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Characteristic volatiles fingerprints in olive vegetable stored at different conditions by HS-GC-IMS

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ABSTRACT

The olive vegetable is popular food owing to its unique flavor. This study innovatively used headspace-gas chromatography-ion mobility spectrometry to evaluate olive vegetables' volatiles under different conditions. A total of 57 volatile compounds were determined from olive vegetables, including 30 aldehydes, 8 ketones, 5 alcohols, 2 esters, 8 hydrocarbons, 1 furans, 3 sulfur compounds. The PCA distinguished the olive vegetable stored at different conditions by volatiles. The gallery plot showed that olive vegetables stored at 4 °C for 21 d produced more limonene, which had a desirable fruity odor. The (E)-2-octenal, (E)-2-pentenal, (E,E)-2,4-heptadienal, 5-methylfurfural, and heptanal in fresh olive vegetables were lowest and increased with storage time. Furthermore, the change of volatiles was the least when the olive vegetable was stored at 0 °C. This study can provide theoretical bases for improving the flavor quality of olive vegetables and developing traditional food for standardized industrial production.

1. Introduction

The olive vegetable is a special pickle in Chaoshan of China, which attracts lots of consumers. It is a kind of unique flavor food made of olives and mustard as the main raw material and other auxiliary ingredients. It is rich in nutrition, chlorophyll, calcium, iodine, and many vitamins (Montano, Casado, De Castro, Sánchez, & Rejano, 2004). Pickles are the most traditional and common way of processing vegetables, which are easy and inexpensive to process, using the products of beneficial microbes and various ingredients to enhance the preservation of the products. Pickles have a unique aroma. In recent years, lots of research on pickles have been reported, mainly including analysis of pickling mechanisms, process improvement, function and composition analysis (Montano, Casado, De Castro, Sánchez, & Rejano, 2004; Ji, Ji, Li, & Lu, 2009; Guan, Zheng, Huang, Xiao, & Xiong, 2020).

However, there is a lack of intensive processing of pickles, commonly processed in small workshops, and the composition of basic composition and flavor characteristics of pickles are not fully understood. Research on the identification and mechanisms of flavor substances is limited (Wu et al., 2015). The unique flavor of pickles is the key to their market competitiveness and consumer preference. Meanwhile, pickles are now becoming increasingly popular as condiments or the raw material of condiments (Liu, Li, Deng, Wang, & Yang, 2009). Flavor is a key indicator to influence the quality of pickles, which largely determines the choice of consumers. It is also an essential indicator for developing new technology and new pickles products. The type and quantity of flavor substances determine the quality of pickles, which is comprehensively assessed by the overall effect of human senses, such as smell and taste.

As a typical pickle, kimchi has certain similarities in terms of nutritional value, pickling mechanism, flavor substances, and composition analysis. Studies have shown that temperature affects the bacteria's

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structure and the kimchi's flavor (He, Chen, Wang, Lin, Ji, Li, & Liang, 2020; Liu, & Tong, 2017). Researchers investigated the changes in microbial quantity and the content of metabolic substrates of traditional Sichuan kimchi at different fermentation temperatures. They found that temperature had a significant effect on the quantities and metabolism of microorganisms during the fermentation of traditional Sichuan kimchi (Wang, Chen, Tang, Li, & Zhang, 2020; Park, Seo, Kim, Na, & Son, 2018; Jiang et al., 2021).

However, there has been a lack of research on volatile compounds in olive vegetables, especially the changes of volatile compounds during preservation. The Headspace-gas chromatography-ion mobility spectrometry (HS-GC-IMS) technique is rapid for determing volatiles (Xu et al., 2022). This method has been wildly used to analyze the food flavor, including evaluating characteristic volatiles of Dongbei Suancai and other traditional fermented food (Zhao et al., 2022; Han, Wang, Zhang, Li, & Gao, 2022; Chen, Li, Liao, Qin, Jiang, & Liu, 2021).

The olive vegetable was selected as the experimental material in this work, and the volatile aroma compounds were identified by HS-GC-IMS technology. The objective of this study was to compare the volatile compounds in olive vegetables stored at different conditions. This could provide a fundamental basis for improving the olive vegetable preservation process.

2. Materials and methods

2.1. Materials and reagents

The olive vegetable was obtained from Peng Sheng Food Co., Ltd. and produced in Guangzhou, China. The reagents and solvents were obtained from Sinopharm Chemical Regent (Shanghai, China).

2.2. Determination of physical and chemical properties of olive vegetable

The moisture and fat content of the samples were determined by the oven drying method (AOAC, 1995), and the Chinese National Food Standard (GB/T 5009.6-2016, 2016), respectively. The total contents of salt and sugar were measured according to the Chinese National Food Standard (GB2714-2015, 2015). The total contents of acrylamide were measured according to the Chinese National Food Standard (GB/T5009.2042014, 2014).

The nitrite was detected according to the method reported by Ding et al (2018). 5 mL of filtrate was taken from the sample and put into the colourimetric tube. Then 2 drops of hydrochloric acid and 40 mL of water were added. After shaking well, 1 mL of naphthalene ethylenediamine hydrochloride and 1 mL of p-aminobenzene sulfonic acid were added into the sample. After standing for 30 min, the wavelength was set at 540 nm by spectrophotometer and the absorbance was measured.

2.3. Volatile components analysis by GC-IMS

Agilent 490 gas chromatograph (Agilent Technologies, Palo Alto, CA, USA) and IMS instrument (FlavourSpec®, Gesellschaft für Analytische Sensorsystem mbH, Dortmund, Germany) was used to analyze the volatiles of samples. The detection conditions were according to our previous study (Xu et al., 2022a). The olive vegetable sample (2 ± 0.02 g) was placed in a 20 mL vial. The sample vials were heated in a incubator at 40 °C for 15 min. Then 500 μL of the volatile sample was injected to the 40 °C injector and the mode is splitless. The flow of nitrogen was programmed from 2 mL min^{-1} (withholding for 2 min) to 100 mL min^{-1} (withholding for 25 min). The qualitative analysis of volatile compounds were according to some aspects, which were the retention index of volatile compounds, the mass spectrum of compounds in the NIST library, and drift time of standards in the GC-IMS library.

2.4. Statistical analyses

All analyses were conducted in triplicate. The relevant results were denoted as mean ± standard deviation (SD). Significant differences were performed by one-way analysis of variance (ANOVA) combined with Turkey's multiple-range test using the SPSS 22.0 (IBM, Armonk, New York).

3. Results and discussion

3.1. Basic physical and chemical properties of olive vegetable

As shown in Table 1, the water content of olive vegetables was 22.7%. The salt content of olive vegetable was 3.10%, which belong to pickled food. According to the newest Dietary Guidelines for Chinese Residents, each Chinese citizen should consume no more than 5 g of salt per day, so it is appropriate to keep the intake of olive vegetables within 100 g per day. The global obesity epidemic and the increasing incidence of metabolic diseases are mainly related to fat intake (Li, et al., 2023). With the improvement of health awareness, humans have begun to pursue low-fat diet recipes, making the development of low-fat food a hot research topic (Azeredo, Tonon, & McClements, 2021). The fat content in olive vegetables was 45.4 g/100 g, the development of low-fat and low-salt olive vegetables would have a specific commercial value. The sugar content in olive vegetables was low, only 0.167 g/100 g. No nitrite and acrylamide were detected in the olive vegetable, indicating that the olive vegetable was in line with Chinese food safety management regulations (An et al., 2021).

3.2. Data analysis of GC-IMS

3.2.1Volatile compound identification in olive vegetable stored at different conditions.

Fig. 1(a) and (b) showed the 3D topographical and 2D visualization of the volatile compounds in olive vegetable stored at different conditions by GC-IMS, respectively, fresh (A); stored at 0 °C for 14 d (B); stored at 4 °C for 14 d (C); stored at 25 °C for 14 d (D); stored at 0 °C for 21 d (E); stored at 4 °C for 21 d (F); stored at 25 °C for 21 d (G). The X-axis and Y-axis represented the ion migration time and the retention time of the gas chromatography, respectively. The whole spectrum showed the total volatiles of the olive vegetable samples. Each point in the spectrum represented one volatile compound in the samples. The higher intensity of volatiles will lead to the colour change to red. The olive vegetable samples presented more signals in the retention time of 100–500 s and the drift time of 1.0–1.8. Fig. 1(b) showed the volatile compounds of olive vegetables stored at different conditions had similar types, but had different signal intensity. In order to clearly compare the difference in these samples, the differential comparison model was used in Fig. 1(c). The Fig. 1(c) used the fresh olive vegetable (A) sample as a reference. The results indicated the difference of the volatile compound content in olive vegetables stored at different conditions. The figure deducted from other samples was white if the content of volatile compounds were the same. The background was red, when the compound content was higher than the reference sample, in contrast, it was blue.

Table 1
Basic physical and chemical properties of olive vegetable.

Index	Content
Water/%	22.7 ± 0.20
Salt/g/%	3.10 ± 0.03
Fat/g/100 g	45.4 ± 0.40
Sugar/g/100 g	0.167 ± 0.01
Nitrite	Nd
Acrylamide	Nd

Nd: not detected.

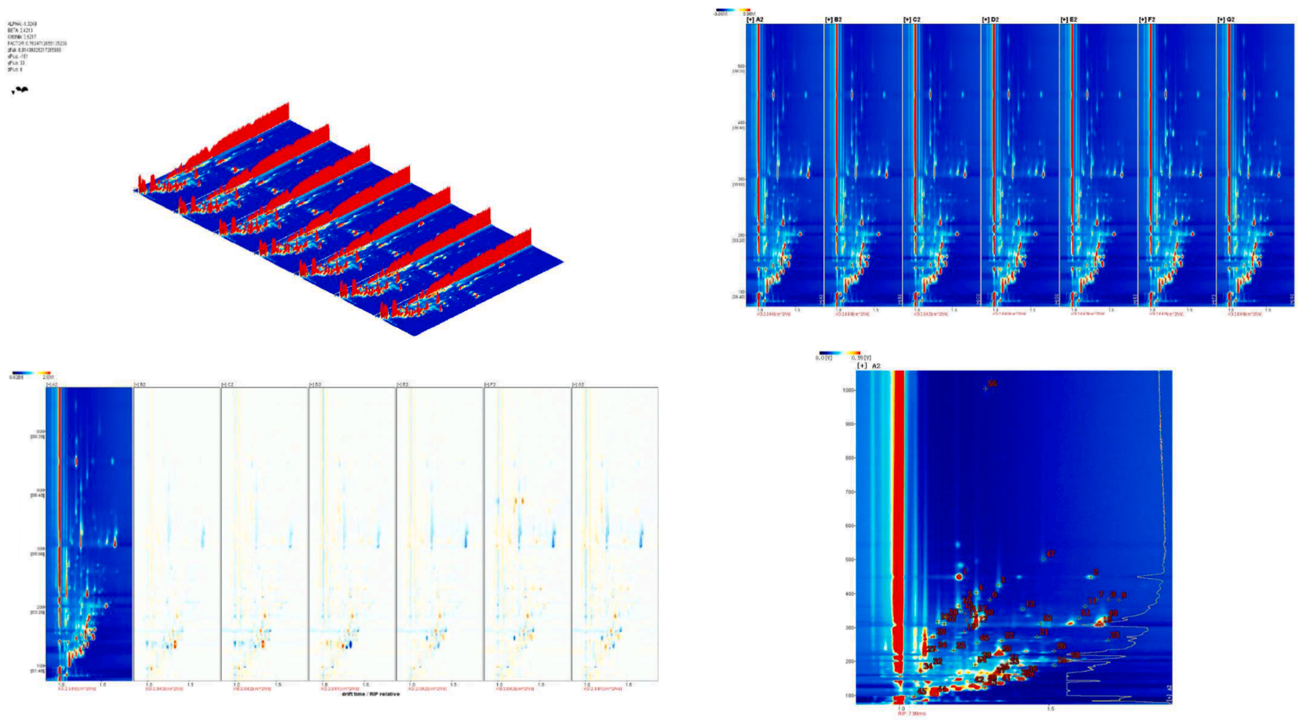

Fig. 1. The three-dimensional spectrum (a), topographic plot (b) and Comparison of the different GC-IMS spectrum of volatile compounds (c) from olive vegetable stored at different conditions, fresh (A); stored at 0 °C for 14 d (B); stored at 4 °C for 14 d (C); stored at 25 °C for 14 d (D); stored at 0 °C for 21 d (E); stored at 4 °C for 21 d (F); stored at 25 °C for 21 d (G), and GC-IMS spectra with detected volatile compounds numbers of samples (d).

Compared to the fresh olive vegetable (A), the signals of samples at the drift time of 1.0–1.8 enhanced, which indicated that the contents of these volatile compounds increased when olive vegetable samples were stored at different conditions. However, the change is the least when olive vegetable was stored at 0 °C.

3.2.2. Qualitative and quantitative analysis of volatile compounds

Fig. 1(d) showed the qualitative results of the olive vegetable samples. Each signal peak in the Fig. 1(d) represented one volatile compound. The detailed volatile compounds shown in the figure were shown in Table S1. 57 kinds of volatile compounds were detected, including 30 aldehydes, 8 ketones, 5 alcohols, 2 esters, 8 hydrocarbons, 1 furans, and 3 sulfur compounds (Table 2). If the compounds showed two signal peaks, they are the monomers and dimers of these compounds. These compounds with a high proton affinity could make ions to form dimers when they moved in the drift trough (Pan et al., 2022). The aldehydes compounds contained alkanals (butanal, pentanal, hexanal, heptanal, octanal, nonanal, 3-methyl-2-butenal, 2-methyl-propanal, 2-methylbutanal, 3-methylbutanal, 5-methylfurfural), alkenals ((E)-2-hexenal, (E)-2-octenal, (E)-2-heptenal, (Z,Z)-2,4-heptadienal, (E,E)-2,4-heptadienal, (E,E)-2,4-hexadienal) and the substituted aldehydes (Phenylacetaldehyde, 2-Furfural, Benzaldehyde). The alcohols compounds contained 3-hydroxy-2-butanone, 1-penten-3-one, 2-butanone, 2-propanone, 2-heptanone, and 1-octen-3-one. The alcohols compounds contained ethanol, 1-octen-3-ol, and 1-pentanol. The esters included ethyl acetate and ethyl phenylacetate. Furans compounds contained 2-pentylfuran. In addition, three kinds of sulfur compound were detected, i.e. diallyl sulfide, diallyl disulfide (M), Diallyl disulfide (D).

Aldehydes usually had low odor threshold, so they played an important role in the flavor of olive vegetable samples. Some aldehydes e.g. pentanal, (E)-2-heptenal were the oxidation products of fatty acids, which had the fatty odor (Chang, Wu, Zhang, Jin, & Wang, 2019). The contents of pentanal, (E)-2-heptenal (M), (E)-2-heptenal (D), and phenylacetaldehyde in fresh olive vegetable reached 1327.26, 2812.81, 3018.19, and 519.07, respectively, which were higher than other samples and these volatiles decreased with storage time. Alkenals were mainly existed with *trans*-forms with ten carbon or less, including (E)-2-octenal, (E)-2-pentenal and (E,E)-2,4-heptadienal all with fatty aroma. The contents of (E)-2-octenal, (E)-2-pentenal, (E,E)-2,4-heptadienal, (Z,Z)-2,4-heptadienal, 5-methylfurfural, and heptanal, were lowest in fresh olive vegetable and increased with storage time. When the olive vegetable were stored for 14 d, the (E)-2-hexenal, 2-methylbutenal, 3-methyl-2-butenal, and benzaldehyde increased with storage temperature. The (E)-2-hexenal (M), (E)-2-hexenal (D), benzaldehyde (M) and benzaldehyde (D) reached 360.58, 280.70, 544.9, and 436.88, respectively, which were highest at olive vegetable stored at 4 °C for 21 d. The benzaldehyde contributed to fruity and woody aroma in olive vegetable, which was the oxidation and hydrolysis product of linoleic acid (Xu et al., 2022b; Chang, Wu, Zhang, Jin, & Wang, 2019). In addition, the contents of butanal, and 3-methylbutanal decreased in olive vegetable when stored for 14 d compared to fresh samples, and then increased when stored for 21 d, therefore, these volatiles were higher in olive vegetable stored for 21 d than that stored for 14 d.

The ketones had higher threshold compared to aldehydes, so they may have relatively lower contribution to the flavor (van Gemert, 2011). Most of ketones were also the oxidation products of oleic and linoleic fatty acids. The 1-octen-3-one with metallic odor and 3-hydroxy-2-butanone with buttery odor had the highest contents in the fresh olive vegetable, and they decreased with the storage time. The content of 1-penten-3-one increased in olive vegetable stored for 14 d compared to fresh olive vegetable. Besides, the content of 2-butanone increased in olive vegetable after stored for 14 d with storage temperature.

Alcohols had a higher threshold compared to aldehydes (van Gemert, 2011). The 1-pentanol had a spice odor, which was mainly derived from lipid oxidation. The fresh olive vegetable had the highest content of 1-pentanol (M) (340.40) and 1-pentanol (D) (114.06) and the content of 1-pentanol decreased with the storage time. Besides, the contents of 1-octen-3-ol increased in olive vegetable when stored for 14 d compared to fresh samples, and then decreased when stored for 21 d, therefore the longer storage time may lead to the decline of 1-octen-3-ol.

Table 2
Volatile compounds of olive vegetable stored at different conditions.

Compound	Formula	MW	RI	Rt [*sec*]	Dt [a.u.]	Signal intensity						
						A	B	C	D	E	F	G
Aldehydes (30)												
Butanal	C4H8O	72.1	590.9	133.977	1.28777	2171.6 ± 6.22^{a}	1913.2 ± 3.09^{c}	1967.64 ± 33.96^{bc}	1389.36 ± 10.38^{d}	2129.13 ± 42.46^{ab}	2214.09 ± 32.69^{a}	2039.03 ± 262.19^{abc}
Pentanal	C5H10O	86.1	686.7	162.533	1.42383	1327.26 ± 19.07^{a}	1102.39 ± 14.07^{b}	995.91 ± 26.19^{c}	985.91 ± 35.06^{c}	965.8 ± 17.99^{c}	876.14 ± 16.96^{d}	865.99 ± 27.47^{d}
Hexanal(M)	C6H12O	100.2	780.7	202.621	1.26762	554.65 ± 10.07^{a}	507.09 ± 9.17^{b}	474.37 ± 17.49^{c}	459.16 ± 12.63 cd	483.63 ± 13.3^{bc}	459.18 ± 16.56^{cd}	435.63 ± 12.7^{d}
Hexanal(D)	C6H12O	100.2	779.6	202.106	1.56462	2094.94 ± 20.58^{d}	2009.66 ± 29.8^{e}	2140.57 ± 21.07^{cd}	2183.21 ± 19.64^{bc}	2110.52 ± 35.85^{d}	2288.68 ± 28.61^{a}	2218.05 ± 18.1^{b}
Heptanal(M)	C7H14O	114.2	895.5	261.34	1.34378	307.89 ± 12.07^{bc}	294.8 ± 10.6^{c}	311.05 ± 6.83^{b}	327.93 ± 4.57^{a}	316.28 ± 2.96^{ab}	330.29 ± 10.24^{a}	326.68 ± 5.59^{a}
Heptanal(D)	C7H14O	114.2	896.6	262.199	1.69843	38.44 ± 1^{d}	36.65 ± 2.42^{d}	46.57 ± 1.52^{c}	50.45 ± 1.82^{b}	45.54 ± 2.23^{c}	57.74 ± 2.08^{a}	56.64 ± 3.44^{a}
Octanal	C8H16O	128.2	1002.4	354.197	1.41351	124.63 ± 2.32^{a}	115.89 ± 2.89^{bc}	113.6 ± 2.81^{bc}	116.99 ± 3.85^{b}	108.29 ± 3.94^{c}	124.25 ± 6.33^{a}	114.57 ± 5.02^{bc}
n-Nonanal	C9H18O	142.2	1103.2	500.985	1.4837	212.93 ± 17.82^{ab}	195.91 ± 5.63^{abc}	211.94 ± 9.74^{ab}	190.76 ± 6.11^{bc}	188.53 ± 17.57^{c}	216.57 ± 13.37^{a}	193.35 ± 6.61^{bc}
(E)-2-Hexenal(M)	C6H10O	98.1	841.1	231.465	1.18277	236.34 ± 14.75^{d}	229.58 ± 10.16^{d}	271.77 ± 13.03^{c}	301 ± 11.06^{b}	233.19 ± 7.49	360.58 ± 6.08^{a}	308.31 ± 1.77^{b}
(E)-2-Hexenal(D)	C6H10O	98.1	839.7	230.779	1.51893	83.86 ± 3.34 ^{g}	97.15 ± 6.7^{f}	151.7 ± 5.1^{d}	182.04 ± 2.33^{c}	116.34 ± 5.87^{e}	280.7 ± 7.88^{a}	209.94 ± 14.73^{b}
(E)-2-Octenal	C8H14O	126.2	1055	424.399	1.33132	405.75 ± 17.09^{bc}	382.52 ± 7.97^{c}	416.49 ± 16.24^{b}	463.82 ± 18.7^{a}	390.71 ± 18.12^{bc}	391.05 ± 13.45^{bc}	414.89 ± 7.92^{b}
(E)-2-Heptenal(M)	C7H12O	112.2	954.1	308.124	1.25695	2812.81 ± 30.38^{a}	2487.08 ± 59.33^{b}	2311.78 ± 67.22^{c}	2286.01 ± 23.06^{c}	2103.43 ± 23.58^{d}	1858.98 ± 66.43^{f}	1987.74 ± 19.57^{e}
(E)-2-Heptenal(D)	C7H12O	112.2	952.6	306.84	1.6719	3018.19 ± 91.45^{a}	2431.9 ± 147.15^{b}	2269.9 ± 193.96^{b}	2349.36 ± 47.95^{b}	1725.64 ± 7.29^{c}	1478.04 ± 101.78^{d}	1708.3 ± 88.9^{c}
(Z,Z)-2,4-Heptadienal	C7H10O	110.2	998.8	349.781	1.20795	171.31 ± 8^{c}	250.64 ± 8.6^{b}	285.38 ± 7.94^{a}	295.68 ± 1.12^{a}	248.29 ± 18.67^{b}	257.85 ± 7.58^{b}	299.5 ± 4.82^{a}
(E,E)-2,4-Heptadienal (M)	C7H10O	110.2	1009.8	363.323	1.2011	571.51 ± 69.31^{d}	629.46 ± 9.57 ^{d}	759.58 ± 8.16^{c}	883.46 ± 21.1^{b}	704.38 ± 19.61^{c}	992.42 ± 61.6^{ad}	965.48 ± 27.75^{a}
(E,E)-2,4-Heptadienal (D)	C7H10O	110.2	1008.6	361.851	1.6225	91.02 ± 21.63^{e}	106.82 ± 6.43^{de}	147.25 ± 3.45^{c}	178.62 ± 9.17^{b}	123.05 ± 10.66^{cd}	236.36 ± 34.21^{a}	215.8 ± 2.98^{a}
(E,E)-2,4-Hexadienal(M)	C6H8O	96.1	910	272.203	1.11673	440.11 ± 2.8^{ab}	426.36 ± 11.44^{c}	428.96 ± 7.28^{bc}	452.05 ± 7.49^{a}	434.56 ± 2.49^{bc}	426.31 ± 4.75^{c}	432.8 ± 8.06^{bc}
(E,E)-2,4-Hexadienal(D)	C6H8O	96.1	910.5	272.639	1.46132	117.56 ± 2.98^{bc}	113.3 ± 3.44^{c}	121.28 ± 2.45^{b}	129.79 ± 2.27^{a}	122.75 ± 4.9^{ab}	119.48 ± 6.47^{bc}	124.38 ± 3.95^{ab}
(E)-2-Pentenal(M)	C5H8O	84.1	741.3	184.704	1.10515	412.28 ± 39.18^{b}	501.21 ± 4.22^{a}	521.34 ± 13.19^{a}	495.8 ± 4.98^{a}	498.6 ± 4.96^{a}	515.43 ± 4.98^{a}	495 ± 1.6^{a}
(E)-2-Pentenal(D)	C5H8O	84.1	741.9	184.956	1.36188	1654.65 ± 118.61^{c}	2179.2 ± 53.36^{b}	2532.36 ± 6.95^{a}	2503.66 ± 59.44^{a}	2433.05 ± 18.79^{a}	2534.25 ± 63.05^{a}	2539.76 ± 51.47^{a}
3-Methyl-2-butenal	C5H8O	84.1	765.6	195.538	1.36192	215.14 ± 3.03^{d}	247.71 ± 3.8^{c}	271.65 ± 3.43^{a}	270.48 ± 1.46^{ab}	262.73 ± 4.73^{ab}	250.29 ± 10.32^{c}	260.81 ± 5.93^{b}
2-Methyl-propanal	C4H8O	72.1	550.9	123.598	1.28777	364.42 ± 15.14^{a}	292.98 ± 5.96^{d}	316.91 ± 8.54^{c}	323.23 ± 5.29^{c}	330.71 ± 7.12^{bc}	344.6 ± 9.28^{b}	336.78 ± 16.94^{bc}
2-Methylbutanal	C5H10O	86.1	654.2	152.197	1.39924	1715.46 ± 14.96^{c}	1813.87 ± 90.03^{c}	1852.2 ± 61.39^{bc}	1958.25 ± 28.69^{b}	2250.96 ± 28.27^{a}	2330.48 ± 140.5^{a}	2201.61 ± 81.04^{a}
3-Methylbutanal	C5H10O	86.1	633.1	145.86	1.41307	1447.98 ± 87.56^{b}	1379.8 ± 92.15^{bc}	1287.65 ± 70.23^{c}	1457.18 ± 25.64^{b}	1836.29 ± 47.78^{a}	1861.76 ± 132.59^{a}	1748.58 ± 90.1^{a}
5-Methylfurfural	C6H6O2	110.1	963.6	316.471	1.12853	231.46 ± 31.98^{d}	321.22 ± 20.09^{c}	388.07 ± 13.99^{b}	390.18 ± 5.59^{b}	395.46 ± 15.98^{b}	431.37 ± 18.79^{a}	461.2 ± 12.95^{a}
2-Furfural(M)	C5H4O2	96.1	820.3	221.164	1.08485	1728.87 ± 30.41^{a}	1706.91 ± 2.71^{a}	1635.98 ± 24.47^{b}	1562.64 ± 5.6^{c}	1651.35 ± 14.46^{b}	1555.13 ± 11.63^{c}	1567.33 ± 8.7^{c}
2-Furfural(D)	C5H4O2	96.1	821	221.507	1.33507	4767.64 ± 197.12^{c}	4948.85 ± 12.72^{ab}	4890.02 ± 45.54^{abc}	4772.14 ± 59.71^{c}	5028.63 ± 23.01^{a}	4758.06 ± 12.16^{c}	4872.83 ± 18.66^{bc}
Benzaldehyde(M)	C7H6O	106.1	957.4	310.934	1.15171	303.65 ± 28.67^{e}	377.63 ± 12.89^{d}	407.06 ± 12.53^{c}	413.62 ± 7.81^{c}	474.84 ± 10.25^{b}	544.9 ± 9.93^{a}	476.17 ± 11.72^{b}
Benzaldehyde(D)	C7H6O	106.1	958.7	312.089	1.47248	186.77 ± 5.35^{e}	264.86 ± 11.02^{d}	329.32 ± 20.71^{c}	328.06 ± 10.34^{c}	346.57 ± 0.98^{c}	436.88 ± 10.03^{a}	393.16 ± 4.39^{b}
Phenylacetaldehyde	C8H8O	120.2	1039.3	402.19	1.25788	519.07 ± 71.75^{a}	400.27 ± 20.72^{b}	386.18 ± 4.64^{bc}	369.11 ± 11.77^{bc}	364.74 ± 8.79^{bc}	334.85 ± 5.63^{c}	353.94 ± 21.01^{bc}
Ketones (8)												
3-Hydroxy-2-butanone (M)	C4H8O2	88.1	706.7	170.235	1.07287	557.41 ± 20.95^{a}	517.58 ± 6.73^{b}	430.59 ± 22^{d}	462.16 ± 3.41^{c}	471.04 ± 16.02^{c}	435.03 ± 16.04^{d}	402.6 ± 0.8^{e}
3-Hydroxy-2-butanone (D)	C4H8O2	88.1	701.6	168.233	1.32964	1523.11 ± 91.28^{a}	1431.48 ± 22.15^{b}	1169.06 ± 70.03^{d}	1336.77 ± 35.36^{c}	1398.75 ± 36.16^{bc}	1189.48 ± 6.62^{d}	1225.98 ± 44.33^{d}
1-Penten-3-one	C5H8O	84.1	675.3	158.827	1.31778	2194.96 ± 187.21^{d}	2784.34 ± 104.65^{bc}	3074.2 ± 19.4^{a}	2900.38 ± 72.36^{ab}	3052.47 ± 34.95^{a}	2685.51 ± 155.63^{c}	2881.44 ± 91.34^{abc}

(*continued on next page*)

Table 2 (*continued*)

Compound	Formula	MW	RI	Rt [*sec*]	Dt [a.u.]	Signal intensity						
						A	B	C	D	E	F	G
2-Butanone	C4H8O	72.1	577.7	130.462	1.24391	2570.39 ± 110.93^{cd}	2065.3 ± 26.99^{e}	2282.19 ± 52.57^{de}	3734.94 ± 72.6^{a}	3043.66 ± 30.44^{b}	2853.34 ± 122.26^{bc}	3224.71 ± 518.11^{b}
2-Propanone	C3H6O	58.1	462.2	103.34	1.12001	11217.27 ± 183.22^{a}	10510.48 ± 25.26^{cd}	10431.28 ± 148.34^{d}	10803.79 ± 95.03^{b}	10896.86 ± 100.18^{b}	10752.43 ± 87.67^{bc}	$10662.05 \pm 272.48^{bcd}$
2-Heptanone	C7H14O	114.2	884	254.364	1.2614	177.46 ± 49.51^{a}	124.65 ± 6.08^{b}	131.94 ± 4.3^{b}	138.96 ± 11.02^{ab}	130.38 ± 3.21^{b}	143.07 ± 25.91^{ab}	139.99 ± 10.62^{ab}
1-Octen-3-one(M)	C8H14O	126.2	977.2	328.758	1.27596	580.83 ± 27.49^{a}	448.69 ± 21.03^{b}	447.35 ± 46.37^{b}	483.86 ± 20.73^{b}	362.66 ± 6.27^{c}	280.03 ± 38.35^{d}	338.06 ± 17.77^{c}
1-Octen-3-one(D)	C8H14O	126.2	974.9	326.612	1.69102	456.03 ± 7.43^{a}	278.74 ± 25.89^{b}	268.94 ± 42.92^{b}	293.12 ± 8.4^{b}	180.65 ± 6.06^{c}	121.79 ± 20.71^{d}	156.16 ± 19.04^{cd}
Alcohols (5)												
Ethanol	C2H6O	46.1	425.5	95.974	1.05228	416.11 ± 52.12^{c}	834.83 ± 7.97^{a}	771.7 ± 15.84^{a}	304.04 ± 6.52^{e}	629.51 ± 9.92^{b}	329.13 ± 14.45^{de}	380.76 ± 84.69^{cd}
1-Octen-3-ol(M)	C8H16O	128.2	978.6	330.078	1.15788	126.21 ± 6.13^{c}	128.96 ± 3.41^{c}	150.08 ± 6.4^{b}	154.76 ± 8.03^{ab}	148.2 ± 11.76^{b}	165.27 ± 2.38^{a}	150.02 ± 5.07^{b}
1-Octen-3-ol(D)	C8H16O	128.2	976.3	327.932	1.60026	82.48 ± 9.51^{abc}	80.37 ± 7.37^{bcd}	91.87 ± 6.53^{ab}	94.55 ± 1.79^{a}	75.8 ± 7.87^{cd}	69.54 ± 3.26^{d}	74.81 ± 6.4^{cd}
1-Pentanol(M)	C5H12O	88.1	752.6	189.643	1.25339	340.40 ± 7.36^{a}	287.09 ± 2.89^{b}	280.8 ± 6.54^{b}	279.25 ± 12.39^{b}	275.52 ± 5.99^{b}	247.89 ± 1.5^{c}	251.91 ± 11.87^{c}
1-Pentanol(D)	C5H12O	88.1	750.4	188.667	1.52174	114.06 ± 3.6^{a}	101.22 ± 2.3^{b}	95.49 ± 3.05^{b}	93.61 ± 1.43^{c}	91.52 ± 1.79^{c}	78.43 ± 6.21^{c}	86.19 ± 5.08^{c}
Esters (2)												
Ethyl acetate	C4H8O2	88.1	597.7	135.819	1.33472	3279.64 ± 458.62^{b}	5928.67 ± 113.16^{a}	6077.22 ± 50.38^{a}	694.38 ± 17.04^{c}	2233.12 ± 95.36^{b}	3188.83 ± 555.88^{b}	2629.33 ± 1716.76^{b}
Ethyl phenylacetate	C10H12O2	164.2	1304.9	1002.833	1.28861	229.56 ± 13.64^{a}	257.77 ± 16.01^{a}	262.74 ± 19.36^{a}	273 ± 30.08^{a}	224.29 ± 46.7^{a}	250.37 ± 14.65^{a}	233.49 ± 18.43^{a}
Hydrocarbons (8)												
Limonene(M)	C10H16	136.2	1024.5	382.165	1.22052	140.71 ± 17.37^{d}	161.56 ± 8.53^{c}	175.39 ± 11.06^{b}	157.79 ± 11.41^{cd}	183.14 ± 6.9^{c}	707.79 ± 6.9^{a}	181.85 ± 5.21^{b}
Limonene(P)	C10H16	136.2	1023.8	381.282	1.30388	119.59 ± 20.25^{cd}	127.47 ± 13.68^{bcd}	132.19 ± 4.83^{bc}	108.33 ± 2.14^{d}	144.21 ± 11.64^{b}	749.59 ± 2.78^{a}	126.86 ± 6.51^{bcd}
Limonene(P)	C10H16	136.2	1024.3	381.87	1.66019	28.33 ± 4.86^{b}	28.78 ± 1.16^{b}	28.02 ± 1.8^{b}	30.93 ± 3.32^{b}	26.89 ± 6.69^{b}	69.87 ± 2.95^{a}	31.57 ± 5.86^{b}
Limonene(P)	C10H16	136.2	1024.3	381.87	1.70016	21.93 ± 2.28^{bc}	26.09 ± 2.66^{bc}	24.7 ± 4.05^{bc}	25.4 ± 2.74^{bc}	19.58 ± 4.46^{bc}	62.04 ± 8.85^{a}	31.1 ± 3^{b}
Limonene(P)	C10H16	136.2	1022.5	379.515	1.73556	30.8 ± 2.88^{b}	28.34 ± 2.79^{b}	27.89 ± 2.55^{b}	23.65 ± 2.14^{b}	24.99 ± 8.24^{b}	66.71 ± 4.09^{a}	28.95 ± 5.06^{b}
Myrcene	C10H16	136.2	988.3	339.182	1.21937	19.86 ± 1.44^{b}	22.29 ± 1.16^{b}	25.13 ± 1.4^{b}	21.66 ± 3.49^{b}	23.22 ± 5.21^{b}	72.44 ± 4.89^{a}	24.66 ± 1.8^{b}
(-)-beta-Pinene	C10H16	136.2	968.7	320.965	1.22405	55.66 ± 8.92^{c}	102 ± 20.48^{b}	52.9 ± 6.94^{c}	69.89 ± 5.04^{c}	53.18 ± 5.92^{c}	185.39 ± 4.63^{a}	57.19 ± 2.53^{c}
alpha-Pinene	C10H16	136.2	927.7	286.129	1.21653	132.1 ± 5.4^{d}	136.88 ± 3.35^{cd}	127.27 ± 2.31^{d}	143.29 ± 8.56^{bc}	136.79 ± 2.12^{cd}	152.14 ± 9.66^{ab}	157.13 ± 3.5^{a}
Furans (1)												
2-Pentylfuran	C9H14O	138.2	988.6	339.467	1.25409	434.36 ± 3.21^{a}	356.54 ± 5.1^{c}	354.07 ± 8.01^{c}	342.27 ± 6.6^{c}	325.91 ± 6.27^{d}	350.56 ± 14.72^{c}	377.74 ± 13.4^{b}
Sulfur compound (3)												
Diallyl sulfide	C6H10S	114.2	844.8	233.354	1.12076	476.72 ± 8.92^{a}	462.53 ± 4.55^{a}	433.81 ± 6.23^{b}	428.43 ± 2.96^{b}	470.6 ± 12.03^{a}	382.67 ± 3.05^{c}	435.36 ± 11.79^{b}
Diallyl disulfide(M)	C6H10S2	146.3	1071.6	449.328	1.20403	2262.85 ± 20.83^{a}	2174.27 ± 12.52^{d}	2194.09 ± 9.03^{cd}	2253.74 ± 23.88^{ab}	2239 ± 23.7^{ab}	2199.88 ± 12.53^{cd}	2221.01 ± 19.1^{bc}
Diallyl disulfide(D)	C6H10S2	146.3	1070.7	447.969	1.64097	396.96 ± 18.47^{ab}	384.64 ± 10.06^{bc}	377.44 ± 21.87^{bc}	412.73 ± 12.1^{a}	397.84 ± 8.76^{ab}	368.66 ± 5.11^{c}	398.06 ± 8.06^{ab}

M: monomer, D: dimer; MW: Represents the molecular weight of the volatile compounds. RI: Represents the retention time in the capillary GC column. RT: Represents the retention index calculated using *n*-ketone C4-C9 as the external standard in the FS-SE-54-CB column. DT: Represents the drift time in the drift tube, fresh (A); stored at 0 °C for 14 d (B); stored at 4 °C for 14 d (C); stored at 25 °C for 14 d (D); stored at 0 °C for 21 d (E); stored at 4 °C for 21 d (F); stored at 25 °C for 21 d (G), Values in each row with different letters are significant difference at $p < 0.05$.

The limonene had a fruity aroma and was a key flavor compound contributed to the odor of vegetables (van Gemert, 2011; Yang et al., 2022). The limonene increased in the olive vegetable after stored for 21 d and reached the highest content stored at 4 °C for 21 d. Besides, the (-)-beta-pinene and myrcene reached the highest contents of 185.39 and 72.44, respectively in the olive vegetables stored at 4 °C for 21 d. The alpha-Pinene increased in olive vegetable after stored for 21 d with storage temperature. In addition, the content of diallyl sulfide and 2-pentylfuran decreased after stored for 14 d compared to fresh samples.

3.2.3. Comparison of fingerprints of volatile compounds

The gallery plot (Fig. 2) was used to distinguish different volatile compounds in olive vegetable samples stored at different conditions. As shown in Fig. 2, the degree of color reflected the intensity of the signal. If the contents of volatile compounds increased, the color would change from blue to red. The different letter of M, D, T and P in parentheses represented the compound's monomer, dimer, trimers and polymers. The Fig. 2 indicated that there were clear differences between different olive vegetable samples. Some intensity of components shown in the red box indicated that these compounds in olive vegetable sample stored at 4 °C for 21 d was much stronger than other samples, including limonene, (−)-beta-pinene and myrcene, they are all hydrocarbons. The results indicated that the olive vegetable stored at 4 °C could produce more alkenes with storage time. The limonene contributed to a pleasant lemon-like aroma, which lead to a desirable odor in the olive vegetable. Many volatile compounds shown in the yellow box indicated that the signal intensity of these compounds in fresh olive vegetable was much higher than other samples, including phenylacetaldehyde, 3-hydroxy-2-butanone, 1-pentanol, 1-octen-3-one, (E)-2-heptenal, and pentanal, and these volatiles decreased with storage time. The contents of 1-octen-3-one, (E)-2-heptenal, and pentanal were higher in olive vegetable samples stored at 14 d than that stored at 21 d. The results indicated the longer storage time may lead to the decline of these volatiles. In the green box, the signal intensity of some volatile compounds in olive vegetable increased with storage temperature in both storage time, including (E)-2-hexenal, 2-furfural, benzaldehyde, 3-methyl-2-butenal, and 1-penten-3-one. In the orange box, the contents of some volatiles in fresh olive vegetable were lowest and increased with storage time, including (E,E)-2,4-heptadienal, (Z,Z)-2,4-heptadienal, 5-methylfurfural, (E)-2-octenal, heptanal, (E)-2-pentenal, and 2-butanone. In the blue orange box, the contents of 2-methylbutanal and 3-methylbutanal in olive vegetable stored at 21 d were higher than that stored at 14 d, however, the content of 1-octen-3-ol was higher in olive vegetable stored at 14 d than that stored at 21 d. The gallery plot indicated that GC-IMS was a effective technique to clearly distinguish different olive vegetable samples.

3.2.4. PCA analysis of GC-IMS

A PCA chart was used to better evaluate the difference in volatile compounds of olive vegetable under different storage conditions. The PCA is an effective multivariable technique for decreasing the dimensionality of original variables to give a general review of sample variations and determine patterns in data by simplifying the influencing factors (Song et al., 2020). Fig. 3 showed the PCA results of volatile compounds in olive vegetable samples. In general, when the first two principal components reached 60%, the PCA model is effective to separate the samples (Chen, et al., 2020). Th PC1 was 53% and PC2 was 18%, which explained 71% of the total variance, which indicated that PC1 and PC2 could explain most of the flavor information. It had a clear separation trend of olive vegetable samples stored at different conditions in the axis of PC1, indicating that these are significantly different. The volatile compounds in olive vegetable changed with the storage time, however, the change is the least when olive vegetable was stored at 0 °C.

4. Conclusion

The volatile aroma compounds of olive vegetables stored at different conditions were compared. This study used the GC-IMS to determine the volatile compounds change in olive vegetables during storage. In this study, we could conclude that PCA of GC-IMS revealed that flavor compounds could distinguish olive vegetable samples stored at different conditions. 57 volatile components were determined from olive vegetable samples. The volatile compounds of olive vegetables include aldehydes, ketones, alcohol esters, hydrocarbons, furans, sulfur compounds. The results of gallery plot indicated that olive vegetables stored at 4 °C could produce more alkenes with storage time. The limonene contributed to a pleasant lemon-like aroma, which lead to a desirable odor in the olive vegetable stored at 4 °C. The contents of some volatiles in fresh olive vegetable were lowest and increased with storage time, including (E,E)-2,4-heptadienal, (Z,Z)-2,4-heptadienal, 5-methylfurfural, (E)-2-octenal, heptanal, (E)-2-pentenal, and 2-butanone. The volatile compounds in olive vegetable changed with the storage time, however, the change was the least when olive vegetable was stored at 0 °C. Therefore, evaluating the effect of different storage conditions on the flavor

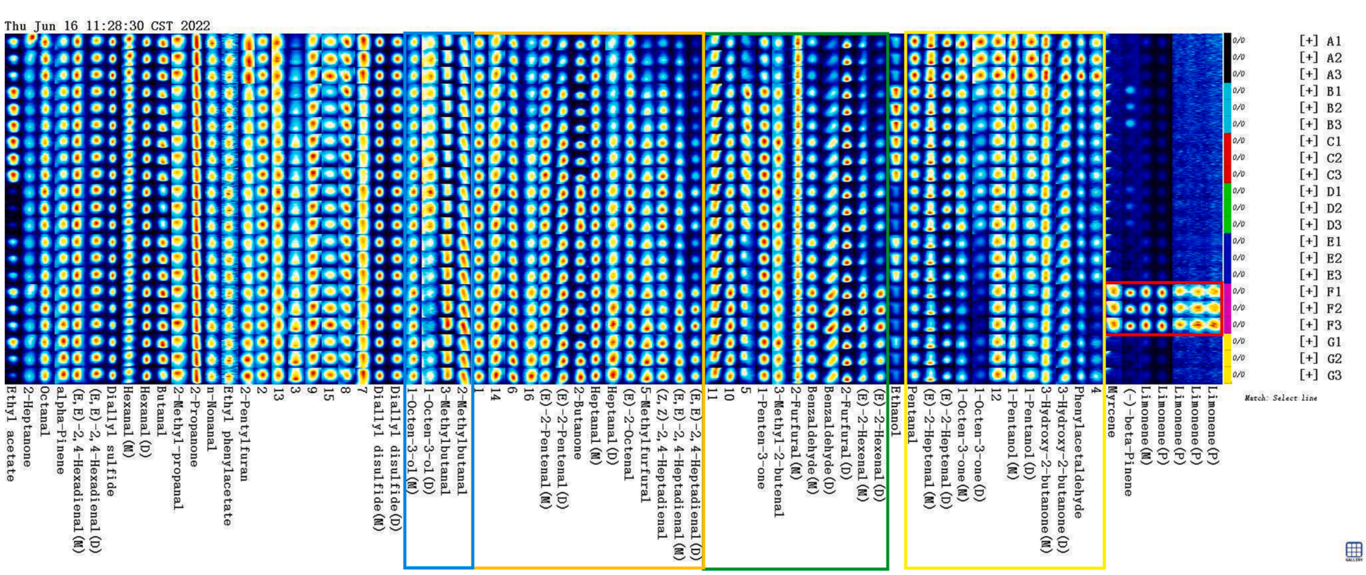

Fig. 2. Gallery plot comparison of volatile compounds in olive vegetable stored at different conditions, fresh (A); stored at 0 °C for 14 d (B); stored at 4 °C for 14 d (C); stored at 25 °C for 14 d (D); stored at 0 °C for 21 d (E); stored at 4 °C for 21 d (F); stored at 25 °C for 21 d (G).

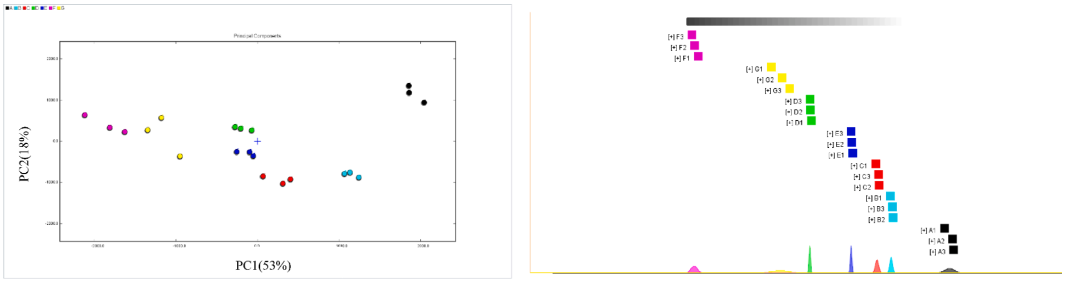

Fig. 3. PCA analysis and "Nearest neighbor" fingerprint analysis of GC-IMS.

compounds of olive vegetable can provide theoretical bases for improving the flavor quality of olive vegetable. Furthermore, this study can provide bases for the development of traditional food and standardized industrial production.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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