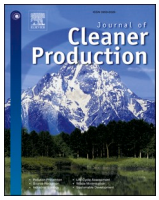
Journal of Cleaner Production 434 (2024) 139961



Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro





Odorous organic gas emission characteristics from cooked food wastes during aerobic decomposition

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ARTICLE INFO

Handling Editor: Panos Seferlis

Keywords:
Odorous organic gases
Cooked food wastes
Leachate
Microbial community structure
Fermentation

ABSTRACT

Emission of volatile organic compounds (VOCs) from food waste is a key trouble for surrounding residents. Herein, the VOC emission patterns from headspace (gaseous) and produced leachate (aqueous) during decomposition of cooked milk cabbage, tilapia fish, and their mixture (Mix) were investigated in self-made reactors. The average amount of gaseous VOCs from fish (2888.7 mg/kg dry matter (DM)) was higher than Mix (2152.9 mg/kg DM) and cabbage (374.2 mg/kg DM). The dominant VOCs were volatile organic sulfur compounds (VOSCs) and oxygenated VOCs, accounting for 65.8 and 32.5 %, 71.8 and 25.2 %, 39.0 and 36.7 % of total VOC yields in fish, Mix, and cabbage, respectively. From leachate, average leaching fluxes of VOCs followed the order of: Mix (2487.9 μg/(L kg) DM) > fish (2374.7 μg/(L kg) DM) > cabbage (368.7 μg/(L kg) DM). VOSCs were also the major VOCs from leachate, accounting for 87.9, 98.6, and 98.5 % of total VOCs in leachate, respectively. *Lactobacillus* and *Weissella* were the dominant genera in cabbage, while *Bacteroides* were dominant in fish and Mix. Physicochemical factors were significantly related to bacterial composition shifts over time. Furthermore, *Lysinibacillus* and *Vibro* were positively correlated with gaseous dimethyl disulfide, while *Lactobacillus*, *Weissella*, *Savagea, Ignatzschineria*, *Acinetobacter*, and *Pseudomonas* showed strong relationship with dominant gaseous and aqueous VOCs. These findings provide significant references for understanding distribution and controlling of VOCs released during cooked food waste fermentation.

1. Introduction

Currently, municipal solid waste (MSW) production is increasing rapidly. According to the World Bank, > 2.01 billion tons of MSW are produced each year since 2016 (Saadoun et al., 2021). In China, annually MSW reaches 23.5 million tons (NBSC, 2020) and approximately 54.5 % is food wastes. Landfill, incineration, and compost are the main MSW disposal methods, and more than half of MSW is sent to landfill (Ding et al., 2021). During landfilling, food wastes with high water content (Portelinha et al., 2020) and organic content (Chew et al., 2018) facilitate rapid reproduction of microorganisms especially pathogens by transferring these degradable components into nutrients. This may cause emission of pathogens and VOCs in landfill (Nie et al., 2019; Zhang et al., 2020, 2023). Emitted VOCs may cause odor nuisances, photochemical smog, and health risks (Luo et al., 2021; Ma et al., 2023). For example, short-term exposure to sulfur compounds, limonene and some esters can acutely irritate eye, nose, throat, and skin of people (Liu et al., 2018). While long-term exposure to VOCs may damage respiratory central nervous system, and cause neurasthenia and cancer (Liu et al., 2016). Therefore, great attention should be paid to these emitted microbial VOCs.

To understand emission mechanism of VOCs from food waste to air during food waste decomposition, the influence factors such as moisture content (Nguyen et al., 2022), raw waste composition (Zhang et al., 2020), air supply (Onwosi et al., 2017), temperature, and primary microbial community (Cui et al., 2022) are considered to simulate food waste decomposition. For example, a significant correlation between emission curve of VOCs and bacterial community is found in different raw wastes (Zhang et al., 2020). The odor emissions during food waste fermentation decrease after adding fungal agent compared with that

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https://doi.org/10.1016/j.jclepro.2023.139961
Received 26 July 2023; Received in revised form 6 November 2023; Accepted 26 November 2023
Available online 1 December 2023
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without adding identical food waste (Zou et al., 2023). When fermentation temperature of fresh food waste increases from 10 °C to 35 °C, VOC yields increase 20 times higher, and the dominant VOCs and bacteria changed from alcohols to esters, corresponding *Weissella* to *Lactobacillu*, respectively (Cui et al., 2022). Unlike raw food waste, cooked fresh food and dining leftovers have different characteristics and microbial community structure, and thus may result in different characteristics of VOC emission (Derqui and Fernandez, 2017; Katajajuuri et al., 2014). However, few studies were conducted on exploring the pollution profile of gaseous VOCs and their relationship with microbial community succession as well as physicochemical property during cooked food waste fermentation.

Besides direct emission of VOCs from food waste, leaching process from food waste to leachate is also an odorous source (Liao et al., 2021). Oxygenated VOCs (OVOCs) and volatile sulfur compounds are the dominant compounds with 2-butanone and H_2S as the main compounds over leachate storage pool (Han et al., 2020; Wang et al., 2021). However, few studies investigate VOC pollution profiles of leachate by Purge and Trap sampler coupled with GC/MS (P&T-GC/MS) even though it has advantages of no organic solvent pretreatment, less sampling, and less matrix interference (Sparkman et al., 2011). Furthermore, contribution of leachate to the odor values, the relationship between VOCs emitted directly from food waste and dissolved in leachate during cooked food wastes decomposition remains unclear.

To address this research gap, the cooked food including milk cabbage and tilapia fish were used as our fermentation targets, which are high consumption by people in the South China. They were decomposed individually or in combination in reactors under aerobic conditions. The profiles of VOCs emitted directly from food wastes and produced leachates during decomposition were identified with GC/MS and P&T-GC/MS, respectively. The succession trends of bacterial community structure were further measured using high-throughput sequencing technique. Principal component analysis (PCA) and spearman correlation analysis was used to analyze similarities and differences of VOCs directly emitted from food waste and dissolved in leachate. Kruskal-Wallis significance test was used to analyze differences of community structure. Furthermore, how physicochemical property and bacterial community structure affect VOC emission were also studied. These findings provide key reference for understanding distribution and controlling VOCs emitted from food waste fermentation.

2. Materials and methods

Experimental materials, instrument analysis methods, data statistical analysis were mainly described in this section.

2.1. Experiment procedure

Three self-made polymethyl methacrylate reactors with the capacity of approximately 56 L each (0.3 m width and 0.8 m height) (Zhang et al., 2020) were used. Before loading waste, a 10-cm height sterilized gravel and a thin glass fiber layer were placed at the bottom of each reactor to form an effective drainage layer. Two kinds of cooked wastes, milk cabbage and tilapia fish, were purchased from a university canteen and divided into three groups, cabbage, fish, and the mixture of cabbage and fish (w/w = 1:1), and each group is 8 kg in weight (Table S1). To accelerate fermentation, all food waste was cut into fragments (0.5 cm × 1 cm) and loaded into reactors. A thermocouple probe was inserted into half depth of waste to monitor its internal temperature in each reactor. During 62-day fermentation, ventilation at a rate of 0.5 L/(min kg) is sufficient to maintain initial aerobic conditions (Wu et al., 2010).

The fermentation gas in the upper level of reactor was collected on days 1–62, using a 2.7-L vacuum stainless Summa canister (Silonite TM, ENTECH, USA). While solid waste (1 g) in different heights and leachate (5–10 mL) at bottom of reactor was collected. The collected solid waste was used for extracting DNA of bacteria and detection of physicochemical properties by using portable multi-parameter analyzer (DZB-718L, China), while the collected leachate samples were divided into two parts for the following analysis: 1) qualitative and quantitative analysis of VOCs with P&T-GC/MS; 2) extracting DNA to study the microbial community.

2.2. VOC analysis with GC/MS

A 7890B GC/5977B MS (Agilent Technology, USA) with a preconcentrator Entech7200 (Entech Instruments Inc., CA, USA) was used to analyze the collected VOCs from upper level of reactor. The sample preparation and analysis, quality control and quality assurance refer to reference (Han et al., 2019). Temperature program, chromatography, mass spectrometry parameter settings are referred to supporting information (SI). Emission fluxes and cumulative emission were calculated refer to reference (Wang and Wu, 2008).

Purge and trap instrument TMR-ATOMX (Agilent Technology, USA) was used with 7890B GC-5977B MS. Standard sample preparation, sample analysis method, quality control and quality assurance were similar to our work (Wang et al., 2023). An appropriate amount of leachate sample was added to a 40-mL amber glass bottle with 1, 4-difluorobenzene and chlorobenzene-d5 as internal standards. The recovery efficiencies of VOCs were in the range of 72–110 %. Instrument setting parameters including column temperature program, ion source, sample preparation are referred to SI.

2.3. Analyses of VOC emission trend with fugacity model

The theoretical volatilization and leaching rates of VOCs from cooked food waste were calculated with fugacity model (Mackay, 2001) and experimental values were also calculated to further determine trend of VOCs. Related parameters are shown in Tables S2 and S3, and specific formulas refer to SI.

2.4. Microbial community analysis

Bacterial genomic DNA was extracted from solid waste and leachate with Rapid Soil DNA Isolation Kit (Sangon, China) and purified with Ezup Column Bacteria Genomic DNA Purification Kit (Sangon, China). Then, the concentration and purity of DNA were detected using NanoDrop 2000 spectrophotometer. Primers 338F (5′-ACTCCTACGGGAGGCAGCAG-3′) and 806R (5′-GGACTACHVGGGTWTCTAAT-3′) were used to amplify the V3–V4 region of the bacterial 16S rRNA gene using the extracted DNA as template. The amplicon was sent to Illumina Miseq platform for sequencing by Majorbio Bio-Pharm Technology Co., Ltd., Shanghai, China.

2.5. Bioinformatics and statistical analyses

The raw data of sequencing were analyzed on the online platform of Majorbio Cloud Platform (www.majorbio.com). Briefly, the effective sequences were clustered into operational taxonomic units (OTUs) at a 97 % similarity level. Then, a bacterial taxonomic classification of each OTU was performed using program against silva 138 database (Yu et al., 2021). Alpha diversity indexes including ACE and Chao indexes were used to calculate community richness, while Shannon and Simpson's index were used to reflect diversity of microbial communities (Li et al., 2020). The differences of bacterial community composition (β diversity) among three groups were measured using Kruskal-Wallis significance test and non-Metric Multidimensional Scaling (NMDS) based on the bray-curtis distance algorithm (Shen et al., 2022). Spearman correlation was used to analyze correlation between VOCs, physicochemical property, and microbial composition at the genus level (Zhang et al., 2020).

3. Results and discussion

The results of gaseous and aqueous VOC characteristic from food waste and bacterial community composition of waste were listed. Further the mechanism of VOC production by microorganisms was discussed.

3.1. VOC emission from cooked food waste

Gaseous VOCs produced by food waste degradation or composting is a significant source of malodor. Total of 59 VOCs were detected by pre-concentrator GC/MS.

3.1.1. The emission characteristics of total VOCs (TVOCs) direct from food waste

During whole fermentation, the emission fluxes of TVOCs reached maximum on day 3 (1343.2 μg/(h kg) DM), day 1 (6393.0 μg/(h kg) DM) and day 3 (6315.5 μg/(h kg) DM) for cabbage, fish and Mix, respectively (Fig. 1A–C), suggesting VOCs were mainly emitted at early fermentation stage. VOC emission fluxes were significantly higher from fish than cabbage during fermentation, and possible reason was that protein content of fish is higher than cabbage (He et al., 2020). Further extending fermentation time of fish and Mix to days 7 and 9, TVOCs first rapidly decreased to 1781.1 and 1837.1 μg/(h kg) DM, and then increased to 5525.8 and 4339.7 μg/(h kg) DM, respectively (Fig. 1B and C). While for cabbage, TVOCs steadily decreased to day 15 (204.2 μg/(h kg) DM) and increased to 549.7 μg/(h kg) DM on day 20 (Fig. 1A). At day 62, emission fluxes of TVOCs were steadily decreased to 259.7, 666.8, and 520.5 μg/(h kg) DM for cabbage, fish and Mix, respectively, due to depletion of degradable substances. Based on PCA of TVOCs produced by three groups, fish and Mix were clustered together and separated from cabbage (Fig. S1), suggesting that TVOCs in Mix were mainly released from fish and there were significant differences of VOCs released from cabbage and fish. Overall, average emission fluxes during 62-day were 468.2, 2890.4, and 2260.7 μg/(h kg) DM for cabbage, fish, and Mix, respectively, and the most VOCs were released at early fermentation stage, especially within the first five days. This indicates that cooked food waste in trash can, garbage transfer vehicle, garbage transfer station and landfill should be disposed of as soon as possible in case of odor emission.

3.1.2. VOC yields and compositions

Specifically, total of 59 VOCs with yields of 2888.7 mg/kg DM were detected in fish, which were 7.7 and 1.3 times higher than that in cabbage and Mix, respectively (Table S4). Based on compound property, VOCs were divided into five groups: volatile organic sulfur compounds (VOSCs), OVOCs, aromatic hydrocarbons (AHs), aliphatic hydrocarbons (AlHs), and halogenated hydrocarbons (HHs). Among them, VOSCs and OVOCs were the most dominant VOCs during whole fermentation, accounting for 7–55 % and 16–78 % of the total emission fluxes in cabbage (Fig. 1D). While their contributions were 45–91 % and 7–54 % as well as 49–92 % and 1–49 % for fish and Mix, respectively (Fig. 1E and F). Other VOCs were minor and altogether shared less than 8 % of TVOCs except for cabbage (14–49 %), which might be that most of AHs (He et al., 2020) and HHs (Komilis et al., 2004) are regarded as xenobiotics and not major released by microbial degradation. For clearly understanding

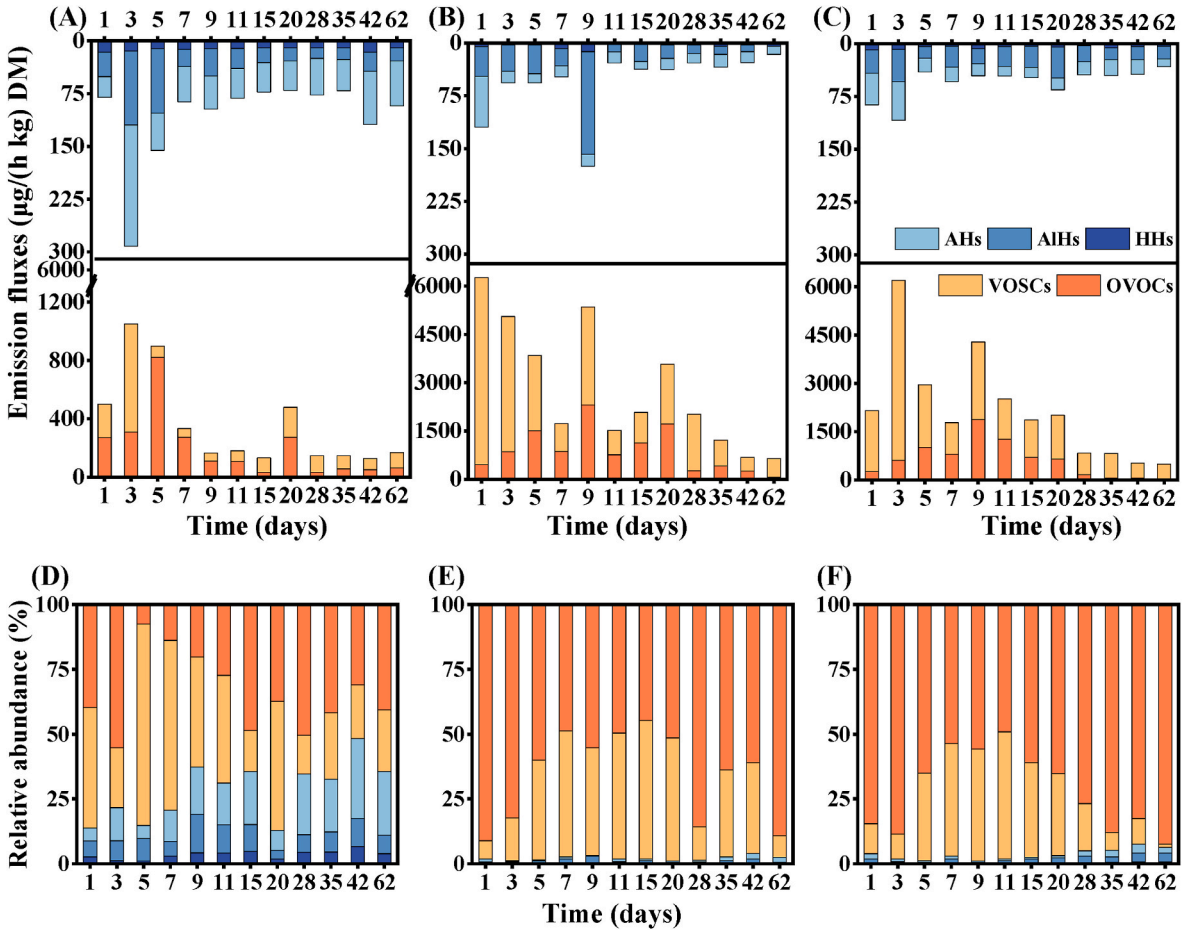


Fig. 1. The volatilization fluxes and relative abundance of VOSCs, OVOCs, AHs, AIHs, and HHs from the headspace of cabbage (A, D), fish (B, E), and Mix (C, F) aerobic fermentation.

VOC emission profiles, pollution profiles of five types of VOCs will be described in detail.

3.1.2.1. VOSCs. The yields of VOSCs from fish were 1899.8 mg/kg, which was 12.8 and 1.2 times higher than that in cabbage and Mix, respectively (Table S4). It is because fish and meat contain higher portion of S-containing amino acids (He et al., 2020; Higgins et al., 2008) than vegetable, which can be degraded into VOSCs by microbes (Cerda et al., 2018).

Dimethyl disulfide (DMDS), dimethyl sulfide (DMS), carbon disulfide (CS_2), and carbonyl sulfide (COS) were the dominant VOSCs emitted directly from food wastes. Among them, DMDS was the most dominant with yields of 112.6, 1874.4, and 1512.7 mg/kg DM and accounted for 75.9, 98.7, and 97.9 % in cabbage, fish, and Mix, respectively. Considering that the odor thresholds of DMDS (47.90 μg/m^3) and DMS (5.89 μg/m^3) are extremely low (Devos and Laffort, 1990), and production of DMDS is high in various food waste composting or degradation (Duan et al., 2014; Nie et al., 2019; Zhang et al., 2020), measures should be taken during food waste decomposition to minimize discomfort caused by odor. Meanwhile, DMDS, DMS, CS_2 and COS join to chemical process of atmospheric aerosol and cloud formation (Mustafa et al., 2017), also needing great attention. Maximum VOSC yield per day was observed on days 3, 1, and 3 for cabbage (17.7 mg/kg DM), fish (139.7 mg/kg DM), and Mix (134.2 mg/kg DM), respectively, then fluctuated to day 20 with cumulative yield reaching 106.6, 1307.0, and 1078.7 mg/kg DM, respectively. After that, VOSC yields were less than 4.4, 44.5, and 32.6 mg/kg DM per day for cabbage, fish, and Mix, respectively. That is, 68.8–71.9 % of VOSC yields in three groups occurred within the first 20 days, suggesting that DMDS yield at early stage is non-negligible whatever types of waste fermentation.

3.1.2.2. OVOCs. Generally, OVOCs are produced by incomplete aerobic fermentation of organic matter. That is, carbohydrates, proteins, and fat in vegetable and meat wastes will be first hydrolyzed to the precursors of OVOCs (e.g., free sugars, amino acids, and fatty acids) (Jinno et al., 2018; Khabibullin et al., 2019), and then broken down by enzymes to form carbonyls, esters, etc (Peterson and Reineccius, 2002). Here, OVOC yields followed the order of: fish (938.6 mg/kg DM) > Mix (543.1 mg/kg DM) > cabbage (139.4 mg/kg DM), with maximum emissions observed on day 9 (55.3 mg/kg DM), 9 (45.0 mg/kg DM), and 3 (19.6 mg/kg DM), respectively (Table S4). It is because fish and Mix contain significantly higher lipid and protein levels than that of cabbage, resulting in significantly higher OVOC yields. Similar to VOSCs, OVOC emissions accounted for 61.4–79.3 % within first 20 days. Specifically, ethyl acetate, 2-butanone, and acetone were the dominant OVOCs with the total yields of 135.4, 929.2, and 535.8 mg/kg DM in cabbage, fish, and Mix, respectively. As reported, high level of ethyl acetate is detected during vegetable waste decomposition (Qu et al., 2022). Acetone is a main intermediate of kitchen waste hydrolysis (Staley et al., 2006). The possible reason for these dominant OVOCs is that volatile fatty acids including acetic acid and lactic acid can be converted to ethyl acetate and ketones (Fan et al., 2019). Furthermore, OVOCs may contribute to ozone formation (Han et al., 2019). Therefore, high emission of OVOCs especially at early fermentation stage may lead to high ozone formation in this study.

3.1.2.3. AHs, AIHs, and HHs. AHs, AIHs and HHs were minor with total yields of 93.0, 50.1, and 64.9 mg/kg DM in cabbage, fish and Mix, respectively, and altogether shared less than 8 % of TVOCs except for cabbage (24 %). Specifically, the yield of AHs of cabbage (49.0 mg/kg DM) was twice that of fish (23.9 mg/kg DM) with toluene and o-xylene as the dominant ones, accounting for 14.0 and 12.4 % of AHs in cabbage, respectively. Besides, naphthalene and toluene were also the dominant AHs in fish (20.3 and 14.9 % of AHs) and Mix (19.9 and 15.0 % of AHs). The maximum emissions of AHs were on the first or third day of each waste.

For AIHs, n-dodecane (5.5 mg/kg DM) and 2,2,4-trimethylpentane (4.5 mg/kg DM) were the top two AIHs in cabbage; whereas, besides n-dodecane (3.6–4.8 mg/kg DM), high levels of 3-methylhexane and 1-hexene also emitted from fish (1.7 mg/kg DM) and Mix (5.0 mg/kg DM), respectively. Unlike VOSCs and OVOCs, the emissions of AIHs main occurred on 20–62 days (accounting for 55–64 %).

Comparatively, yields of HHs were the lowest (0.4–4.7 % of TVOCs). Among seven detected HHs, 1,2,4-trichlorobenzene was the most abundant (accounting for 54–83 % of total HHs). However, for 1,2,4-trichlorobenzene and AHs including naphthalene, their concentration did not change much over time, suggesting that they may be exogenous substances rather than microbial metabolites regarding they are difficult to be degraded by microorganisms (He et al., 2020).

3.2. The leaching characteristic of VOCs from food waste to leachate

Leachate produced by food waste degradation or composting is also a significant source of malodor (Liao et al., 2021; Safianowicz et al., 2018) and aqueous VOCs in leachate were qualitatively and quantitatively analyzed with P&T-GC/MS in this study. Total of 32 VOCs including 3 VOSCs, 15 AHs, and 14 HHs were detected (Table S5). With the extension of decomposition time from days 1–20 and to 62, VOC emission fluxes increased from 125.1 to 938.9 μg/(L kg) DM and then decreased gradually to 144.3 μg/(L kg) DM in cabbage (Fig. 2A). Differently, VOC emission fluxes reached maximum on day 1 (7272.3 μg/(L kg) DM), and then decreased rapidly to 104.8 μg/(L kg) DM on day 3 for fish (Fig. 2B). For Mix, the maximum emission fluxes of VOCs occurred on day 11 (4870.6 μg/(L kg)DM), which was earlier than cabbage but later than fish (Fig. 2C). The average emission fluxes within first 15 days (200.7 μg/(L kg) DM) were lower than that in the latter 34 days (620.6 μg/(L kg) DM) in cabbage. Generally, volatility of a compound depends on its saturated vapor pressure, which depends on temperature (Cabrerizo et al., 2011). The average temperature in reactor with cabbage within 3–15 days was (27.5 °C) higher than 20–62 days (24.6 °C) (Fig. S2A). Further, PCA of VOC emission characteristic from leachate showed that major days in fish and Mix had variance with cabbage on PC2 (except day 3, 20, 28, and 46 in fish, except day 1, 3, 15, and 46 in Mix), which is consistent with PCA of VOCs emitted directly from cooked food waste (Fig. S3).

In addition, the order of average 62-day leaching fluxes followed as Mix (2487.9 μg/(L kg) DM), fish (2374.7 μg/(L kg) DM) and cabbage (368.7 μg/(L kg) DM). The highest VOC emission fluxes from leachate were observed in Mix, which might be caused by the low average temperature during fermentation. Similarly to VOC emissions direct from cooked food waste, VOSCs in leachate were dominant during fermentation, accounting for 87.9–98.5 % of TVOCs in all leachates, followed by AHs (1.0–9.5 %) and HHs only accounted for 0.3–2.6 % (Fig. 2D–F). Among VOSCs in leachate, DMDS was also the most abundant, accounting for 40.3, 78.7, and 65.8 % of VOSCs in cabbage, fish, and Mix, respectively, followed by DMS. Butyl mercaptan only accounting for 1.0–7.9 % among VOSCs (Fig. S4), the possible reason was that mercaptan are converted into thioethers (Xi et al., 2023). For AHs in leachate, 1,2,4-trimethylbenzene and naphthalene were the two highest proportions, accounting for 22.2 and 16.7 %, 21.4 and 9.0 %, as well as 23.3 and 8.6 % in cabbage, fish, and Mix, respectively (Fig. S4).

3.3. Comparison of VOCs released to air and dissolved in leachate

The distribution characteristics of VOCs released to air and dissolved in leachate were also evaluated by fugacity model and experimental results. The trends of VOCs were characterized by the mean ratios of VOC volatilization rates divided by VOC leaching rates during fermentation (Fig. 3). The simulated ratios of VOSCs (including DMDS and DMS) and HHs (including 1,2,4-trichlorobenzene and chlorotoluene) were at 10^{*4} in three kinds of waste. Most values of AHs were greater

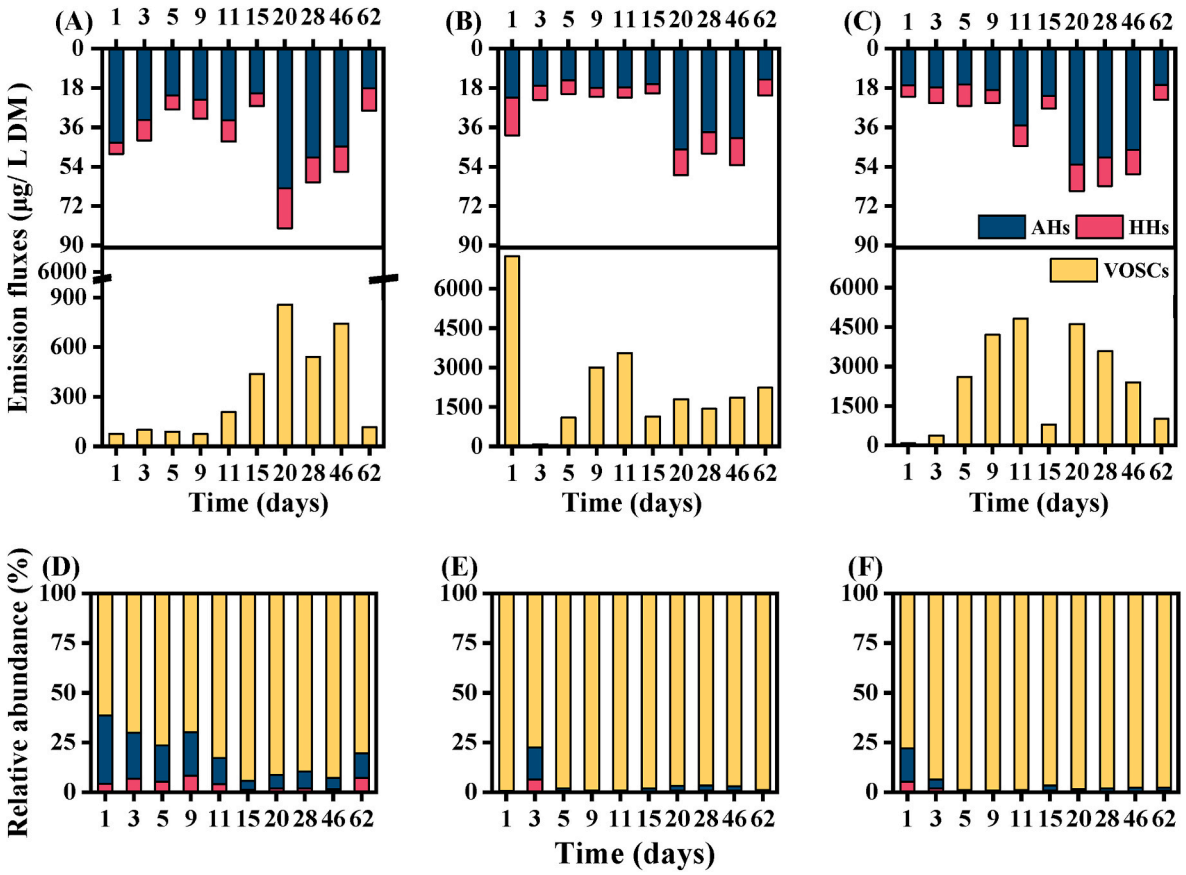


Fig. 2. The leaching fluxes and relative abundance of VOSCs, AHs, and HHs from the leachate produced by the aerobic fermentation of cabbage (A, D), fish (B, E) and Mix (C, F).

than $10^{\wedge}5$ except for styrene ($10^{\wedge}4$) in cabbage and fish, while values of 1,2,4-trimethylbenzene, styrene, m-xylene, p-xylene, toluene, and benzene in Mix were at $10^{\wedge}4$. The possible reason for this is that the total leaching flow in Mix was the largest caused larger value D_L. For experimental results on VOC volatilization rates divided by VOC leaching rates, average ratios of VOSCs (including DMDS and DMS) and AHs (including 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene, p-xylene, cumene, n-propylbenzene, and ethylbenzene) were followed by fish > cabbage > Mix. These results were consistent with the theoretical results that ratios in fish were the maximum. As known, volatilization of VOCs is related to Henry's coefficient, which is positively correlated with temperature (Zhang et al., 2022). The average temperature during fish fermentation was the highest within initial 28 days (Fig. S2A). However, the ratios of AHs (including styrene, m-xylene, toluene, and benzene) and HHs (including 1,2,4-trichlorobenzene and chlorotoluene) were not highest in fish, suggesting that besides temperature, the media where the pollutants presented also affect fate of the pollutants to a certain extent. VOCs, mainly are VOSCs and AHs, produced from food waste tend to volatilize in accordance with theoretical results. The difference between the simulation ratios and the measured ratios was 0–3 orders of magnitude, the possible reason was that some parameters, mass transfer coefficient K_V, molecular diffusivity in air B_{MA}, etc., may have deviation this experiment. Secondly, the environment of food waste was complex media.

To further clarify association between VOCs released to air and dissolved in leachate, the same VOC species were selected and analyzed by PCA. As Fig. S5 shows, trend was responsible for most of the variance (PC1), explaining volatilization and leaching distribution characteristics of VOCs of each type food waste were different, especially for cabbage. PC2 explained 40.2 % of total variance, suggesting distribution characteristics of VOCs from fish and Mix were similar but different with cabbage, whatever in volatilization or leaching, which corresponds to the above VOCs distribution in air and leachate in three groups. Then, the relevance between volatilization and leaching VOCs was further evaluated (Fig. 4). AHs in gaseous phase including benzene, p-xylene, cumene and toluene were negatively correlated with that in aqueous phase, styrene was positively correlated with that in aqueous phase ($p \leq 0.05$) for cabbage (Fig. 4A); while AHs in aqueous phase including 1,2,4-trimethylbenzene, styrene, n-propyl benzene was positively correlated with that in gaseous phase ($p \leq 0.05$) for fish (Fig. 4B). Naphthalene and toluene in aqueous phase had a negative correlation with those in gaseous phase for Mix ($p \leq 0.05$) (Fig. 4C). This suggested that AHs from food wastes and corresponding leachates were closely related to each other, and the trends of styrene and toluene were same in different type food wastes. Besides, the relationship between the dominant gaseous VOSCs and other gaseous VOCs was evaluated. In fish, DMDS had a significantly positive correlation with naphthalene ($p \leq 0.05$) (Fig. 4B). While in Mix, DMDS had a positive relationship with AHs (m-xylene, p-xylene, and toluene) ($p \leq 0.05$) (Fig. 4C). For DMS, it was significantly correlated with cumene in fish (Fig. 4B), as well as AHs (1,2,4-trimethylbenzene, styrene, p-xylene, toluene, benzene) and AIHs (chloroform, 1,2,4-trichlorobenzene) in cabbage (Fig. 4C). This suggests that specific microbial metabolism produces various VOCs. By analysis the correlation between the predominant VOSCs in gaseous and aqueous phases, we found that DMDS in gaseous phase also had a significant correlation with AHs (1,2,4-trimethylbenzene, styrene, 1,3,5-trimethylbenzene, n-propylbenzene and cumene) in aqueous phase (Fig. 4C). The possible reason is that the change of VOCs in a specific environment. The above results suggest that in complicated media, volatilization and leaching VOCs produced by cooked food wastes had complex association. Overall, these correlations were of meaning to our in-depth understanding of VOC emission and odor control in food waste related

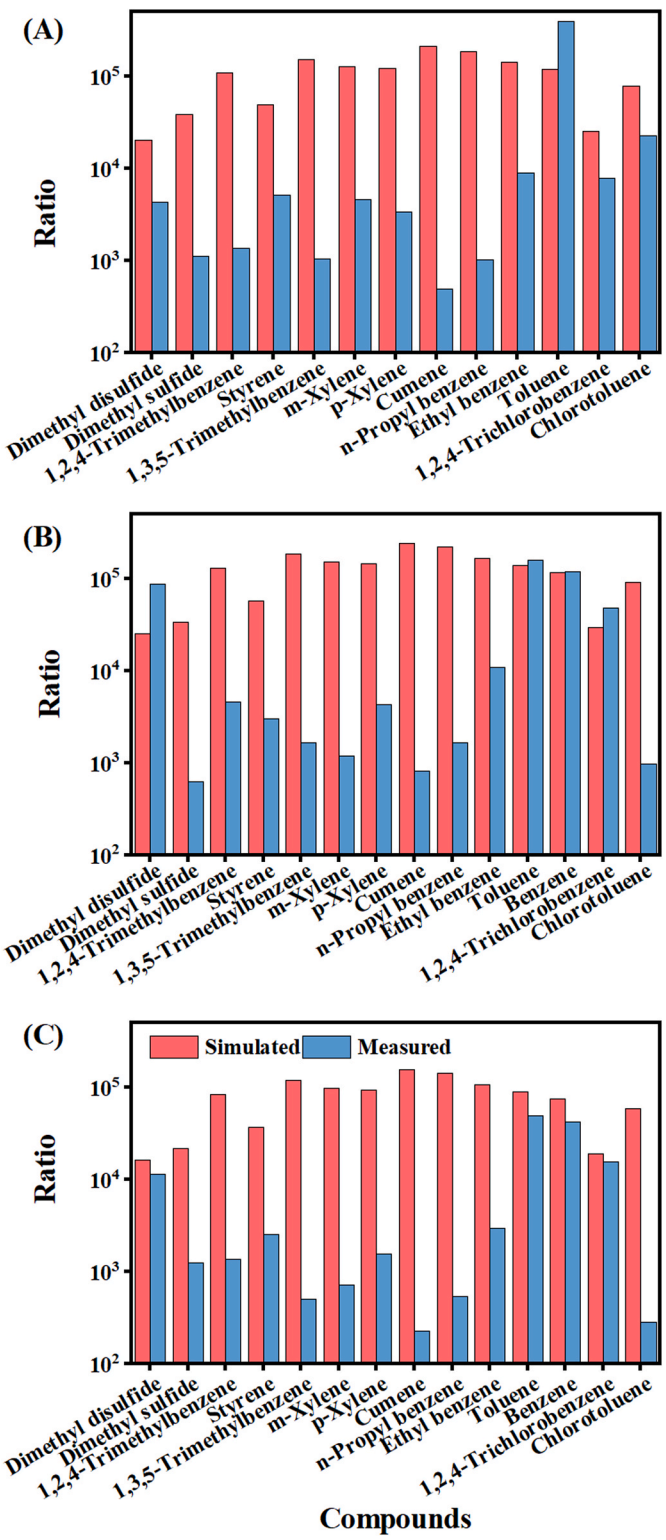


Fig. 3. Theoretical simulated and experimental measured mean ratio of VOC volatilization rates divided to VOC leaching rates during fermentation of cabbage (A), fish (B), and Mix (C).

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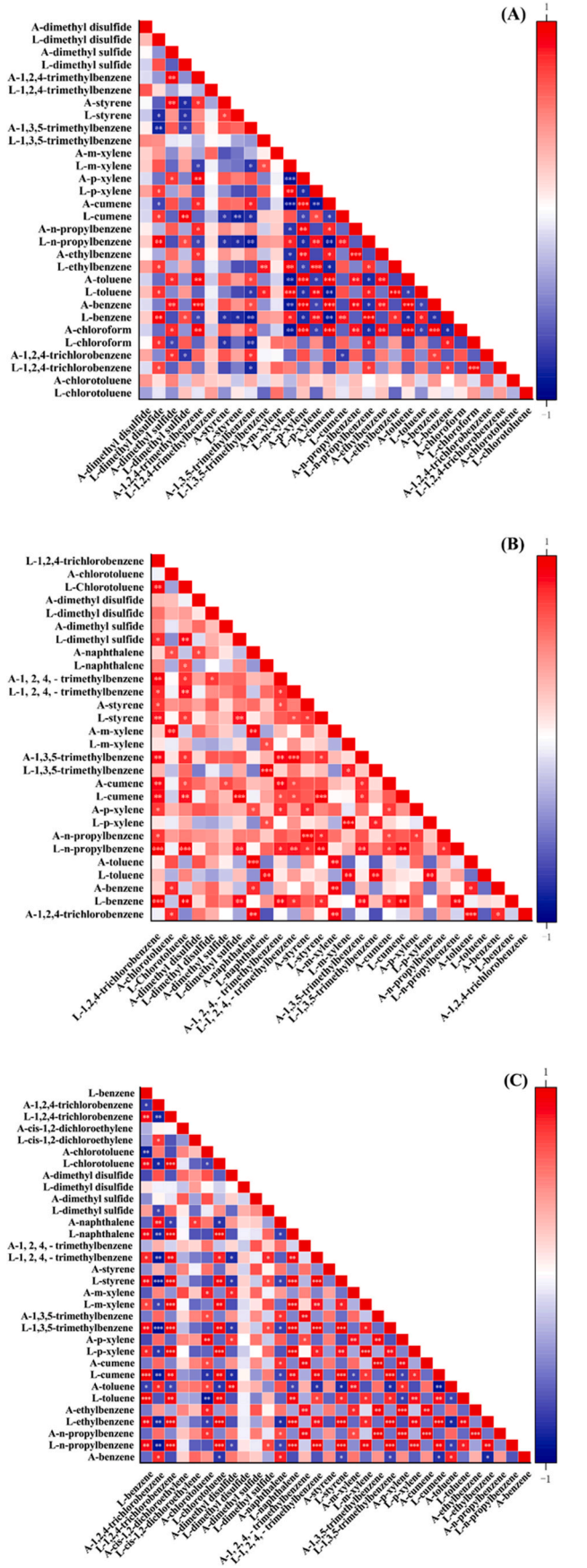
3.4. Bacterial diversity

VOCs produced from cooked food wastes are closely related to the microbial community during their fermentation (Cui et al., 2022). Therefore, to characterize the shifts in microbial community structure over time, DNA mixture of food waste and leachate was used for high-throughput sequencing. To investigate the bacterial community diversity and richness, the alpha indexes were evaluated (Table S6). The ACE and Chao indexes of cabbage and Mix increased from days 1–62, indicating that the bacterial community was becoming richness during cabbage and Mix fermentation. However, for fish, the community richness fluctuated slightly. The average value of the Shannon, ACE, and Chao indexes followed the order of: cabbage > Mix > fish. One possible reason is that high concentration of VOCs may have a toxic effect on the bacterial community. The harsher the environment, the simpler the community structure (Li et al., 2020).

3.5. Bacterial community composition

Fig. 5 shows the top 30 bacterial genera detected in three kinds of waste. Among them, *Bacteroide* and *Savagea* as well as *Bacteroide* and *Acinetobacter* were the two most abundant genera in fish and Mix, respectively. While in cabbage, *Lactobacillus* and *Weissella* were the most abundant genera. In specific, *Bacteroides* accounted for 38.4 and 26.9 % on average in fish and Mix, respectively. The highest abundance of them were observed on days 20 (81.7 %) and 15 (71.0 %) in fish and Mix, respectively. This is because *Bacteroides* grow well at pH 7–8 (Xiong et al., 2019), and in this study, pH at middle and later stages of fish and Mix decomposition was close to neutral and higher than that earlier stage (Fig. S2B). Comparatively, pH in cabbage were significantly lower than that in fish and Mix (Fig. S2A), *Bacteroides* only appeared at day 15 when pH increased to 6.36 and only accounted for 0.8 %. One possible reason for lower pH at early stage of cabbage fermentation (pH 4.09–4.33 from days 1–5) is that it had high abundance of *Lactobacillus* (accounting for 9.7–46.7 % in the first five days). *Lactobacillus* can produce organic acids, resulting in rapid pH reduction during fermentation (Wang et al., 2019). Besides, lactic acid bacteria *Weissella,* which was accounted for 11.1–58.0 % in the first five days, could also produce acetic acid. Previous studies also demonstrated that *Lactobacillus* and *Weissella* are usually found at early stage of food waste composting along with low pH (Sundberg et al., 2013; Tran et al., 2019). In Mix, *Acinetobacter* was also dominant (accounting for 26.1–44.7 %) in the first 9 days, obviously higher than that in cabbage (accounting for 1.2–8.6 %) and fish (accounting for 0–31.3 %). It is because *Acinetobacter,* with ability to decompose cellulose, is a strictly specific aerobic gram-negative bacterium that grows easily at pH 5.5–6.5 (He et al., 2019; Lopez-Dominguez et al., 2019). Since the average pH value was 6.02 of Mix leachate is more suitable for *Acinetobacter* growth, it was dominant in Mix within first 9 days. While for fish, as the second most abundant genera, *Savagea* was firstly observed on day 5 and then increased with the increased decomposition time with average proportion of 10.0 %. Besides, *Lysinibacillus* involved in the conversion of dissolved organic matter (Ma et al., 2022) also accounted for 8.0 % in fish and was mainly observed at the early fermentation stage (days 1–9). *Pseudomonas,* which can effectively degrade organic waste to acetic acid and butyric acid (Palaniveloo et al., 2020) was not negligible in cabbage was 7.2 % and Mix was 6.6 %.

To further understand the changes of bacterial community structure, non-metric multidimensional scaling (NMDS) analysis was constructed. As shown in Fig. 6, bacterial community in cabbage was completely separated from that in Mix and fish, both of which were clustered together. These suggest that the bacterial composition in Mix and fish were very similar, but different from cabbage. These results were consistent with PCA of gaseous and aqueous VOCs, further demonstrating that VOCs were produced by microbial decomposition food waste. Using a one-way ANOVA for comparison, 10 bacterial genera were statistically different ($p < 0.05$) between cabbage and fish, while eight bacteria genera were significantly different ($p < 0.01$) between three kinds of waste (Fig. S6). No matter in two-group or three-group comparison, *Pseudomonas* and *Comamonas* were statistically higher in



(caption on next column)

Fig. 4. Correlation analysis between VOCs in gaseous and aqueous phases: (A) cabbage, (B) fish, (C) Mix. "*, **, ***" represent the $0.01 \leq p \leq 0.05$, $0.001 \leq p \leq 0.01$, and $p \leq 0.05$, respectively. The color of the square represents the correlation, the red represents the positive correlation, and the blue represents the negative correlation. "A-styrene" represents styrene in gaseous phase. "L-styrene" represents styrene in aqueous phases. Other names have similar meanings. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

cabbage, while *Savagea* and *Lysinibacillus* were higher in fish. However, whether these dominant bacteria being related to VOC emissions deserve further discussion.

3.6. Correlations between bacterial community composition and physicochemical properties as well as VOCs

To determine whether physicochemical properties of wastes or dominant VOCs associated with the bacterial community structure in wastes, distance-based redundancy analysis (db-RDA) was performed. As Fig. S7 shows, physicochemical properties of waste including oxidation-reduction potential (ORP), pH, temperature (T) and electric conductivity (EC), as well as gaseous VOCs including DMDS, DMS, CS_2, COS, 2-butanone, ethyl acetate and acetone are the primary factors accounting for the variation of bacterial community structure. The pH, EC had a negative correlation with bacterial community in cabbage and a positive relationship with bacterial community in most fish (except for days 1 and 3) and Mix (except for days 1, 3, and 5). This was consistent with above-described bacterial community behavior under different pH values. While OPR showed positive correlation with bacterial community in cabbage as well as the early edge of bacterial community of fish (days 1 and 3) and Mix (days 1, 3 and 5), and the positive selection effect was stronger at early stage than middle and late stages of fermentation except on the 9th day in cabbage. It is because at early stage, ORP was obvious higher than latter in three wastes (Fig. S2D), suggesting adequate oxygen was conducive to aerobic microbial growth, once the oxygen was limited may be inclined to the growth of anaerobic bacteria.

The correlation heat map between the top 20 genera and physicochemical properties of waste was analyzed (Fig. S8). The pH and EC were significantly positive correlated with *Koukoulia*, *Bacteroides*, *Savagea*, *Tissierella*, *Ignatzschineria*, and *dysgonomonas* ($p \leq 0.05$), and this result was consistent with above mentioned the popularity of *Bacteroides* and *Savagea* in both cabbage and fish, indicating that pH and EC played a key role in the succession of bacterial communities during fermentation. Different from pH and EC, ORP, which reflects macroscopic redox property of all substances, showed an opposite correlation with bacterial communities. Besides, *Pseudomonas* and *Comamonas* were positively related to T, suggesting that higher temperature is favorable for their growth and further affected metabolism of food waste.

From heat map (Fig. 7), the correlation between the top 20 genera and dominant gaseous as well as dominant aqueous VOCs was further discussed. The gaseous n-dodecane, CS_2, and COS were positively correlated with *Lactobacillus* and *Weissella*, *Acinetobacter* and *un_Enterobacteriaceae,* the dominant genera in cabbage and Mix, respectively ($p \leq 0.001$). OVOCs including acetone and 2-butanone showed great correlation with *Lysinibacillus,* which agree with the finding that *Lysinibacillus* can produce ketones through the utilization of glucose (Zheng et al., 2022). DMDS was positively related with *Lysinibacillus and Vibrio*, and DMS was positively related with *Pseudomonas* ($p \leq 0.001$) (Fig. 7A). It has been reported that *Pseudomonas* can produce DMS and DMDS under laboratory culture conditions (Elkahoui et al., 2015; Morales et al., 2005).

For aqueous VOCs, DMDS, DMS in leachate were positively related with *Bacteroides* and *Tissierella* ($p \leq 0.05$). AHs in aqueous phases including 1,3,4-trimethylbenzene, styrene, 4-isopropyltoluene, and butylbenzene in leachate were positively correlated with *Pseudomonas* ($p \leq 0.05$) (Fig. 7B). The same VOCs in different phases were

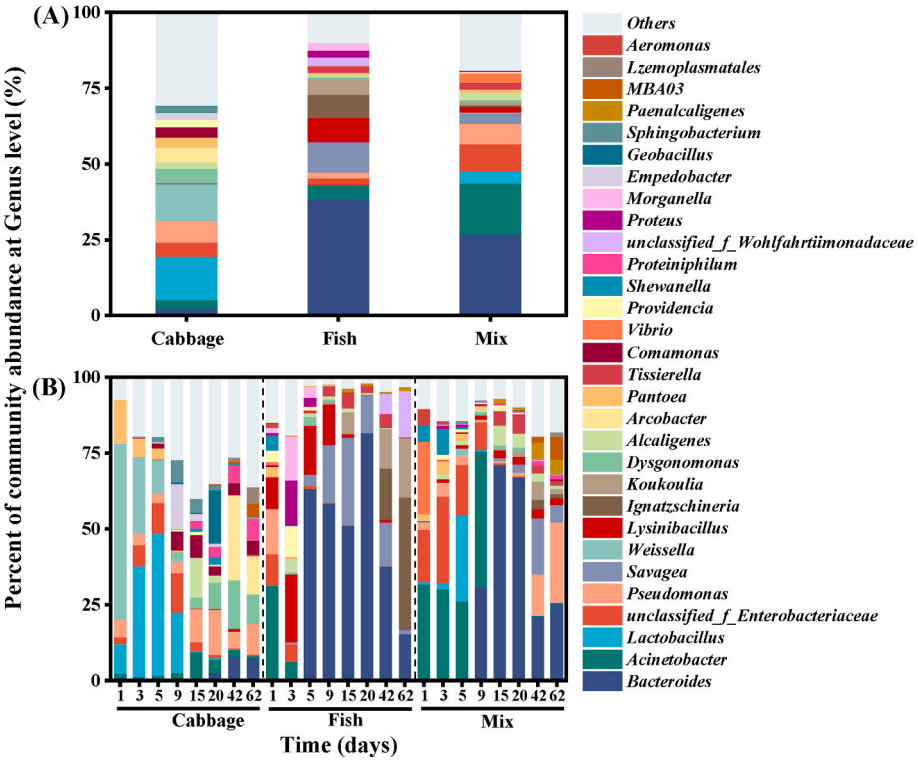


Fig. 5. Relative inter-group (A) and intra-group (B) species abundance at the genus level during the aerobic fermentation of cabbage, fish, and Mix.

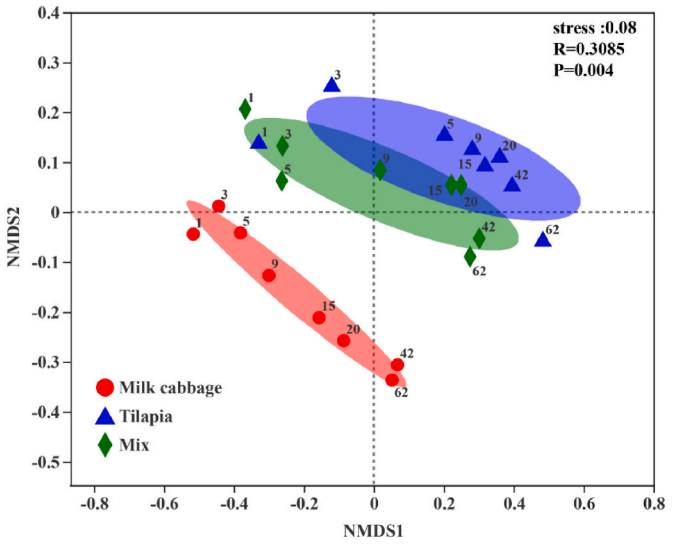


Fig. 6. NMDS ordination based on Bray-Curtis similarities of OTU at different decomposition days of cabbage, fish, and Mix.

significantly correlated with different genera, the possible reason was that the physicochemical properties of waste, which affects the distribution of VOCs produced. Therefore, for the controlling of odorous gas, further work needed to focus on verifying the relationship between the dynamic change of VOCs and microbial succession.

4. Conclusions

During decomposition, more than 50 and 30 kinds of VOCs emitted directly from cooked food wastes (gaseous VOCs) and produced leachates (aqueous VOCs), respectively. The total volatilization yields followed the order of: fish > Mix > cabbage; and average leaching fluxes followed the order of: Mix > fish > cabbage. DMDS, ethyl acetate, 2-butanone, acetone, carbon disulfide and DMS were dominant gaseous VOCs; and DMDS, DMS, butyl mercaptan, 1,2,4-trimethylbenzene were dominant aqueous VOCs. The trends of VOC tend to volatilize from waste to air, and a significant association between gaseous and aqueous VOCs was observed. *Lactobacillus* and *Weissella*, *Bacteroide* and *Savagea* as well as *Bacteroide* and *Acinetobacter* were the most abundant genera in cabbage, fish, and Mix, respectively. *Lysinibicillus* and *Vibrio*, *Bacteroides* and *Tissierella* were strongly related with gaseous and aqueous DMDS, respectively.

Furthermore, the gaseous VOSCs and OVOCs released from fish waste were much higher than cabbage waste, possibly due to high content of sulfur-containing protein and low porosity of fish. Therefore, when dealing with meat waste, straw and other fillers can be added to increase the C/N ratio and increase the oxygen content to reduce the release of gaseous and aqueous odorous gases. VOSCs, the main VOCs released of the three food wastes, were significantly positively correlated with *Lysinibacillus* and *Vibrio*, meaning that related bacteria agent can be appropriately added to accelerate fermentation. These results will provide further insights into the diversity and variability of VOCs and the succession laws of bacterial community structure in waste bins, waste transfer stations, landfill sites, and food waste related industries. However, this study had some limitations. Firstly, when VOC emission trend was analyzed using Fugacity model, the used classical parameters such as B_{MA}, K_V have deviation in self-made reactor and actual environment, which may affect the accuracy of theoretical value and universality of this study. Secondly, only cabbage and fish were chosen as fermentation targets, which might also affect the universality of this work. Further study is still needed to make the related knowledge can be improved.

CRediT authorship contribution statement

Zuming Wang: Data curation, Formal analysis, Writing – original draft, Conducted majority experiments, data analysis and paper

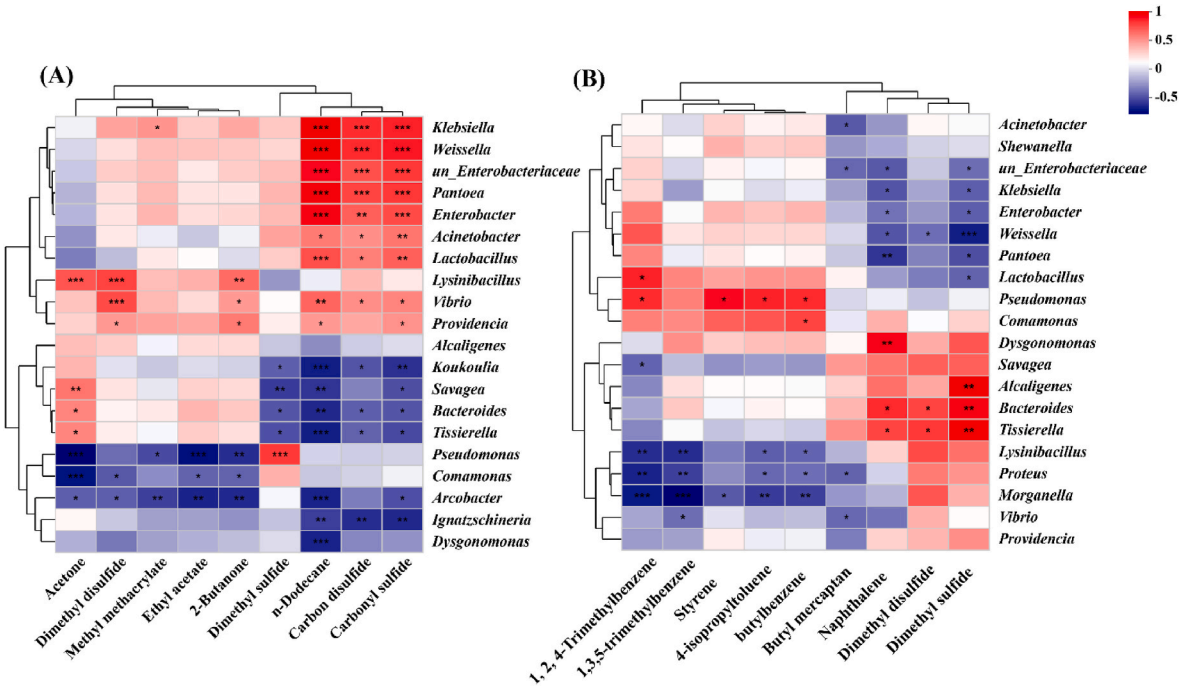


Fig. 7. Spearman correlation analysis the relationship between relative abundance of top 20 genera and dominant gaseous VOCs (A) as well as dominant aqueous VOCs emitted from the food waste (B). "*, **, ***" represent the $0.01 \leq p \leq 0.05$, $0.001 \leq p \leq 0.01$, and $p \leq 0.001$, respectively and correlation coefficient icon was on the right, the deeper the color, the greater the correlation. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

drafting. **Zhishu Liang:** Methodology, Data curation. **Guiying Li:** Supervision, Writing – review & editing. **Taicheng An:** Conceptualization, Supervision.

Declaration of competing interest

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

Data availability

No data was used for the research described in the article.

Acknowledgments

This work was funded by National Natural Science Foundation of China (U1901210 and 41877363), Local Innovative and Research Teams Project of Guangdong Pearl River Talents Program, China (2017BT01Z032), and Young S&T Talent Training Program of Guangdong Provincial Association for S&T (GDSTA), China (2022QNRC23).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.139961.

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