fermentation

Review

Recent Progress in Electronic Noses for Fermented Foods and Beverages Applications

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Abstract: Fermented foods and beverages have become a part of daily diets in several societies around the world. Emitted volatile organic compounds play an important role in the determination of the chemical composition and other information of fermented foods and beverages. Electronic nose (E-nose) technologies enable non-destructive measurement and fast analysis, have low operating costs and simplicity, and have been employed for this purpose over the past decades. In this work, a comprehensive review of the recent progress in E-noses is presented according to the end products of the main fermentation types, including alcohol fermentation, lactic acid fermentation, acetic acid fermentation and alkaline fermentation. The benefits, research directions, limitations and challenges of current E-nose systems are investigated and highlighted for fermented foods and beverage applications.

Keywords: E-nose; gas sensor; fermented food; alcohol; lactic acid; acetic acid; alkaline

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1. Introduction

Due to uncertainties and unpredictable crisis situations (e.g., war conflicts) and global pandemics, the demand for fermented food and beverages has been significantly increasing because they are easy to cook and have a long shelf life [1]. In the production of fermented foods and beverages, different types of microorganisms, such as bacteria, yeast and mold, have been used to modify the chemical composition, resulting in changes in taste, smell, color and nutrients. For example, fermentation with probiotic microorganisms such as a lactic acid bacterium in products such as yogurt, kefir and kimchi can increase the nutritional value, for example, by reducing cholesterol and promoting healthy digestive function [2]. In countries, the fermentation technique has been developed specifically from local wisdom to large-scale manufacturing industries by selecting pure microbial strains, resulting in unique fermented foods and beverage products, such as pla ra (fermented fish) in Thailand, Chinese kombucha, Indonesian tempeh, natto and miso in Japan, Korean kimchi, etc. In the fermentation process, microorganisms can produce a variety of enzymes to modify food precursors. For example, the fermentation of natto employs bacteria of the genus *Bacillus* to create a fibrous texture that is sticky and stretchy [3]. The fermentation of coconut water (nata de coco) is produced via the bacterium *Acetobacter xylinum* [4]. Fermented tofu employs *Monascus purpureus* [5]. Different microorganisms make products with distinctive flavor compounds, nutrients and shelf lives.

To assess the quality of fermented food and beverages, such as the physical quality, nutrition value, microbiological quality, safety and sensory quality, there have been several types of equipment and techniques developed, ranging from spectroscopies to sensory evaluation techniques [6–10]. Equipment and methods enabling non-destructive measurement, rapid analysis and on-site testing with low operating costs and simplicity have received great interest in recent years. One of the effective tools for qualitative food and beverage identification meeting these criteria is an electronic nose (E-nose). There is a global trend in the development of E-nose systems instead of standard equipment such as GC, GC/MS, SPME/GC-TOFMS, GC-IMS, etc., in the qualitative analysis of food and beverages [11–20]. Although there are a set of review articles on the E-nose in food and beverage applications, comprehensive reviews focusing on fermented food and beverages based on E-nose technology are still limited.

In this review, the advancement of E-nose technologies for fermented food and beverage applications is presented. Fermented food and beverages can be classified according to the end product into four major groups as follows: (I) alcoholic fermentation, such as beer, sourdough, makgeolli, vodka, whiskey, wine, brandy and cider [21,22]; (II) lactic acid fermentation, such as yogurt, fermented milk, cheese, kefir, sour pork, sour sausage, salami, pepperoni, kimchi, sauerkraut and pickles (olives and cucumbers) [23,24]; (III) acetic acid fermentation, such as vinegar, kombucha, and pu-er tea [25,26]; and (IV) alkaline fermentation, such as fish sauce, jeotgal, fesikh, soy sauce, soy paste, miso, natto and tempeh [27,28]. In addition, each major group can be split into subcategories according to the main raw ingredients of each type of fermented food and beverage, such as cereals, fruits, herbs, milk, meat, vegetables, legumes, etc. An overview of the entire literature review in this article is displayed in Figure 1.

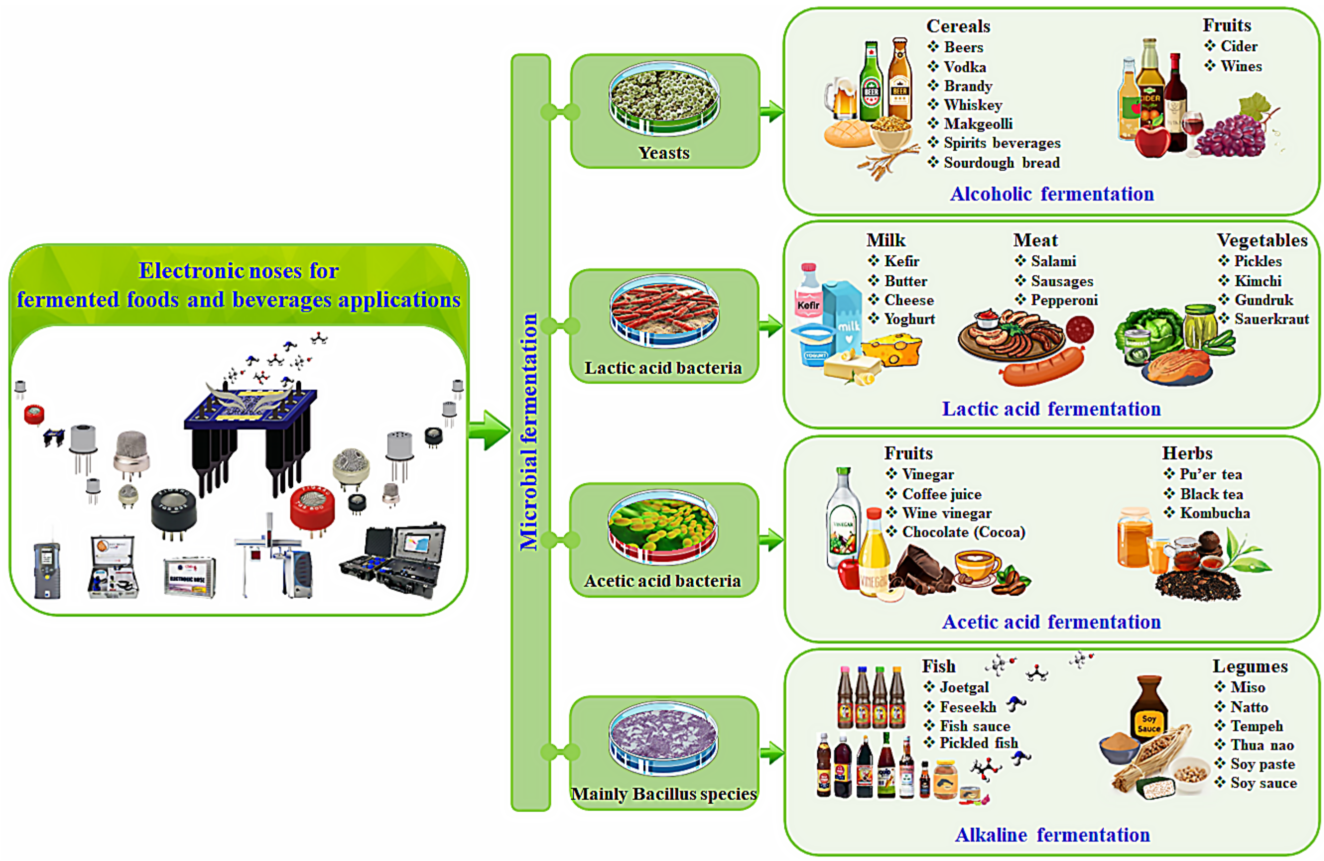

Figure 1. Applications of E-nose for fermented food and beverage.

2. History and Basic Principle of E-Nose

One of the first reports on the E-nose was in 1982, originated by Dodd, G. H. and Persaud, K. from the University of Warwick, England [29]. They employed three Figaro semiconducting gas sensors as transducers. The results showed fine discriminations between odors according to the mammalian olfactory system. This pioneering work has inspired several research groups to develop E-nose systems for various applications [30,31]. The concept of the E-nose is based on mimicking the human olfactory system, as shown in Figure 2. The mechanisms, odor recognition and limitations of human olfactory perception at a molecular level were first investigated by Buck and Axel from Columbia University, USA, in 1991 [32]. Their discovery helped to establish new ways/ideas to develop an E-nose system instead of using complex human olfactory perception for multifunctional purposes. The human olfactory system can be considered to consist of three main parts: (I) the odor-receiving part, consisting of olfactory receptor glands and scent delivery systems [33], (II) the nervous system for the transmission of signals between the brain and the rest of the body and (III) a decision system that is able to recognize, identify and act on the sense of smell experienced via the brain. The mechanism of smell perception is very complex. On the basis of psychophysical testing, humans are able to discriminate > 1 trillion olfactory stimuli [34]. However, the emotions and age of humans have a significant effect on odor recognition and classification [35–37]. Moreover, toxic agents in the sample and testing time are crucial obstacles in the ability to identify smells via the human olfactory system. The E-nose has thus become one of the powerful tools as an alternative to human evaluation of scent in samples.

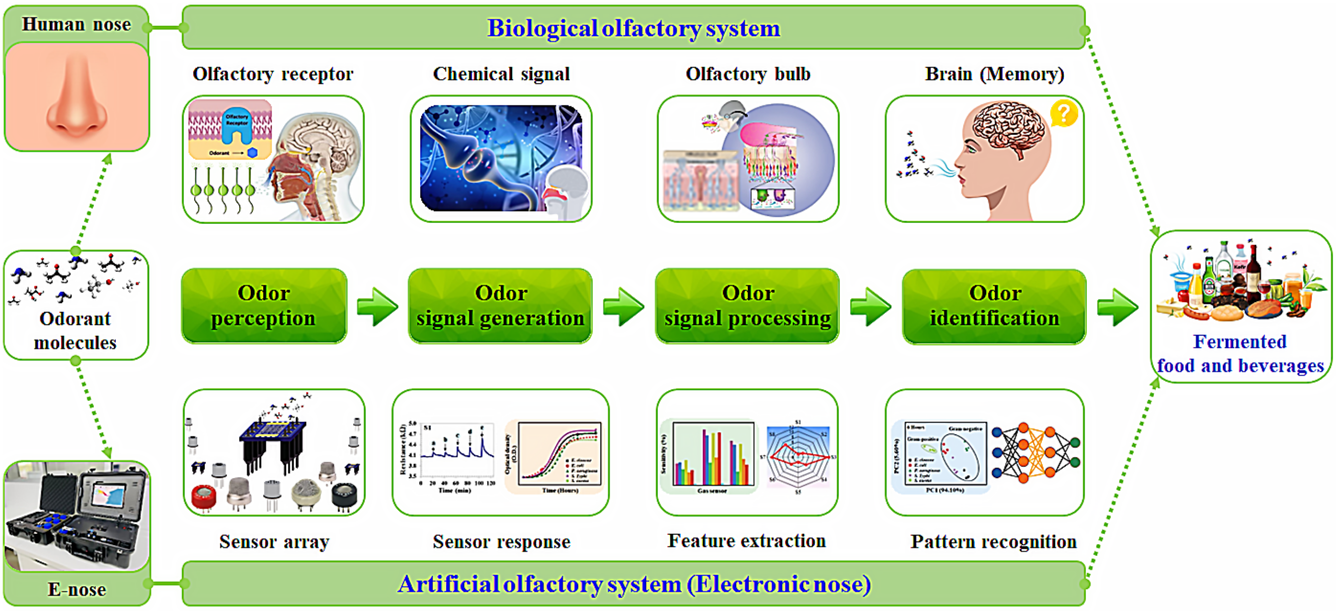

Figure 2. Schematic diagram of E-nose device versus biological olfactory system.

A comparison between an E-nose system and the biological olfactory system is displayed in Figure 2. In the E-nose system, the olfactory receptor section consists of the odor delivery unit (including the pipes, pumps and valves), which creates a path for aroma delivery into the sensor chamber. Numerous gas sensors, a so-called sensor array, are the heart and most important component of the olfactory receptor. Various sensing materials, such as conducting polymers [38,39], carbon-based nanomaterials [40–42], metal oxides [43–46] and nanocomposites [47,48], have been used to adsorb odor molecules based on both physisorption and chemisorption. When the odor molecules adsorb on the sensing material surface, they lead to charge transfers, volume expansion, ion exchange or interaction with ion species that can cause changes in the electrical conductivity/resistivity of the sensing materials. The electrical signals generated by various sensors are converted from analog to digital via an A/D converter and modified via signal processing, such as noise reduction or signal amplification. The data are stored on a local computer/online platform for further analysis. Due to the multivariate data obtained from the gas sensor array of the E-nose system, data analysis is usually performed via supervised/unsupervised machine learning algorithms with statistical methods such as principal component analysis (PCA) [49–51], hierarchical cluster analysis (CA) [52,53], analysis of variance (ANOVA) [54], linear discriminant analysis (LDA) [55], partial least squares discriminant analysis (PLS-DA) [56], simple visualization techniques [57], multivariate data analysis [58], artificial neural networks

(ANNs) [59–61], artificial intelligence (AI) [62] and F-test [63]. A photograph and schematic diagram of a prototype portable E-nose system are displayed in Figure 3.

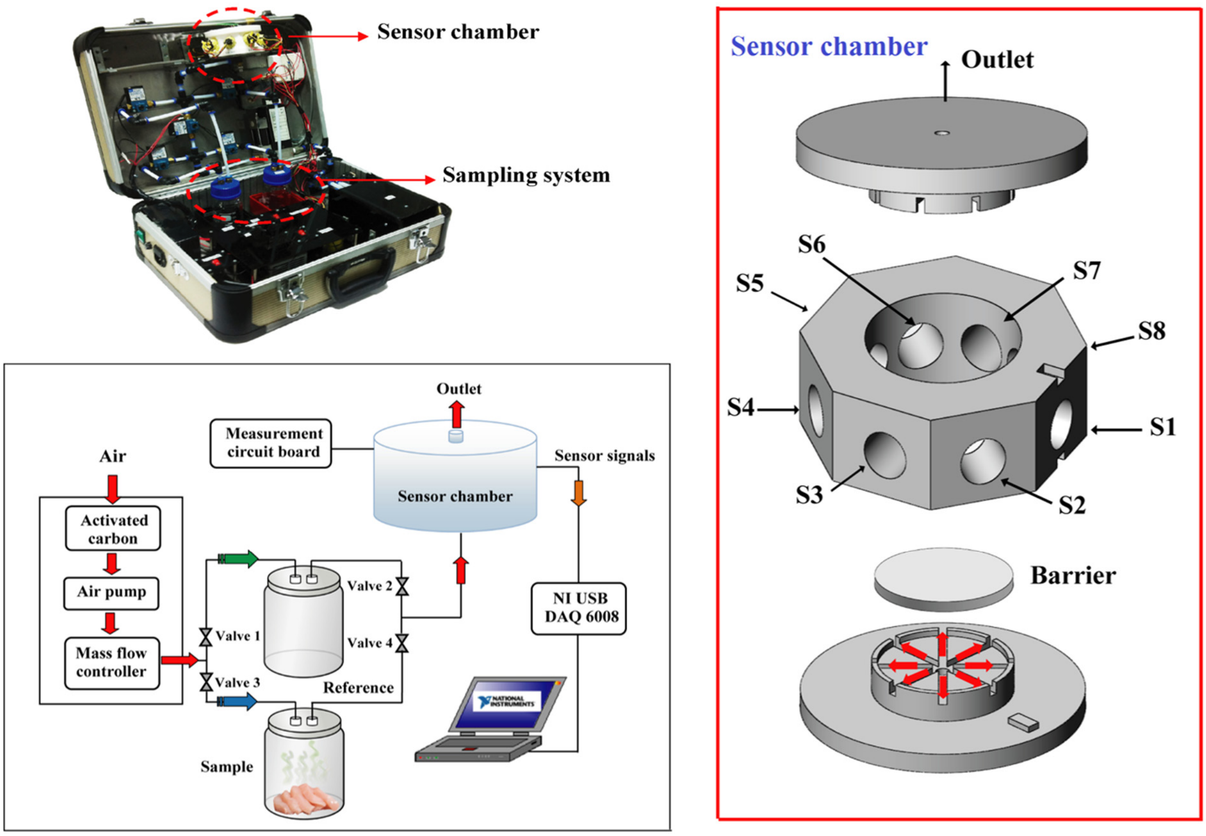

Figure 3. Photograph and schematic diagram of a prototype portable E-nose system. (Reproduced with permission from Elsevier [16]).

Nowadays, E-nose systems based on a diversity of gas sensor arrays are applied in all major sectors, such as agriculture and forestry [64–66], industrial processes [67], environmental toxin/pollutant analysis [68–70], space stations [71,72], medical/healthcare [73–76], authentication of a person [77], medicine/pharmaceuticals [78], forensic science [79], military [80], toxicology/security [81,82] and the food and beverage industry [83–85]. Another emerging direction in the field could be to develop mobile and affordably priced devices for people who suffer from anosmia (loss of smell) [86,87]. With a focus on applications in the food and beverage industry, E-nose systems have been used for both direct/indirect identification via odor analysis for multiple purposes, such as product quality inspection [88], batch-to-batch uniformity studies [89], contamination detection [90], spoilage detection [91–93], adulteration detection [13,94], the detection of pathogenic bacteria [95,96], the study of storage conditions/shelf life [97–100] and the creation of specific sensory profiles [101,102]. In terms of food business competition, they have been used to analyze aromas and compare them with competitor products [103,104], evaluate the impact of changes in the production process and components that affect organoleptic characteristics [105,106] and compare different food formulations [84,107]. Moreover, E-nose systems have showed high performance in identifying the quality of many products, including wine [108], beer [109], coffee [110], carbonated drinks [111], dairy products [112,113], pork [114], beef [115,116], chicken [117], fish [118–120] and shrimp [121,122]. However, the sensors in E-nose systems may have a drift effect. Due to the aging of the sensors, measurements performed at different time intervals have a slight bias [123–125]. From the past to the present, research is still ongoing for the development of E-noses with high precision and accuracy, on online platforms, and for quantitative identification.

3. E-Nose for Alcoholic Fermentation

Alcoholic fermentation is the anaerobic transformation of fructose and glucose into mainly ethanol and carbon dioxide. Microorganisms associated with the fermentation of alcoholic beverages, such as yeast, play an important role in the fermentation process of sugar to produce alcohol, especially species in the genus *Saccharomyces*, which is commonly used in the production of alcoholic beverages from various carbohydrates, i.e., the production of beer using *Saccharomyces cerevisiae* and *S. carlbergensis* and the production of whiskey, wine, vodka, brandy and bread using *S. cerevisiae* [126].

Currently, the E-nose has been mainly used for quality evaluation in the production of alcoholic products, starting from the fermentation/incubation process to the storage of the end product. For example, 13 MOS gas sensors were used to detect the concentration of ethanol in seven types of beers of various brands. The results from four types of ANN models (MLR, MNLR, RF and ELM) showed good performance of the E-nose for detecting the levels of alcohol contained in beverages [127]. Viejo et al. reported the early detection of beer faults by using nine MOS gas sensors integrated with AI [60]. Different beer flavor profiles based on the E-nose showed a comparable accuracy to that of a standard expensive method, namely, near-infrared spectroscopy (NIR). Berna et al. presented an E-nose application for wine spoilage prediction. Twelve MOS gas sensors were selected to detect 4-ethylphenol (4EP) and 4-ethylguaiacol (4EG), which are taints produced by Brettanomyces yeasts [128]. The results showed that the E-nose was effective in screening wines for ethylphenol taints. Bread and bakery products can be considered a group of alcoholic fermentation products. There are many key aroma compounds, such as acetaldehyde, methanol, 1-propanol and 3-methyl-1-butanol that can be used for the identification of bread freshness [129–133]. However, the challenge of the E-nose is still the quantitative identification of VOCs. Therefore, the quantitative analysis of E-noses has usually been compared with standard spectroscopies, such as UV-Vis, GC and GC-MS [134–138]. The combination of the E-nose and GC-MS for qualitative and quantitative analysis during the wheat bread-making process is displayed in Figure 4. The variety of E-nose applications to detect odors from food and beverage products caused by alcoholic fermentation is summarized in Table 1.

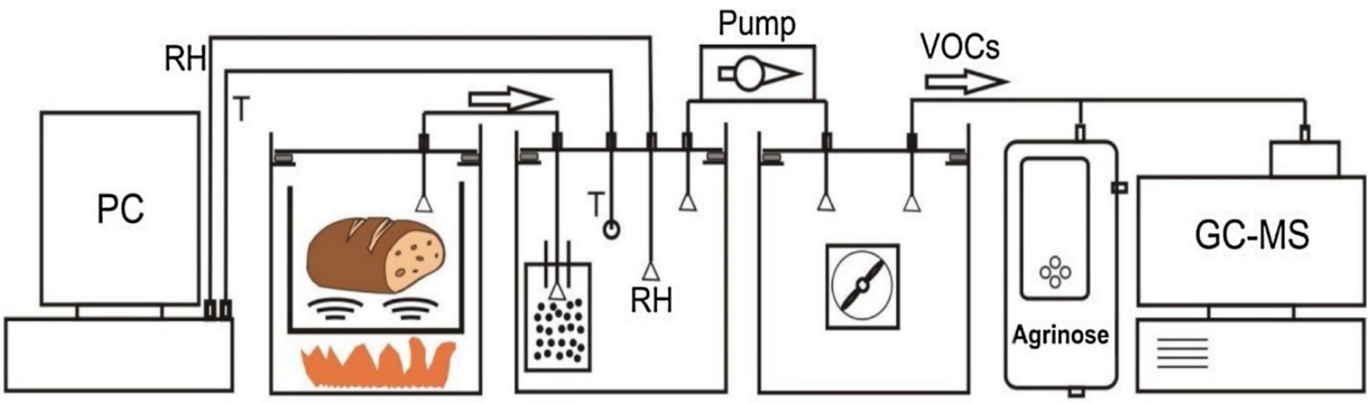

Figure 4. Schematic diagram of E-nose system combined with GC-MS for monitoring VOC emissions during the bread production process. (Reproduced with permission from Elsevier [137]).

Table 1. The E-nose for alcoholic fermentation.

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
Fermented cereal and grain products					
2014	Thailand	Draft beer (To monitor the draft beer fermentation process)	7 MOS Model: Lab-made E-nose (PCA)	General combustible, alcohol, natural, methane, LPG, propane, carbon monoxide, air contaminants	[139]
2019	Brazil	Beer (To detect the alcohol content of beers)	13 MOS Model: Lab-made E-nose (MLR, ELM, RF, MNLR)	Ethanol, propane, butane, methane, isobutane, benzene, NH_3, toluene, CH_4, hexane, LPG, CO, H_2, H_2S, methyl mercaptan, trimethylamine	[127]
2020	Australia	Beer (To assess beer quality)	9 MOS Model: Lab-made E-nose (ANN)	Ethanol, methane, carbon monoxide, hydrogen, hydrogen sulfide, ammonia, benzene/alcohol/ammonia, carbon dioxide	[140]
2021	Australia	Beer (To detect beer faults in the brewing process)	9 MOS Model: Lab-made E-nose (ANN)	Alcohol, methane (CH_4), carbon monoxide (CO), hydrogen (H_2), ammonia/alcohol/benzene, hydrogen sulfide (H_2S), ammonia, carbon dioxide (CO_2)	[60]
2014	Korea	Korean rice wine (makgeolli) (To detect volatile compounds from initial amino acid profiles in Korean rice wine (makgeolli) model)	MS-EN Model: SMart Nose 300 (PCA, PLSR)	Isobutanol, isoamyl alcohol, 2-methyl butanol, 2-phenylethanol, isobutyl acetate, isoamyl acetate, 2-methylbutyl acetate, 2-phenylethyl acetate, ethyl caproate, ethyl caprylate, ethyl caprate	[135]
2016	Brazil	Spirit beverages (To classify odors from alcoholic beverages, as well as butanol and methanol vapors)	5 MOS Model: Lab-made E-nose (PCA, MLP, SOM, CA)	Aguardiente, butanol, tequilas, vodkas, whiskey, methanol	[141]
2016	Poland	Spirit beverages (To discriminate raw spirits, vodkas and whisky samples)	Fast GC e-nose Model: Alpha MOS (PCA, DFA, SIMCA)	tert-Butylmethylether, ethyl acetate, 2-methyl-1-butanol, 2-methyl-1- propanol, n-butanol, 2,3-pentanedione, methyl butanoate, pentanol, 1-hexanol, (Z)-4-heptenal	[136]
2017	Poland	Whisky (To analyze and evaluate type of whisky)	MOS Model: Alpha MOS (PLS-DA)	2-Propanol, n-butanol, furfural, ethyl isovalerate, alpha-pinene, decanal, eugenol, geosmin, gamma-decalactone, 12-methyltridecanal	[138]
2006	India	Bread (To cluster bread odor data according to the state of freshness of bread)	4 MOS Model: Lab-made E-nose (SOM network)	Methane, butane, propane, alcohol, toluene, xylene, LP-gas/propane, CO, CO_2, H	[132]

Table 1. *Cont.*

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
2019	Italy	Bakery products (To investigate the volatilome of different bakery products, obtained from mature and immature grains and transformed by sourdough)	10 MOS Model: PEN2 (CAP analysis)	Ethyl alcohol, 3-hydroxy-2-butanone, acetic acid, octanoic acid, heptanoic acid, 3-methyl butanoic acid, heptanal, acetophenone, ethylbenzene, 2-methyl-3-decen-5-one, 2,4-bis (1-methylethyl) phenol, heptanal	[131]
2019	China	Sourdough-based Chinese steamed bread (To identify a key aroma compound in type I sourdough-based Chinese steamed bread)	14 MOS Model: ISENSO INTELLIGENT (PCA, PLS-DA)	Ethyl acetate (fruity), ethyl lactate (caramel-like), hexyl acetate (fruity), (E)-2-nonenal (fatty) and 2-pentylfuran (fruity), acetic acid, 2-methyl-1-propanol, ethanol, 1-pentanol, pentanal, (E)-2-octenal, hexanal	[129]
2020	Iran	Sourdough (To investigate effective variables to produce desirable aroma in sourdough)	6 MOS Model: Lab-made E-nose (LSD test)	Alcohol, LPG, natural gas, carbon monoxide, ammonia, sulfide, benzene, H2S, toluene, acetone, hydrogen	[130]
2020	Poland	Bread (To test the suitability of bread for consumption during four-day storage)	8 MOS Model: Lab-made E-nose (PCA)	Ketones, acids, aromatic comp., terpenoids, sulfur comp., esters, alcohols, hydrocarbons	[133]
Fermented fruit products					
2008	Australia	Wine (To predict red wine spoilage)	12 MOS Model: FOX-3000, Alpha MOS (PLS regression, RMSECV)	Ethanol, 3-methylbutanol, 4-ethylphenol, 4-ethylguaiacol	[128]
2012	Spain	Wine (To identify the aging of red wines)	9 MOS Model: Lab-made E-nose (PCA, Tucker3 analysis)	2-Furaldehyde, guaiacol, eugenol, cis- and trans-whiskeylactone	[134]
2021	Australia	Wine (To assess volatile aromatic compounds in smoke-taintedcabernet sauvignon wines)	9 MOS Model: Lab-made E-nose (LSD, PCA, ANN)	Ethanol, methane, carbon monoxide, hydrogen sulfide, ammonia, alcohol, benzene, carbon dioxide, hydrogen	[142]
2021	Australia	Wine (To detect smoke taint in Pinot Grigio wines)	9 MOS Model: Lab-made E-nose (LSD, PCA, ANN)	Ammonia, alcohol, benzene, carbon dioxide, hydrogen, ethanol, methane, carbon monoxide, hydrogen sulfide,	[61]
2022	Australia	Wine (To assess and classify flaws and faults in wine)	9 MOS Model: Lab-made E-nose (HSD, SE)	Alcohol, methane, hydrogen, ammonia, benzene, carbon dioxide, hydrogen sulfide	[143]

4. E-Nose for Lactic Acid Fermentation

Lactic acid bacteria are a group of Gram-positive, non-sporulating bacteria that play an important role in food fermentation in anaerobic, carbohydrate-containing environments [144]. The production of fermented food with lactic acid bacteria results in different aroma compounds according to the type of raw material and the duration of the fermentation process. For example, fermented milk products such as yogurt, fermented milk and cheese give the specific odor properties characteristic of acidic flavor and other substances, including carbon dioxide, acetic acid, diacetyl and acetaldehyde, during the fermentation process [145]. The metabolic pathway of VOC generation during milk fermentation by *Lactobacillus pentosus* is displayed in Figure 5. However, insufficient or abnormal flavor may occur during the fermentation process with lactic acid bacteria due to changes in pH. The quantity of lactic acid strongly affects the flavor. During the fermentation process, unpleasant flavors, including maltiness, metallic flavor, methyl sulfide flavor, green flavor and fishy flavor, can frequently occur and need to be monitored.

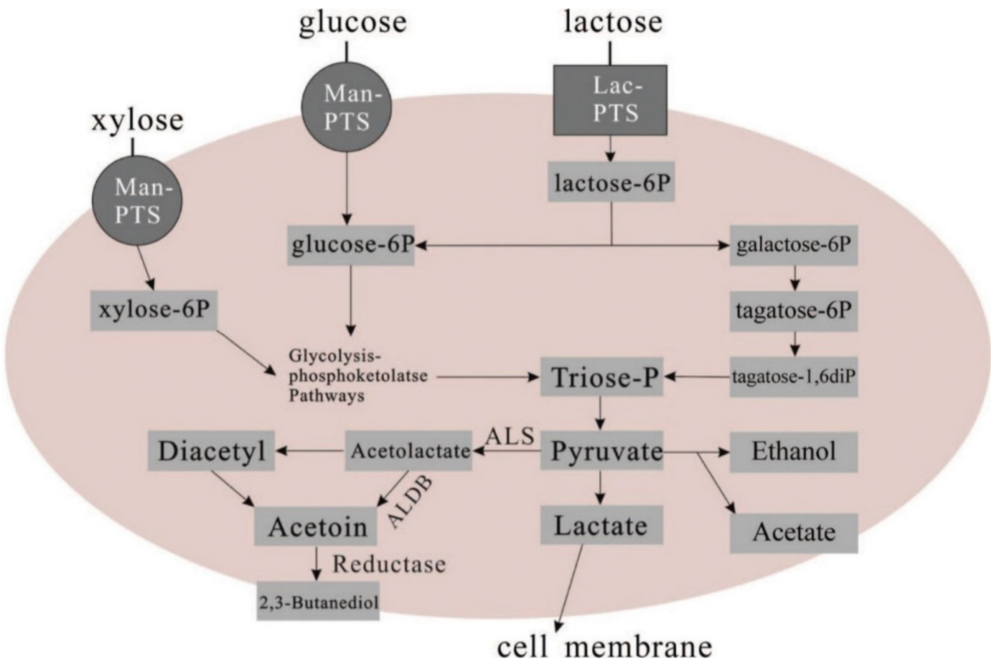

Figure 5. Generation of VOCs in carbohydrate metabolism during milk fermentation by *Lactobacillus pentosus*. (Reproduced with permission from Elsevier [145]).

Finding a correlation between different odors in multiple product conditions and the duration of the lactic acid fermentation process has become a common way to develop a new product and control the quality standard. There are many lactic bacteria in different groups associated with fermented food processing, such as *Lactobacilli*, *Lactococci*, *Leuconostoc*, *Pediococci and Streptococci* [146], that can give specific flavors. Most studies on E-noses aim to detect the characteristic odor alteration according to a distinct change in the growth of bacteria during the fermentation process. For example, an E-nose based on MOS was used to identify four yeast species (*Pichia anomala*, *Pichia kluyveri*, *Hanseniaspora uvarum* and *Debaryomyces hansenii*) in white fresh soft cheese [147]. The results showed a good prediction of cheese spoilage by using PLS models. The Heracles E-nose with LDA models was employed to monitor odor changes during milk fermentation with Lactobacillus species [148]. An E-nose with a mass spectrometer showed good differentiation of aged cheddar cheese aromas [149]. Odor measurement was performed using an E-nose with two FID detectors to investigate the characterization of steamed cheese and to assess the quality decay of steamed cheese during storage [150]. A POLFA odor sensor and Tukey's HSD test were selected to classify cheese types, production areas and cheese ages [106]. Most of the odors could be readily distinguished using an E-nose based on MOS to detect the ripening of Danish blue cheese [151], to analyze stirred yogurt with cheddar cheese added to milk [152] and to detect unsatisfactory products for the quality control of yogurt [153].

In fermented meat products such as fermented pork, sour sausage and salami, it has been found that VOCs released from these foods are usually acetic acid, ethyl butanoate, hexanal, methional, 1-octen-3-ol, benzeneacetaldehyde and 4-methyl-phenol [154]. The vast majority of research studies have used MOS in the E-nose system for a variety of purposes, including the manufacturing evaluation process to evaluate the lipid oxidation of Chinese-style sausage during processing and storage [155], the detection of the flavor substances in Ningxiang pork [156], the identification of the flavor profile for dry fermented sausages with different NaCl substitutes [157], the construction of volatile profiles and taste properties of Harbin red sausages [158] and the discrimination of different Mediterranean salami [159].

In products fermented from fruits and vegetables, such as kimchi, sauerkraut and pickled fruit, a GC-MS odor analysis revealed several VOCs related to different salt concentrations in the fermentation process. For the first stage of sauerkraut fermentation, VOCs such as esters, aldehydes and ketones were found at low salt concentrations. Acids, alcohols, isothiocyanates and hydrocarbons became dominant when the salt concentration was higher [160]. Many applications of E-noses for lactic acid fermentation have been found, i.e., the detection of the bacterial diversity during the fermentation process and the flavor quality of zha-chili samples [161], assessments of the abnormal fermentation defects of Spanish-style table olives [162], investigation of the lactic acid bacteria in the fermentation of Chinese northeast sauerkraut [163], the classification of the geographical origin of kimchi [164] and the detection of saponin in submerged fermentation with *Tremella aurantialba* (*T. aurantialba*) [165]. A list of E-nose systems for lactic acid fermentation applications is summarized in Table 2.

Table 2. E-noses for lactic acid fermentation.

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
Fermented milk products					
2003	USA	Cheddar cheese (To evaluate the differentiation of aged Cheddar cheese aroma)	E-nose based on GC (Kenner, LA, USA) Model: Lab-made E-nose (CA, PCA)	4-Mercapto-4-methyl-pentan-2-one	[149]
2005	Denmark	Danish blue cheese (To detect the ripening of Danish blue cheese)	12 MOS Model: FOX-3000, Alpha MOS (PCA, PLS-R)	2-Methyl ketones, benzaldehyde, 3-methyl-butanal, 2-hexanol	[151]
2014	China	Yogurt (To analyze stirred yogurt with cheddar cheese added to milk)	18 MOS Model: FOX-4000, Alpha MOS (PCA)	Pentanal, dimethyl trisulfide, ethyl butanoate, methanethiol, dimethyl disulfide	[152]
2019	Slovakia	Slovak cheese (To characterize steamed cheese and assess quality decay of steamed cheese during storage)	E-nose with two FID detectors Model: Heracles II, Alpha M.O.S. (PCA)	Acetaldehyde, 1-propanal, propanoic acid, ethyl hexanoate, furfural, butan-2-one, isovaleric acid, 1-hexanol or α-pinene	[150]
2020	China	Yogurt (To detect unsatisfactory products for the quality control of yogurt)	18 MOS Model: FOX-4000, Alpha MOS (PCA, BPNN, RF)	Satisfactorily favored yogurt samples and three subsets of yogurt samples with unsatisfactory favors (cooked, lack of yogurt favor, vinegarish)	[153]
2020	Hungary	Milk (To monitor odor changes during milk fermentation with Lactobacillus species)	Heracles GC e-nose Model: Heracles Neo 300 ultra-fast GC analyzer (LDA, ANOVA tests, Tukey HSD test)	2-octanol (expressing fatty or oily aroma), decane (sweet aroma), 2,4-heptadienal (fatty aroma), compound of pyridine, 2-pentyl (fatty and tallowy aroma)	[148]

Table 2. *Cont.*

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
2020	Slovakia	Parenica cheese (To investigate the quality of smoked and unsmoked Parenica cheeses after seven days of storage)	GC-FID e-nose Model: Heracles II; Alpha M.O.S. (PCA)	Carboxylic acids, alcohols, aldehydes, ketones, esters, sulfur compounds, furans/furanones, phenols, terpenes	[166]
2021	Japan	Cheese (To classify cheese types, production areas and cheese ages)	POLFA odor sensor Model: POLFA, Karumoa Co., Ltd. (Tukey's HSD test)	VOCs from five kinds of processed cheeses (two types of normal processed cheese, one type containing aged cheese, two types containing blue cheese, one type of natural cheese)	[106]
2022	Palestine	White fresh soft cheese (To identify and quantify four yeast species in white fresh soft cheese)	8 MOS Model: Lab-made E-nose (PCA, PLS)	Four species of yeast (*Pichia anomala, Pichia kluyveri, Hanseniaspora uvarum, and Debaryomyces hansenii*)	[147]
Fermented meat products					
2003	Italy	Salami (To analyze and discriminate different Mediterranean salami)	5 sensors based on SnO_2 sol–gel thin films Model: Lab-made E-nose (PCA)	β-Pinene, α-pinene, limonene, hexanal, benzaldehyde, 1-hexanol, 2-ethyl, 2-heptanone	[159]
2017	China	Chinese-style sausage (To evaluate lipid oxidation of Chinese-style sausage during processing and storage)	10 MOS Model: PEN3 (SVM, ANN, HCA)	Aromatic compounds, nitrogen oxides, ammonia, benzene, hydrogen, methane, sulfur compound, alcohols	[155]
2021	China	Ningxiang pork (To detect flavor substances in Ningxiang pork)	10 MOS Model: PEN3 (PCA, LDA)	Aldehydes, alcohols, ketones, olefins, nitrogen oxides, methyl compounds, aldol, sulfur compounds, aromatic hydrocarbons	[156]
2021	China	Dry fermented sausage (To detect the flavor profiles of dry fermented sausages with different NaCl substitutes)	10 MOS Model: PEN3 (PCA, HCA)	Aldehydes, ketones, alcohols, acids, esters, terpenes	[157]
2021	China	Harbin red sausages (To study volatile profiles and taste properties of Harbin red sausages prepared with traditional and conventional processing methods)	10 MOS Model: PEN3 (PCA, PLS)	Hexanol, furfuryl alcohol, hexanal, furfural, 5-methyl furfural, 2-methyl-2-cyclopenten-1-one, 3-methyl-2-cyclopenten-1-one, 3-ethyl-2-hydroxy-2-cyclopenten-1-one, 1-indanone, guaiacol, 2-methoxy-4-methylphenol, 2,3,5-trimethyphenol, o-cresol, 4-ethyl-2-methoxyphenol, 2,5-dimethylphenol, 2,4-dimethylphenol, 2-methoxy-4-propylphenol, octane, naphthalene, heptanoic acid, 1-octen-3-ol, 2-nonanone, methyl 2-furoate	[158]

Table 2. *Cont.*

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
Fermented vegetable products					
2021	China	Zha-chili (To detect the bacterial diversity during fermentation process and flavor quality of zha-chili samples)	10 MOS Model: PEN3 (PCA, PCoA, CA, ANOSIM)	Ammonia, aromatic compounds, aldehydes, ketones, methane, sulfur compounds, terpenes, organic sulfur compounds, alcohols, ketones	[161]
2021	Spain	Green olives (To assess abnormal fermentation defects of Spanish-style table olives)	11 MOS Model: Lab-made E-nose (PCA, PLS-DA)	VOCs from five types of table olives (control, zapateria, putrid, butyric and musty), carboxylic acids, alcohols, phenols, esters	[162]
2021	Korea	Kimchi (To classify geographical origin of kimchi by volatile compound analysis)	Smart Nose SA, Marin-Epagnier, Switzerland Model: Lab-made E-nose (PCA, DFA)	Total of 69 kinds of kimchi samples (Korean and Chinese kimchi)	[164]
2021	China	Chinese northeast sauerkraut (To investigate lactic acid bacteria for fermentation of Chinese northeast sauerkraut)	10 MOS Model: PEN3 (PCA, HCA)	2-Ethylhexanol, 3,5-dimethylbenzaldehyde, 3,5-ditert-butylphenol, alcohols (2-nonanol, 2-heptanol), esters (ethyl crotonate, ethyl butanoate, ethyl lactate, ethylhexanoate, ethyl 3-phenylpropanoate and ethyl acetate), terpenes (geraniol, nerolidol, dihydrocarveol), polyphenols, nitriles, lactones, isobutanol, methyl acetate, 2-methylpentaldehyde	[163]
2021	China	Tremella aurantialba (To detect saponin in submerged fermentation with *Tremellaaurantialba (T. aurantialba)*)	8 MOS Model: Lab-made E-nose (PLSR)	Saponin compounds, aldehydes, esters, alcohols, heterocyclic, aromatic compounds, ketones	[165]

5. E-Nose for Acetic Acid Fermentation

The majority of bacteria for fermented food in this group are acetic acid bacteria, which can oxidize ethyl alcohol and other organic matters to acetic acid, especially *Acetobacter aceit*, *A. xylinum*, *A. peroxidans and A. pasteurianus* strains. Acetic acid bacteria are commonly used in the production of vinegar, nata and tea fungus. Among the foods fermented in the acetic acid fermentation process, vinegar is one of the most popular for quality grading via odor testing. In the vinegar production process, the most dramatic changes in VOCs during the fermentation process were ethanol, 3-methyl-1-butanol, acetic acid and ethyl acetate [167]. Therefore, gas sensors that can detect these key VOCs play an important role in the application of acetic acid fermentation. The nano-ZnO thick film is a popular MOS for sensing these VOCs. The E-nose based on this gas sensor array was used to establish a fingerprint database of Chinese vinegars [168] and identify them individually for their quality control [169]. A surface acoustic wave (SAW) sensor array that relies on the resonance frequency-sensing mechanism was also used for acetic acid monitoring and can determine the volatile profiles of cider vinegars [170]. However, the most popular MOS gas sensors are still those that are able to detect, discriminate and recognize different kinds of vinegars and herb products in online/offline platforms [171–177]. A schematic diagram of an IoT-based E-nose system for VOC detection is shown in Figure 6. To detect VOCs emitted from fermentation, the MOS is still a popular sensing material in E-nose systems. Based on PCA coupled with CA, they were used to analyze odor substances for quality control in black tea during the fermentation process [178,179] and monitor the fermentation process of Sri Lankan low-country tea [89,180]. Not only MOS but also polymer/CNT gas sensors can be integrated into an E-nose for tracking odors of Thai sweet fermented rice (Khao Mak) during the fermentation process [181]. A list of E-nose applications for acetic acid fermentation is summarized in Table 3.

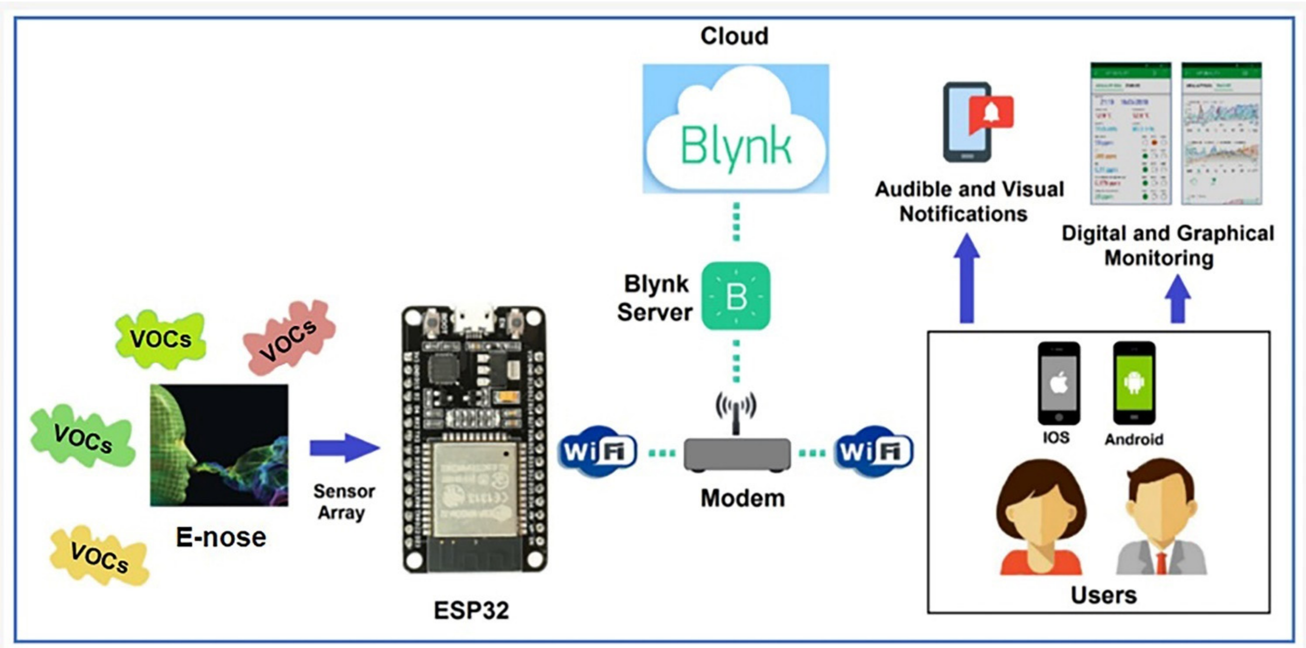

Figure 6. Schematic diagram of IoT E-nose system [172].

Table 3. E-noses for acetic acid fermentation.

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Refs.
Fermented fruits products					
2006	China	Chinese vinegars (To establish a gas-sensing fingerprint database of Chinese vinegars for quality control)	Nine nano ZnO thick film gas sensors Model: Lab-made E-nose (PCA, CA, LVQ)	Acetic acid and vinegar samples (aromatic vinegar, mature vinegar, rice vinegar, white vinegar, fruit vinegar)	[168]
2008	China	Chinese vinegars (To establish a gas-sensing fingerprint database of Chinese vinegars and identify them individually for their quality control)	Nine doped nano-ZnO thick film gas sensors Model: Lab-made E-nose (BP-ANNs, KNN)	Aromatic vinegar (Hengshun Xiangcu, Beigushan Xiangcu, Yanhui Xiangcu, Jinyou Xiangcu, ZhengjiangXiangcu, Longmen Xiangcu and Weichunyun Xiangcu), maturevinegar (Shuita Chencu, Donghu Chencu, Beigushan Chencu, Zhengjiang Chencu and Weichunyuan Chencu), Rice vinegar (Jinbiao Micu and Longmen Micu), white vinegar (Meiweixia Baicu and Longmen Baicu), fruit vinegar (Haitian Pingguocu)	[169]
2012	China	Vinegar (To identify vinegar online)	6 MOS Model: Lab-made E-nose (RBF, K-means)	Five kinds of Chinese vinegar (vinegarriver white city, river city rice vinegar, Liubiju vinegar, apple vinegar, Zilin old vinegar	[174]

Table 3. *Cont.*

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Refs.
2013	Korea	Cider vinegars (To determine the volatile profiles of cider vinegars)	Surface acoustic wave (SAW) sensor Model: zNose (PCA)	Acetic acid, ethyl alcohol, 1-hexanol, furfural, benzaldehyde, ethyl acetate, n-butyl acetate, isobutyl acetate, isoamyl acetate, hexyl acetate	[170]
2014	China	Vinegar (To evaluate the aroma of vinegars and control their quality)	10 MOS Model: PEN3 (La, LDA, PCA)	Volatile components in vinegars (acids, esters, aromatics compounds, alcohols, sulfur compounds, alkenes)	[173]
2016	Korea	Vinegar (To discriminate aged vinegars based on taste and aroma profiles)	MS-E-nose Model: SMart Nose300; Alpha M.O.S. (PCA)	Aroma profiles of aged vinegars (Chinese Shanxi extra-aged vinegar, Japanese black vinegar, Italian balsamic vinegar)	[177]
2017	China	Vinegar (To discriminate different flavors of vinegar)	8 MOS Model: Lab-made E-nose (PCA, LDA, MLP)	Five different flavors of vinegar (white vinegar, mature vinegar, rice vinegar, balsamic vinegar and apple vinegar)	[171]
2020	China	Chinese vinegar (To identify Chinese vinegar varieties)	10 MOS Model: Lab-made E-nose (PCA, SNV, LDA, KNN)	VOCs from five different kinds of Chinese vinegars (Zhenjiangxiangcu, Zhenjiangchencu, Baoningcu, Hengshunxiangcu and Shanxichencu)	[175]
2021	China	Zhenjiang aromatic vinegar (To differentiate different grades of Zhenjiang aromatic vinegar (ZAV))	10 MOS Model: PEN3 (PCA, PLS-DA)	Furfural, acetic acid, tetramethylpyrazine, phenethyl acetate, phenylethyl alcohol, acetic acid, isobutyric acid, ethyl phenylacetate, 3-hydroxy-2-butanone, 2,3-butanedione, 2-acetylfuran	[176]
Fermented herb products					
2011	China	Green tea (To discriminate the green tea quality)	8 MOS Model: Lab-made E-nose (PCA, KNN, ANN)	Hexenols, pentanal, hexanol, pinene	[182]
2014	Italy	Chinese teas (To discriminate the tealeaf quality)	10 MOS Model: Win Muster v.16 (PCA, CA, LSD)	Aromatic compounds, broad-range compounds, polar compounds, nitrogen oxides and ozone, ammonia, aromatic compounds, aldehydes, ketones, hydrogen, alkanes, less polar compounds, methane, sulfur compounds, terpenes, alcohols	[183]
2016	China	Pu-erh teas (To discriminate two types of Pu-erh teas)	12 MOS Model: Agrinose (PCA)	Alcohol, methoxyphenlic compounds,	[137]
2018	Turkey	Black tea (To monitor the quality control in black tea during fermentation process)	13 MOS Model: Lab-made E-nose (LDA, Bayes, KNN-3 methods)	(E)-2-hexenal, hexenal, (E)-geraniol, linalool, linalool oxide II (cis, furanoid), benzeneacetaldehyde, linalool oxide I (trans, furanoid), 3,7-dimethyl-1,5,7-octatrien-3-ol, benzaldehyde, methyl salicylate	[178,179]

Table 3. *Cont.*

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Refs.
2019	China	Chinese liquors (To discriminate Chinese liquors)	10 MOS Model: Lab-made E-nose (FDPCA, DPCA, PCA, KNN)	Six types of Chinese liquors (Maotai, Gujinggongjiu, Yingjiagongjiu, Haizhilan, Fenjiu, Kouzijiao)	[184]
2022	Sri Lanka	Sri Lankan tea (To monitor the fermentation process of Sri Lankan low-country tea)	4 MOS Model: Digi-Nose (PCA, SVM)	Diethyl phthalate, dimethyl phthalate, Diisononyl phthalate, 2-hydroxyisobutyric acid, 3-penten-2-ol	[89,180]

6. E-Nose for Alkaline Fermentation

Alkaline fermented foods play an important role in many local food cultures around the world. Bacillus spp. bacteria are a major group of microorganisms that react with protein-rich raw materials to produce alkaline fermented food [27]. Additionally, other secondary microorganisms, such as LAB, staphylococci and micrococci, are also involved in this process [185]. Alkaline fermentation involves the breakdown of proteins that release essential peptides and amino acids. Amino acids are broken down into alkaline compounds, such as ammonia, that can raise the pH level (8–10) of the fermented foods. Soybeans are considered one of the common precursors for traditional alkaline fermentation, which can be found in different foods, including Thua-nao of Thailand, Japanese Natto, Indonesian tempeh, Nepalese and Indian Kinema, Chungkookjang of Korea and Chinese Douchi. Generally, in alkaline fermented foods and seasoning agents, it has been found that the released VOCs have distinctive aromas, such as caramel, floral, smoky, malt and the aroma of boiled sweet potato [186]. Volatile substances emitted as gas molecules from each fermented food sample are used to assess the chemical diversity of these molecules, such as acids, alcohols, aldehydes, ketones, esters, furanones, pyrazines and S-compounds [187]. The origin of VOCs generated by the entire fermentation process involves both the raw material and the initial culture used, as well as the parameters used during production. Therefore, odor measurements can be focused on the targeted VOCs released by specific raw materials in each category. For example, MOS sensor arrays were made to analyze ethanol content in soy sauce for halal food certification [188], to investigate the sensory profile of fermented soybean paste (doenjang) [189] and to monitor the fermentation process of tempeh [190]. In addition, odor detection was carried out to determine the quantity and quality of volatile compounds in soybean varieties [191] and to select Chinese soybean paste based on flavor profiles [192] by using HS-SPME-GC/MS with an E-nose.

In addition to the fermented foods mentioned above, there are also alkaline fermented foods from another group of raw materials, such as fish sauce and fermented fish [193]. In the fermentation of fish sauce, enzymes in the body and the intestines of solid fish are gradually activated and changed to liquid, where salt acts as a preservative to prevent the spoilage of the fish. The odor of fish sauce is caused by protein digestion by the digestive juices in fish into peptides, which may be further broken down into amines, ketoacids, ammonia, carbon dioxide, fatty acids, ketones and aldehydes. The amounts of alkalis and acids gradually increase over the fermentation period until approximately nine months, after which they decrease rapidly when the chemical change in fish muscle ends [194]. The amounts of volatile acids and bases still remain after the completion of fermentation. The major characteristics of fish sauce are the unique smell and taste obtained from bacterial activity in the fermentation process. Researchers thus focus on these compounds in their studies on the process of fish sauce production to meet standards, including those for the unique features of each fish sauce. Several studies have shown that the MOS E-nose can classify odors that occur in each stage of fermentation, such as the analysis of traditional fermented fish flavor [195] and the identification of the volatile fingerprint of fermented mandarin fish at different fermentation stages [196], as shown in Figure 7. In the commercial fish sauce industry, the control of total nitrogen content is very crucial for the quality of fish sauce during the fermentation process. Studies have shown the classification of fish sauce based on the level of total nitrogen content [197], which will be useful in selecting the quality of fish sauce. Although there are many different brands of the product, the flavor may be similar; i.e., the 46 commercially available fish sauce products can be grouped into only three major groups [198]. Therefore, the E-nose is still necessary for quality control and the design of new flavors. A list of E-nose applications for alkaline fermentation is summarized in Table 4.

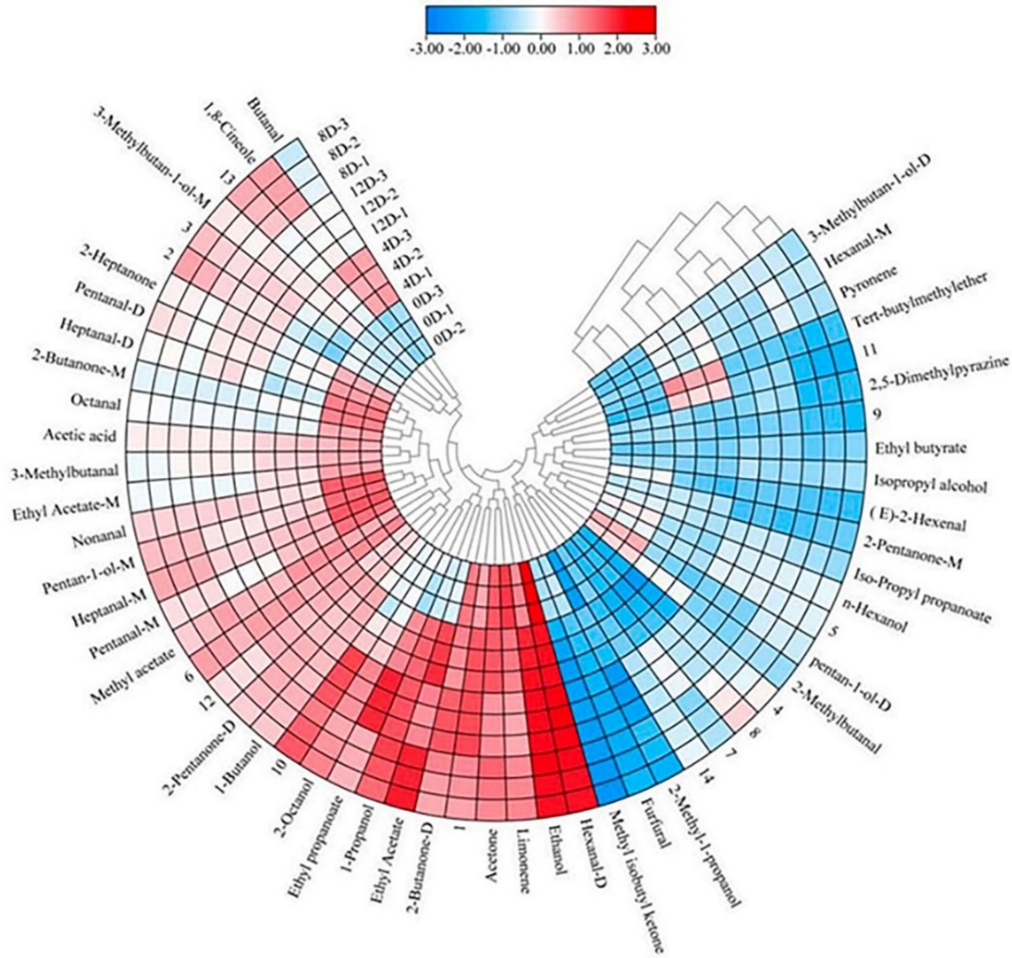

Figure 7. VOC profiles of fermented Mandarin fish samples [196].

Table 4. E-noses for alkaline fermentation.

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
Fermented fish products					
2009	China	Fermented fish (To analyze traditional fermented fish flavor)	18 MOS Model: FOX-4000, Alpha MOS (PCA, SQC)	VOCs from traditional fermented fish	[195]
2014	Thailand	Fish sauce (To classify fish sauce based on the level of total nitrogen content)	6 MOS Model: Lab-made E-nose (PCA, LVQ)	Nitrogen content from fish sauce	[197]

Table 4. *Cont.*

Time	Country	Local Name (Objectives)	Type of E-nose (Pattern Recognition)	Target VOCs	Ref.
2018	Japan	Fish sauce (To classify different smells from 46 commercially available fish sauce products)	18 MOS Model: FOX-4000, Alpha MOS (CA)	VOCs from 46 fish sauce products in several countries (Japan, Thailand, Vietnam, China, Philippines, Italy)	[198]
2021	China	Chinese fermented Mandarin fish (To establish the volatile fingerprint of fermented mandarin fish at different fermentation stages)	18 MOS Model: FOX-4000, Alpha MOS (PCA, PLS-DA, HCA)	Hexanal-D, nonanal, limonene	[196]
Fermented legumes products					
2018	Indonesia	Tempeh (To monitor the fermentation process of tempeh)	8 MOS Model: Lab-made E-nose (PCA, LDA, SVM, KNN)	CO, methane, ethanol, propane, isobutane, hydrogen, ethanol, ammonia, ethanol, toluene, hydrogen sulfide	[190]
2017	Korea	Soy sauce (To analyze ethanol content in soy sauce for halal food certification)	Model: Smart Nose 300 (DFA)	Ethanol in 24 different kinds of soy sauce	[188]
2017	Korea	Soybean paste (To investigate the sensory profiling of fermented soybean paste (doenjang))	Model: Heracles II, Alpha MOS (PCA)	Ethanol, ethyl acetate, ethyl iso-butyrate, pyridine, ethyl 2-methylbutyrate, 1-hydroxy-2-propanone, 1-penten-3-one, 2-ethylfuran, ethyl furan, ethyl butyrate, ethyl isovalerate, 2-methyl-1-butano	[189]
2019	USA	Soybean (To determine the quantity and quality of volatile compounds in soybean varieties)	Model: HERCALES GC Flash electronic nose (PCA)	Ethyl-2-methyl butyrate, 2-methyl propanal, 2-propanol	[191]
2022	China	Chinese soybean paste (To select Chinese soybean paste based on flavor profiles)	Model: HS-SPME-GC/MS with E-nose (LDA, PLSR, SVR)	Esters, total acids, reducing sugar, salinity, amino acid nitrogen	[192]

7. Summary and Future Trends

In summary, E-nose systems have been successfully applied in fermented food and beverage applications. Combined with machine learning algorithms, E-nose systems can be effectively used in qualitative identification for multipurpose objectives. Most gas sensors used in E-nose systems for fermented food and beverage applications are MOS sensors due to their low cost, ease of use, long-term stability and commercial availability. However, the challenge of the E-nose is its limitation in quantitative analysis. E-nose systems have always been coupled with spectroscopies such as GC-MS for both qualitative and quantitative identification in fermented foods and beverages. This is an expensive method, causing the limitation of real-time and online E-nose usage. Nowadays, there are some commercially available E-nose systems, such as Heracles Neo (Flash GC/E-nose), that are able to identify chemical compounds composing the smell and to perform quantitative analysis. In the field of fermented food and beverage applications, most researchers have focused on the development of prototype E-noses by altering the size, delivery system, online/offline platform and algorithms for classification/prediction, not gas sensors. Actually, the gas sensor is the heart of the E-nose instrument because it is sensitive to/selective for functional groups emitted from fermented foods and beverages. Therefore, the integration of highly selective gas sensors, such as electrochemical gas sensors, optical gas sensors and nanocomposite-based gas sensors, with MOS sensors in E-noses may be useful and enable qualitative and quantitative identification of compounds in fermented foods and beverages. The development of highly selective gas sensors for key VOCs emitted from fermented food and beverage products is one of the interesting future trends, as well as the use of a cloud platform that allows researchers to put the huge datasets of signals from MOS sensors into an international database. It will be very useful for both qualitative and quantitative identification by E-noses and for creating and transferring aroma data via the internet (Internet of Smell, IoS) in the future.

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