



Analysis of the volatile compounds in Fuliji roast chicken during processing and storage based on GC-IMS

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ABSTRACT

To investigate the flavor changes of Fuliji roast chicken during processing and storage, the volatile organic compounds (VOCs) during processing (fresh, fried, stewed and sterilized) and storage (1 month, 2 months and 4 months) were determined by gas chromatography ion mobility spectrometry (GC-IMS). A total of 47 kinds of VOCs were identified across seven sampling stages, including aldehydes, hydrocarbons, alcohols, ketones, esters, ethers and heterocyclic compounds. More diverse range of aldehydes, alcohols, ketones and esters have been detected compared to acids, ethers and heterocyclic substances. Fingerprints directly reflect the pattern of VOCs at different stages of growth and decay, revealing that frying and stewing are key processes in flavor formation, and that sterilization and storage processes lead to flavor loss in Fuliji roast chicken. Hexanal, nonanal, octanal, 2-heptanone, 3-octanol, 1-octene-3-alcohol, 1-pentanol and ethyl acetate were mainly generated during the frying process. Benzaldehyde, nonanal, octanal, methyl-5-hepten-2-one, 2-methyl-3-heptanone, 1,8-Cineole, linalool, butyl acetate, ethyl propionate, ethyl acetate, coumarin, 2-furfuryl methyl disulfide and 2-pentyl furan were mainly generated during the stewing process. After sterilization, the content of octanal-D, 2-heptanone-D, 2-Methyl-3-heptanone, pentan-1-ol-D decreased, resulting in the reduction of aroma, lemon flavor and oil flavor of Fuliji roast chicken. Seven flavor markers, including hexanal-D, nonanal-M, octanal-M, heptanal-D, acetone, 3-octanol and ethyl acetate-D, were identified in the evolution of the aroma profile of Fuliji roast chicken.

1. Introduction

Fuliji roast chicken, among the traditional marinated chicken products in China, is a representative category with a long history and is a national geographical indication product. Fuliji roast chicken has a delicious flavor, with aromas of meat and ingredients. Yang et al. (2021) analyzed the flavor of six commercial smoked chicken products in China by gas chromatography-mass spectrometry (GC-MS). A total of 89 components were identified in all samples, it was found that the diversity of taste of smoked chicken is mainly due to differences in cooking culture. Yao et al. (2020) investigated the flavor profile on 5 different regional Chinese smoked chicken samples through GC-IMS and identified 34 flavor compounds, in which n-nonanal, heptanal, n-nonanal,

heptanal, furfural, and hexanal were the main common flavor compounds. Xu et al. (2021) explored the flavor and taste of soft-boiled chicken at different post-mortem aging time based on GC-IMS and multivariate statistical analysis. It was found that post-mortem aging was an important process affecting flavor. Innucleotides, seven low-molecular weight water-soluble peptides and eight volatile aromas were identified as the main components affecting flavor.

The unique sauce flavor of the marinated chicken products is one of the important reasons why they are favored by consumers. For a long time, the characteristic flavor of braised chicken products, especially Dezhou braised chicken, has been widely investigated by researchers. Duan et al. (2015) used comprehensive two-dimensional gas chromatography-high-resolution time-of-flight mass spectrometry (GC

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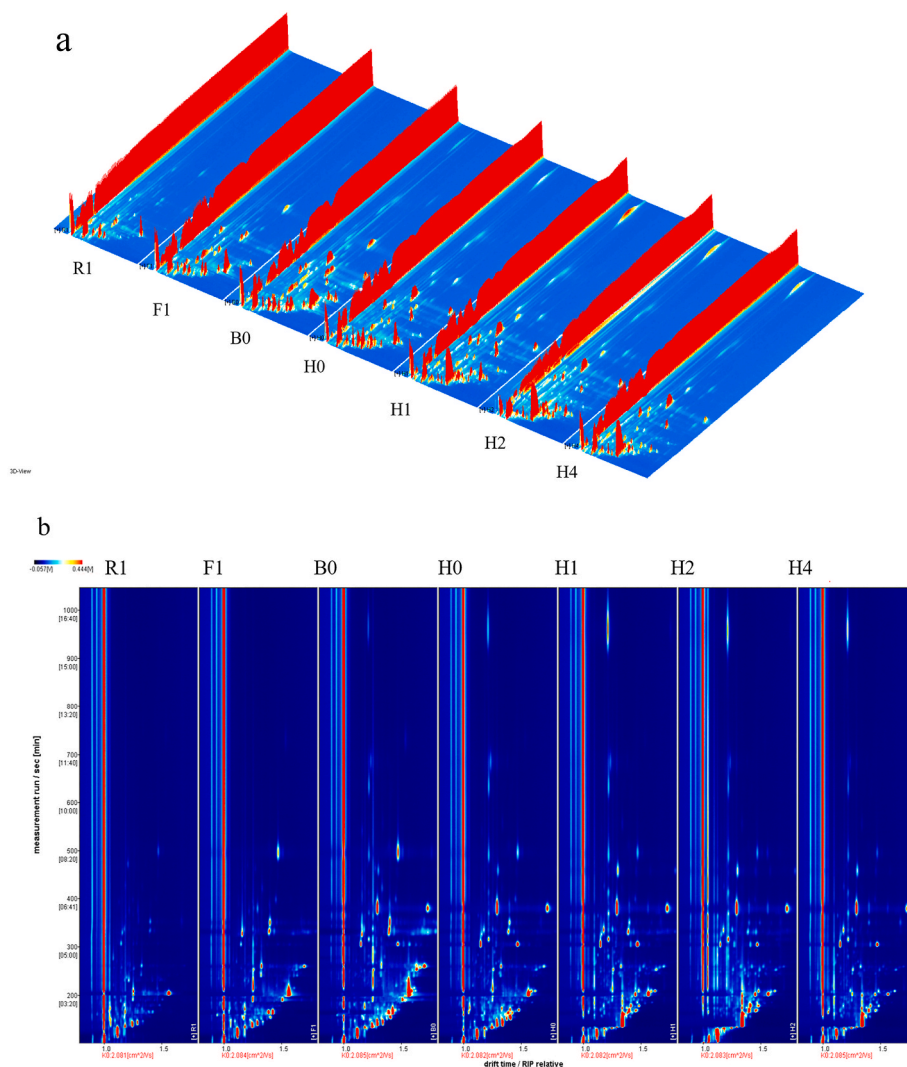


Fig. 1. Three-dimensional spectrum (a), two-dimensional spectrum (b) of the volatile substances in the Fuliji roast chicken at different processing and storage stages. R1: raw chicken or fresh; F1: end of frying; B0: end of stewing; H0: commercial sterilization; H1: storage for 1 month; H2: storage for 2 months; H4: storage for 4 months.

× GC-TOFMS) to analyze the volatile organic compounds (VOCs) in the Dezhou braised chicken. Wang et al. (2020) studied the volatile flavor compounds after the thermal processing of the Dezhou braised chicken at different temperatures. Zhang et al. (2021) used headspace solid-phase microextraction gas chromatography-mass spectrometry (HS-SPME/GC-MS) to analyze the effect of the different sugar smoking times on the flavor profiles of the chicken drumsticks.

Gas chromatography-ion mobility spectrometry (GC-IMS) is a new detection technology with a high visualization of the detection results compared with GC-MS, GC-O-MS. GC-MS is the most common technique for the analysis of volatile compounds and is widely used. However, GC-MS analysis generally requires enrichment and concentration of the sample prior to analysis, the composition may change and the long detection times may not meet the rapid detection requirements of many analytes. In addition, the MS instrument operates requires at vacuum work, He is expensive as a carrier gas and data processing is cumbersome (Feizi et al., 2021). GC-O-MS is effective in selecting aromatically active compounds from complex mixtures but also requires vacuum work and a lot of repetitive and time-consuming work (Song and Liu, 2018). GC-IMS is a powerful technique for the separation and sensitive detection of volatile organic compounds for application in the classification of food products, the identification of food freshness, quality control of the production process (Wang et al., 2020). It offers fast response times, high

sensitivity, ease of operation and low cost. The sample does not need to be enriched and concentrated to maintain true flavor, the IMS instrument operates at atmospheric pressure, using N₂ as the carrier gas at low cost, while visual fingerprinting is possible and data processing is simple for rapid detection (Hernández-Mesa et al., 2019). Martín-Gómez et al. (2019) established the fingerprint of the volatile components of Iberian ham by GC-IMS and realized the rapid and accurate identification of its authenticity. Zhang et al. (2020) used GC-IMS to analyze the changes in volatile flavor compounds during the storage of dry-cured fish. More recently, Yao et al. (2022) analyzed flavor formation during production of the Dezhou braised chicken with GC-IMS.

Nowadays, visual data on the flavor changes during the processing and the storage of Fuliji roast chicken are still lacking, limiting its standardized and large-scale development. In this study, the VOCs of Fuliji roast chicken during processing and storage were analyzed using GC-IMS, and a visual fingerprint of VOCs of Fuliji roast chicken was constructed to identify the change patterns, with the aim of providing a theoretical basis and technical support for the modern processing of Fuliji roast chicken.

Table 1 (continued)

Species	Number	Compound Name	Processing stage			Storage stage			
			R1	F1	B0	H0	H1	H2	H4
					224.08 ± 10.05 ^c	217.6 ± 37.44 ^c	493.7 ± 21.53 ^a	509.35 ± 37.96 ^a	285.62 ± 29.29 ^b
	38	Ethyl propanoate-M	22.09 ± 1.34 ^d	17.59 ± 0.85 ^e	16.4 ± 0.79 ^e	21.59 ± 2.62 ^d	59.17 ± 1.28 ^a	33.48 ± 1.78 ^c	48.68 ± 3.30 ^b
	39	Ethyl Acetate-M	96.83 ± 5.81 ^c	141.54 ± 3.32 ^b	156.4 ± 4.29 ^a	146.15 ± 6.82 ^b	18.99 ± 3.57 ^e	16.11 ± 1.96 ^e	29.45 ± 6.07 ^d
	40	Ethyl Acetate-D	157.86 ± 8.37 ^e	432.76 ± 24.94 ^d	1749.81 ± 208.55 ^c	2826.24 ± 209.72 ^b	7471.28 ± 115.95 ^a	7405.28 ± 201.53 ^a	7255.68 ± 192.69 ^a
	41	Propyl acetate	12.01 ± 1.19 ^e	19.73 ± 2.43 ^e	85.13 ± 6.39 ^c	384.46 ± 42.15 ^a	85.51 ± 3.89 ^c	125.83 ± 7.54 ^b	52.86 ± 2.57 ^d
Ethers	42	Butyl sulfide	41.10 ± 6.29 ^f	35.70 ± 2.95 ^f	272.50 ± 12.33 ^c	316.25 ± 7.57 ^d	777.54 ± 10.63 ^a	639.31 ± 18.80 ^b	499.71 ± 7.31 ^c
	43	1,2-Dimethoxyethane	76.02 ± 12.07 ^e	84.57 ± 5.23 ^{de}	88.80 ± 5.98 ^d	81.44 ± 6.42 ^{de}	204.5 ± 5.51 ^b	115.84 ± 2.18 ^c	300.27 ± 10.63 ^a
Heterocycles	44	Coumarin	161.85 ± 14.55 ^f	150.02 ± 11.79 ^f	1014.13 ± 148.27 ^e	1329.42 ± 80.43 ^d	3676.34 ± 257.47 ^a	2541.62 ± 146.57 ^b	2126.65 ± 31.68 ^c
	45	2-ethyl furan	224.16 ± 14.28 ^b	123.26 ± 13.13 ^d	50.23 ± 9.57 ^e	236.14 ± 47.61 ^b	175.86 ± 30.52 ^c	254.93 ± 16.75 ^b	324.33 ± 23.24 ^a
	46	2-furfuryl methyl disulfide	83.03 ± 5.79 ^e	78.16 ± 5.18 ^e	574.8 ± 37.32 ^{bc}	602.89 ± 31.35 ^b	733.25 ± 14.50 ^a	556.76 ± 17.42 ^c	443.55 ± 9.04 ^d
	47	2-pentyl furan	23.33 ± 5.35 ^f	72.67 ± 8.35 ^e	203.02 ± 9.67 ^a	82.65 ± 5.33 ^d	150.99 ± 4.29 ^b	147.97 ± 3.64 ^b	97.49 ± 4.03 ^c

Notes: All the values are means ± SD, n = 5. Monomer and dimer formed in the IMS drift tube were represented by symbols “M” and “D” respectively. a-g means within the same row with different superscript showing significant differences ($P < 0.05$). R1: raw chicken or fresh; F1: end of frying; B0: end of stewing; H0: commercial sterilization; H1: storage for 1 month; H2: storage for 2 months; H4: storage for 4 months.

2. Materials and methods

2.1. Sample collection and pre-experiment processing methods

HuaiBei Ma chicken (180 days old, weighing about 1.5 kg), as raw material, was processed and stored with the industrialization process of the Fuliji roast chicken. The raw chicken was evenly coated with diluted honey and fried at 180 °C for 2–3 min until the chicken skin turned golden yellow. Then the fried chicken was stewing in stuffy soup, immersed in the soup with spices, heated and boiled for 10 min, then treated with slow fire for 1 h, pickled and soaked for 2 h. Finally vacuum packaging was carried out after cooling to room temperature, and commercial sterilization processing was completed. The sterilized roast chicken was stored at room temperature for 1, 2 and 4 months. Samples of chicken meat were collected for analysis after seven of the key stages during processing and storage. The processing stages included R1 (raw chicken or fresh), F1 (end of frying), B0 (end of stewing), and H0 (commercial sterilization), and the storage stages covered H1 (storage for 1 month), H2 (storage for 2 months), and H4 (storage for 4 months). The samples were taken for analysis during the aforementioned processing and storage stages. After the samples were taken back, they were prepared in a 4 °C freezer (chicken breast meat), five chickens were randomly selected from the seven stages, and their breasts (with skin) were chopped by the meat grinder to provide samples for analysis. The minced meat was vacuum packed, labeled with R1-1, R1-2, R1-3, R1-4, R1-5; F1-1, F1-2, F1-3, F1-4, F1-5and H4-1, H4-2, H4-3, H4-4, H4-5 respectively represent five samples collected in each of the seven stages, and stored in a –80 °C freezer for subsequent testing.

2.2. GC-IMS analysis parameter

The volatile components were analyzed by HS-GC-IMS (FlavorSpec®, Gesellschaft für Analytische Sensorsysteme mbH, Dortmund, Germany, Department of Shandong HaiNeng Science Instrument Co., Ltd., Shandong, China). Slightly modified the analytical method of Li et al. (2022). A 2 g sample of the ground chicken breast was accurately weighed and transferred into a 20 mL headspace vial that was subsequently incubated at 60 °C for 20 min, with an incubation speed of 500 rpm. Whereafter, 500 µL of volatile gas was collected from the headspace bottle and injected into the injector automatically at 65 °C without

split flow. The volatile gas was separated by a fused silica capillary column (FS-SE-54-CB-1 15 m × 0.53 mm, 1 µm) and combined with IMS at 45 °C. We used high purity nitrogen as the carrier gas/drift gas. The initial carrier gas flow rate was set at 2 mL/min. It was maintained at 2 mL/min within 0–2 min, and the carrier gas velocity increased linearly from 2 mL/min to 100 mL/min within 2–20 min, maintained 100 mL/min for 10 min. The total running time was 30 min. The drift gas flow rate was set to 150 mL/min. The column temperature was set at 60 °C, and the separation was ionized in an IMS ionization chamber at 45 °C. GC-IMS analysis was conducted in quintuplicate. GC-IMS data were obtained in positive mode using LAV software and chemical compounds of the samples were identified by using GC-IMS library search software. Both sets of software were provided by G.A.S. (Dortmund, Germany). 2-Butanone, 2-Pentanone, 2-Hexanone, 2-Heptanone, 2-Octanone and 2-Nonone (Sinopharm Chemical Reagent Beijing Co., Ltd, China) were used as external references to calculate the retention index (RI) of volatile compounds.

2.3. Statistical analysis

Five independent batches of ground chicken breast samples (replicates n = 5) were conducted and all measurements were conducted in quintuplicate for each sample. The software VOCal of the GC-IMS was used to perform a qualitative analysis according to the comparison of the RI (the retention index) and Dt (the drift time) of the substance with the NIST and IMS spectral libraries in the database. A Reporter plug-in was used to draw and compare the spectral differences between samples (with two-dimensional top view, three-dimensional spectra, and difference spectra). A Gallery Plot plug-in was used to draw the fingerprints to visually and quantitatively compare the differences in the volatile organic compounds between different samples. The changes of Volatile components of Fuliji roast chicken at different processing and storage stages was statistically analyzed using principal component analysis (PCA), partial least squares discriminant analysis (PLS-DA), and variable importance in projection (VIP) scores, based on the concentration of peak volume during different times of processing and storage stages. The significance of different times of processing and storage stages was assessed using VIP analysis, and compounds with VIP scores of ≥1.0 exhibited substantial distinctiveness. The heat map of volatile compounds and VIP scores were drawn by MetaboAnalyst v5.0. The PCA was

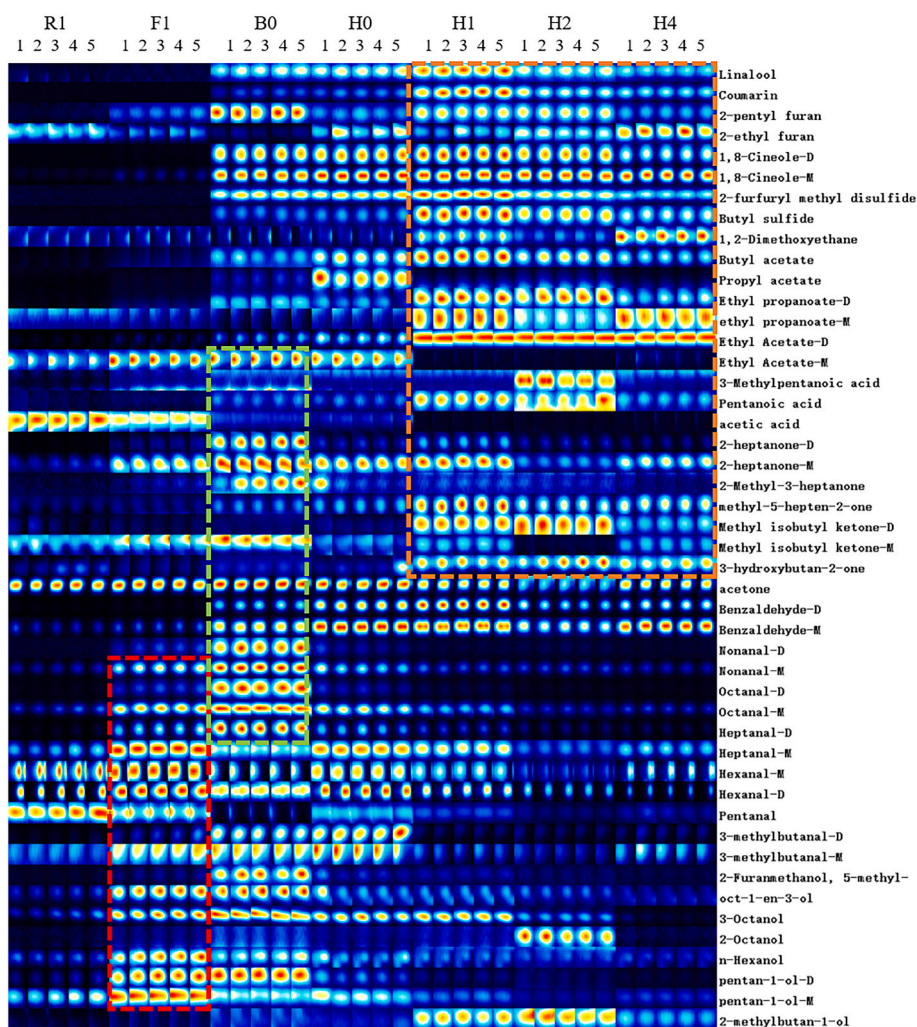


Fig. 2. Gallery Plot fingerprint of volatile substances during processing and storage in the Fuljii roast chicken, which contains most of the important data, are marked with a rectangle (red, green and orange) respectively. R1: raw chicken or fresh; F1: end of frying; B0: end of stewing; H0: commercial sterilization; H1: storage for 1 month; H2: storage for 2 months; H4: storage for 4 months. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

performed by SIMCA 14.1 version (Umetrics, Umea, Sweden).

3. Results and discussion

3.1. Comparative analysis of the GC-IMS spectra of the volatile substances in the Fuljii roast chicken

Fig. 1 (a) shows the GC-IMS three-dimensional spectrum (retention time R_t , drift time D_t , and peak intensity), Fig. 1 (b) shows the two-dimensional top view (retention time R_t and drift time D_t) of the Fuljii roast chicken during processing and storage. As shown in the figure, the volatile components of the special marinated chicken product were different at various stages of processing and storage compared with the GC-IMS three-dimensional spectra of the Fuljii roast chicken at different processing and storage stages. The differences and changes in the volatile substances could be more clearly reflected through the two-dimensional top view. In the two-dimensional top view, the ordinate was the R_t when VOCs were separated and the abscissa was the D_t relative to the reactive ion peak (red vertical line at 1.0 on the abscissa) on both sides of the reactive ion peak. Each point represented a volatile organic compound. The blue was the background, and different colors represented the different concentrations of VOCs of the substance, where the white represented a lower concentration, the red a higher concentration, and a darker color a higher concentration of the compound. Fig. 1 shows that a few volatile components existed in raw chicken and the flavor substances changed a lot in the subsequent processing and storage processes.

3.2. Qualitative and quantitative analyses of the volatile substances in the Fuljii roast chicken

Based on the GC-IMS retention time and the signal intensity, qualitative and quantitative analyses of the volatile components in the Fuljii roast chicken were carried out at different processing and storage stages (Table 1). As shown in Table 1, 47 VOCs could be identified in the Fuljii roast chicken in this study, including 13 aldehydes, 8 ketones, 11 alcohols, 6 esters, and 3 acids, 2 ethers, and 4 heterocycles. During the separation process, benzaldehyde, heptanal, hexanal, nonanal, octanal, isovaleral, 2-heptanone, 4-methyl-2-pentanone, eucalyptol, 1-pentanol, propane ethyl acetate, and ethyl acetate existed as both monomers and dimers, similar to the results observed by Wang et al. (2021) when using GC-IMS to separate the volatile flavor compounds from the Jingyuan mutton. When performing qualitative analysis at high concentrations, the same substance will generate multiple signals or spots representing its monomers, dimers or even multimers due to different concentrations of compounds (Arce et al., 2014).

Aldehydes have a lower threshold and contribute more to the overall flavor of the meat. In meat and meat products, aldehydes are mainly derived from lipid oxidation. For example, hexanal, nonanal, octanal, heptanal, pentanal, benzaldehyde, and so forth are the products of lipid oxidation (Rasinska et al., 2019). During the processing, the content of aldehydes (except *n*-valeraldehyde) increased significantly after frying. These aldehydes were formed from the oxidation of chicken lipids on the one hand and the oxidation of unsaturated fatty acids in oil on the other hand (Zhang et al., 2015). Meinert et al. (2007) also reported that lipids

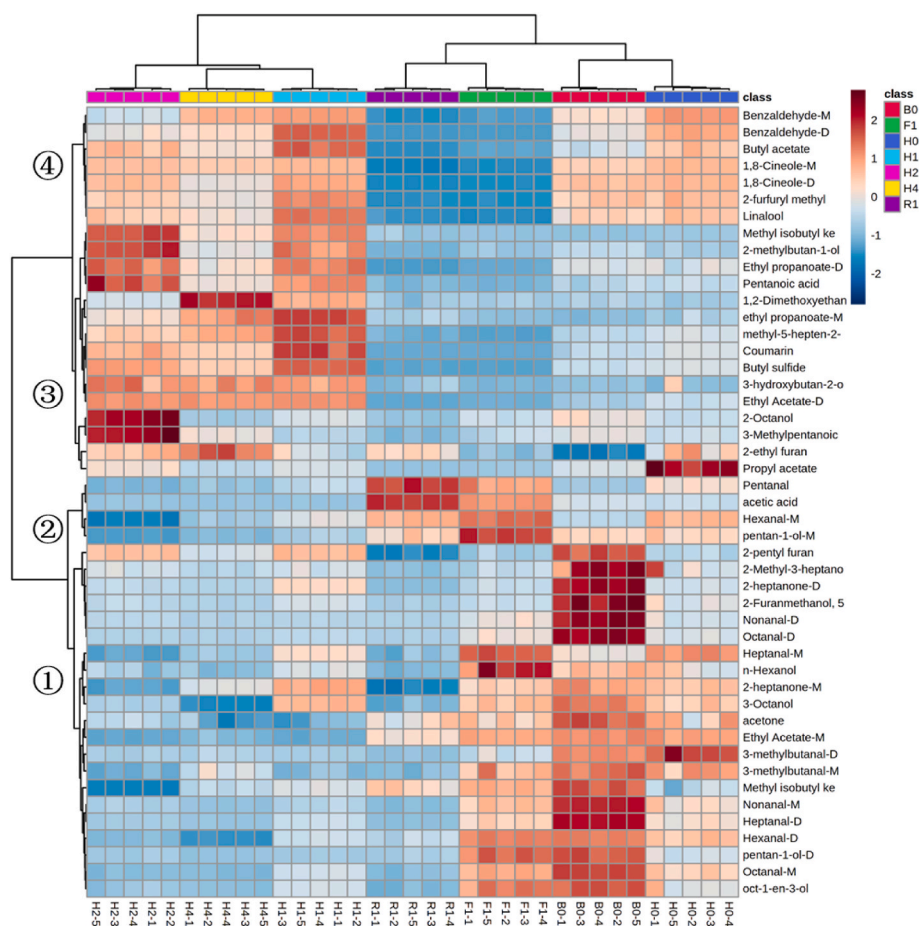


Fig. 3. Heat map clustering of volatile substances during processing and storage in the Fujili roast chicken. R1: raw chicken or fresh; F1: end of frying; B0: end of stewing; H0: commercial sterilization; H1: storage for 1 month; H2: storage for 2 months; H4: storage for 4 months.

were oxidized to form aldehydes during the frying of the meat. Domínguez et al. (2014) pointed out that aldehydes containing 6–10 carbons were the main VOCs in the cooked meat and thus played an important role in the meat aroma. Jin et al. (2021) found that aldehydes were the main volatile flavor compounds in Chinese local high-quality broilers, of which hexanal was the most important one. In this study, the content of hexanal was significantly higher than that of other aldehydes during the processing and during the first 2 months of storage. In general, hexanal could be generated from the oxidation of oleic acid, linoleic acid, and arachidonic acid, as well as the degradation of other unsaturated aldehydes (such as 2,4-decadienal (Hammouda et al., 2017)). The diversity of this synthetic route determined the dominance of the hexanal among the volatile components (Domínguez et al., 2014). During storage, especially when the storage period reached 4 months, the content of the aldehydes decreased significantly. The aldehydes produced by the lipid oxidation could form adducts with proteins (Lynch et al., 2001), which might be an important reason for the decrease in the content of aldehydes at the end of storage in this study.

Alcohols are a class of aromatic compounds produced by lipid oxidation and Strecker degradation in meat and meat products (Qian et al., 2021). Table 1 showed that the contents of 3-octanol, 1-octen-3-ol, n-hexanol, and 1-pentanol increased significantly after frying, indicating that the lipid oxidation and Strecker degradation reactions were violent in the frying stage. Among alcohol substances, the threshold value of unsaturated alcohol was relatively low, and its influence on the flavor of meat products was higher than that of saturated alcohol. Among them, 1-octen-3-ol, secondary alcohol with a mushroom-like odor, was considered to be an important source of the characteristic flavor of cooked chicken fat (Qi et al., 2017). Jin et al. (2021) reported that

similar to hexanal, 1-octen-3-ol was also the main volatile flavor compound in Chinese local high-quality broilers. In this study, the contents of camphor and eucalyptol increased significantly after stewing, indicating that these alcohols mainly came from spices. In the processing of the braised meat products, spices were an important source of alcohol. Qin et al. (2020) found that alcohols such as linalool could be identified in the star anise broth.

Ketones are formed by the auto-oxidation or the β -oxidation of fatty acids. They are stable in nature, with a long-lasting aroma and generally a floral aroma, and have an important impact on the formation of meat flavor (Wang et al., 2021), generally considered a precursor to the formation of fatty flavors associated with meat products (Guo et al., 2021). In this study, ketones were greatly enriched after the stewing process, which was consistent with the conclusion reported by (Yang et al., 2021) that the chicken produced large amounts of ketones during the high-temperature cooking. Among the ketones, 2-heptanone was mainly produced by the oxidation of linoleic acid, which could improve the flavor of the meat and meat products to a certain extent. Domínguez et al. (2014) pointed out that 2-ketones in ketones had a great influence on the aroma of the meat and meat products due to their abundant presence and special aroma.

Esters are mainly derived from the esterification of alcohols and free fatty acids (Li et al., 2016). Domínguez et al. (2019) pointed out that the lower threshold of esters had a great influence on the overall aroma of the meat. In this study, ethyl acetate was the most abundant ester compound during the processing and storage. Table 1 clearly shows that acetic acid, the precursor compound of ethyl acetate, existed in raw meat. The acetic acid content showed a downward trend after the processing steps such as frying and stewing, and the corresponding ethyl

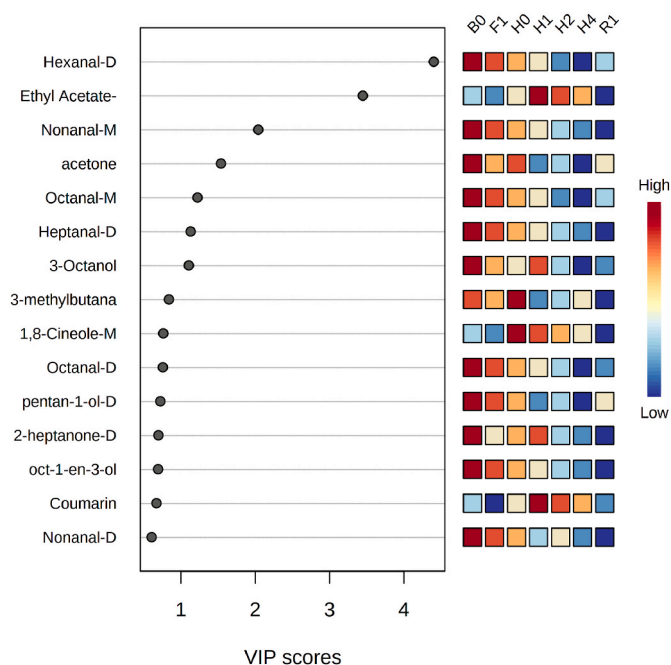


Fig. 4. Variable importance in projection (VIP) scores of volatile compounds in Fuliji roast chicken samples during processing and storage period. Important flavors (VIP ≥ 1.0) screened by PLS-DA during different times during processing and storage period. The colored boxes on the right of represented the relative concentrations of the flavor compounds during different times during processing and storage period. R1: raw chicken or fresh; F1: end of frying; B0: end of stewing; H0: commercial sterilization; H1: storage for 1 month; H2: storage for 2 months; H4: storage for 4 months.

acetate content continued to increase, which might be an important pathway for the ethyl acetate production in this study. At the same time, the Maillard reaction during frying might also be an important pathway for the formation of ethyl acetate (Qian et al., 2021). The fact that spices such as the star anise also contain esters such as ethyl caproate, which play a role in coordinating the fatty, sweet, and fruity flavors in stewed chicken, cannot be ignored (Rasinska et al., 2019).

Besides, the heterocyclic compounds are also an important flavor substance during the processing and storage of the Fuliji roast chicken. Coumarin has a hay fragrance and has been detected and identified as an important flavor substance in the Dezhou braised chicken (Duan et al., 2014). In this study, the coumarin content increased significantly during the stewing process, indicating that coumarin mainly came from the spices. Furans were produced by the Maillard reaction and Strecker degradation and typically exhibited sweet, burned, and toasty flavors (Ge et al., 2020). Furans are essential for the meat flavor (Qi et al., 2017). 2-Pentylfuran has a botanical aromatic odor with a relatively low threshold (Qi et al., 2017), which has an important impact on the meat flavor.

3.3. Fingerprint analysis of the volatile substances in the Fuliji roast chicken

Fig. 2 shows the fingerprint spectrum of the volatile flavor compounds in the processing of the Fuliji roast chicken. The same column in the figure represents the composition of the volatile flavor substances of a sample, the same row represents the signal peak of a certain volatile substance in the sample during processing and storage, and the color of the signal peak represents the concentration of the substance. Fig. 2 can not only be used as a database of the volatile flavor compounds but also intuitively and quantitatively compare the change rules of the VOCs in different processing and storage stages. In the area marked with red rectangle, the content of the flavor compounds such as hexanal, nonanal,

octanal, heptanal, 2-heptanone, 3-octanol, 1-octen-3-ol, 1-pentanol, ethyl acetate, and so on increased significantly after frying compared with the fresh chicken and the fried samples, indicating the important role of the frying process in the formation of these flavor compounds. Among the flavor compounds generated in the frying process, aldehydes and ketones mainly came from lipid oxidation, among which the unsaturated fatty acids of chicken and exogenous vegetable oils could both be oxidized and participate in the generation of aldehydes and ketones. Alcoholic flavor compounds were associated with lipid oxidation and Maillard reactions. Ethyl acetate at this stage might come from the esterification reaction and the Maillard reaction. After the stewing process, in the area marked with green rectangle, the contents of benzaldehyde, nonanal, octanal, isovaleral, methylheptanone, 2-methyl-3-heptanone, eucalyptol, linalool, butyl acetate, ethyl propionate, ethyl acetate, coumarin, methylfurfuryl disulfide, 2-pentylfuran, and other flavor substances increased significantly. In the stewing process, aldehydes and ketones were also mainly the products of lipid oxidation, while alcohols, esters, and heterocyclic flavors came mainly from the combination of the characteristic flavors of the spices and the chicken tissue at this stage. In the subsequent commercial sterilization process, the content of most flavor substances decreased, indicating that the high-temperature sterilization caused damage to the flavor. In addition, the flavor substances such as aldehydes, ketones, and alcohols were lost to varying degrees during the storage process. It was worth noting that in the orange marked area, a large number of ketones, acids, esters and other flavor substances were increased significantly during storage for 1–4 months, and the maximum concentration of flavor substances was 1–2 months. Most of the flavor substances in chicken were reduced after storage for 4 months. The increase of a large number of ketones, acids, esters and other flavor substances and the disappearance or reduction of some alcohols and aldehydes resulted in the formation of bad flavor during the storage process, which affected the consumer acceptability of the Fuliji roast chicken. So, how to reduce the loss of the VOCs of Fuliji roast chicken during the sterilization and storage stages is what we need to do in the future.

3.4. Cluster and similarity analysis of volatile substances in Fuliji roast chicken

As shown in Fig. 3, the volatile flavor compounds were clustered vertically. The peak volume of each volatile flavor was marked by different color in the heat map. The darker the red, the higher the peak volume, and the darker the blue, the lower the peak volume. The samples clustered into the same category showed a high degree of correlation. The shorter the Euclidean distance, the higher the similarity of samples (Xu et al., 2019). From the vertical clustering, it could be roughly divided into four categories since the complexity of flavor substances before they eventually aggregated into one group.

The first category mainly corresponded to aldehydes, alcohols and some ketones, such as heptanal-M, hexanal-D, nonanal-D, nonanal-M, octanal-D, octanal-M, heptanal-D, 3-methylbutanal-D, 3-methylbutanal-M, 3-Octanol, oct-1-en-3-ol, n-Hexanol, 5-methyl-2-Furanmethanol, pentan-1-ol-D, 2-heptanone-M, methyl isobutyl ketone-M, acetone, 2-Methyl-3-heptanone, 2-heptanone-D (Fig. 3 ①). These flavor compounds were mainly produced and enriched in the stages of frying and stewing. The contents of aldehydes (except n-valeraldehyde) increased obviously after frying. Among the alcohols, the contents of 3-octanol, 1-octene-3-ol, hexanol and 1-pentanol increased significantly after the end of frying, among them, 1-octen-3-ol was considered as one auto-oxidation product of unsaturated fatty acids (Xu et al., 2020), and the reaction was violent in the frying stage due to lipid oxidation and Strecker degradation reaction. Among ketones, 2-heptanone was mainly produced by the oxidation of linoleic acid, which can improve the flavor of meat and meat products to a certain extent (Domínguez et al., 2014).

The second category involved aldehydes, alcohols and acids, including hexanal-m, pentanal, pentan-1-ol-M and acetic acid, which

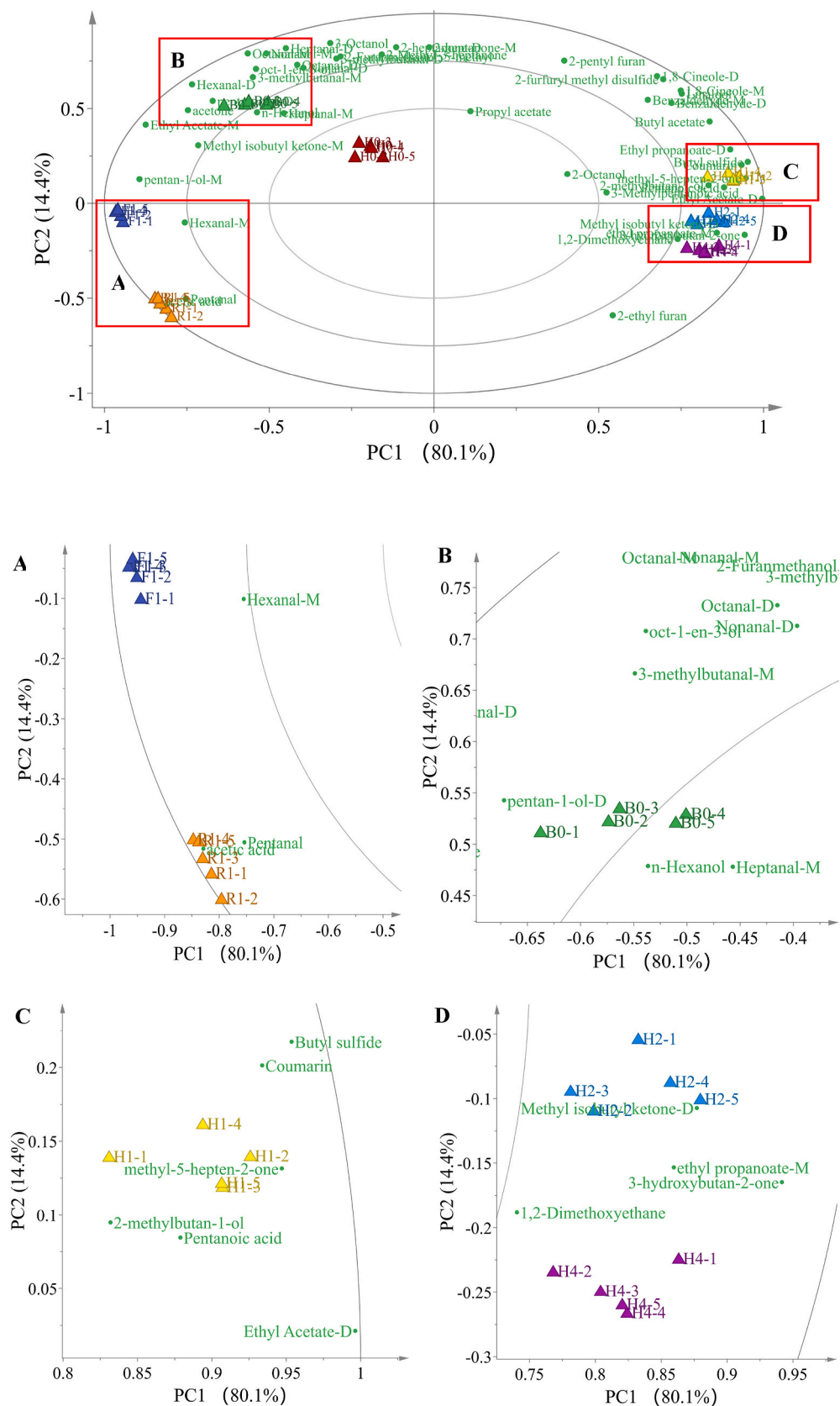


Fig. 5. Biplot of principal component analysis (PCA) and different variance of volatile compounds in Fulji roast chicken samples during processing and storage period. A, B, C and D partial magnification of biplot. R1: raw chicken or fresh; F1: end of frying; B0: end of stewing; H0: commercial sterilization; H1: storage for 1 month; H2: storage for 2 months; H4: storage for 4 months.

were the main volatile compounds in fresh chicken (Fig. 3 ②). The of these volatile compounds gradually decreased in the subsequent processing process. The third category mainly included ketones, acids, esters, ethers and heterocyclic compounds, which were enriched during storage, including methyl-5-hepten-2-one, 3-hydroxybutan-2-one, methyl isobutyl ketone-D, 5-methyl-2-Furanmethanol, 2-Octanol, 2-methylbutan-1-ol, pentanoic acid, 3-Methylpentanoic acid, ethyl propanoate-D, ethyl propanoate-M, ethyl Acetate-D, propyl acetate, butyl sulfide, 1,2-Dimethoxyethane, coumarin, 2-ethyl furan (Fig. 3 ③). Wang et al. (2020) found 2-ethyl-furan was identified as a key volatile flavor compound in Chinese fish sauce. It was very important for meat flavor. In the storage process after commercial sterilization, a series of complex reactions such as Maillard reaction occurred, which increased the content of these volatile substances. The fourth category mainly included benzaldehyde-M, benzaldehyde-D, 1,8-Cineole-M, 1,8-Cineole-D, Linalool, Butyl acetate, 2-furfuryl methyl disulfide (Fig. 3 ④). Bitter almond, flower and candy flavor become more intense with the increase of fourth category VOCs content (Odor description query reference <http://www.flavournet.org>).

In order to find out the main markers in different stages, the potential flavor compound markers in different processing stages were screened. These samples produced a total of 47 volatile flavor components. The effects of seven different stages on flavor compounds were determined by VIP method, and the potential labeled flavor components were screened (Fig. 4). When the VIP score of flavor components was equal to or higher than 1.0, it was used as a marker to distinguish the effects of different processing stages (Al-Dalali et al., 2022). High VIP score increased the possibility of compounds being identified. Fig. 4 summarized the important flavor compounds identified by PLS-DA, namely hexanal-D, nonanal-M, octanal-M, heptanal-D, acetone, 3-Octanol, ethyl Acetate-D, of which 7 markers distinguished the volatile substances effects of the sample from processing to storage. As expected, stewing and frying were the main stages of flavor production. Most of the selected markers belong to aldehydes and alcohols, indicating that these components came from lipid oxidation. In the storage process after commercial sterilization, the selected markers were represented by ethyl acetate.

Combined with biplot, this study attempted to determine the possible location of key flavor compounds and which processing stage had the greatest impact on the flavor characteristics of Fuliji roast chicken processing line. Therefore, due to the complexity of different volatiles, principal component analysis (PCA) was carried out for further analysis.

Principal component analysis can be used to directly reflect the differences of volatile substances in Fuliji roast chicken at different processing and storage stages (Fig. 5). It can be seen from Fig. 5 that the cumulative contribution rate of the first two principal components after dimensionality reduction was 94.5%, which can be better characterize the characteristics of the original data. Fig. 5 intuitively showed that the distance between parallel samples in the same processing stage was close, while the distance between samples in different processing stages was far. Fig. 5 directly reflected the similarity and difference of flavor in Fuliji roast chicken at different processing and storage stages. In Fig. 5, among the different samples from seven key processing points can be better resolved by reporting score plots combined with loading plots, since in such a way it's more easily display correlations between the 47 volatile compounds and the samples. Process stage H1 samples H1-1, H1-2, H1-3, H1-4, H1-5 were mainly scattered in the I quadrant. Process stage H0 and B1 were mainly scattered in the II quadrant. Process stage F1 and R1 were mainly scattered in the III quadrant. Process stage H2 and H4 were mainly scattered in the IV quadrant. The flavor substances of the five parallel samples at each stage were all in the same quadrant, with a wide distribution of flavor substances in the different stages, indicating that there were no significant differences in the volatile flavor components of Fuliji roast chicken at the same processing stage and significant differences at different processing and storage stages.

4. Conclusions

In this study, GC-IMS was used to analyze the changes of the VOCs in Fuliji roast chicken during seven stages of processing and storage. A total of 47 volatile substances were identified, including aldehydes, hydrocarbons, alcohols, ketones, esters, ethers and heterocyclic substances. It was found by fingerprint that frying and stewing were the key control stages of flavor formation, and sterilization and storage led to the loss of VOCs in Fuliji roast chicken. With the aid of heat map, VOCs were clustered into four categories, further identifying seven flavor markers in the evolution of the aroma profile in Fuliji roast chicken during processing and storage. In addition, the results obtained by the PCA method combined with chemometrics showed that the different stages of the samples were in relatively separate spaces and could be distinguished. GC-IMS offers the advantages of high sensitivity, no sample pretreatment, operability at atmospheric pressure and rapid detection for the analysis of flavor compounds in poultry products such as Fuliji roast chicken.

CRedit authorship contribution statement

Hui Zhou: Conceptualization, Methodology, Data curation, Formal analysis, Writing – review & editing. **Wei Cui:** Conceptualization, Supervision, Writing – original draft, Data curation. **Yafei Gao:** Investigation, Validation. **Ping Li:** Software, Supervision. **Xinyuan Pu:** Methodology, Writing – review & editing. **Ying Wang:** Conceptualization, Formal analysis. **Zhaoming Wang:** Conceptualization, Supervision, Project administration. **Bao cai Xu:** Validation, Project administration, Funding acquisition.

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.

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