization

One of the most important rheological properties of honey is viscosity, influenced by several factors such as temperature, moisture content, and composition [102]. Different types of honey have widely different viscosities due to the possible presence of colloids and differences in the moisture contents [103]. In general, the viscosity increases when de-

creasing the moisture content [102]. Additionally, most honeys are glucose-supersaturated solutions, so crystallization may spontaneously occur at room temperature with a crystallization peak between 10 and 15 °C, forming a glucose monohydrate [103].

In 2013, important research about viscosity of honey was conducted in four types of honeys (Brazilian, Australian, Chinese, and Romanian Acacia honey), chosen for their different crystallization rates and compositions. For the viscosity measurements, the crystallized samples were incubated constantly at 14 °C. The viscosity of Chinese and Brazilian honeys increased after 24 h processing due to the rapid crystal growth. However, the viscosity change in Acacia honey was slower due to the lower rate of crystal growth and the initial viscosity [102]. Interestingly, although the viscosity of the Australian honey was high compared to the Brazilian and the Chinese honey, the change in the viscosity was higher than the Acacia honey as time progressed. Based on the literature, honey crystallizes faster with a greater glucose content (above 30%). Despite this, Acacia honey with a glucose content of 28.5% was found to have a lower crystallization rate than the Brazilian, Chinese, and Australian honeys. "Glucose content of the honey sample has no consistency in predicting the crystallization tendency and honey crystallizes more rapidly when this glucose/fructose ratio is less than 1.14". Additionally, at low temperatures (14 °C), the crystallization phenomenon is limited by the viscosity, which means that as the viscosity of the honey increases, the crystallization rate decreases [102].

As we mentioned above, the viscosity influences the rate of crystallization, because it directly alters the diffusivity of glucose through the honey solution [102]. The solubility of glucose is increased when the temperature is also increased, while this lowers the honey's tendency to crystallize. Lowering the temperature also lowers the diffusion coefficient of glucose, resulting in delayed crystallization. However, as we mentioned before, there is an optimal temperature for the crystallization rate; this temperature is found to be between 10 and 15 °C [102]. The most important factor for the crystallization rate in honey is the glucose/water ratio [103]. However, the presence of fructose in honey is an especially important factor, because it inhibits glucose crystallization. Furthermore, the crystal growth rate was found to be related to the glucose content and inversely related to the viscosity of the honey [102]. Glucose has been investigated for its ability to degrade polymers [104,105], so it is worth mentioning it as a possible factor for polymeric migration in foodstuffs.

4.2.2. Viscosity/Heat

In a related article, it was shown that dynamic viscosity was affected by the heating of raw rape honey at various temperatures (30, 40, 50, 60, 70, and 80 °C). The viscosity after 20 °C started to significantly decrease at 30, 40, and 50 °C for all individual samples. The effect of temperature was remarkable for temperatures up to 30 °C but was not significant above that temperature [106].

4.2.3. Viscosity/Water Activity

In another study, 35 samples of *Apis mellifera* L. honey were collected, stored at room temperature, and kept in dry, airy conditions to wait for yeast and mold analyses. There was no direct relationship between the moisture content levels and the presence of these microorganism, but it was revealed that the moisture content influences honey viscosity [24]. The molal concentration of the main sugars determines, in a significant way, the water activity of the honey, mainly by the monosaccharides fructose and glucose and not so much by the disaccharides maltose and/or sucrose. In a research work regarding water activity focused on 49 crystallized and redissolved honey samples from Argentina, the water activity of the crystallized honey samples was higher than the redissolved ones upon heating. When the temperature was increased, the solubility of glucose was also increased, so its tendency toward crystallization was lower [102]. As the glucose concentration increased, all sugar solutions became increasingly viscous, and the mobility of the sugar molecules decreased as the viscosity increased. As crystallization occurred at room temperature, the glucose

concentration in the liquid phase was lowered, and thus, the water activity increased as the viscosity was lowered [103].

4.2.4. pH

The acidity of honey is partly responsible for its characteristic taste. Honey is considered mildly acidic due to the minor acid content (mainly amino acids and organic acids), with an average pH of 3.9. Honey from tropical countries has lower acidity due to its water content, resulting in increased fermentation and a further decrease in the pH. More acidic values (pH < 3.24) indicate, in some cases, the existence of impurities in the samples or improper storage [107].

As mentioned before, acidic foods, especially liquids, contribute to plastic oxidation and degradation [48,49]. Researchers observed that, when the moisture level is higher, the development of microorganisms is greater, so the total acidity increases due to fermentation. Osmophilic yeasts ferment glucose and fructose in honey, forming alcohol and carbon dioxide, where, in the presence of oxygen, alcohol is broken down into acetic acid and water, further decreasing the pH of the honey. However, a high level of acidity is not always due to fermentation by microorganisms. Acidity in honey is dependent on many factors, such as floral sources, amount of minerals, and gluconic acid [24].

The temperature also contributes to the pH of honeys. Recently, it was reported that, by increasing the heating temperature of honey, the pH decreases rapidly due to the chemical reaction between sugars and amino acids [23]. According to a honey study, the researchers found that the pH at 20 °C of the honey types tested was quite lower compared to the general honeys, due to an enzymatic action that caused a great amount of organic acids. The honeys had different botanical and geographical origins, such as Acacia (native to Hungary), orange (United Mexican States), lavender (Spain), blueberry (Canada), litchi (People's Republic of China), and coffee (Guatemala) [91].

In another study of Yatei honey, stored at 15–38 °C and at refrigeration temperatures 7–10 °C using glass and PP translucent containers, it was found that the pH decreased after 15 days. Later, in time, the acidity increased in the samples stored at room temperature [99].

4.2.5. Color

Honey color is influenced by the honey contents (phenolics [108], carotenoids, sugars, minerals, and pollens); water content; floral and geographical origin; temperature; age; time; and conditions of storage [94,109]. The same types of honey at different ages have differences in L* and chroma [109]. An example of a color determination measurement study in honeys was made in 2021, where 31 Acacia (*Robinia pseudoacacia*), 10 Linden (*Tilia* spp.) 7 chestnut (*Castanea sativa*), 7 honeydew, 7 sunflower (*Helianthus annuus*), 5 silkgrass (*Asclepias syriaca*), and 6 rapeseeds (*Brassica napus*) honeys were stored in darkness at room temperature and measured based on the UV–Vis transmission spectra. [109]. Examples of different honey colors is shown in the following Figure 2.

In another study, it was confirmed that the Maillard reaction occurs in unheated Polish-originated honeys, where the BPF was strongly correlated with antioxidant activity. Floral, Acacia (*Acacia* Mill.), buckwheat (*Fagopyrum esculentum* Moench.), heather (*Calluna vulgaris* Hull), linden (*Tilia cordata* Mill.), rapeseed (*Brassica napus* L.), and multiflorous honey samples were collected during the 2017 season in the Warmia and Mazury region in the northeastern part of Poland and were kept in a jar in darkness at room temperature (20–22 °C) before analysis. Dark honeys such as buckwheat and heather showed better antioxidant activity and higher melanoidin contents compared to light honeys. Additionally, linden and rapeseed had greater values in the L* and b* parameters, and a* was higher in buckwheat and heather honeys [96].

Figure 2. Examples of different colors of honey from Greece (**left**) and Mostar, Bosnia, and Herzegovina (**right**, royalty-free stock photo) [110].

4.2.6. Optical Properties

Different types of honey usually have different RI, even among the same types of honey samples. For example, red pine honeys from different locations of Mugla Province in Turkey have RI between 1.4944 and 1.4996, depending on the moisture and maturity degree of the honey [92]. For Moroccan honey samples, the RI was found to be 1.49028 for the eucalyptus type and 1.49300 for the herbal type of honey [100]. For bitter and sweet honey from the Mediterranean coast of Algeria, the RI was found to be 1.4845 and 1.4915, respectively [101].

The RI and water content in honey samples are correlated with each other [95]; this is the reason why we have to know the RI value of a honey sample, because a high amount of water causes a major loss of flavor and quality in the honey due to the higher fermentation rate [101]. Generally, when we have a loss of the water content, the RI increases [111]. In some cases, the RI increases with the solid content of honey, and the moisture content is negatively correlated to the RI; if honey is less solid, light moves faster through it [108]. The RI is an accurate indicator of the total soluble solid concentration in liquid samples such as honey [111].

Long periods of storage and heating conditions do not only influence the color and optical density of honey but the RI as well. Specifically, the RI was slightly reduced at higher temperatures (40 °C) compared to 20 °C in Egyptian clover honey samples [22]. In another study, Água-mel, a traditional Portuguese honey, was thermally processed, causing a loss of moisture and exponential increase of the RI from 1.38478 at time zero to 1.47285 after 400 min [111]. The RI definition can also help sugar profile investigation in honeys [112], such as Saudi honey examination through a HPLC system equipped with refractive index and diode array detectors [113]. However, an RI detector is typically used for carbohydrates detection [112].

As mentioned before, the optical properties of rape honey can be modified by changing the morphology of the crystalline structure when honey is heated between 60 and 80 °C, causing a lightening of the honey [106]. The browning intensity of different monofloral honey types such as Acacia, orange, lavender, blueberry, litchi, and coffee is affected by the botanical origin of honeys during the heating process at 100 °C for 24 h, resulting in different UV absorbance ratios [16].

4.3. Factors That Affect Honey Properties

The main factors that could affect honey properties are based on their storage conditions. Those are identified to be three enlisted based on their significance:

4.3.1. Storage Time

The storage time is a critical parameter in food preservation. Usually, as the storage time is increased, food preservation is gradually affected due to the development of mi-

croorganisms. Additionally, the storage time affects polymer migration into food from their plastic packaging. As an example, mentioned previously, dairy products stored in polystyrene containers present higher migration rates of styrene through time, while the fat content intensifies the migration rate even more compared to storage time [79].

Regarding honey, long periods of honey storage influence the color, optical density, and RI [22]. Generally, honey storage initiates melanoidin formation, resulting in reducing sugars and polyphenols [114]. Specifically, in one study, important volatile compounds such as terpenes were present in higher concentrations in fresh citrus honeys compared to stored honeys; at the same time, it was observed that, during prolonged storage, lower levels of monosaccharides and higher levels of maltose occurred [115]. In another study, it was found that, in almost all the samples, cis- and trans-linalool oxide and hotrienol increased over 540 days, and in total, 24 volatile compounds showed higher abundances during storage [116]. However, the quality of honey is not significantly affected by the storage time for up to two years if the honey is properly harvested, extracted, and preserved at room temperature [117]. Lastly, in bracatinga honeydew honey, long storage conditions cause the formation of brown pigments and, consequently, the degradation of the honey quality [118].

4.3.2. Temperature

The moisture content, free acidity, and electrical conductivity of raw rape honey are not affected by storage temperatures; freezing temperatures maintain the freshness and color of honey, even though the viscosity is increased. Nevertheless, the storage of honey at room temperature causes color changing [31,119].

Raising the honey temperature reduces its viscosity and water content. Heating or cooling has been found to affect liquid foods by changing their viscosity, pH, acidity, and antioxidant activity [23]. Heat treatment leads to the development of antioxidant activity due to the formation of Maillard reaction products, affecting human health in a positive way [120].

In particular, the heating of raw rape honey for 15 min at a temperature range of 50–80 °C significantly altered its chromatic parameters (increased its L* and b* but decreased its a*) and decreased its dynamic viscosity [106].

The heat treatment of honeys from the Warmia and Mazury region in Poland enhanced the antioxidant activity in all the samples, the melanoidin content, and the TPC of the honey [91,96]. Additionally, the Maillard reaction and melanoidin formation occurred in unheated Polish-originated honeys [96]. Additionally, honeys heated at 100 °C for 24 h presented an increased amino-free group of all the honeys. The sugar contents of lavender, litchi, coffee, Acacia, and blueberry honeys increased with time, but for Acacia and blueberry, it decreased after 6 h of heating. Orange honey showed an increase at 12 h [91]. Additionally, the pHs of different honeys tested at 20 °C were lower compared to the general honeys, due to large amounts of organic acids produced by enzymatic action [91]. Lastly, during the heating process, the browning intensity of the honey types were affected by the botanical origins of different monofloral honeys, resulting in the different UV absorbances at 284 and 420 nm [91].

In another study, it was found that, by increasing the temperature (up to 65 °C) and the heating time, the color of the Jujube honey from Iran was darkened, and the phenolic content in the honey increased with the linear increment of heat (50, 60, and 70 °C) over a period of 12 days [94]. Additionally, studies of Yatei honey, stored at 15–38 °C and in refrigeration temperatures of 7–10 °C using glass and PP translucent containers demonstrated increased acidity and decreased pH at 15 days. The acidity remained unchanged in the samples stored in the refrigerator while increased in the samples stored at room temperature [99]. Furthermore, it was found that the moisture content of the samples was affected by the air humidity; specifically, the moisture content of the samples stored at room temperature increased, while those stored in the refrigerator presented stable moisture content values. Additionally, honey stored at room temperature and kept in a dry place (South Arabia)

presented an increased total acidity with a higher moisture level, because fermentation was increased due to the greater development of microorganisms [24,107]. Why research is interested in acidity is because acidic foods, especially liquids, contribute to plastic oxidation and degradation [121,122]. Crystallization can lower the glucose concentration in the liquid phase of the honey, thus increasing the water activity, which can potentially allow the multiplication of naturally occurring yeasts cells, resulting in honey fermentation [103]. Lastly, it was shown that the thermal processing of Água-mel (traditional Portuguese Honey) caused a gradual decrease in the sugars, forming brown pigments [111].

As we mentioned before, the heating of honey affects its color. In a related study, the heating of raw rape honey for 15 min at a temperature range of 50–80 °C significantly altered its chromatic parameters, increasing the L* and b* but decreasing the a*. Heating in these temperatures did not significantly degrade the quality of the honey. When the heating was between 25 and 45 °C, the fraction of the smallest crystals was increased from 65.1% to 72.2%. The share of the large crystals decreased from 2.1 to 0.6%, changing the morphology of the crystalline structure of the honey. This occurs especially when heating honey between 60 and 80 °C, where the optical properties of the honey will be modified, such as increasing its lightening [106]. In a commercial floral honey incubated at 50 and 60 °C for 12 days and 70 °C for 10 days, it was observed that the antioxidant activity and BPF [123] and TPC [94] were increased with the treatment temperature and time, which, despite the positive effects of antioxidants on human health, the BPF is not desirable for consumption [120].

In pH studies, the effect of heating time on the browning index was studied in commercially available honey types. Initially, the color of Acacia, orange, lavender, and blueberry honey was glossy and bright clear yellow–orange, where the color of litchi and coffee honey was glossy, bright, clear, and dark brown. Acacia honey had the strongest whiteness and b*, due to its highest levels of L* and b* values. Blueberry honey was strong in its a*, followed by lavender, in contrast to coffee honey, which had strong blackness and weak a* and b* colors due to the lowest L*, a*, and b* values. After honey was heated at 100 °C for 24 h, the UV absorbance and the browning intensity were measured over time at 284 and 420 nm, respectively. The progression of the initial browning in each honey was proven, while the UV absorbance at 284 nm was gradually increased [91].

Lastly, in another study, it was found that, increasing the temperature (50, 60, and 70 °C) and the heating time (up to 12 days), the color of the Jujube honey from Iran was darkened due to the BPF increment, while the phenolic content in the honey was increased. Darker honey samples are associated with greater levels of polyphenols, possibly due to the Maillard reaction that produces different compounds.

4.3.3. Humidity, Water Activity, and Moisture Content

The moisture content is very important for the preservation of honey properties. The moisture content of honey influences the flavor, preservation, viscosity, specific weight, crystallization, and palatability and contributes to the development of fermenting microorganisms [24]. As we know, the higher the moisture level, the greater the development of microorganisms, which increases the total acidity of the honey [24]. The moisture content in honey is typically between 16% and 18%. However, some honeys may have a moisture content as low as 13%, whereas, in others, it can reach up to 29% [24,102,103].

Yatei honey is produced by Apis mellifera. It is a special honey, less viscous and more acidic, with a particular aroma. Some honey samples were obtained from the central zone of the Province of Misiones and examined during different storage conditions. The results for the room temperature storage showed that the humidity of the samples in glass containers increased until reaching a value of 26.5% at 90 days and, in plastic containers, a value of 26.8%. These values remained stable at 120 and 180 days of storage, respectively. For samples preserved in refrigeration, the humidity values were stable, regardless of the type of packaging [99]. Generally, time and temperature significantly affect the humidity of the honey, which is a very important parameter for the honey quality, delineating the stability against fermentation and granulation [23].

5. Recent Evidence and Spectroscopic Analytical Techniques on the Presence of MPs/NPs in Honey

Raman and FTIR spectroscopy analyses have contributed a lot to food quality, including honey analysis, MP/NP analysis, and general food analysis. In the last two decades, there have been a lot of spectroscopic investigations for the classification and study of honey's ingredients and properties but very few regarding the presence of MPs.

5.1. Honey Characterization through Spectroscopic Techniques

The characterization and classification of honey have been accomplished by fluorescence spectroscopy processed with a factor analysis and partial least squares-discriminant analysis methods (PLS-DA) [25].

Recently, food analyses, especially honey characterization, are mostly successful through Raman [31] and FTIR [30] analyses, laser-induced breakdown spectroscopy [124], photoacoustic spectroscopy [32], and general optical spectroscopy methods, often combined with a multivariate statistical analysis [33]. FTIR has been used mainly for water content measurements and sugar (fructose and glucose) identification in honey [26,27]. A Raman spectroscopy analysis was able to discriminate honey samples from different geographical origins due to different sugar fingerprints [28,29]. Previously, using a multivariate analysis, ATR-FTIR spectroscopy could also discriminate Anatolian honey samples of different botanical origins [125]. The classification of the botanical origin of honey was achieved, as for other transparent or semitransparent products, using the visible light spectra transmitted through a relatively thin layer of honey samples [126]. Additionally, chemometric methods (PCA and PLS) and an artificial neural network were applied with Raman spectroscopy as a rapid method for the quantification of glucose, fructose, sucrose, and maltose contents in honey samples [34]. A combination FTIR–Raman spectroscopy method was used for mass percentage determination of fructose and glucose, evaluated with a standard HPLC-based method [127]. Consequently, Raman and FTIR analyses in different honeys all over the word were conducted for the prediction of the phenolic compounds and antioxidant activity [128]. Sugars and phenolic compounds of honey powders were identified with the use of GC-MS, FTIR spectroscopy, and X-ray diffraction, combining the advantages of each method: the separation and detection of the components, vibrations of the components, and their interactions within the samples as spectra and microstructure analyses, respectively [35]. Additionally, chemometrics have been used for the determination of physicochemical properties in the honey, samples combined with ATR-FTIR [129].

Raman and FTIR analyses were also used for the adulterant [130,131] and authenticity analyses of various honeys [18,131,132]. For instance, Raman spectroscopy combined with air PLS-LDA was successfully applied for adulterant detection in honey, such as high fructose corn syrup and maltose syrup, where the Raman spectroscopy fluorescence background was reduced by the PLS pretreatment [133]. Additionally, Raman spectroscopy in combination with HPLC techniques was used for the verification of honey authenticity [134], aiming to create an authenticity pattern recognition analysis protocol [135]. Lately, honey adulterant and authenticity were studied with other spectroscopy methods, such as Visible–Near-infrared spectroscopy [136], fluorescence spectroscopy, chemometrics [16], and UV–Vis spectroscopy [137]. Finally, machine learning algorithms combined with Raman spectroscopy is an expeditious growing field in food science, such as, for example, the study of adulterant in Suichang native honey [138].

5.2. Spectroscopic Determination of MPs/NPs in Honey and Foodstuffs

5.2.1. Chemical Compounds in Honey—General Overview

Are honey bees eventually at risk due to exposure in MPs? A lot of different chemical compounds have been detected in honey samples, such as styrene [36], plasticizers (phthalates [36,139] and DEHA [140]), bisphenol A [36,139], TBOEP (flame retardant) [140], and microplastics [9,10]. In studies where honey bees were exposed to PS MPs, showed sublethal effects. There were no changes to the weight, but alterations in the expression

of antioxidative detoxification and immune system-related genes in bees' guts, where PS MPs interacted with gut bacteria due to their accumulation and degradation inside of the hindgut [141,142], but still, a broad field of research remains to be studied.

The extraction of MPs from honey has not been studied thoroughly. Only a couple of studies have been done so far, mostly in laboratory conditions. The detection, isolation, and origin determination of synthetic microparticles in honey samples is not an easy challenge for researchers. Large numbers of natural and synthetic foreign particles have been found in honey samples from different parts of the world (Germany, France, Italy, Spain, and Mexico) [143]. The origins of the synthetic particles found in honey were studied, resulting in a large intake of foreign particles coming from honey harvesting, processing, and packaging. However, synthetic fibers and fragments were already present in the honey bees' feed, body, and wings, transferred to the hive from the blossoms [144,145].

5.2.2. Spectroscopic Detection of MPs/NPs in Honey

Raman and FTIR spectroscopy have been successfully used the last few years for the detection and identification of MPs in foodstuffs such as honey [146]. In a recent study, honey samples from Switzerland were investigated for the presence of MPs. The majority of isolated fibers identified were cellulose and PET with textile origins, detected by light microscopy and chemically characterized by Raman and FTIR spectroscopy [10]. Another study revealed that LDPE, HDPE, PP, and polyacrylamide were identified with FTIR in honey, produced in Ecuador [9]. In parallel, some studies detected "plasticizers" such as phthalates, migrating from plastic containers to the honey [139]. There is a plurality of evidence for plastic-related chemicals contaminating honey through plastic packaging migration [140]. For instance, a DLLME–GC–MS method was developed to evaluate the migration of chemical compounds from PET and PS containers to honey, where fifteen target compounds were found and quantified [139].

5.2.3. Spectroscopic Detection of MPs/NPs in Foodstuffs

Approximately 11.6 billion MPs and 3.1 billion NPs (nylon and PET) were detected in a single cup of tea. Particularly, 2.3 million particles in sizes 1−150 µm and 14.7 billion submicron plastic particles were estimated to be released in a single cup of tea. Tea was filtered with a 2.5-µm pore cellulose filter, which was dried and scanned with a FTIR/ATR analysis [54].

A combinational approach of FPA-based micro-FTIR spectroscopy and micro-Raman spectroscopy was used for the detection and characterization of MPs in mussels originating from 12 different countries. Due to their filter-feeding nature, mussels are at a high risk of contamination by MPs. MPs such as PP, PE, polystyrene (PS), and PET were detected and characterized [147–149]. Furthermore, Raman spectroscopy provided evidence for the uptake of plastic microparticles by plant species [65,150]. The detection of MPs in white wine resulted in findings of synthetic PE stoppers under micro-Raman spectroscopy [151].

The NR staining method for polymeric particles is another approach widely used for the detection of MPs/NPs also in combination with Raman and FTIR spectroscopy [151–153]. It can be used for a variety of plastics, such as PP, PE, PET, and PS of different sizes (100–1000 µm) and shapes (sphere, fiber, film, and flake), improving the identification time [154,155]. Lastly, Raman and FTIR spectroscopy are further used in the detection of MPs/NPs on filter membranes for their analytical capabilities and, also, for their low-cost usage [156,157]. The imaging and mapping of surfaces for the detection and visualization of MPs is constantly under development and continuous improvements [158–161].

The ongoing research and development in the spectroscopic methods of detection and analysis can potentially be beneficial to the research field studying the migration of MPs in food.

6. Discussion

It is well-known that plastic packaging is ecotoxic and unhealthy for humans [162] and, generally, for the environment [63]. MPs are globally detected in freshwaters and

drinking water, with PE and PP detected at a higher rate, followed by PS, PVC, and PET [163] in honey [9,10] and, generally, in food. Their isolation complexity is mainly based on their broad size range, various chemical composition, shapes, and surface properties such as charge and hydrophobicity [163]. MP degradation rates, transport mechanisms, long-term accumulation, and all the possible potential consequences of MP deposition in the environment are important fields that have to be studied for the protection of human health [63]. We already know that MPs/NPs have been proven as risk factors for the growth, survival, and reproduction in nematodes and earthworms [164]. There are land-to-sea pathways that deposit MPs/NPs (40–50 nm) in the soil and oceans and can be absorbed and transferred by terrestrial plants. These particles can interfere with plant growth, causing oxidative, genotoxic, and antioxidant system damage [63]. They have also been found in edible species (fruits and vegetables [150]) commonly consumed, supposing a probable transfer to humans by consumption [63]. The latest research studies showed the presence of MPs in commercial fish, which raises the concern about the human health risk [165], especially in human digestive tract cells [166]. MPs were found in the human placenta [167] and can cause damage in mice kidneys [168] and human liver [169], and it is considered possible that MPs can cause genotoxic potential in human peripheral blood lymphocytes [170].

Some kinds of foods such as honey are very important for the human diet. Honey consumption is beneficial to the human body due to vitamins, phenolic and flavonoid contents, and antioxidant substances, which offer antibacterial benefits [86,97]. However, honey could be contaminated by MPs [9,10], plasticizers [36,139,140], and other dangerous chemical substances such as bisphenol A [36,139]. Honeybees were found to be active samplers for MPs, where particles were detected on their body and wings [145]. It is assumed that the MP presence in honey could possibly be traced back to either honeybees or to microplastic migration from honey packaging or to the honey preparation and processing procedures, while, in any case, the damage is the same. Concluding, notwithstanding that science has proven that MPs exist as a global problem, there are some important research gaps mainly concerning human health [37]. For this reason, there is also the need for a global, generalized, easy, and ecofriendly common protocol for the extraction and identification of MPs/NPs, starting from the air and soil [171] and moving onward to the rest of the food samples, such as honey.

7. Conclusions

Following this detailed review, there were many conclusions derived. A variety of different types of plastics have been used for food packaging. There is evidence of plastic migration in food samples and, particularly, in honey products. Honey is classified into multiple different types with different properties such as the viscosity, pH, and moisture content. Storage conditions such as temperature, humidity, and light affect the acceleration of plastic packaging degradation. Particularly, since honey can be kept in extended storage periods, it potentially allows higher degradation rates of its plastic packaging. Unfortunately, there is a lack in this research field; the literature regarding MP migration to honey is very limited. Although spectroscopic and analytical techniques such as Raman, FTIR, HPLC, and GC-MS are in the foreground of the detection and characterization of MPs/NPs in honey, a universal way of isolation and quantification has not yet been developed.

Research should be done on other ways to protect human health, such as reducing the usage of plastic packaging or on developing new packaging materials that are completely resistant to the aforementioned stress conditions, to eliminate the possibility of MP/NP migration. Consumers should pay more attention to how a product is packaged and in what conditions it was stored.

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Abbreviations

The following abbreviations are used in this manuscript:

MPs	microplastics
NPs	nanoplastics
LDPE	low-density polyethylene
HDPE	high-density polyethylene
PET	polyethylene terephthalate
PP	polypropylene
PC	polycarbonate
PVDC	polyvinylidene chloride
PS	polystyrene
DEHA	Di-2-ethylhexyl adipate
TBOEP	tris(2-butoxyethyl) phosphate
UV	ultraviolet
L*	lightness
α*	redness
b*	yellowness
BPF	brown pigment formation
TPC	total phenolic content
RI	refractive index
FTIR	Fourier-transform infrared spectroscopy
PCA	principal component analysis
PLS	partial least squares
LDA	linear discriminant analysis
ATR	Attenuated total reflectance
GC	Gas chromatography
PY	pyrolysis
MS	mass spectrometry
DLLME	Dispersive liquid-liquid microextraction
HPLC	High-performance liquid chromatography
FPA	focal plane array
NR	Nile red
XPS	extruded polystyrene
SEM	scanning electron microscopy
FCMs	Food Contact Materials

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