


## Review Article

# Rapid and Visual Flavor Analysis Using Gas Chromatography-Ion Mobility Spectrometry (GC-IMS) in Meat Products: Research Progress and Future Trends

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The flavor profile of meat and its processed products is highly volatile and subject to significant changes during storage, processing, and transportation. It is therefore crucial to monitor the flavor of meat to evaluate sensory quality and protect consumer health and safety. Gas chromatography-ion mobility spectrometry (GC-IMS) has become increasingly popular due to its advantages of being nondestructive, rapid, and capable of trace detection. The analysis of volatile organic compounds (VOCs) using this technique is continuously applied in the assessment of food freshness, origin traceability, and adulteration detection. GC-IMS has emerged as a promising tool for accurate monitoring and characterization of VOCs in food, particularly in the field of meat flavor analysis. Its applications include meat product authentication, adulteration detection, processing and storage-related flavor changes, and freshness monitoring. This paper provides a comprehensive overview of the working principle of GC-IMS and its applications in meat flavor analysis, while exploring future trends and potential limitations associated with the technique.

## 1. Introduction

Abundant in nutrients and offering a distinctive flavor, meat and its various processed products play an essential role in providing the required energy for individuals' daily activities [1]. They are not merely a part of the human diet but also an indispensable and vital aspect of human life. The meat industry encompasses a diverse range of meats and their corresponding processed products, all of which undergo various processing techniques. The specific storage conditions coupled with intricate processing techniques can result in pronounced differences in the quality and price of meat products. Issues related to meat quality and safety, including adulteration, altered flavors, and loss of freshness, can deceive consumers and pose risks to human health and well-

being. As a result, ensuring the safety and quality of meat products is of utmost importance. Consequently, investigating the flavor components in meat products serves as a foundation for more in-depth research. Instrumental analysis, a flavor analysis technique built upon sensory analysis, offers benefits such as objective evaluation criteria and more precise conclusions.

Gas chromatography-ion mobility spectrometry (GC-IMS) technology melds the high separation power of gas chromatography with the swift response time of ion mobility spectrometry, thereby introducing a unique approach for gas-phase ion detection [2]. This technique is employed to identify trace quantities of volatile and semivolatile organic compounds (at parts per billion volume levels) across a range of matrices [3]. By combining the two techniques,

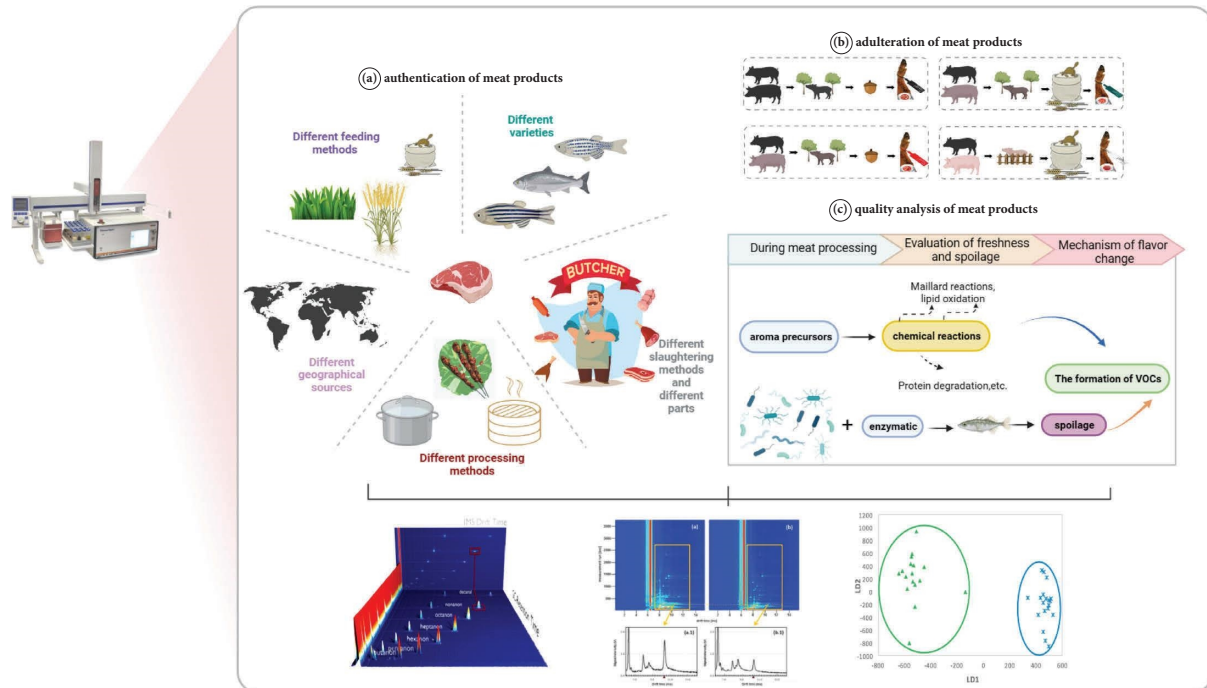


FIGURE 1: The applications of GC-IMS in the analysis of meat and meat products.

enhanced selectivity and peak capacity can be attained, given that the high degree of orthogonality between GC and IMS bolsters their efficacy [4]. In recent years, due to continuous technological advancements, GC-IMS has expanded beyond its traditional military applications, which include chemical agent detection, explosive detection, and drug detection. It has also been extensively applied in areas such as food quality analysis [5]. Specifically, IMS is already playing a crucial role in the fields of proteomics, lipidomics, and metabolomics [4, 6]. Indeed, several studies have explored the potential of GC-IMS technology in various aspects of food analysis. This includes food traceability, adulteration detection, and quality and safety monitoring of various food products [7, 8] (Figure 1). However, there is a discernible shortage of comprehensive, professional, and systematic reviews about GC-IMS, particularly in relation to the sensory characteristics of meat products. This review aims to provide a comprehensive overview of the principles of GC-IMS, flavor analysis techniques, and the technology's benefits and drawbacks concerning flavor analysis for meats and processed products. Such insights could be crucial for assessing meat freshness, detecting adulteration, or tracing meat products origins, thereby ensuring higher standards and greater transparency in relation to meat quality.

## 2. Principle of IMS and GC-IMS

Ion Mobility Spectrometry (IMS) is a technique utilized for the characterization and analysis of substances based on the specific velocities of gaseous ions derived from a substance when subjected to an electric field and a supportive gas atmosphere. When coupled with other techniques like capillary or multicapillary columns, gas chromatography

(GC), and liquid chromatography (LC), IMS can provide enhanced selectivity of the analyte, thus improving the accuracy and scrutiny of results. This combination of techniques leverages the distinct advantages of each method, resulting in a more precise and comprehensive analysis of the substance being investigated [9].

The drift tube serves as the core component of IMS instruments, with its fundamental structure depicted in Figure 2. During the heating and vaporization process, carrier gas transports sample molecules to the ionization zone. Here, the sample molecules undergo radiation from the  $^{63}\text{Ni}-\beta$  ionization source, leading to the formation of various product ions [10]. The ionization procedure is essentially propelled by proton or electron transfer reactions. These reactions occur following collisions between neutral sample molecules and reactant ions. This results in the accumulation of ions at the forefront of the ion gate, which is closely linked to the ion gate grids [11]. Under the action of the electric field, the analyte ions migrate to the right end against the reverse drift gas [12]. The drift time of ions in the electric field is related to the mobility of ions, and the calculation formula of the drift velocity is as follows:

$$\text{Drift velocity: } \mathbf{v}_d = K E, \text{ Mobility: } K = \frac{d}{t_d E}, \quad (1)$$

where  $E$  is the electric field strength,  $d$  is the drift path length, and  $t_d$  is the drift time.

IMS enables the detection of complex samples even at atmospheric pressure [13]. Furthermore, it facilitates the resolution of complex matrices, thereby enhancing the analytical capabilities of these techniques [14]. The qualitative analysis of complex samples using GC-IMS requires consideration of multiple factors, such as the characteristics

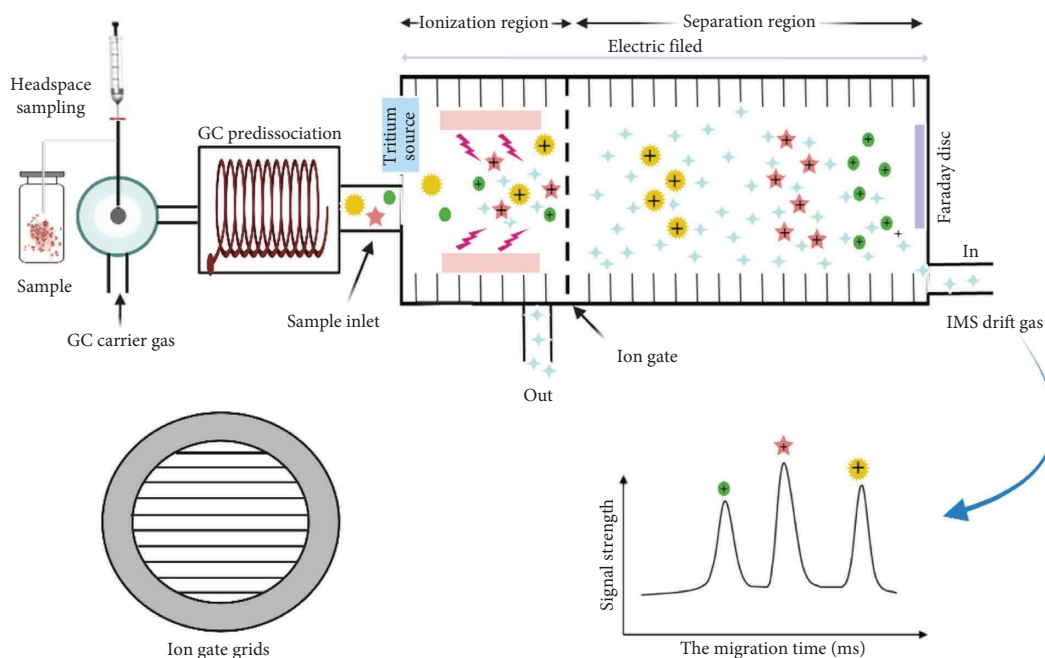


FIGURE 2: The working principal diagram of GC-IMS.

of product ions, the potential for overlapping peaks, and the limitations of existing databases [9]. In recent years, GC-IMS has gained popularity due to its nondestructive, rapid, and high-sensitivity detection capabilities, rendering it an indispensable technique for flavor profiling. The application of GC-IMS in monitoring the flavor of meat products is illustrated in Figure 3 and summarized in Table 1.

The huge dataset of GC-IMS requires accurate chemometric processing. The Laboratory Analytical Viewer (LAV) software is used to process the IMS data, with the integration region being utilized to extract information on signal intensity, retention, and drift time of characteristic substances, which are then converted to .csv format for further analysis. The identification of the substances was completed with the calibration of the standards in the GC-IMS library. In subsequent chemometric analysis of the data, supervised discriminant analysis methods such as Partial Least Squares-Discriminant Analysis (PLS-DA) and Orthogonal Partial Least Squares-Discriminant Analysis (OPLS-DA) can prevent overfitting of data caused by unsupervised discriminant analysis methods. The PLS-DA model maximizes the differences between groups based on preclassification, resulting in better separation than principal component analysis (PCA). OPLS-DA, on the other hand, can separate irrelevant information from the original matrix to maximize differences between groups and is typically used to identify substances that differ between groups. Before analysis, data must undergo background noise subtraction to prevent deviations in the original data and may require normalization, if necessary. Typically, 80% of samples are used for training, with the remaining 20% used for validation in chemometric models. Establishing a postclassification model for VOC in real samples can aid in the rapid identification of samples. Analyzing and categorizing collected sample data,

and then utilizing these classification results to identify new real samples, constitute a postclassification model. This approach assists in the rapid identification of volatile organic compound samples by leveraging previously known sample classification information, thereby facilitating quicker categorization and identification of new samples.

### 3. Application of GC-IMS in the Authentication and Adulteration of Meat Products

**3.1. Authentication of Meat Products.** The VOCs in meat can be influenced by factors such as diet, breed, geographical location, and processing techniques. Specific VOCs detected by GC-IMS have been extensively utilized for the identification and authentication of meat products from the same breed, serving as potential markers to differentiate various growth environments and processing methods.

To address the gap in flavor classification for different puffer fish species, the researchers have developed an accurate classification method which utilized the combination of GC-IMS and intelligent sensory systems, such as electronic nose and electronic tongue, to distinguish various breeds of cultured puffer fish based on their unique flavor profiles [15]. By matching the dimer or trimer signal of the compound with the retention index and drift time in the GC-IMS library, the target compounds can be determined. However, due to the large and complex nature of the GC-IMS dataset, PCA was then generally applied to reduce the dimensionality of the data and classify it, which helps to identify the main factors or components that explain the most significant variance in the dataset. This technique provides important insights into the flavor differentiation between puffer fish species and can contribute to the development of quality control standards in the fishing and

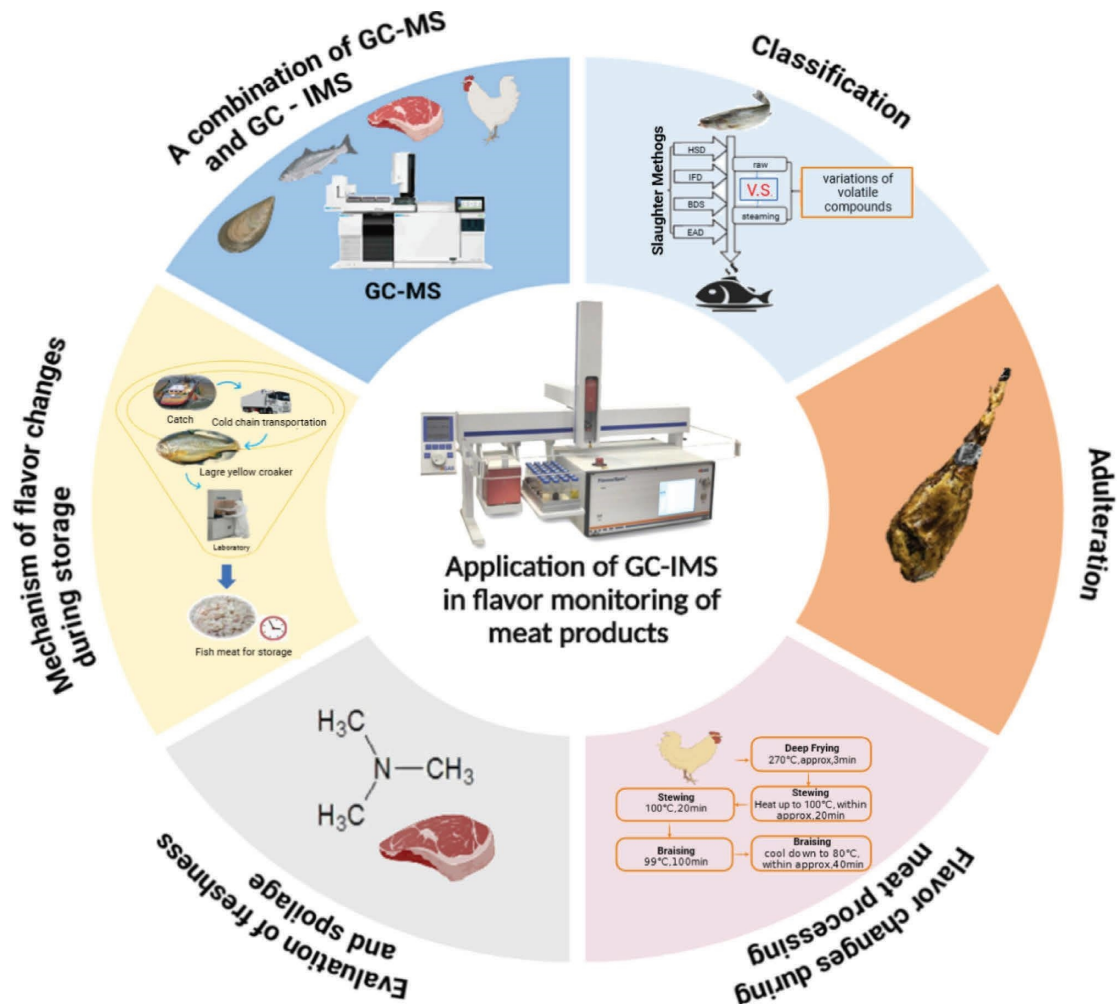


FIGURE 3: Application of GC-IMS in flavor monitoring of meat products.

restaurant industry. In a study [18], a similar approach was used to distinguish and characterize the flavor of different varieties of donkey meat using GC-IMS combined with multivariate analysis. The researchers first obtained the volatile compound profiles of multiple samples of donkey meat using GC-IMS and then applied multivariate analysis techniques such as PCA to analyze the VOCs of the samples. In recent years, researchers have been actively exploring appropriate slaughtering techniques by GC-IMS to maximize the retention of the excellent flavor of the meat. This is an important area of research as the slaughtering process can significantly influence the flavor profile of meat. By using GC-IMS, researchers can analyze the VOCs present in meat samples obtained from different slaughter methods. This allows them to identify and quantify the compounds that contribute to the desirable flavors, such as Maillard reaction products, lipid oxidation products, and specific aroma compounds. Previous study indicated that the slaughtering process is closely linked to changes in meat pH, which in turn is a key factor influencing the Maillard reaction [38]. However, the evaluation of the effects requires accurate technical certification. Wang et al. [16] employed a combination of GC-IMS and electronic nose technology to

establish VOC fingerprints, enabling qualitative analysis of VOCs in sea bass through the NIST and IMS databases of GC-IMS. Combined with the heat map, the differences of VOC changes caused by different slaughtering treatments were clearly visualized. This study aims to determine the optimal slaughtering method for preserving the flavor of sea bass, thus providing a theoretical foundation for further exploration in this area. Wang et al. [17] used the GC-IMS technology to establish the flavor profiles of Jingyuan lambs at different ages. They utilized a differential comparison model of GC-IMS 2D spectra to analyze the changes in flavor. This model involved taking a specific group of samples as a reference for all volatile compounds and then quantifying the changes in flavor by subtracting the color of the reference point. The colors on the map represented the differences, with white indicating no difference, red indicating values above the reference point, and blue indicating values below the reference point. To provide a more accurate description of substances that could not be precisely determined in the topographic map, the authors combined the differential comparison model with fingerprinting. They constructed a PCA model based on the differences in signal intensity of VOCs in lambs of different ages, successfully

TABLE 1: Applications of GC-IMS in meat flavor analysis.

Functions	Application sample	Conclusion	References
Authentication	Pufferfish	Identify different breeds of cultured pufferfish	[15]
	Sea bass	Changes of VOCs in sea bass under different slaughtering treatments	[16]
	Jingyuan lambs	The flavor profiles of Jingyuan lambs at different ages were established	[17]
	Donkey meat	Distinguish and characterize the flavor of different kinds of donkey meat	[18]
	Squid	The obvious differences of specific flavor compounds of squid with different processing methods were clarified	[19]
Adulteration detection	Salmon	Distinguish salmon from different origins	[20]
	Hams	The combination of GC-IMS and electronic nose distinguishes three grades of ham	[21]
	Pork samples	Distinguish different feeding conditions of pigs	[22]
Flavor analysis during meat processing	Ham samples	Identification of adulterated ham with different feeding conditions	[5]
	Iberian hams	Effectively categorize Iberian ham samples into different quality grades based on the feeding mode of pigs	[23]
	Fish fillets	Determine the flavor characteristics of different frying conditions	[24]
	Sturgeon	Detecting changes in flavor substances in raw and cooked sturgeon meat	[25]
	Crayfish	Effectively identified the flavor substances in the enzymatic hydrolysate	[26]
	Salted duck	Distinguish the change of characteristic flavor between different thermal treatment temperatures and different aging time salted duck	[27, 28]
	Grass carp surimi	Screening out the best washing method for the flavor of grass carp surimi	[29]
	Dezhou braised chicken	Identified the flavor substances in chicken meat during stewing and processing	[30]
	Bacon	Analysis of the changes of aroma components of bacon in four processing stages	[31]
	Pigeon	Exploring the influence of marinating and frying on the flavor of braised pigeon	[32]
Evaluation of freshness and spoilage	Chicken	Detecting the composition of flavor substances in Piao chicken boiling, frying, roasting, and cooking	
	Dry-cured pork belly	Obtain the volatile fingerprint results of all processing phases of traditional dry-cured pork belly	[33]
Evaluation of freshness and spoilage	Fish	GC-IMS can evaluate the degradation of fish caused by spoilage microorganisms	[34, 35]
	Meat products	Monitoring volatile biogenic amines in headspace vapor of muscle food by GC-IMS can reflect the deterioration degree	[36, 37]



distinguishing samples from different months. By using positive ketone C4–C9 as an external standard, the retention index was calculated, and the drift time of the retention index and the standard in the GC-IMS library were compared to identify the volatile compounds; the authors were able to identify 66 VOCs and obtain information about their characteristic features and peak intensity. This approach provided a new and effective method for exploring the optimal flavor profile in Jingyuan lamb.

Cooking methods and processing temperatures play a significant role in the flavor development of meat products. Several factors, including moisture content, pH, and chemical reactions, are influenced by these cooking techniques and temperatures, ultimately affecting the formation and release of volatile aroma compounds. It is essential to understand that even for the same meat species, the flavor composition can vary greatly with different cooking methods. Selecting the appropriate cooking technique can help to maximize the desired flavor characteristics of meat products. The impact of processing methods on squid flavor remains uncertain; therefore, several studies have utilized the GC-IMS technique to analyze the flavor compounds in boiled, steamed, and sous vide-treated squid samples. The results of GC-IMS analysis reveal that squid samples processed by sous vide exhibit a more extensive topographic map signal distribution compared to other processing groups, with higher signal strength. Furthermore, heat map cluster analysis demonstrated that the sous vide-treated samples possess distinct volatile components not found in other samples. Among all treatment groups, ester composition is found to be the highest while aldehyde content is relatively lower, contributing to the ideal flavor profile of the squid [19]. The flavor profile of fish is known to be influenced by various factors, including its country of origin. One interesting method that has shown promise in this area is GC-IMS. This technique is capable of classifying fish with complex origins and even those with indistinguishable characteristics. VOCs could serve as potential markers for differentiating salmon from various origins. Interestingly, the study found a certain degree of regularity in the VOCs among salmon from different regions [20]. Salmon samples can be categorized based on their VOC signal intensity, with some flavors such as 2-butanone, pentanone, and (E)-2-hexenal displaying higher signal intensities in certain samples while being barely detectable in others, which may be attributed to the influence of environmental salinity. The research by Qian et al. [21] described the use of a portable electronic nose coupled with GC-IMS for the detection of VOCs, which is capable of differentiating among three grades of hams based on the GC-IMS analysis results of VOCs in ham. The study identified the main flavor substances and selected a suitable electronic nose sensor array accordingly. The GC-IMS technique revealed variations in volatile compound levels among different types of ham, resulting in different response curves from the electronic nose. This approach can be useful in effectively differentiating between various grades of ham. Additionally, the study conducted by Li et al. [39] emphasized the impact of geographical origin on the flavor profile of hams, specifically

highlighting the application of GC-IMS in distinguishing dry-cured hams from various regions. These hams are characterized by their smoky aroma attributed to phenolic compounds.

*3.2. Adulteration Detection of Meat Products.* High-value or expensive meat products in the market are susceptible to adulteration or false advertising. Chemical analysis can be used to verify the quality and origin of such products, but there is a need for fast and reliable analytical methods. Therefore, to guarantee the quality and safety of all types of meat, it is imperative to develop a fast, sensitive, and accurate meat adulteration detection system. GC-IMS, as a novel and rapid detection method, has the potential to accurately identify the VOCs of meat products, making it a promising choice.

GC-IMS has already been extensively utilized for the rapid differentiation of Iberian pigs, as illustrated in Figure 4. The pork quality of Iberian pigs varies significantly due to differences in their feeding and rearing methods. Purebred Iberian pigs are raised in a free-range environment and feed exclusively on acorns. The production of Iberian ham, a dry-cured delicacy made from the meat of these pigs, holds significant economic importance in Spain. There are four grades of Iberian ham, each distinguished by a different colored label. The highest grade ham, indicated by a black label, is made exclusively from purebred Iberian pigs. Due to the unique flavor profile and significant commercial value of Iberian ham, it is unfortunately common for hams fed with other ingredients to be inaccurately labeled as acorn-fed ham, creating consumer fraud. Different feeding methods result in varying fatty acid compositions of pork. Pork from free-range pigs exhibits a higher proportion of unsaturated fatty acids compared to that from confined pigs. It is challenging to distinguish captive pigs fed with high unsaturated fat feed for sale as grazing Iberian pigs. The utility of IMS in detecting VOCs in fat samples for determining Iberian pig feeding regimes has been demonstrated. Pork samples were automatically introduced and detected by IMS. Different feeding conditions of pigs could be distinguished based on the IMS fingerprint spectrum of fat samples, and PCA analysis was used to classify unknown samples, demonstrating the potential of IMS in detecting adulteration in meat products [22].

By comparing the GC-IMS spectral signals of two distinct ham samples, differentiation between the hams was achieved through two methods. The first method utilized the GC-IMS fingerprint and a data matrix was constructed. However, this approach is based on all signals which results in slow data processing, detailed preprocessing, and a large amount of calculation. The second approach involves selecting significant individual signals or markers from the spectral data and verifying the identified compounds by comparing their retention and drift times to actual standards in the GC-IMS database. Remarkably, the spectral signals of the acorn-fed ham samples exhibit a higher abundance compared to those of the forage-fed ham samples. The resolution of 100% was achieved through selective analysis

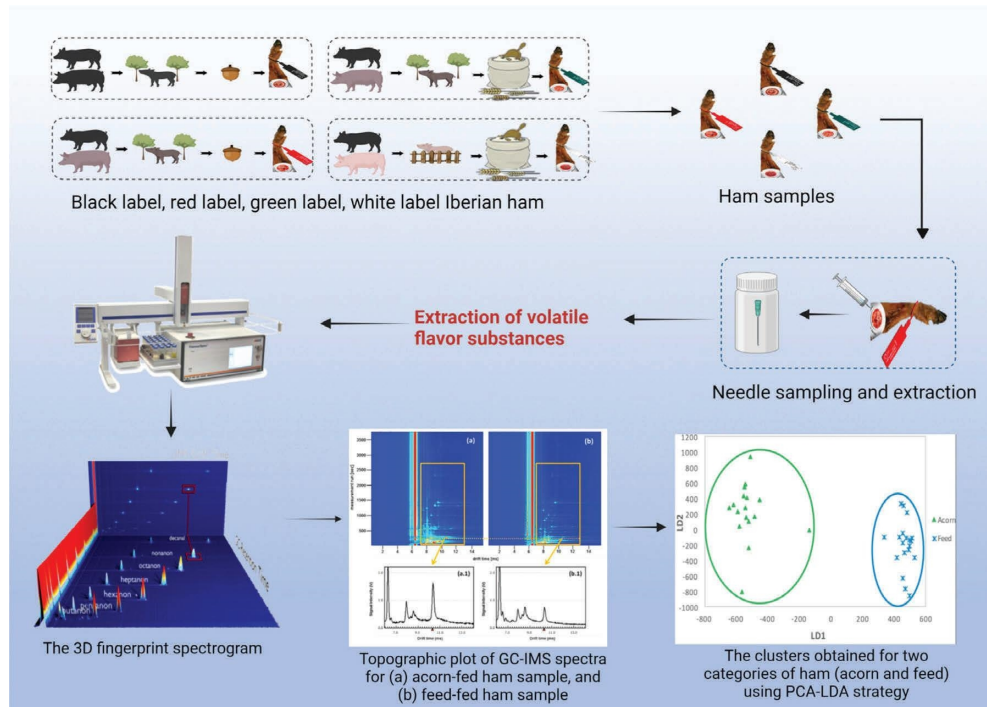


FIGURE 4: Identification of adulterated Iberian ham using GC-IMS. Note: GC-IMS 2D fingerprint and PCA-LDA plot are derived from Arroyo-Manzanares et al. [5].

of significant signals or markers present in the spectrum. This specific tagging method is faster and more efficient. Additionally, the experimental findings indicate that the GC nonpolar column offers a shorter analysis time compared to its polar counterpart, rendering it a superior option [5]. This study provides a great reference for the field of GC-IMS adulteration authentication. The targeted analysis of the spectrum is sufficient to obtain higher resolution and achieve the perfect classification of the sample.

Martín-Gómez et al. [40] have recently proposed a novel nondestructive sampling method that enables the determination of authenticity and quality of acorn-fed Iberian ham during curing, by impregnating pork fat samples with aseptic disposable needles and classifying Iberian ham without compromising its integrity. The VOCs can be obtained through headspace extraction (HS) by directly heating the needle and determined via GC coupled with different detectors, such as MS or IMS. This demonstrates the potential of GC-IMS in preventing label fraud. Martín-Gómez et al. evaluated the potential of two methods, GC-IMS and GC-MS, to distinguish defective Iberian hams [41]. Fifty hams from acorn-fed pigs were nondestructively sampled and classified as defective or nondefective. GC-IMS, with its higher sensitivity and trace analysis capabilities, detects more characteristic compounds in comparison to GC-MS chromatograms. GC-MS achieved more rapid identification of ham by labeling characteristic compounds, whereas GC-IMS did not exhibit significant differentiation among individual compounds. However, a single VOC component may not be sufficient to distinguish all samples, and only formic acid can serve as an

indicator of defective ham. The remaining compounds are present in small quantities due to the unique feeding method of Iberian pigs, making it challenging to identify them through analysis of a single VOC. GC-IMS and GC-MS employ partial least squares discriminant analysis to achieve verification classification rates of 80% and 100%, respectively, demonstrating the potential of these instrumental methods as a complementary approach to traditional olfactory techniques. In the study conducted by Segura-Borrego et al. [42], a nondestructive sampling method was utilized in conjunction with GC-MS and GC-IMS to compare the separation potential of the two detectors, aiming to provide a more efficient and convenient approach for identifying the feeding mode of Iberian pigs. Generally, GC-IMS offers superior sensitivity and speed compared to HS-GC-MS in detecting a broader range of VOCs. This can be advantageous in quickly screening and identifying the VOCs emitted from samples and quantitative reference can be provided by resolution, sensitivity, selectivity, response time, reproducibility, and other indicators. On the other hand, HS-GC-MS is known for providing more precise qualitative results in the analysis of VOCs in Iberian ham. This is due to the availability of comprehensive GC-MS databases and the ability to integrate single peak targeting analysis methods. By utilizing these databases and targeting specific compounds, GC-MS analysis can offer detailed and accurate identification of individual volatile compounds in the samples. However, GC-IMS is not suitable for targeted analysis of single compounds, as it lacks comprehensive standard libraries and may exhibit multiple characteristic peaks for a single

compound. In conclusion, GC-IMS can provide a broad overview of the VOC patterns emitted by the samples, allowing for efficient screening and differentiation between different feeding modes. On the other hand, HS-GC-MS can be used for targeted analysis of specific compounds when more detailed and accurate identification is required. By combining these two techniques, researchers can benefit from the strengths of both methods. GC-IMS can quickly detect and compare overall VOC patterns, providing valuable insights into the differences between feeding modes. HS-GC-MS can then be employed to target specific compounds of interest, offering more precise qualitative analysis.

Although GC-IMS is widely utilized in food traceability and fraud prevention, its vast dataset presents numerous challenges such as nonlinearity and peak overlap, which hinder the accurate acquisition of required information from samples. For this reason, a comprehensive GC-IMS signal preprocessing pipeline is proposed, along with the introduction of a GC-IMS feature data extraction method that enhances both data quality and subsequent performance of peak detection and clustering algorithms. The signal preprocessing pipeline effectively categorizes Iberian ham samples into different quality grades based on the feeding mode of pigs and accurately distinguishes between the two types of ham. In addition, cases such as donkey meat adulterated with horse meat and mutton adulterated with pork can be identified as adulteration based on GC-IMS in combination with chemometric methods. This demonstrates the technical feasibility of utilizing this method for quality control and fraud detection purposes [23]. Despite the challenges, GC-IMS remains a valuable tool in food traceability and fraud prevention. Its ability to rapidly and accurately identify the composition of food samples is essential for ensuring the authenticity and safety of food products. As technology continues to advance, these challenges are expected to be further addressed, making GC-IMS an even more powerful tool in the future.

#### **4. Application of GC-IMS in the Quality Analysis of Meat Products**

*4.1. Flavor Changes during Meat Processing.* The treatment processes involved in meat processing, such as dry-curing, brining, and boiling, have a profound effect on the composition and quantity of volatile compounds. These volatile compounds play a crucial role in creating the characteristic aroma and flavor profiles of meat products. During these treatment processes, various chemical reactions occur among aroma precursors present in the meat. These reactions can include Maillard reactions, lipid oxidation, and degradation of proteins, among others. These reactions lead to the formation of a diverse range of volatile aroma compounds, which contribute to the unique taste and aroma of different meat products. Different processing methods yield distinct taste and flavor profiles for meat products. GC-IMS is widely employed in flavor analysis during meat processing [24–26, 43].

The Maillard reaction, also known as the carbonyl-amine reaction, is a crucial method for generating flavor in food through heating. Heat treatment can significantly impact the extent of the Maillard reaction. Chen et al. [43] conducted a study where they heated fish scale gelatin hydrolysate (FSGH) with sugar to prepare Maillard reaction products (MRPs). The changes in volatile components during the heating process were evaluated using GC-IMS. The results of GC-IMS showed that the types and quantity of VOCs produced by ribose-FSGH MRPs increased after heating, and the production of some heterocyclic compounds also increased their antioxidant activity, whereas before the Maillard reaction, only 15 flavor substances had been detected. According to the study conducted by Chang et al. [24], using fish fillets fried at a temperature of 140°C as a reference and employing a difference-in-difference comparative model, it was discovered that different processing conditions were closely associated with variations in the VOC content. Factors such as thermal degradation of polyunsaturated lipids due to high temperature and the Maillard reaction were identified as important influencers impacting the flavor of fish fillets. Specifically, at a frying temperature of 160°C, the VOC content was found to be higher compared to the other groups, resulting in the fish fillets possessing a deep-fried aroma. However, it was concluded that higher temperatures and longer frying times led to detrimental effects on the VOCs, thereby negatively affecting the flavor of the fish fillets. An interesting result was also found in the study of Li et al. [25]. During the process of detecting changes in flavor substances in raw and cooked sturgeon meat using HS-SPME-GC-MS-O and GC-IMS, it was observed that with few exceptions, the compounds detected on the gallery plot were markedly different from those identified by GC-MS. The significant variance may be attributed to the differences in separation and extraction principles between solid-phase microextraction and headspace injection, as well as variations in detection methods. Additionally, GC-IMS detected the most alcohols because of its high sensitivity to substances with strong proton affinity. The number of volatiles detected by both detection methods was almost the same. The results of this study showed that GC-IMS was able to identify more novel compounds than GC-MS and that the PSL-DA model in conjunction gave a more thorough depiction of the variations in sturgeon meat flavor caused by various processing methods. This study combined the advantages of GC-MS and GC-IMS to restore the relatively complete flavor library of sturgeon meat to the greatest extent. In addition to the above treatment process, in the process of improving the flavor of crayfish by enzymatic hydrolysis, GC-IMS effectively identified the flavor substances in the enzymatic hydrolysate [26].

In addition to aquatic products, GC-IMS is also used to evaluate the flavor changes of livestock and poultry meat during processing. The study conducted by Xie et al. [27] demonstrated the effectiveness of GC-IMS in evaluating the flavor changes of duck meat at different thermal treatment temperatures. The study found that low-temperature heat treatment had little effect on the flavor of duck meat.



Additionally, the aging process can have a significant impact on the composition of VOCs in meat products. GC-IMS can be used to monitor these changes and provide valuable insight into the overall flavor profile of meat products during aging. Some scholars established GC-IMS flavor fingerprints of water-boiled salted ducks at different aging times to differentiate the changes in characteristic flavor compounds and determine the optimal aging time for water-boiled salted ducks. 47 volatile aroma compounds were identified through GC-IMS; 1-octen-3-ol, *n*-nonanal, 2-heptanone, isopropyl acetate, and ethyl propionate are the main volatiles produced during the aging process, which enrich the flavor characteristics of water-boiled salted ducks. The combination of GC-IMS analysis and electronic tongue demonstrates that a 24-hour maturation period is more appropriate for practical production [28]. Similarly, based on GC-IMS, Xu et al. [44] pointed out that soft-boiled chicken matured for 2 hours had the highest overall score, as well as the best flavor, sensory score, and umami score. In contrast, the 1-hour sample exhibited a more pronounced bitter taste characteristic. There was no significant difference in sensory scores between samples aged for 8 hours.

The GC-IMS technique is useful not only for evaluating flavor changes in food products but also for assisting food producers in identifying the optimal process for food handling and processing. By comparing the flavor profiles after different treatment processes, GC-IMS can help maximize the retention of the most desirable flavors in meat products. This makes it a valuable tool in food processing applications, where producers are looking to achieve the best possible flavor in their products. In the deodorization process of grass carp surimi, it is crucial to determine the optimal rinsing method to achieve high-quality processing outcomes. To tackle this challenge, Xiao et al. [29] employed a combination of multiple sensory techniques, namely, GC-IMS, GC-MS, and E-nose, to screen for the most favorable washing methods that enhance the flavor profile of grass carp surimi. The GC-IMS method was able to distinguish the volatile compounds present in raw grass carp and those treated with different washing methods. Additionally, the combined use of GC-MS and GC-IMS provided a more comprehensive aroma profile, allowing for a more in-depth analysis of the flavor characteristics. The utilization of GC-IMS in analyzing the characteristic flavors of meat products offers several advantages in understanding the mechanisms underlying flavor changes during meat processing. As meat products undergo processing, there are alterations in precursor substances and VOCs. GC-IMS provides a powerful analytical technique to identify and measure these changes in flavor compounds, allowing researchers to gain insights into the chemical transformations that occur during meat processing. This information is crucial for understanding how different processing methods or conditions affect the flavor profile of meat products and can aid in optimizing processing parameters to achieve desired flavor outcomes. Therefore, GC-IMS can help in enhancing our understanding of the flavor development and changes that occur during the processing of meat products. To investigate the emergence of key flavor compounds and

examine the impact of various processing methods on flavor, GC-IMS technology was utilized to track characteristic flavor compounds throughout the seven critical stages of Dezhou braised chicken production. GC-IMS was successfully employed to identify the flavor compounds in chicken meat during stewing and processing. Among various processing stages, stewing and seasoning are crucial for the development of Dezhou grilled chicken's unique flavor profile [30]. GC-IMS was employed for both qualitative and quantitative analysis of the flavor compounds in Wuding chicken. The results showed that the addition of spices increased the type and content of volatile flavor precursors in chicken samples, with 1-pentanol, acetone, 2-butanone, valeraldehyde, and heptanal being the most abundant. These compounds are beneficial to enhancing the taste and quality of Wuding chicken. In comparison to GC-IMS, GC-MS was able to detect a total of 60 flavor components, which is 39 more than what was detected by GC-IMS. This could be attributed to the fact that GC-IMS is still an emerging technology for flavor analysis and detection, lacking a relatively comprehensive database. However, it does provide more intuitive differentiation between groups [45].

A study analyzed the changes in aroma components of Zhenba bacon during four processing stages. GC-IMS and GC-MS detected 44 and 40 VOCs, respectively. The VOCs produced by lipid oxidation in bacon are the most abundant during the curing and smoking process. During the curing process, there is an increase in both lipid content and quantity. Additionally, during smoking, phenols are produced in large quantities which promote the formation of flavor compounds in Zhenba bacon [31]. During the investigation into the impact of marination and frying on the flavor profile of braised pigeon, GC-IMS was able to detect certain unique VOCs such as pentanal, methional, and ethyl acetate that were not detected by GC-MS [32]. In the three Chinese cooking methods of Piao chicken, namely, boiling, frying, and roasting, GC-MS and GC-IMS analysis revealed that the VOCs in chicken samples were similar after each method but with varying concentrations. The majority of these substances were found to be volatile [46]. Aheto et al. [33] employed GC-MS and GC-IMS techniques to acquire the volatile fingerprint outcomes for all processing stages of traditional dry-cured pork belly. In this process, GC-MS was utilized to provide the names and proportions of volatile components, while GC-IMS was employed to visualize topographic maps of individual volatile components generated during processing for analyzing differences between test samples. The GC-IMS-generated topographic map successfully distinguished pork samples from fresh, cured, and commercial ones.

The formation of aldehydes in meat processing mainly depends on lipid oxidation and Maillard reaction, and aldehydes are important intermediates in the two reactions. Unsaturated aldehydes are the main volatile flavor compounds formed. Aldehydes with low perception threshold, high content, and strong volatility, such as hexanal and nonanal, are the main VOCs in meat processing. However, the concentration of aldehydes increases significantly after meat products are processed and matured, possibly due to

enhanced lipid oxidation and decomposition [47]. For example, when the concentration of hexanal is high, the food will emit a fatty acid-like odor [48]. Therefore, hexanal can be used for monitoring meat spoilage [49]. Hexaldehyde and heptanal are mainly derived from the oxidation of linoleic acid and arachidonic acid, while octanal and nonanal are mainly produced by the oxidation of oleic acid [50]. Aldehydes can also be derived from carbonyl groups generated by protein oxidation. Benzaldehyde is a Strecker aldehyde that is derived from the degradation products of phenylalanine through the Strecker degradation reaction, but the production of benzaldehyde also contributes to the off-flavor of meat products [51]. The threshold for ketones, also as lipid oxidation products, is much higher than for aldehydes. Ketones and aldehydes can react with amino compounds as carbonyl compounds during processing and storage of meat products. Ketones play a coordinating role in the overall flavor of meat products and have a positive effect on the volatile flavor of meat products [52]. And some aliphatic ketones are produced in the cooking process of meat products, for example, dimethyl ketone is the main contributor to the flavor of meat products [53]. Alcohol is produced by thermal oxidation brought by cooking, and lipoxygenase and peroxidase in polyunsaturated fatty acids degrade linoleic acid in meat tissues, most of which have pleasant odors such as sweetness and fruity, which can increase the volatile flavor of meat products [54]. Previous study demonstrated that the level of 1-octene-3-ol, a degradation product of linoleic acid or arachidonic acid, increased significantly after meat cooking [55]. Generally, the concentration change of 1-octen-3-ol reflects the degree of rancidification of meat products and provides mushroom-like aroma to meat products [56]. During the fermentation process of meat products, a special alcohol, linalool, is produced. It comes from spices added during the fermentation process of meat products [57]. It has distinct floral and citrus aromas and can be identified in high concentrations, making a significant contribution to the flavor of meat products. Alcohols can also be reduced and coagulated with acids to produce aldehydes and esters, respectively, reducing the concentration of alcohols, for example, during drying. Esters are produced through esterification reactions between fats or proteins and alcohols and by transesterification of triglycerides with fatty acids in ethanol. Esters formed in stews may originate from esterification of alcohols and carboxylic acids [58]. Furan is the product of Maillard reaction, which has meat aroma, can be formed at lower temperature and shorter time, and gradually increases with the increase of oxidation number [52].

Taken together, processing factors play a crucial role in unraveling the mechanism of aroma formation in meat products. The application of GC-IMS technology enables rapid detection and identification of high-resolution fingerprint maps during meat processing.

**4.2. Evaluation of Freshness and Spoilage of Meat Products.** GC-IMS is a reliable tool for detecting microbial spoilage in fish. The decomposition of nutrients and the

metabolic activity of spoilage bacteria can lead to the production of VOCs. In a study conducted by Jia et al. [34] to evaluate the spoilage potential of dominant bacteria in spoiled silver carp, GC-IMS was employed to detect VOCs present in chill-stored fish meat inoculated/noninoculated with spoilage bacteria. In this research, 25 VOCs were detected, mainly composed of alcohols, aldehydes, ketones, esters, and sulfur compounds. By analyzing the distribution intensity of these VOCs, the dynamic change process of the compound can be observed, thereby revealing the spoilage characteristics of silver carp and facilitating a deeper understanding of the spoilage process. Likewise, Li et al. [35] assessed the accumulation of VOCs in sea bream meat inoculated/noninoculated with spoilage bacteria (*Pseudomonas versuta*, *Shewanella putrefaciens*, and *Aeromonas sobria*) during cold storage. The signal intensities of 1-propanol, 1-pentanol, 1-hexanol, 3-octanol, 3-methylbutanol, and hexanal in the three inoculation groups were significantly higher than those in the noninoculated group. Also, different concentrations of *Salmonella typhi* in pork samples can be distinguished based on the varying intensities of VOCs. The combination of GC-IMS and electronic nose technology successfully detected, identified, and quantified VOCs with different bacterial concentrations in pork samples. The signal and color intensities of samples with varying levels of pollution exhibit significant differences, which can be effectively utilized for the identification of food-borne pathogenic bacterial contamination in meat products [59].

Foodstuffs are susceptible to enzymatic and microbial degradation during preparation or storage, leading to the formation of volatile compounds, particularly through decarboxylation reactions that break down amino acids into biogenic amines. Since amines exhibit favorable ionization chemistry and possess characteristic migration spectra in atmospheric pressure ionization sources, the monitoring of volatile biogenic amines present in the headspace vapor of muscle foods can serve as an indicator for assessing the degree of deterioration. Therefore, the rapid and precise detection of biogenic amines is crucial for assessing the freshness and spoilage of meat products. GC-IMS technology stands out among monitoring methodologies due to its distinctive benefits. The technology has been adeptly employed for the direct detection of trimethylamine in meat products, a volatile amine indicative of spoilage. By measuring its concentration, GC-IMS facilitates a quantitative assessment of the spoilage process, providing a comprehensive picture of food quality over time [36, 37]. However, only a single volatile amine can be monitored, which is relatively limited. Espalha et al. [60] have developed a novel method for detecting seven types of biogenic amines in fish by GC-IMS. The analytical process was conducted at 23°C, followed by heating to 40°C to examine the volatile compounds of each amine. Distinctive fingerprints were successfully obtained for each type of biogenic amine, and on the third day of measurement, an unpleasant odor was detected indicating spoilage of the fish. This study highlights the potential application of GC-IMS quantitative analysis in this field.

**4.3. Mechanism of Flavor Change during Storage.** The mechanism of flavor change in meat products that GC-IMS can detect revolves around the identification of VOCs that are either generated or lost through various chemical, enzymatic, and microbiological processes during storage. This analytical capability allows GC-IMS to identify the specific compounds associated with fresh meat, as well as those related to spoilage or fermentation, providing a picture of how the flavor profile of the meat changes over time under different storage conditions.

In the course of meat processing and refrigeration, the oxidation of lipids results in a highly diverse composition of flavor compounds. Deng et al. [61] utilized a combination of GC-MS and GC-IMS technologies to identify 53 and 30 flavor components in white pig and black pig bacon, respectively. The research indicated that the main determinant of flavor variation during cold storage at 4°C is the breed of pork. Notably, white pig bacon exhibited greater susceptibility to storage techniques compared to black pigs. The extent of thermal oxidation in meat fat varies with different heating times, which generates key precursor substances that impact the flavor profile of meat. The differentiation of volatile compounds in meat fat during various heating durations serves as a fundamental basis for further exploration into the flavor framework of meat products. A study utilizing GC-IMS was conducted to identify the flavor components of pork belly at varying cooking times. The results revealed the primary flavor constituents of high-fat pork belly exposed to different heating durations. The analysis of flavor components in pork belly at different cooking times and the identification of key volatile compounds revealed the impact of oxidation products from flavor precursors such as oleic acid, linoleic acid, and stearic acid on the flavor of pork belly. The key volatile compounds were further clarified by using PCA and the relative odor activity value [62].

Using aroma as an evaluation indicator, previous study examines the quality changes of sea cucumber peptide powder (SCPP) under various storage conditions. It also measures the tissue-specific distribution and changes of aroma substances in SCPPs during different storage periods under hygroscopic and microbial conditions. The degree of change in peak signal intensity is used to determine whether moisture absorption affects VOCs in SCPPs [63]. Therefore, GC-IMS can enhance quality control and inventory monitoring by elucidating the flavor change mechanism and facilitating regulation of physical and chemical characteristics during storage of aquatic products. By integrating the examination of acid value, peroxide value, fennel value, conjugated diene value, and various flavor substances GC-IMS, a variable approach was employed to investigate the changes in flavor profile of large yellow croakers during different storage periods [64]. The findings indicated that aldehydes exhibited higher levels than ketones and esters during the storage process. Moreover, n-nonanal content demonstrated a decreasing trend while 3-methyl butanol (trimer), 3-methyl butanol (dimer, D), 3-pentanone (D), and 3-pentanone (monomer) contents increased. After 120 days of storage, the maximum number of flavor components was

achieved, which are crucial to the alterations in fish flavor. The flavor characteristics of *Thunnus obesus* sashimi treated with cold plasma (CP) were assessed using E-nose and GC-IMS techniques. The GC-IMS fingerprint was utilized to qualitatively characterize all the flavor information of tuna, revealing the impact of CP treatment on its preservation. This study provides a comprehensive understanding of the VOCs in bigeye tuna from both macroscopic and microscopic perspectives [54]. Similarly, GC-IMS can be utilized for comparing the freshness of fish fillets stored at different temperatures. Shi et al. [65] compared all fingerprints of fresh tilapia fillets at 4°C and 25°C using GC-IMS non-targeted analysis approach based on all spectral signals and analyzed the content changes of 14 target compounds. It is concluded that flavor changes are closely related to fish spoilage.

## 5. GC-IMS Technology Combined with Other Flavor Detection Technologies

Compared with traditional analysis methods, GC-IMS can achieve rapid screening and identification, high sensitivity, selective detection of target compounds, and user-friendly operation. GC-IMS is a robust analytical technique for probing volatile and semivolatile compounds [66], which are key contributors to the flavor profiles of meat and meat products. However, the complexity of these profiles necessitates a more integrative approach. By integrating GC-IMS with complementary analytical techniques, researchers can achieve a comprehensive analysis that provides a more nuanced and in-depth understanding of the intricate flavors inherent to these products.

In the analysis of food flavors using GC-IMS in conjunction with GC-MS, the order of their analysis can vary. Typically, when comparing samples within or among three groups, an initial analysis of flavor compounds using GC-MS is commonly conducted first. Following this, GC-IMS is employed as an additional tool to analyze supplementary flavor compounds. The simultaneous analysis capability of GC-MS and GC-IMS is mainly utilized for multiple processing methods of the same sample, providing mutual confirmation for the identification of flavor components by both techniques. This approach ensures comprehensive and accurate results. When comparing more than three groups of samples, the screening function of GC-IMS is often utilized initially to identify volatile components with significant flavor characteristics before proceeding with further analysis using GC-MS.

In a study on the analysis of VOCs following enzymatic hydrolysis of oysters, HS-SPME and solvent-assisted flavor evaporation (SAFE) were combined with GC-MS and GC-IMS to detect the flavor profiles in different oyster samples. Compared with SAFE-GC-MS and GC-MS, GC-IMS demonstrated superior capability in identifying a wider range of VOCs, particularly alcohols and esters. Meanwhile, GC-MS was found to be more effective in detecting aldehydes [67]. When analyzing VOCs in grilled lamb shashliks by GC-IMS, HS-SPME-GC-MS, and SPME-Arrow-GC×GC-TOF-MS, a total of 81 flavor signals were

detected by GC-IMS, with aldehydes being the most significant. It was noteworthy that GC-IMS successfully identified (1-methylheptyl)-benzene and 2-ethenyl-naphthalene, which had not been detected before. Additionally, it was found that GC×GC-TOF-MS detected the highest number of chemical types, identifying twice as many as GC-MS did. In this study, the strong qualitative and quantitative capability of GC-MS, the high separation and resolution of GC×GC-TOF-MS, and the low detection limit and high sensitivity of GC-IMS have provided a novel approach for characterizing meat flavor [68]. Therefore, the integration of these two technologies can enable a more comprehensive detection of VOCs and enhance the reliability of experimental results. The exploration of data fusion from different complementary analysis techniques represents a novel direction in the field of food certification. These two methods are also complementary, as they rely on the distinct properties of their respective instruments. A study on food flavor analysis revealed that certain small-molecule C2~C7 compounds, such as acetone, 2-butanone, acetoin, and pentanal, were undetectable by GC-MS; however, this issue was resolved through the use of GC-IMS. However, the normal pressure and constant temperature detection environment of GC-IMS renders it unresponsive to high molecular weight VOCs such as alkanes. The disparity in flavor analysis emphasis between GC-MS and GC-IMS is attributed to their distinct priorities. While GC-MS prioritizes qualitative and quantitative accuracy, GC-IMS emphasizes broad identification of sample components [69]. For rapid screening and certain specific applications, GC-IMS is a very useful tool, especially for on-site analyses or when a rapid response is required.

Gas chromatography-olfactometry-mass spectrometry (GC-O-MS) combines the analytical power of gas chromatography-mass spectrometry (GC-MS) with human olfactory sensory perception, making it possible to provide a direct assessment of the organoleptic properties of specific compounds. GC-O-MS is particularly beneficial in the food, flavor, and fragrance industries, enabling scientists to understand how the odor of a particular compound influences the overall odor properties of a product. For instance, it has been used to identify VOCs during poultry roasting and to identify key aroma compounds [70, 71]. The anethole identified by GC-O combined with aroma extract dilution analysis method was first discovered in dzo beef, which may become a typical marker of dzo beef [72]. However, the subjective nature of olfactory testing can affect the consistency and reproducibility of results. GC-O-MS may provide more subjective data in identifying specific odor compounds because it incorporates the human sense of smell, but in scientific terms GC-O-MS is not as useful in identifying odor compounds in the food industry as it is in the flavor industry. Compared to GC-MS and GC-O-MS, the accuracy of GC-IMS in determining aromatic compounds may be slightly lower. GC-MS offers high accuracy by combining gas chromatography with mass spectrometry, providing precise identification of compounds based on their molecular structures and mass measurements. GC-O-MS further enhances accuracy by integrating olfactory detection alongside

mass spectrometry, enabling comprehensive analysis including human sensory evaluation. In contrast, GC-IMS primarily relies on ion mobility spectrometry for compound identification, offering rapid screening and characterization of VOCs; however, its accuracy in identifying compounds may require additional validation when compared to GC-MS and GC-O-MS.

Gas Chromatography-Time of Flight Mass Spectrometry (GC-TOF-MS) provides extremely high mass resolution and mass accuracy, allowing in-depth analysis of complex samples. It provides detailed mass spectra that allow very precise identification and quantification of aromatic compounds. In terms of resolution, time-of-flight mass spectrometry offers very high mass resolution and mass accuracy, and GC-IMS is not as good as GC-TOF-MS at separating complex mixtures, but GC-IMS is also very effective at dealing with simple to moderately complex samples that require rapid screening or on-site detection. However, there is a significant difference in the identification of VOCs between GC-IMS and GC-TOF-MS. GC-IMS can identify acidic compounds, which are not found in the results of GC-TOF-MS [68]. Therefore, in practice, the choice of technique depends on the specific application, the required analytical speed, sensitivity and accuracy, and the ease of operation.

Electronic nose and electronic tongue are sensor arrays that provide partial specificity, facilitating rapid identification of food flavors. Different sensors have different responses to different gas components. Electronic nose and electronic tongue mimic the human olfactory and taste system, with low detection limit and high sensitivity, making the sensory evaluation of flavor more objective. They have become widely used in volatile flavor analysis of food, often in combination with other techniques. For instance, electronic nose and GC-IMS were used to evaluate the flavor of *Penaesus sinensis* inoculated with *Bacillus subtilis* subsp during fermentation [73]. E-nose, GC-MS, and GC-IMS were combined to distinguish four different sources of red-cooked chicken [74]. Although their principles were different, the results showed that they could effectively distinguish the changes of volatile components in the samples. Also, GC-IMS, GC-MS, and electronic nose were used to explore the flavor regulation mechanism of insoluble dietary fiber on surimi gel [75]. Generally, GC-IMS and GC-MS are employed for the comprehensive detection and identification of VOCs. However, due to the limitations of the GC-IMS database, less than half of the VOCs can be successfully identified.

## 6. Summary and Prospects

GC-IMS has emerged as a highly successful and widely adopted technique in the field of on-site trace compound detection, owing to its inherent advantages of simplicity, rapidity, convenient operation, and low detection thresholds. The versatility of GC-IMS extends across various applications, including food authentication, adulteration identification, assessment of food freshness, detection of food flavors, and monitoring of food processing and storage

conditions. Moreover, GC-IMS facilitates the generation of distinct flavor profiles that enable comparative analysis of diverse samples. This capability allows for the categorization or monitoring of variations in substance content across different time periods and variances in VOCs between various times or samples. In the area of meat analysis, GC-IMS plays a crucial role in meat authentication, meat adulteration identification, and monitoring quality changes. Meanwhile, signal intensity, represented by colors in the atlas, provides visual insight into differences in flavor substance content among samples. Furthermore, the affordability of GC-IMS compared to alternative methods, coupled with its portability under atmospheric temperature and pressure, simplifies its implementation in meat laboratories and industries. These attributes collectively position GC-IMS as a pivotal tool for enhancing food quality analysis and ensuring consumer safety in the food industry. In the GC-IMS characteristic fingerprint extraction method, there exists no definitive recommendation for the targeted analysis approach based on the selected feature signal and the nontargeted analysis approach based on all spectral fingerprint signals. Although targeted analysis eliminates the processing of a large amount of tedious data, it may not always provide the highest sample resolution. The nontargeted analysis is based on all spectral signals, including the mutated signals, while also ensuring that no potentially important artificial information is omitted. Therefore, the targeted labeling approach is likely to gain more traction in future meat product flavor analysis. Nonetheless, further research is warranted to augment its classification accuracy.

Although the advantages previously mentioned are significant, it is crucial to highlight that the development of GC-IMS technology remains in its early stages, which is primarily attributed to the following aspects. Firstly, the primary limiting factor is the absence of a comprehensive database for volatile compounds. Such a deficiency impedes the technology's full potential, as the identification and analysis of unknown compounds largely depend on the availability of extensive databases for comparison. Currently, it is comparatively inadequate in characterizing unknown substances when compared to GC-MS due to the more comprehensive fragmentation information accessible in its NIST database. Therefore, the primary challenge remains the development of a comprehensive and exhaustive database to support qualitative analysis. Secondly, GC-IMS is sensitive to sample matrix effects, where different matrices can lead to variations in signal intensity, retention times, and peak shapes, affecting result comparability and reproducibility. Standardizing sample preparation protocols, including extraction methods and matrix matching, is essential to ensure consistent sample matrices for GC-IMS analysis. This reduces the impact of matrix effects and enhances result accuracy and reproducibility. Preparing the same matrix facilitates easier comparison of results between different samples and ensures the reliability and reproducibility of the analysis outcomes. Thirdly, the majority of volatile compounds can be successfully separated by leveraging GC pre-separation; however, the identification of volatile compounds with similar mobility still presents

limitations in IMS. Furthermore, there is ample room for improvement in enhancing the separation efficiency of IMS. Additionally, another key challenge will be to miniaturize IMS devices while maintaining their performance. Taken together, due to the diverse range of volatile compounds present in meat products, further development of GC-IMS technology is necessary to improve its specialized functions and explore new application fields.

## Data Availability

The review data used to support the findings of this study are available on request from the corresponding authors.

## Conflicts of Interest

The authors declare that there are no conflicts of interest related to this article.

## Authors' Contributions

Shiliang Jia was responsible for conceptualization, investigation, and writing and editing the original draft. Zhifang Jia and Yicheng Ding were responsible for visualization and writing and editing. Jun An was responsible for project administration, visualization, and reviewing the manuscript. Jie Chang and Yanbo Wang were responsible for supervision and reviewing the manuscript. Xuxia Zhou was responsible for funding acquisition and reviewing the manuscript. All authors revised and approved the final version of the manuscript.

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