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Research Article

Low-Pressure Plasma Treatment Increased the Quality and Characteristic Flavor of Lyophilized Lemon Slices

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Low-pressure plasma (LPP) treatment was implemented as a nonthermal technology to preserve the organoleptic and nutritional qualities of lyophilized lemon slices. Effects of LPP treatment on the basic quality and flavor characteristics of lyophilized lemon slices stored for 7 d were evaluated. Fresh lyophilized lemon slices were prepared as a reference. The total titratable acids and sugars were in the range of 1.00~1.11 mg/mL and 190.21~197.37 mg/mL. The total phenolic content and Vc gradually decreased during storage from 1.74 to 1.33 mg/mL and 0.53 to 0.31 mg/mL. LPP had minor effects on total sugars and total titrate acids but prevented the storage losses of vitamin C (Vc) and phenols. A total of 35 volatile organic compounds were identified with C10 monoterpenoids being the major compounds. C6-C9 aldehydes corresponding to a green grassy aroma gradually decreased with increasing storage time. The gallery plot confirmed the different compositions of volatile organic compounds in LPP-treated samples. LPP-treated lemon slices had a higher content of preferred aromatic substances (terpinen-4-ol, α-terpineol, α-terpinene, and limonene) with a citrus-like flavor. These results demonstrated the positive effect of LPP treatment on retaining the flavor characteristics of lemon slices.

1. Introduction

Lemons (*Citrus limon*) belong to the *Citrus* genus within the Rutaceae family. As a functional fruit, lemons are rich in limonene, vitamin C, and multiple trace elements such as calcium, magnesium, and sulfur [1]. Lemon peels are rich in pectin and volatile organic compounds and possess antioxidization and antibacterial properties [2]. Research has indicated that the small quantity of citral in lemons is responsible for the flavor quality and distinctive aroma of lemon oil. The lemon essential oil is extracted from the peel and has been proven to be beneficial in the alleviation of symptoms associated with anxiety [3]. Furthermore, citric acid in lemons is effective in reducing the risk of recurrent stones [4]. Currently, the most commonly used strategies for fruit processing in the fruit industry include juicing, pickling, and lyophilizing [5–7]. Among these treatments, lyophilization is a way to completely preserve the nutritional value of fruits [5]. In China, lyophilized lemon slices are popular amongst consumers due to their extensive use in the preparation of beverages.

Low-pressure plasma (LPP) treatment is an emerging nonthermal processing technique in the food industry, which can utilize the media surrounding food to produce photoelectrons, ions, free radicals, and other active substances [8]. It has been shown that plasma technology can improve the functional value of food by increasing the concentration of bioactive compounds and reducing antinutritional components [9]. Furthermore, cold plasma technology can inhibit the growth of microorganisms and improve the quality of food and has been a potential technology to be applied in the food industry [10, 11]. Research has demonstrated that industrial processing techniques, such as cutting, peeling, and slicing, can lead to microbial contamination by pathogens entering the internal tissues of the fruit [12]. LPP can be an alternative for microorganism control that is more environmentally friendly and healthy than chemical methods such as the use of chlorine or SO_2 [13, 14] and overcomes the deficiencies of traditional physical sterilization such as heat (moist heat and dry heat), radiation (UV and microwave), and filtration [15]. Heating is considered as the most commonly used method worldwide. However, thermal treatments are known to induce the caramelization reaction, which negatively influences the appearance, taste, and nutrition of the fruit, resulting in a loss of quality [16]. Nonthermal processing techniques have attracted an increasing attention in the food industry in order to maintain quality and nutritional value as much as possible. There are many studies focused on the inactivation of contaminating bacteria effect of low-pressure plasma; however, limited research efforts have been aimed at investigating the effect of LPP on the quality of fruit products [17–19]. The application of LPP in the food industry is still in an emerging stage.

Recently, gas chromatography-ion mobility spectrometry (GC-IMS) has been extensively applied in the separation and detection of volatile organic compounds [20]. Compared with gas chromatography-mass spectrometer (GC-MS), GC-IMS technology is able to combine the virtues of both the strong separation ability of gas chromatography and higher sensitivity of ion migration spectrum and can quickly detect trace volatile organic compounds without any sample pretreatment, maximizing the loss of volatile substances during material pretreatment [21, 22]. In addition, the three-dimensional map of different volatile organic compounds obtained by GC-IMS technology can be transformed into the fingerprint of breeze flavor, which is beneficial to recognize the difference in volatile organic compounds content in the samples more intuitively.

A particular focus has been put on nonthermal processing technologies, which are designed to eliminate the adverse effects of heat on food products. Cold plasma generates reactive oxygen species (ROS) in air mixtures, and the production of ozone not only has a highly antimicrobial effect but also causes strong oxidative stress on enzyme inactivation. LPP treatment will significantly reduce protease activity and, however, increase texture profile and color properties. In addition, it can be carried out at room temperature and atmospheric pressure, making it secure and energy-saving. Color, texture, and flavor are important indicators of lemons. Since the relative humidity of the environment can weaken the LPP performance, it may be a suitable tool to maintain the quality of lyophilized lemon slices. Literature reported a controversial conclusion on the effects of cold plasma on polyphenols and flavonoids, which may determine the flavor of lemons. Herein, we investigated the effect of LPP on the basic quality parameters (titratable acids, total sugar, total phenol, vitamin C, and color index) and flavor profile of lyophilized lemon slices. GC-IMS was used to evaluate the effects of LPP treatment on the volatile organic compounds in lyophilized lemon slices.

2. Materials and Methods

2.1. Materials. Lemons were purchased from a fruit market in Guangzhou (China), which belong to the *Citrus* genus within the Rutaceae family. Folinphenol, gallic acid, and acetic acid were obtained from Lvyin Biotechnology Co., Ltd. (Guizhou, China). Deionized water, acetone, hydrochloric acid, acetone, and phenolphthalein were purchased from Pengcai Fine Chemical Co., Ltd. (Langfang, China).

2.2. Sample Preparation. The fresh lemons (approx. 60 g/per lemon) were cut into circular slices with a diameter of 5 cm and thickness of 0.5 cm after careful washing. Then, the lemon slices were lyophilized for 48 h in a freeze-dryer. Next, the lyophilized lemon slices were divided into three groups: G1, G2, and G3. G1 are fresh lyophilized lemon slices without any treatment. G2 are lyophilized lemon slices stored at room temperature for 7 days. G3 are lyophilized lemon slices exposed to low-pressure plasma treatment and then stored at room temperature for 7 days. Two parallel circular aluminum electrodes with a 75 mm distance between them formed the plasma source (Figure 1). Samples were placed between the electrodes. The low-pressure plasma treatment was conducted as described in our previous work [24]. Briefly, the lyophilized lemon slices were treated at a discharge voltage of 40 kV root mean square (RMS) at 0.4 mbar for 1.5 min. All the treatments were carried out in triplicate.

2.3. Titratable Acid (TA) Content. Approximately, 10 g of lyophilized lemon slices from each group were ground into powder and dissolved in 100 mL of distilled water. Two drops of 1% phenolphthalein indicator were added to the solution. A precalibrated 0.05 mol/L sodium hydroxide solution was titrated with the lemon solution (approximately 10 mL). Distilled water was also titrated and used as a blank control. The TA of the sample was calculated according to the following formula, where the equivalent weight of malic acid is 67.04.

$$\text{Titratable acid}\left(\frac{\text{mg}}{\text{mL}}\right) = \frac{(\text{ml base titrant}) \times (\text{N of base in mol/L}) \times \text{equivalent weight of acid}}{\text{sample volume in ml}}. \quad (1)$$

2.4. Total Phenol Content. The method used to extract the polyphenols from the lemon slices was previously described by El-Serafy and El-Sheshtawy [25] and with some modifications. Based on the previous reports, ethanol is the best extracting solvent for determining the total phenol content [26]. Therefore, 98% ethanol was used to extract the phenols from 10 g of powdered samples collected from each group (G1, G2, and G3). Approximately, 1.0 mL of the sample and 0.2 mL of Folin–Ciocalteu reagent were mixed with a magnetic stirrer in a beaker. After 2 min, 1.5 ml of 10% sodium carbonate solution and deionized water were added. The mixtures were incubated at room temperature for 75 min. The absorbance of the mixtures was detected with a spectrophotometer (Shenzhen Pukang Electronics Co., Ltd., Shenzhen, China) at 765 nm. The total phenolic content of each sample was calculated after comparison with a gallic acid standard curve.

2.5. Total Sugar Content. The total sugar content was determined using the phenol sulfuric acid method [27] with a little modification. Approximately, 0.25 g of powdered samples from each group were mixed with 100 mL distilled water and 20 mL concentrated hydrochloric acid. After reflux heating in a boiling water bath (100°C) for 3 h, the mixture was analyzed using an enzyme marker analyzer (Shenzhen Pukang Electronics Co., Ltd., China). The total sugar content was calculated by comparison with the standard curve.

2.6. Vitamin C (Vc) Content. The Vc content of the lyophilized lemon slices was determined using high-performance liquid chromatography (HPLC), according to the method described by Lafarga et al. [28] with some modifications. Approximately, 1 g of powdered samples from each group (G1, G2, and G3) were mixed with 6 mL distilled water and 6 mL ethanol (12%) in a beaker. After stirring for 30 s, the mixture was transferred to a 100 mL volumetric flask. Next, 5 mL of 2 mol/L acetic acids were added, and distilled water was used to fill the remaining volume of the flask. Then, the prepared solution was transferred to an HPLC to measure the vitamin content on a reversed-phase Supelcosil™ LC18 (5 μm) stainless-steel column (250 × 4.6 mm i.d., Supelco, USA). An isocratic solvent system was used (0.1 mL/L of sulfuric acid, pH 2.5–2.6). The flow rate was fixed at 1 mL/min, and the UV-Vis photodiode array detector was set at 254 nm. The identification of vitamin C was performed by comparing the retention time and the obtained spectra to those previously obtained with a standard.

2.7. Color Index of Lyophilized Lemon Slices. The color index of the lyophilized lemon slices was measured using the colorimeter CR400 (Konica Minolta Co., Ltd., Japan). The colorimeter was calibrated with a black and white standard plate prior to any sample measurement. The color index (L^*, a^*, and b^*) was determined at three random surface areas of each sample.

2.8. Analysis of the Volatile Component. The volatile component analysis of lyophilized lemon slices was conducted using an Agilent 490 gas chromatograph (Agilent Technologies, Palo Alto, CA, USA) and IMS instrument (FlavourSpec®, Gesellschaft für Analytische Sensorsystem mbH, Dortmund, Germany). Headspace solid-phase microextraction (HS-SPME) was used to extract the volatile organic compounds from lyophilized lemon slices. Approximately, 0.5 g of ground lemon powder was placed into a 20 mL headspace bottle and incubated at 25°C for 30 min. After that, 100 μL of the analyte was injected into the injector (85°C) for analysis.

GC-IMS conditions are FS-SE-54-CB-115MID column (15m × 0.53 mm), column temperature of 60°C, and high purity N_2 as the carrier gas. The flow program of N_2 was performed as follows: 2 mL/min for 2 min, increased to 10 mL/min within 8 min, to 100 mL/min within 10 min, and to 150 mL/min within 10 min. The analytes were ionized in an ionization chamber by a 3H ionization source with a positive ion mode and then transferred to a 9.8 cm drift tube at a 500 V/cm constant voltage with a 150 ml/min nitrogen flow at 45°C.

The volatile compound contents were estimated using the internal standard quantitative method, which was expressed as an n-hydrocarbon (C_7-C_{30}) equivalent according to the following equation (2) [29]:

$$W_i = f^{'} A_i A_z \times W_z, \quad (2)$$

where W_i represents volatile I; W_z represents the mass of the prepositive hydrocarbon of volatile compound I; A_i and A_z represent the peak area of volatile compound I and the normal hydrocarbon before volatilization, respectively; f' represents the correction factor of volatile compound I. The f' value was assumed as 1 in this study.

2.9. Statistical Analysis. One-way analysis of variance (ANOVA) of the data was performed with Duncan's test using IBM SPSS Statistic software, and a least significant difference (LSD) with a confidence interval of 95% was applied to compare the means. All analyses were conducted in triplicate, and data are given as mean ± standard deviation (SD).

3. Results and Discussion

3.1. Physical and Chemical Properties

3.1.1. Total Titratable Acids, Total Sugars, Total Phenols, and Vc Content. Titratable acidity reflects the quality index of food, as the organic acid content directly affects the flavor,

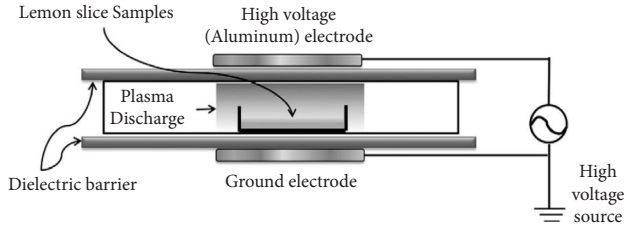


Figure 1: Schematic of the experimental setup for DBD plasma system (derived from [23]).

color, and storage stability of fruit. The total titratable acids of G1, G2, and G3 samples were in the range of 1.00~1.11 mg/mL (as shown in Figure 2(a)). The total sugar, which includes soluble monosaccharides and oligosaccharides in fruit, reflects on the sensory quality, organizational morphology, and nutritional value of products. Meanwhile, total sugar content is a crucial factor that affects the choice of consumers, especially for diabetic consumers. As shown in Figure 2(b), no significant differences ($P > 0.05$) were observed in the total sugar content from group G1 (197.37 mg/mL), G2 (190.21 mg/mL), and G3 (196.76 mg/mL). It is well known that Vc is essential for the normal functioning of the human body. Recent research has reported that the health-promoting effect of Vc might be associated with a low expression of proinflammatory cytokines in the spleen [30]. In Figure 2(c), we found that Vc gradually decreased during storage, while the content of Vc in LPP-treated samples was 4% higher than that in G2. The total phenolic content of fruit is a reference value in antioxidant research and plays an important role in the redevelopment of fruit products [31]. It was found in Figure 2(d) that the total phenol content was 1.74 mg/mL in G1, 1.33 mg/mL in G2, and 1.68 mg/mL in G3. Vc and phenols are reported to be chemically unstable and easily decomposed when exposed to heat, light, and oxygen [32]. Therefore, the phenol and Vc contents in the fresh samples (G1) were higher than that of samples stored for 7 d (G2). Also, the samples treated by LPP showed a higher content of phenol and Vc than that of G2 samples. Farias et al. reported that cold plasma reduced the PPO activity by 46% of apple cubes and 50% of apple juice [33]. Besides, the reactive species of plasma can cause the degradation of the cell membrane and enhance the release of phenolic compounds. There are contradictory reports about the effect of cold plasma on food antioxidants. Referring to Vc, cold plasma creates reactive species in the medium to oxidize Vc into deoxyascorbic acid. In addition, the exposure time is too short to cause the degradation of vitamin content. In this study, we proposed that LPP treatment can reduce the loss of antioxidants such as phenols and Vc that occur in lyophilized lemon slices during storage.

3.1.2. Color Index and Rehydration Rate. The appearance of the fruit is also a very important factor to evaluate the overall quality. In many cases, appearance and color play a significant role in the pricing and selling of fruit. The $L^*.a^*.b^*$ value of lyophilized lemons was determined before and after rehydration using a chronometer, and the results are shown in Figure 3. According to colorimetric guidelines, different parameters can indicate the presence of different colors in samples. The detailed guidelines for colorimetry are as follows: a higher a^* value indicates that the color is closer to pure red, and a lower a^* value indicates a color closer to pure green; a higher b^* value indicates that the color is closer to pure yellow, while a lower b^* value indicates that the color is closer to pure blue. As is well known, the storage conditions (temperature, time, light, oxygen, and moisture content) or the processing would affect the appearance of lemon slices. In this way, the differences between the samples from the three groups were compared using the colorimetric guidelines. In terms of fresh G1 samples (as shown in Figure 3), no significant difference was observed in the L^* value for the samples before ($L^* = 30.63$) and after rehydration ($L^* = 34.63$). However, a decrease in a^* and an increase in b^* were found, which was lower than that of samples after rehydration. It is interesting to note that the brightness (L value) was significantly enhanced after rehydration in fresh or LPP-treated lyophilized lemons after 7-d storage. In contrast, the yellowness (b^* value) showed an opposite trend. The lightness of lyophilized lemons may be explained as a result of the loss of water and the stability of pigments. In terms of LPP treatment, the penetration depths of plasma and the chemical reactions initiated by reactive species are probably the main reasons for changes in color and other physicochemical properties [34]. In this study, LPP treatment was too short to generate a significant change in the color of lyophilized lemons.

3.2. Analysis of Volatile Organic Compounds

3.2.1. GC-IMS Topographic Plot. GC-IMS technology is used for the separation and detection of volatile organic compounds [20]. A three-dimensional (retention time, migration time, and peak intensity) spectrum map of the organic matter by comparing the three groups of the lyophilized lemon slice is presented in Figure 4(a), and a 2D topographic top view plot of GC-IMS is shown in Figure 4(b). In Figure 4(b), each point represents one type of volatile organic compound. The color represents the concentration of the substance, where white indicates a lower concentration and red indicates a higher concentration (the darker the color, the greater the concentration). Most of the signals were distributed in retention times of 100–950 s and a drift time of 1.0–1.75 with a high separation degree. The retention times of some of the ions were between 500 s and 800 s, mainly because these compounds had a low polarity, and it is known that nonpolar compounds tend to retain longer on nonpolar columns than polar compounds [35]. The comparison spectrum showed the difference in the volatile compound content of lyophilized lemon slices more intuitively. Compared to the fresh samples (G1, left), the red plot in G2 (middle) and G3 (right) indicated higher volatile organic compounds, more concentrated in the range of 550–950 s. The blue plot indicated a lower concentration of volatile organic compound than the fresh sample, more detected in the range of 100–200 s. It can be concluded that

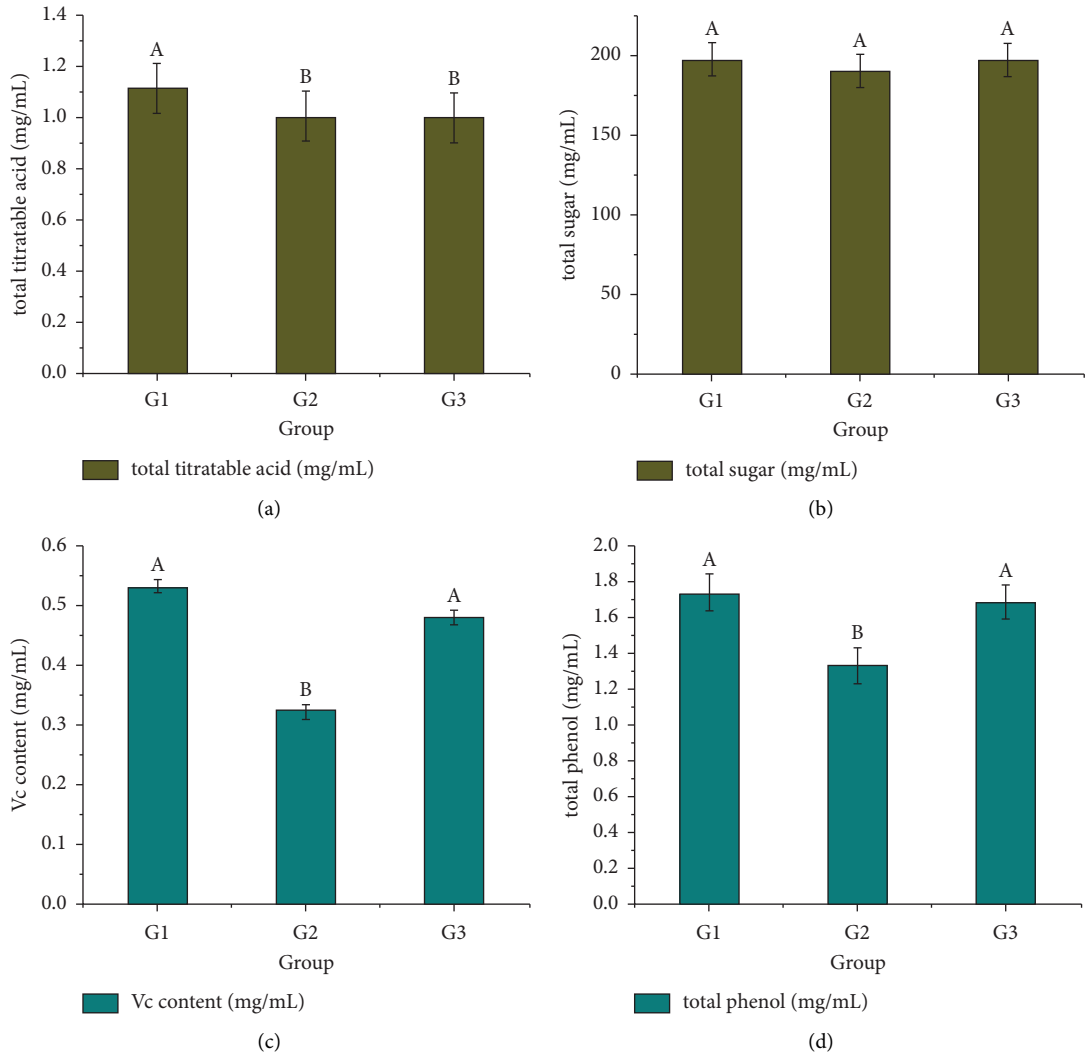


Figure 2: Comparison of physical and chemical properties of lyophilized lemon slices: (a) the total titratable acid content of lyophilized lemon slices in different treatment groups (G1: lyophilized lemon slice at 0 d; G2: lyophilized lemon slices at 7 d; G3: lyophilized lemon slices with LPP treatment at 7 d); (b) the total sugar content of lyophilized lemon slices in different treatment groups; (c) the vitamin C content of lyophilized lemon slices in different treatment groups; (d) the total phenol content of lyophilized lemon slice in different treatment groups.

the concentration of small molecules decreased with storage time, while the concentration of larger molecules, such as terpenes, increased with storage time. The distinct differences observed from the 3D and 2D spectrum require an in-depth statistical analysis.

3.2.2. Fingerprint Analysis of Volatile Organic Compound. A total of 35 aroma compounds were detected by GC-IMS analysis in lemon slices, and the results are shown in Table 1. The results indicated that of the total aromatic compounds, olefins accounted for 77.43%, alcohols accounted for 14.69%, aldehydes accounted for 2.12%, ketones accounted for 5.4%, and lipids accounted for 0.36%. These results show that olefins (limonene accounted for 51.58% of the total) are the main aroma substances found in lemons, followed by ketones, alcohols, and aldehydes with lower proportions. The detected volatile substances were lower than in previous studies of lemon slices subjected to different drying methods [32], probably due to the different processing or the different sources of lemon. As some individual compounds presented at different concentrations, several signals or spots were generated which represent the formation of corresponding dimers [36].

Figure 4 was transformed into a gallery plot (Figure 5). In the fingerprint, each row represents all the signal peaks selected from a lyophilized lemon slice sample, while each column represents the signal peaks of the same volatile organic compounds in different lyophilized lemon samples. The letters *M* and *D* following some substances represent the monomer or dimer of the same substance, respectively, and the numbers refer to the peaks of unidentified components. As shown in Figure 5, the concentration of volatile organic compounds can be obtained using the color depth of the plot. Through longitudinal comparison, the rule of the flavor characteristics of different substances is more intuitive. In

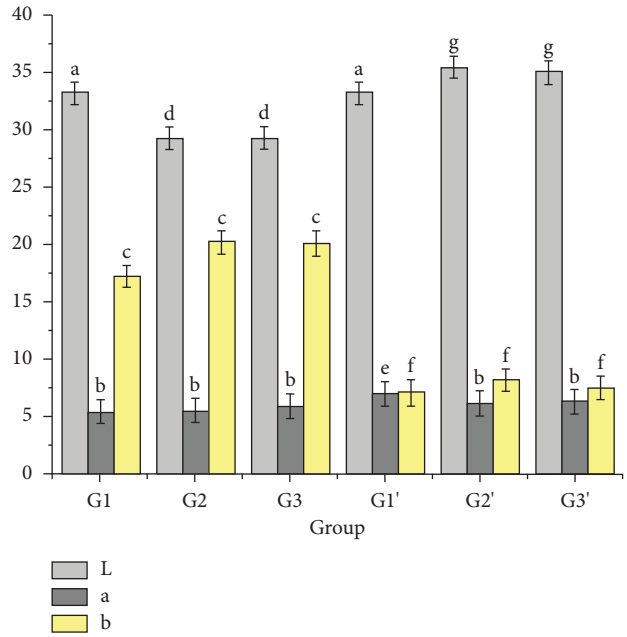
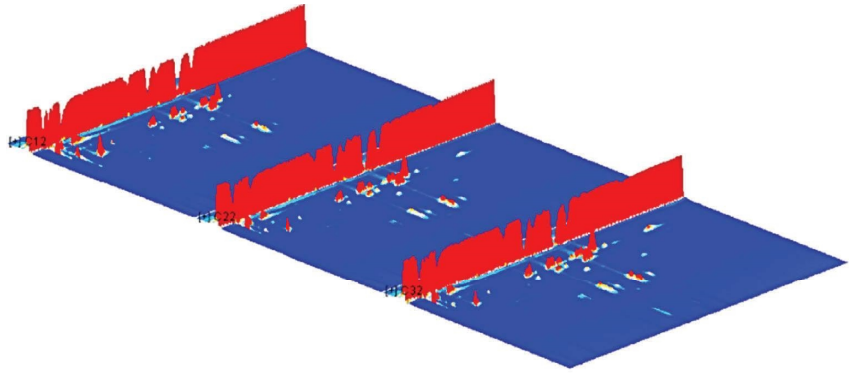
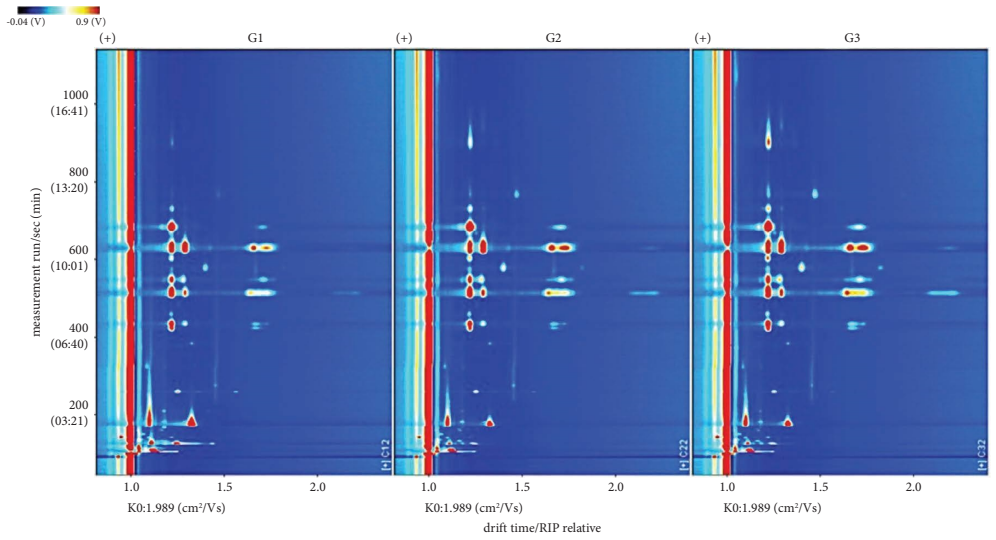


Figure 3: The $L^*.a^*.b^*$ values of lyophilized lemon slices from different groups before and after rehydration (G1, G2, G3: before rehydration; G1′, G2′, G3′: after rehydration).

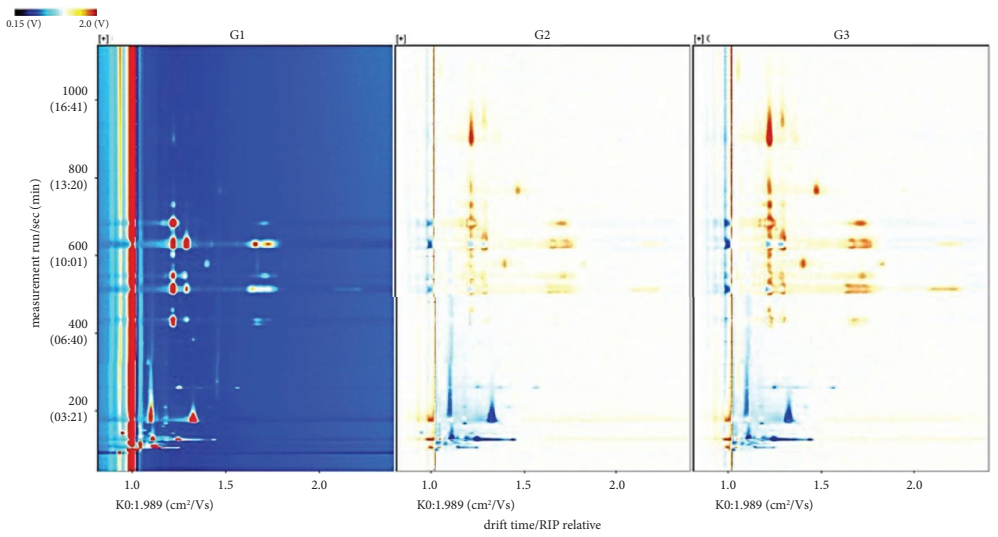


(a)



(b)

Figure 4: Continued.



(c)

Figure 4: GC-IMS spectrum of volatile organic compound in lyophilized lemon slices from different groups (left: G1; middle: G2; right: G3): (a) three-dimensional spectra; (b) the two-dimensional topographic plot; (c) comparison spectra (*Y*-axis: the retention time of the gas chromatography; *X*-axis: the ion migration time).

Table 1: GC-IMS integration parameters of volatile organic compounds in the lyophilized lemon slice[1].

No.	Compound	CAS	Molecular formula[1]	RI	Rt (sec)	Relative content (%)			Aroma
						G1	G2	G3	
1	Terpinen-4-ol	C562743	$C_{10}H_{18}O$	1172.6	902.103	1.13 ± 0.16	2.98 ± 0.14	4.39 ± 0.05	Fresh, woody
2	*α*-Terpineol	C98555	$C_{10}H_{18}O$	1197.8	952.53	0.27 ± 0.08	0.49 ± 0.06	0.66 ± 0.05	Fresh, woody
3	Linalool	C78706	$C_{10}H_{18}O$	1109.3	775.935	0.30 ± 0.04	0.60 ± 0.02	0.79 ± 0.02	Fruity
4	n-Nonanal	C124196	$C_9H_{18}O$	1107.0	771.332	0.32 ± 0.03	0.67 ± 0.04	0.87 ± 0.01	Grass
5	Limonene	C138863	$C_{10}H_{16}$	1036.8	631.271	7.18 ± 0.12	7.23 ± 0.16	6.38 ± 0.05	Citrus
6	Limonene	C138863	$C_{10}H_{16}$	1036.6	630.981	9.91 ± 0.07	10.21 ± 0.19	9.17 ± 0.09	
7	Limonene	C138863	$C_{10}H_{16}$	1037.1	631.849	3.14 ± 0.10	3.83 ± 0.08	3.73 ± 0.02	
8	Limonene	C138863	$C_{10}H_{16}$	1034.9	627.507	3.92 ± 0.12	5.00 ± 0.16	5.13 ± 0.02	
9	Limonene	C138863	$C_{10}H_{16}$	1035.7	629.244	0.32 ± 0.03	0.41 ± 0.16	0.46 ± 0.05	
10	*α*-Terpinene	C99865	$C_{10}H_{16}$	1023.3	604.515	1.00 ± 0.07	1.28 ± 0.15	1.48 ± 0.03	Lemon, lime
11	Octanal	C124130	$C_8H_{16}O$	1011.8	581.538	0.84 ± 0.08	1.39 ± 0.13	1.50 ± 0.01	Fruity
12	Octanal	C124130	$C_8H_{16}O$	1011.2	580.186	0.14 ± 0.03	0.24 ± 0.05	0.26 ± 0.03	
13	*β*-Pinene	C127913	$C_{10}H_{16}$	975.3	513.807	7.84 ± 0.25	8.00 ± 0.16	7.33 ± 0.06	Turpentine taste
14	*β*-Pinene	C127913	$C_{10}H_{16}$	974.8	512.876	2.78 ± 0.02	3.04 ± 0.04	2.88 ± 0.005	
15	*β*-Pinene	C127913	$C_{10}H_{16}$	975.3	513.807	5.80 ± 0.03	7.03 ± 0.03	7.45 ± 0.009	
16	*β*-Pinene	C127913	$C_{10}H_{16}$	975.8	514.738	1.07 ± 0.09	1.36 ± 0.07	1.63 ± 0.09	
17	*α*-Pinene	C80568	$C_{10}H_{16}$	931.2	436.376	2.99 ± 0.06	2.95 ± 0.03	3.10 ± 0.04	
18	*α*-Pinene	C80568	$C_{10}H_{16}$	930.8	435.749	0.85 ± 0.005	0.87 ± 0.01	0.94 ± 0.005	
19	*α*-Pinene	C80568	$C_{10}H_{16}$	929.7	433.866	0.47 ± 0.03	0.49 ± 0.02	0.65 ± 0.004	
20	*α*-Pinene	C80568	$C_{10}H_{16}$	929.7	433.866	0.16 ± 0.01	0.16 ± 0.01	0.21 ± 0.004	
21	Heptanal	C111717	$C_7H_{14}O$	901.5	384.403	0.22 ± 0.01	0.20 ± 0.02	0.19 ± 0.008	Floral, sweet
22	Furfural	C98011	$C_5H_4O_2$	852.7	324.687	0.42 ± 0.003	0.27 ± 0.01	0.25 ± 0.02	Caramel
23	Hexanal	C66251	$C_6H_{12}O$	793.5	259.6	0.70 ± 0.02	0.91 ± 0.07	0.38 ± 0.008	Green
24	4-Methyl-2-pentanone	C108101	$C_6H_{12}O$	733.8	209.514	0.14 ± 0.02	0.09 ± 0.001	0.08 ± 0.001	Green
25	Pentanal	C110623	$C_5H_{10}O$	699.3	182.2	0.08 ± 0.001	0.18 ± 0.09	0.10 ± 0.004	Almond
26	1-Butanol	C71363	$C_4H_{10}O$	674.9	167.839	0.10 ± 0.001	0.04 ± 0.001	0.04 ± 0.001	Fruity, sweet
27	2-Methylbutanal	C96173	$C_5H_{10}O$	666.3	164.181	0.03 ± 0.001	0.03 ± 0.001	0.02 ± 0.001	Musty
28	3-Methylbutanal	C590863	$C_5H_{10}O$	656.7	160.054	0.12 ± 0.01	0.07 ± 0.001	0.06 ± 0.001	Green
29	Ethyl acetate	C141786	$C_4H_8O_2$	613.7	141.624	0.22 ± 0.001	0.09 ± 0.001	0.08 ± 0.001	Fruity
30	1-Propanol	C71238	C_3H_8O	579.1	126.807	1.70 ± 0.06	0.81 ± 0.01	0.83 ± 0.006	Fruity, floral
31	1-Propanol	C71238	C_3H_8O	581.5	127.853	1.79 ± 0.07	0.37 ± 0.02	0.44 ± 0.007	
32	1-Propanol	C71238	C_3H_8O	577.4	126.11	0.14 ± 0.005	0.01 ± 0.001	0.01 ± 0.001	
33	Acetone	C67641	C_3H_6O	542.5	111.119	3.17 ± 0.07	2.93 ± 0.02	2.33 ± 0.03	Fragrance
34	Ethanol	C64175	C_2H_6O	529.4	105.541	0.73 ± 0.06	0.92 ± 0.06	0.76 ± 0.02	Green
35	Ethanol	C64175	C_2H_6O	526.6	104.321	2.83 ± 0.04	2.29 ± 0.02	2.07 ± 0.004	

MW represents molecular weight, RI represents reserved index, and Rt represents retention time. [1]different letters represent significant differences, $p < 0.05$.

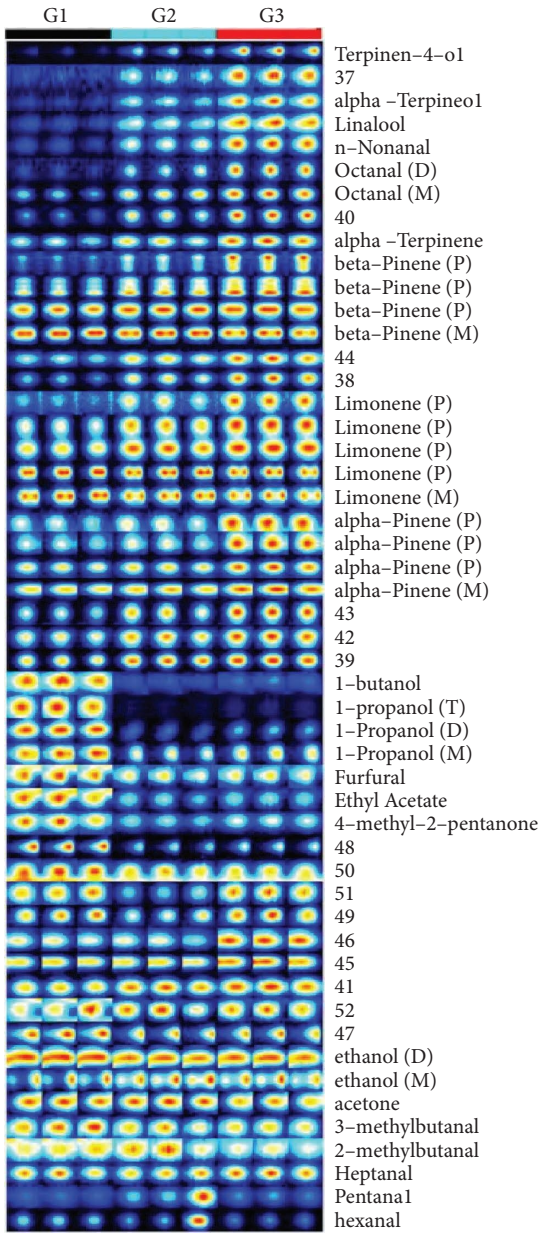


Figure 5: Gallery plot of volatile organic compounds in lyophilized lemon slices of different groups (M, D, T, and P in parentheses following the substance name represent the monomer, dimer, trimer, and polymer of the substance, respectively).

Figure 5, the concentration of red squares detected in samples follows the order: G3 > G2 > G1. It was found that G3 had the highest concentration of 4-terpineol, α-terpineol, linalool, nonaldehyde, octyl aldehyde, α-terpinene, β-pinene, limonene, and α-pinene. In the previous report, aldehydes (nerolides and geranyl aldehyde) and esters (nerolides and geranyl acetate) are responsible for the aroma quality of lemons [37]. Therefore, LPP showed no negative effect on the flavor characteristics of lyophilized lemon slices. We also found that the concentration of compounds in the green squares decreased or even disappeared in G1 and G3, which included compounds such as 1-butanol, 1-propanol, furfural, ethyl acetate, and 4-methyl-2-pentanone. In terms of orange squares (unidentified compound), there was also a small difference in the amount of the substance in the three groups of samples.

Four terpenes including α-terpinene, limonene, β-pinene, and α-pinene were identified in the lemon slices, accounting for 47%–52% of all volatile compounds. These C10 monoterpenoids accumulate in special oil glands of citrus fruit pericarps and are the main contributors to lemon flavor and aroma properties [38]. The process of freeze-drying facilitated the accumulation of these compounds by an increase of 9% and 7% in G2 and G3 compared with G1. α-Pinene and β-pinene might get degraded in the presence of ozone to give molozonide which will further disassociate to form formaldehyde [39]. However, in this study, we did not observe a significant reduction of pinene, probably due to a short treatment time. Limonene might degrade in the presence of hydroxy ions to γ-terpinene and α-terpinene, while γ-terpinene is resistant to further degradation by LPP [40]. In addition, six alcohols such as linalool, terpinen-4-ol, and α-terpineol, which contribute to the unique and desirable flavors and aromas of lemon, were also identified. Linalool and terpinen-4-ol are crucial volatile flavor compounds with citrus fruits and rose aroma notes [41]. The GC-IMS results suggested that these characteristic alcohols were increased during the drying process and retained after LPP treatment. It has been reported that terpinen-4-ol imparted significant reduction during distillation and extraction because of its thermosensitive nature [42]. In contrast, the increase of these compounds observed in this study was a result of the oxidative degradation of limonene exposed to reactive species induced by LPP, and they were maintained due to the low temperature during the whole processing. However, LPP treatments reduced the flavor characteristics of fresh green orange peels corresponding to some C6-C9 micromolecule aldehydes such as hexanal, which are the key contributors of the fresh green orange peels with strong volatility and low threshold. In general, LPP increased the content of characteristic volatile compounds of lemon slices with a citrus-like flavor.

4. Conclusions

In this work, the effect of LPP treatment on the physicochemical properties and flavor characteristics of lyophilized lemon slices was investigated. LPP had no significant influence on the physicochemical properties (color index, titratable acid, total sugar, total phenol, and Vc) of the lyophilized lemon slices. In terms of flavor characteristics, GC-IMS detected over 30 types of volatile organic compounds in lyophilized lemon slices, including alcohols, alkenes, aldehydes, ketones, and other compounds. Combining the gallery plot, we found that the concentration of compounds such as 4-terpenol, α-terpinol, linalool, nonaldehyde, octyl aldehyde, α-terpinene, β-pinene, limonene, and α-pinene was higher in the LPP-treated samples than in the reference samples. The similar composition of key odor substance in LPP-treated lemon slices with original lemon slices indicated the positive effect of LPP treatment on the retention of the flavor characteristics of lemon slices. In

addition to efficient sterilization, LPP demonstrated a positive effect on the quality preservation of lyophilized lemon slices. Therefore, this study shows that LPP promises application in the food industry.

Data Availability

The data supporting the current study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest.

Authors' Contributions

Linlin Li wrote the original draft, reviewed and edited the manuscript, and investigated the study; Ningjing Zhou wrote the original draft and investigated the study; Chiara Sanmartin reviewed and edited the manuscript, and curated the data; Charles Brennan reviewed and edited the manuscript; Gengsheng Xiao collected resources, developed software, and conceptualized the study; Lukai Ma developed methodology, investigated the study, curated the data, and administrated the project; Ying Xiaoguo supervised and investigated the study; Dongjie Liu supervised and investigated the study.

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References

[1] C. A. Ledesma-Escobar, F. Priego-Capote, and M. D. Luque de Castro, "Effect of sample pretreatment on the extraction of lemon (Citrus limon) components," *Talanta*, vol. 153, pp. 386–391, 2016.

[2] H. Jiang, W. Zhang, Y. Xu, L. Chen, J. Cao, and W. Jiang, "An advance on nutritional profile, phytochemical profile, nutraceutical properties, and potential industrial applications of lemon peels: a comprehensive review," *Trends in Food Science and Technology*, vol. 124, pp. 219–236, 2022.

[3] Z. Ozer, N. Teke, G. B. Turan, and A. N. Bahcecik, "Effectiveness of lemon essential oil in reducing test anxiety in nursing students," *Explore*, vol. 18, no. 5, pp. 526–532, 2022.

[4] P. Ruggenenti, M. R. Caruso, M. Cortinovis et al., "Fresh lemon juice supplementation for the prevention of recurrent stones in calcium oxalate nephrolithiasis: a pragmatic, prospective, randomised, open, blinded endpoint (PROBE) trial," *eClinicalMedicine*, vol. 43, Article ID 101227, 2022.

[5] V. Dal-Bó and J. T. Freire, "Effects of lyophilization on colorimetric indices, phenolics content, and antioxidant activity of avocado (Persea americana) pulp," *Food Control*, vol. 132, Article ID 108526, 2022.

[6] N. M. Fayek, M. A. Farag, and F. R. Saber, "Metabolome classification via GC/MS and UHPLC/MS of olive fruit varieties grown in Egypt reveal pickling process impact on their composition," *Food Chemistry*, vol. 339, Article ID 127861, 2021.

[7] X. Gao, T. Feng, E. Liu et al., "Ougan juice debittering using ultrasound-aided enzymatic hydrolysis: impacts on aroma and taste," *Food Chemistry*, vol. 345, Article ID 128767, 2021.

[8] N. U. Sruthi, K. Josna, R. Pandiselvam, A. Kothakota, M. Gavahian, and A. Mousavi Khaneghah, "Impacts of cold plasma treatment on physicochemical, functional, bioactive, textural, and sensory attributes of food: a comprehensive review," *Food Chemistry*, vol. 368, Article ID 130809, 2022.

[9] S. Saremnezhad, M. Soltani, A. Faraji, and A. A. Hayaloglu, "Chemical changes of food constituents during cold plasma processing: a review," *Food Research International*, vol. 147, Article ID 110552, 2021.

[10] F.-G. Chizoba Ekezie, D.-W. Sun, and J.-H. Cheng, "A review on recent advances in cold plasma technology for the food industry: current applications and future trends," *Trends in Food Science and Technology*, vol. 69, pp. 46–58, 2017.

[11] K. Knoerzer, A. B. Murphy, M. Fresewinkel, P. Sanguansri, and J. Coventry, "Evaluation of methods for determining food surface temperature in the presence of low-pressure cool plasma," *Innovative Food Science and Emerging Technologies*, vol. 15, pp. 23–30, 2012.

[12] A. M. Adam, B. Yadav, A. Prasad, B. Gautam, Y. Tsui, and M. S. Roopesh, "Salmonella inactivation and rapid cooling of fresh cut apples by plasma integrated low-pressure cooling," *Food Research International*, vol. 147, Article ID 110464, 2021.

[13] R. Zhou, A. Rezaeimotlagh, R. Zhou et al., "In-package plasma: from reactive chemistry to innovative food preservation technologies," *Trends in Food Science and Technology*, vol. 120, pp. 59–74, 2022.

[14] J. N. Jackowetz and R. Mira de Orduña, "Survey of SO2 binding carbonyls in 237 red and white table wines," *Food Control*, vol. 32, no. 2, pp. 687–692, 2013.

[15] B. Bharti, H. Li, Z. Ren, R. Zhu, and Z. Zhu, "Recent advances in sterilization and disinfection technology: a review," *Chemosphere*, vol. 308, Article ID 136404, 2022.

[16] L. Shao, Y. Zhao, B. Zou, X. Li, and R. Dai, "Ohmic heating in fruit and vegetable processing: quality characteristics, enzyme inactivation, challenges and prospective," *Trends in Food Science and Technology*, vol. 118, pp. 601–616, 2021.

[17] M. Sriraksha, S. B. Ayenampudi, M. Noor, R. Sn, and A. K. Chakka, "Cold plasma technology: an insight on its disinfection efficiency of various food systems," *Food Science and Technology International*, vol. 29, no. 4, pp. 428–441, 2023.

[18] M. Ahangari, Y. Ramezan, and M. R. Khani, "Effect of low pressure cold plasma treatment on microbial decontamination and physicochemical properties of dried walnut kernels (Juglans regia L.)," *Journal of Food Process Engineering*, vol. 44, 2021.

[19] M. D. F. D. Linhares, E. G. Alves Filho, L. M. A. Silva et al., "Thermal and non-thermal processing effect on açai juice composition," *Food Research International*, vol. 136, Article ID 109506, 2020.

[20] D. Liu, L. Bai, X. Feng et al., "Characterization of Jinhua ham aroma profiles in specific to aging time by gas chromatography-ion mobility spectrometry (GC-IMS)," *Meat Science*, vol. 168, Article ID 108178, 2020.

[21] C. Li, S. Al-Dalali, Z. Wang, B. Xu, and H. Zhou, "Investigation of volatile flavor compounds and characterization

of aroma-active compounds of water-boiled salted duck using GC-MS-O, GC-IMS, and E-nose," *Food Chemistry*, vol. 386, Article ID 132728, 2022.

[22] X. Guo, W. Schwab, C. T. Ho, C. Song, and X. Wan, "Characterization of the aroma profiles of oolong tea made from three tea cultivars by both GC-MS and GC-IMS," *Food Chemistry*, vol. 376, Article ID 131933, 2022.

[23] S. K. Pankaj, N. N. Misra, and P. J. Cullen, "Kinetics of tomato peroxidase inactivation by atmospheric pressure cold plasma based on dielectric barrier discharge," *Innovative Food Science and Emerging Technologies*, vol. 19, pp. 153–157, 2013.

[24] W. Pan, S. Benjakul, C. Sanmartin et al., "Characterization of the flavor profile of bigeye tuna slices treated by cold plasma using E-nose and GC-IMS," *Fishes*, vol. 7, no. 1, p. 13, 2022.

[25] R. S. El-Serafy and A. A. El-Sheshtawy, "Effect of nitrogen fixing bacteria and moringa leaf extract on fruit yield, estragole content and total phenols of organic fennel," *Scientia Horticulturae*, vol. 265, Article ID 109209, 2020.

[26] Q. D. Do, A. E. Angkawijaya, P. L. Tran-Nguyen et al., "Effect of extraction solvent on total phenol content, total flavonoid content, and antioxidant activity of Limnophila aromatica," *Journal of Food and Drug Analysis*, vol. 22, no. 3, pp. 296–302, 2014.

[27] T. Masuko, A. Minami, N. Iwasaki, T. Majima, S. Nishimura, and Y. C. Lee, "Carbohydrate analysis by a phenol-sulfuric acid method in microplate format," *Analytical Biochemistry*, vol. 339, no. 1, pp. 69–72, 2005.

[28] T. Lafarga, G. Bobo, I. Viñas, L. Zudaire, J. Simó, and I. Aguiló-Aguayo, "Steaming and sous-vide: effects on antioxidant activity, vitamin C, and total phenolic content of Brassica vegetables," *International Journal of Gastronomy and Food Science*, vol. 13, pp. 134–139, 2018.

[29] L. Zhang, S. Liu, Y. Chen, J. Guo, C. Li, and J. Zhang, "Metatranscriptomic approach reveals the functional and enzyme dynamics of core microbes during noni fruit fermentation," *Food Research International*, vol. 141, Article ID 109999, 2021.

[30] S. K. Jain, M. Y. Dar, S. Kumar, A. Yadav, and S. R. Kearns, "Role of anti-oxidant (vitamin-C) in post-operative pain relief in foot and ankle trauma surgery: a prospective randomized trial," *Foot and Ankle Surgery*, vol. 25, no. 4, pp. 542–545, 2019.

[31] K. Srinivasa Rao, P. K. Chaudhury, and A. Pradhan, "Evaluation of anti-oxidant activities and total phenolic content of Chromolaena odorata," *Food and Chemical Toxicology*, vol. 48, no. 2, pp. 729–732, 2010.

[32] Q. Xu, H. Pan, Y. Shui et al., "Effect of different drying technologies on the characteristics and quality of lemon slices," *Journal of Food Science*, vol. 87, no. 7, pp. 2980–2998, 2022.

[33] T. R. Farias, S. Rodrigues, and F. A. Fernandes, "Effect of dielectric barrier discharge plasma excitation frequency on the enzymatic activity, antioxidant capacity and phenolic content of apple cubes and apple juice," *Food Research International*, vol. 136, Article ID 109617, 2020.

[34] N. N. Misra, O. Schlüter, and P. J. Cullen, *Cold Plasma in Food and Agriculture: Fundamentals and Applications*, Academic press, Elsevier Ltd, San Diego, CA, USA, 1st edition, 2020.

[35] X. B. Li, C. H. Guo, Y. H. Qi et al., "Identification of volatile organic compounds in muscle tissues of different species based on Headspace-Gas-Chromatography Ion-Mobility spectrometry," *Legal Medicine*, vol. 59, Article ID 102132, 2022.

[36] Y. Yang, B. Wang, Y. Fu et al., "HS-GC-IMS with PCA to analyze volatile flavor compounds across different production stages of fermented soybean whey tofu," *Food Chemistry*, vol. 346, Article ID 128880, 2021.

[37] B. Acoglu and P. Yolci Omeroglu, "The effect of drying processes on pesticide residues in orange (Citrus sinensis)," *Drying Technology*, vol. 39, no. 13, pp. 2039–2054, 2021.

[38] M. Shorbagi, N. M. Fayek, P. Shao, and M. A. Farag, "Citrus reticulata Blanco (the common Mandarin) fruit: an updated review of its bioactive, extraction types, food quality, therapeutic merits, and bio-waste valorization practices to maximize its economic value," *Food Bioscience*, vol. 47, Article ID 101699, 2022.

[39] X. Zhang, Z. Chen, H. Wang, S. He, and D. Huang, "An important pathway for ozonolysis of alpha-pinene and beta-pinene in aqueous phase and its atmosphericimplications," *Atmospheric Environment*, vol. 43, no. 29, pp. 4465–4471, 2009.

[40] N. A. Comelli, E. N. Ponzi, and M. I. Ponzi, "Isomerization of α-pinene, limonene, α-terpinene, and terpinolene on sulfated zirconia," *Journal of the American Oil Chemists' Society*, vol. 82, no. 7, pp. 531–535, 2005.

[41] H. Liu, J. Wen, Y. Xu et al., "Evaluation of dynamic changes and formation regularity in volatile flavor compounds in Citrus reticulata 'chachi' peel at different collection periods using gas chromatography-ion mobility spectrometry," *LWT*, vol. 171, Article ID 114126, 2022.

[42] B. B. Mikkelsen and L. Poll, "Decomposition and transformation of aroma compounds and anthocyanins during black currant (ribes nigrum L.) juice processin," *Journal of Food Science*, vol. 67, no. 9, pp. 3447–3455, 2002.