Science of the Total Environment 717 (2020) 137175

Contents lists available at ScienceDirect

Science of the Total Environment

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

Malodorous gases production from food wastes decomposition by indigenous microorganisms

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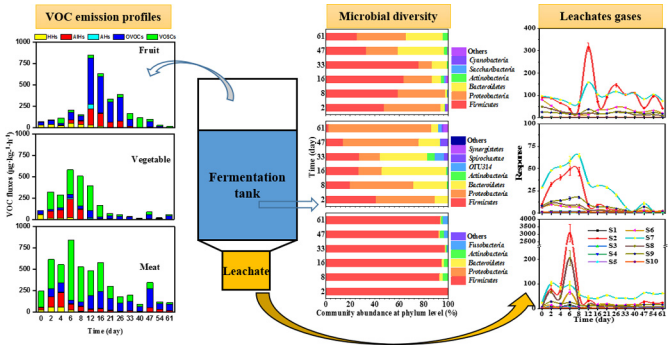
HIGHLIGHTS

- Higher VOC emissions were detected during the decomposition of meat wastes.
- The key odor pollutants were 2-butanone and ethyl acetate from fruit wastes.
- Dimethyl sulfide & dimethyl disulfide were key VOCs from meat & vegetable wastes.
- The bacterial communities changed significantly during food wastes decomposition.
- Significant positive correlation between VOCs and bacteria were observed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 11 December 2019
Received in revised form 4 February 2020
Accepted 6 February 2020
Available online 08 February 2020

Keywords:
Food wastes
Decomposition
Malodorous gas emission
Microbial diversity

ABSTRACT

Volatile organic compounds (VOCs) produced during the degradation of food wastes may harm to the health of people and create annoyance in adjacent communities. In this work, the VOCs emitted from the decomposition food wastes including fruit, meat and vegetable, and their microbial communities were measured in three individual 57-L reactors for 61 days. Total of 232.8, 373.5, and 191.1 $\mu g \cdot kg^{-1} \cdot h^{-1}$ VOCs with oxygenated VOCs (57.6%), volatile organic sulfur compounds (VOSCs, 58.6%) and VOSCs (54.9%) as the main group were detected during fruit, meat and vegetable fermentation, respectively. 2-Butanone (55.1%) and ethyl acetate (13.8%) were the two most abundant VOCs from fruit wastes, while dimethyl sulfide (68.0 and 26.6%) and dimethyl disulfide (89.2 and 10.1%) were in vegetable and meat wastes. The predominant Firmicutes represented 93.0–99.9% of the bacterial communities of meat decomposition, while Firmicutes and Proteobacteria were the dominant phyla throughout the fruit digestion process. Proteobacteria (16.9%–83.6%) was the dominant phylum in vegetable wastes, followed by Bacteroidetes, Firmicutes, and Actinobacteria. Malodorous VOCs emissions were highly affected by microbial activity, the abundant *Weissella, Leuconostoc* and *Enterobacteriaceae* in vegetable wastes showed correlation with carbon disulfide and dimethyl sulfide, while dominant *Peptococcus, Bacteroides, Lactobacillales* and *Peptoniphilus* in meat wastes was related to dimethyl disulfide. Overall, significant differences and correlation between VOCs emission profiles and bacterial communities among different food wastes decomposition were observed. These data contribute to a more comprehensive understanding the relationship between microbial community dynamics and malodorous VOCs emission.

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https://doi.org/10.1016/j.scitotenv.2020.137175
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1. Introduction

Huge amount of domestic wastes has been produced in China, and > 50% of these wastes are food wastes (Ma et al., 2019). It is estimated that the total volume of food wastes generated in China was approximately 108 million tons/per in 2018, which is higher than India (72 million tons) (Thi et al., 2015), United States (34.8 million tons) (EPA., 2018), and European Union (34.8 million tons) (Zheng et al., 2020). Excess food wastes are not just a waste of resources and energy, but also causing serious environmental pollution and complaints during their decomposition process, considering that they are rich in biodegradable organic matters. Thus, food wastes pose seriously adverse effects on the surrounding residents and become a problem non-negligible (Staley et al., 2006).

Previous studies have demonstrated the emission of volatile organic compounds (VOCs) from wastes, but most were focused on a single substance in a laboratory or a digestion plant and landfill (Carriero et al., 2018; Zheng et al., 2020). Few studies have measured the composition and evolution of VOCs from laboratory-controlled decomposition of food wastes. For instance, the emission of oxygenated VOCs (OVOCs) including ethanol, methanol, ethyl acetate, methyl acetate, 2-butanone and acetaldehyde were detected during the aerobic decomposition of orange wastes (Wu and Wang, 2015). Further, up to 409.9 mg kg^{-1} volatile organic sulfur compounds (VOSCs) were detected during aerobic decomposition of food wastes collected from typical urban residential communities in an incubator for a period of 41 days (Wu et al., 2010). Komilis et al. (2004) found that yard wastes decomposition primarily produced terpenes, alkylated benzenes, ketones and alkanes, while food wastes primarily produced sulfides, acids and alcohols (Komilis et al., 2004). However, the comparison of VOCs emission profile from the three largest food wastes including fruit, vegetable and meat was long been overlooked and needed more attention.

Obviously, the emission of these malodorous gases and the decomposition of the food wastes usually are inseparable from the activities of microorganisms. Many previous studies have demonstrated that various indigenous microbes in wastes play a key role in wastes decomposition (Bareither et al., 2013; Slezak et al., 2015). For instance, identified with partial 16S rDNA sequencing, Noseda et al. (2012) found that the dominant bacteria at the end of the shelf life of fillets were generally Gram-negative bacteria, most of which belong to the genera of *Serratia* and *Pseudomonas* (Noseda et al., 2012). By investigating bacterial metabolic function as well as community structure during solid wastes decomposition in lab, Yang and song (2019) found that bacterial community structure and diversity were different in anaerobic acid phase, aerobic phase, and methanogenic phase (Yang and Song, 2019). In a single-stage bioreactor, the municipal organic solid wastes were anaerobically digested and the result showed that the bacterial community was consisted of at least 21 bands of bacteria and archaea at the steady state (Wan et al., 2013). Further, Odeyemi et al. (2018) characterized the bacterial community composition in food spoilage, and a bacterium identified as *Shewanella Baltica* was isolated from mussels with high enzymatic activity for H_2S production (Odeyemi et al., 2018). However, few studies investigated the function of bacteria in the emission of VOCs (Chen et al., 2017; Zhu et al., 2010), let alone evaluate the relationship between the microbial community structure and malodorous gas emission during food wastes decomposition processes.

In this study, therefore, three types of non-recyclable and biodegradable household garbage including fruit, vegetable, and meat were decomposed in the lab. The main objectives are: (1) to study the components and variation of malodorous gases produced during the decomposition of these different food wastes; (2) to investigate the evolution of bacterial community structure at the same time; (3) to interpret the relationship between the malodorous gas emission and bacterial community in the fermentation of food wastes. These data provide insights into the relationship between malodorous gas emission and microbial community dynamics, and advice on malodorous gases control.

2. Materials and methods

2.1. Experimental design

The decomposition experiments of three kinds of food wastes were carried out in three identical self-made reactors (Fig. S1). The reactor (0.3 m i.d × 0.8 m height) consisted of a polymethyl methacrylate tank for food wastes fermentation and a cylindrical container for leachate collection. A plate uniformly distributed with 0.5 cm holes was placed between the tank and container to prevent the clogging of the reactor. Besides, gravel (5 kg) and glass fabric were placed at the base of the tank to form an effective drainage layer during leachate collection.

Food wastes, which were discarded by shopkeepers, were collected from Tangde food markets in Guangzhou, China on August 24, 2018. Food wastes include vegetable trimmings, fruit peelings, meat scraps, excess or spoiled prepared food and were classified into three groups: vegetable, fruit and meat as described in Table S1, and decomposed in three separate reactors. To accelerate the decomposition process, larger pieces of food wastes were shredded by a sterile knife into approximately 2 cm pieces in size. A flange was used to seal the fermentation reactor, and an air pump was used to continuously introduce air into reactor with an aeration rate of 0.2–0.5 $L \cdot min^{-1} \cdot kg^{-1}$. To maintain enough leachate, 100 mL of deionized water was added periodically to the reactor and re-circulated by a peristaltic pump after the leachate samples were collected. The whole experiments last for 61 days.

2.2. Analysis of VOCs using GC–MS

Biogases generated during food wastes decomposition were sampled with 2.7-L stainless Summa canisters (ENTECH Instruments Inc., SiloniteTM) from reactors. VOCs samples were collected at days 0, 2, 4, 6, 8, 12, 16, 21, 26, 33, 40, 47, 54, 61. Then, VOCs were qualitatively and quantitatively analyzed using an Entech 7200 pre-concentrator (Entech Instruments Inc., CA, USA) equipped with gas chromatography–mass spectrometry (7890B GC-5977B MS, Agilent Technology, USA). The detailed sampling and analysis information as well as quality assurance and quality control are provided in supporting information and previous works (Han et al., 2019; Liu et al., 2019). The emission fluxes (μg kg^{-1} h^{-1}) were calculated as previous works (Wang and Wu, 2008).

2.3. Malodorous gas analysis from leachate using electronic nose (E-nose)

Malodorous gases emitted from leachate were analyzed using AIRSENSE PEN3.5 E-nose (Schwerin, Germany) for gas detection and odor data acquisition, and the date of sampling is consistent with VOCs sampling. The E-nose consists of a sampling unit and the gas detection system containing the array of 10 Metal Oxide Semiconductor sensors, which are differentially sensitive to each characteristic volatile compound. The sensors which composed the sensor array system are as follows: S1, aromatic; S2, broad range; S3, aromatic and ammonia (arom-ammonia); S4, hydrogen; S5, aromatic and aliphatics (arom-aliph); S6, broad range and methane (broad-methane); S7, sulfur organic; S8, broad range and alcohol (broad-alcohol); S9, sulfur and chlorinate (sulfur-chlor); and S10, methane and aliphatics (methane-aliph) (Benedetti et al., 2008).

For the analysis, 10 mL of leachate was sampled in a 40-mL glass vial and capped with a poly tetra fluoro ethylene septum. After that, vials were incubated at 30 °C for 10 min to achieve the headspace equilibrium. The headspace gases were injected into E-nose carried by air at a constant flow rate of 400 mL min^{-1} for 60 s, and the sensor signals were recorded each second. The sensor system was purged with filtered air for 120 s to reestablish the instrument baseline between the two sample injections. The sensor response, G/G0 (G and G0 stand for the conductance of MOS connected with sample and clean gas, respectively), is expressed as resistivity (Ohm) and changed accordingly to

the composition of volatile compounds (Barbosa-Pereira et al., 2019). Data were acquired with the pattern recognition software (WinMuster, Airsense Analytics GmbH., Germany), and the average of sensor response was used for the subsequent statistical analysis.

2.4. DNA extraction and sequencing

Leachate was first fully re-circulated into the reactor by a peristaltic pump and then sampled from the bottom of the reactor on days 2, 8, 16, 33, 47 and 61. The samples were then centrifuged at 15000 rpm for 10 min and used for isolating the total genomic DNA with the Rapid Soil DNA Isolation Kit (Sangon Biotech, Shanghai, China) following the manufacturer's instruction. The concentration of isolated DNA was determined using a Nano-Drop 2000 spectrophotometer. Polymerase Chain Reaction (PCR) amplification of the 16S rRNA gene was performed using 16 s rRNA primers (515F:5′-GTGCCAGCMGCCGCGG-3′ and 806R: 5′-GGACTACHVGGGTWTCTAAT-3′), which were specific to the V4 regions (Song et al., 2017). Additional details such as PCR amplification are provided in the supporting information. Sequencing was performed using the Illumina Miseq platform at Majorbio Bio-Pharm Technology Co., Ltd., Shanghai, China.

2.5. Statistical analysis

Statistical and correlation analyses were performed using IBM SPSS Statistics 19.0. All statistical tests were considered significant at $P < .05$. Heat maps and canonical correlation analysis (CCA) were conducted in R 3.53 with the package “pheatmap” and “vegan”, respectively. Principal component analysis (PCA) was performed using Canoco version 5.0 software based on the Bray-Curtis distance (Chen et al., 2016).

3. Results and discussion

3.1. Physiochemical properties of the food wastes

During the process of aerobic decomposition of food wastes, the temporal trends of temperature in wastes and pH in leachates were recorded. As Fig. S2a shows, the temperatures for all three kinds of food wastes were in the range of 25 °C–33 °C during the whole 61-day decomposition process and were higher than that of the ambient air. Besides, the temperature temporal trends were very similar in meat and vegetable (26 °C–33 °C) and lower than that of fruit. One possible explanation is that the mesophilic microorganisms are more active at favorable moisture and nutrient conditions in fruit, resulting in more heat being generated by decomposition than the other two wastes. This is supported by the higher diversity of microbial community (Table S2), higher abundance of *Lactobacillus* (Fig. S3) and higher emissions of VOCs (Fig. 1) in fruit wastes than the other two wastes. Previous study also revealed that proper moisture and nutrient are also vital for the health of the microorganisms that help with the composting process (Guo et al., 2019; Wang and D'Odorico, 2013). In summary, although the temperature of the wastes and temporal changes differed among these reactors, the temperatures during whole period are representative of mesophilic environments.

For the pH evolution in the resultant leachates, their trends were different from each other widely (Fig. S2b). The pH in the fruit wastes leachate was approximately 3.8 at the beginning and increased with time to a top at 5.2 on day 61. The possible reason for this is that fruits are rich in a variety of organic acids, which can be released from fruit tissue during the fruit decomposition process (Cardeal et al., 2005). By comparison, the pH in the meat wastes leachate decreased slowly and continuously. This is because meat wastes are rich in fat, protein and lipid, which are energy-dense and can be rapidly acidified during their hydrolysis. Therefore, an ideal pH could be maintained for acid organic fermentation (Kurade et al., 2019).

For the vegetable wastes leachate, the pH was 5.0 at the beginning and increased with time to 8.7 on day 61. The evolution trends of pH in the vegetable and fruit reactors are consistent with previously described pH behavior during the decomposition of municipal solid wastes (Bareither et al., 2013). Generally, during the food wastes decomposition process, the microorganisms, which propagate rapidly as the wastes begin to decompose, are closely related to the change of pH values. For example, hemicellulose in the sludge is easy to be hydrolyzed to produce fermentable sugars by microbes and further transformed into organic acids, and thus lowered the pH value of the leachate (Kumari and Singh, 2018). Nevertheless, as the composting process progresses and increased temperature, the organic acids will be gradually volatilized and resulted in an increase of the pH. At the same time, the ammonia produced by the nitrogen-containing organic matters (Xue et al., 2019) will also raise the pH to approximately 9 in the vegetable. Another possible reason is that the cellulose and lignocellulose contents in biomass are difficult to be hydrolyzed into fermentable sugars (Sun et al., 2015). Further, high cellulose and lignocellulose contents in vegetable would also inhibit metabolites from acidification and the formation of volatile fatty acids, which, therefore, indirectly responsible for the increase of the pH (Zhang et al., 2018).

3.2. The emissions of VOCs

3.2.1. The compositions of the released VOCs

In this study, 41 kinds of VOCs including 9 halogenated hydrocarbons (HHs), 7 aliphatic hydrocarbons (AlHs), 11 aromatic hydrocarbons (AHs), 10 OVOCs and 4 VOSCs were identified and quantified during the 61-day decomposition process of three kinds of food wastes (Table S3). As Fig. S4 shows, total of 39 kinds of VOCs were quantitatively detected in fruit wastes, and their total concentration increased slightly and then jumped up to the highest (41,750.6 ppb), and then decreased gradually thereafter. Among them, n-hexane, 2-butanone, dimethyl disulfide, vinyl acetate, ethyl acetate, acetone, n-pentane, methyl methacrylate, 1,2-dichloroethene (Z), and 1,2-dichloroethene (E) were the top 10 released VOCs with high average flux in fruit wastes during the whole period. Ethyl acetate, 2-butanone and esters were also observed as major VOCs emitted from orange wastes or citrus fruit flesh (Umano et al., 2002; Wu and Wang, 2015). Also, this profile of emitted VOCs was quite similar that in municipal solid wastes (Staley et al., 2006) and waste treatment facilities (Lehtinen et al., 2013). These VOCs could be produced from the microbial metabolisms of primary components such as pectin, protein, cellulose and sugar in fruit (Brat et al., 2003). Further, as compared to the other two wastes, more esters were released during the early period of fruit decomposition. This is because esters are important components of fruit aroma, which is also highly abundant in fruit substances and wastes (Wang et al., 2016). However, the release of ethyl acetate and vinyl acetate decreased sharply after days 16 and 26, respectively. This result was consistent with previous study, revealing that the emissions of methyl acetate and ethyl acetate were below detection limits at beginning and then increased immediately with a maximum at days 1–8, and finally decreased sharply until leveling off after day10. These suggested that these VOCs were emitted from aerobic decaying fruit wastes rather than inherent (Wu and Wang, 2015), and the emitted VOCs were related to microbial communities.

For the vegetable wastes, 40 kinds of emitted VOCs were quantitatively detected (Fig. S5). Unlike the fruit wastes, the total concentration of the released VOCs from vegetable wastes was very high during the initial period with the highest total concentration occurring on day 6 (15,774.9 ppb) and then decreasing gradually and sharply. Specifically, dimethyl disulfide, dimethyl sulfide and carbonyl sulfide were the top 1, 3 and 5 highest VOCs released with high concentration during days 2 to 16 than other days. It is reasonable, since dimethyl disulfide and dimethyl sulfide have been identified to be important contributor to the aroma of many food products including vegetables (Blank, 2002). In

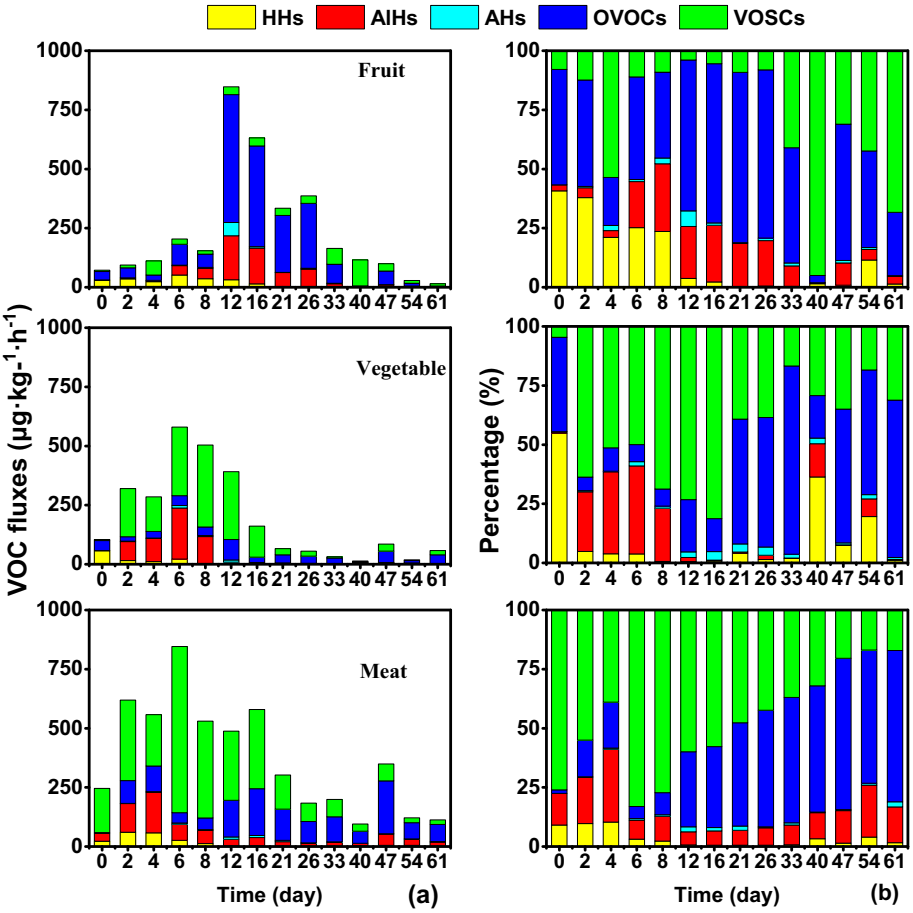

Fig. 1. Temporal trends of different groups of VOCs emissions (a) fluxes and (b) percentage emitted from three types of wastes.

addition, the concentration of released 2-butanone was also very high during this same period. Nevertheless, after day 16, OVOCs became the dominant groups, with methyl methacrylate, methyl isobutyl ketone, 2-hexanone and acetone ranked in the top 10 highest concentration of released VOCs. This is agreement with a previous study, showing that OVOCs from biomaterials such as vegetable and leaf litter could also be formed through the biological conversion of tissue cells by enzymes or microorganisms (Rappert and Müller, 2005). Besides, AlHs with higher concentration were observed on days 2–8 and the dominant compound was n-pentane (top 2). Overall, all aforementioned compounds are typical products of microbial decomposition (Garcia-Alcega et al., 2017; Korpi et al., 2009).

As Fig. S6 shows, similar to vegetable wastes, the total concentration of VOCs investigated and released from meat wastes was very high and up to the highest (19,938.6 ppb) on day 6 and then decreased gradually thereafter with the exception on day 47. Higher emission of VOCs at the initial stage from meat wastes mainly due to their degradation via microbial activity. Among these released VOCs, dimethyl disulfide was the top one. Differently, the emission of dimethyl disulfide from meat wastes was higher than that in vegetable wastes, while dimethyl sulfide was reversed. A possible explanation for higher VOSCs emissions can be due to high sulfur-containing amino acids and peptides in meat wastes. Previous studies also revealed that VOSCs could be released from the degradation of organic materials (animal manures, sewage sludge and plants) by microbes (Wu et al., 2010). With the prolonging of meat wastes decomposition time, the main VOCs changed gradually from dimethyl disulfide and acetone to dimethyl disulfide and n-pentane. A number of VOCs including acetone and methyl ethyl ketone were also measured during fish decaying processes (Ceballos et al., 2019). In addition, the total VOCs and OVOCs such as acetone, methyl methacrylate, ethyl acetate, 2-butanone, methyl isobutyl ketone in meat wastes have higher emission than in vegetable text, due to higher energy-dense of meat. Furthermore, higher emitted levels of OVOCs from meat wastes also indicated higher ozone formation potential (Han et al., 2019).

3.2.2. The emission profiles

To more specifically quantify the malodorous gases produced per kilogram of wastes, we converted the concentration into emission flux. As Fig. 1a demonstrates, the emission fluxes of VOCs released from food wastes varied with decomposition time. In general, during the whole decomposition process, the overall trend of the released VOCs first increased, and began to decrease after reaching a peak. For the total and individual VOCs, their emission fluxes during the first 33 days were significantly higher than that of the last 28 days, indicating that most of the wastes could be decomposed at earlier stage. These results were consistent with previous studies that the orange wastes and food wastes gave off considerable amounts of VOCs at the early disposal (Wu et al., 2017; Wu et al., 2010). However, the total VOCs emitted from the fruit wastes during days 12–61 were significantly higher than those in the first 11 days. The highest and lowest emission fluxes were observed on days 12 and 61, respectively. That is, the decomposition of the fruit wastes mainly occurred during the early period, which may result from the inhibition of the decomposition process under acidic conditions (Wang et al., 2018). The possible reason for this is that the decomposition of these wastes is mainly caused by microorganisms, who grow rapidly with abundant nutrients at the initial stage, and the proliferation of microorganisms gives rise to a large number of VOCs subsequently (Ferguson et al., 2018). However, nutrients will be gradually consumed by microorganisms for their growth and multiplication, in turn leading to the decrease of VOCs emission.

Among these VOCs, OVOCs contribute significantly to the emitted VOCs during the whole decomposition process of fruit wastes, while VOSCs were the main components of VOCs from vegetable and meat wastes, which was consistent with previous studies (Wu and Wang, 2015; Wu et al., 2010). For fruit wastes, the proportion of HHs was >20% during days 0–8, while it was <5% after day 8. Comparatively, during days 6–26 with relatively large emission flux, the proportion of VOSCs was less than other days (Fig. 1b).

Unlike the decomposition of fruit wastes, the lowest and highest total VOCs from vegetable wastes were observed on days 40 and 6, respectively. The increase of flux on day 47 may be due to temperature response since the ambient temperature reaches a peak on day 40. In addition, over 90% of the emission occurred during the first 21 days from vegetable wastes. Whereas, the emission fluxes of the total and most AIHs and AHs were approach to zero after 12 days, suggesting that the emission of VOCs from the food wastes mainly occurred during the early decomposition period. From day 2 to 16, the emission of VOSCs accounts for >50% of the total emission, while OVOCs becomes the dominant group after day 16 (Fig. 1b). This result suggested that considerable amounts of VOSCs emitted during early disposal of organic wastes contributed to malodor from waste treatment facilities, and thus should be taken serious consideration, considering that municipal wastes may stay in dustbins or transfer into stations up to a week before reaching landfills or incinerators (Statheropoulos et al., 2005). High fluxes of VOSCs in vegetable wastes may be related to a number of sulfur compounds in the vegetables such as sulfoxides, S-containing amino acids, and sulfonium compounds (Lewis and Papavizas, 1970). Comparatively, the emission profiles of the VOCs from meat wastes varied with incubation time (Fig. 1a) and were similar to vegetable wastes. That is, a peak of the emitted VOCs was found on day 6, with the lowest emission found on day 40. The proportion of VOSCs began to decrease after day 6, accompanied by the increase of the OVOCs (Fig. 1b). Previous studies revealed that VOSCs could be released from anaerobic or incomplete aerobic fermentation of organic materials (Garcia-Alcega et al., 2017; Zhang et al., 2013). In addition, the substantial emission of VOSCs would also happen even under aerobic conditions. For instance, Scaglia et al. (2011) revealed that poor O_2 transfer resulted from insufficient aeration was believed as an important reason for malodorous gas production during wastes composting process (Scaglia et al., 2011). As time goes on, the abundance of OVOCs is higher than that of VOSCs. This was probably due to the OVOCs were metabolites generated mainly from fatty or amino acid precursors, which was degraded at the later stage of fermentation. As reported previously, OVOCs can be released from both living and dead plants, and emitted transiently upon cell wounding (Derendorp et al., 2011).

Further, the percentages of different groups of VOCs emitted from these wastes were also analyzed by the sum of emission fluxes on all sampling day. As Fig. 2a shows, from fruit decomposition reactor, the percentages of VOSCs, HHs, AlHs and AHs were only 14.8%, 7.1%, 18.1% and 2.4%, respectively, while OVOCs was the dominant VOCs. In contrast, VOSCs, OVOCs and AlHs were the dominant VOCs emitted from vegetable and meat wastes, and contributed to 58.6%, 16.6%, and 20.2% of the total VOCs in the vegetable decomposition reactor, and 54.9%, 27.9% and 12.6% of the total VOCs in the meat fermentation, respectively.

In addition, the main species from the main group were further analyzed (Fig. 2b). From fruit wastes, OVOCs was the main group, among which 2-butanone was the most dominant species (accounting for 55.1% of OVOCs). Ethyl acetate (13.8%) and vinyl acetate (12.1%) were followed by. Although VOSCs were the dominant VOCs group in both vegetable and meat wastes, the compositions were different. Dimethyl disulfide (68.0%) and dimethyl disulfide (26.6%) were dominant VOSC species from vegetable wastes; whereas dimethyl disulfide (89.2%) was the most dominant species in meat. Quite similar to this result, Yi and Wang. (2011) also showed that dimethyl disulfide was the main VOSCs released from vegetable soils in Guangzhou (Yi and Wang, 2011). This was consistent with previous studies which also indicated that VOSCs and OVOCs were the major contributors of the emission of VOCs during the decomposition of yard wastes (Staley et al., 2006), dead plant materials or organic matter, such as plant litter (Isidorov and Jdanova, 2002; Svendsen et al., 2018). These showed that meat and vegetable wastes are important sources of malodorous gases.

3.2.3. The leachate gas characterized using E-nose

The leachate was also collected during the decomposition of these food wastes and the malodorous gases released from leachate were detected using E-nose (Fig. S7), since leachate is an important source of odor nuisance. For fruit and vegetable wastes leachates, the variation of signals was found to be similar, broad range and sulfur organic sensors displayed high response intensities, while broad range was higher in fruit wastes leachate. The sensors of broad range, sulfur organic and broad alcohol displayed high response intensities for the gas released from meat wastes leachate. These data demonstrated that the trend of leachate gas emission is consistent with the VOCs released by the upper layer of wastes. This may be related to the relationship between the biological activity and odor molecule production measured by an E-nose (D'Imporzano et al., 2008).

Further, the PCA analysis of E-nose data showed that the most significant classification of the gases was released from leachate according to the type and date of fermentation (Fig. 3). In this case, the potentials of several sensors that compose the E-nose were considered for the classification of leachate gas. All sensors had positive correlations with the first ordination axis, while only sulfur organic, sulfur-chlor and broad-methane had major positive correlations with the second ordination axis, but the others had negative correlations. The variance explained by the first and second principal component was 66.2% and 17.9%, respectively. All sensors demonstrated highly significant separation of the leachate gas from the whole fermentation cycle of fruit and the other wastes. Fruit wastes contain fewer sulfides, while vegetable and meat wastes contain more sulfides. Further, these results confirm the discrimination of VOCs from the fruit and vegetable wastes obtained with GC–MS data and this illustrated that the E-nose could be a useful tool for detection of the difference of VOCs composition of the composting waste gases (Delgado-Rodríguez et al., 2012).

3.3. Microbial diversity and dynamics

The rich nutrient, suitable pH and high moisture content of food wastes were beneficial for the growth and survival of a large range of microorganisms (Casaburi et al., 2015). Therefore, in this work, the bacteria ware sampled from leachate and analyzed using the sequencing of 16S rRNA gene amplicons. A total of 1,031,821 high-quality reads were generated from 18 samples and clustered into 1158 OTUs (Table S4). Based on the Shannon biodiversity index, assessment of rarefaction curves indicated that all the curves tended to reach plateaus (Fig. S8), suggesting that the obtained sequencing data were believed adequate to cover the vast majority species of microorganism community. These results were further supported by the observed values (>99%) from Good's coverage estimator (Table S2).

As Fig. 4a shows, Firmicutes and Proteobacteria are the most dominant phyla in the core microbiota of the digesters of fruit wastes. The abundance of Firmicutes increased from 47.4% to 76.0% as the incubation time prolonging from day 2 to day 33, then declined to 25.5% as degradation time progressed to day 61. In contrast, Proteobacteria declined from 46.5% to 10.9% when the days increased 2 to 33, and then increased to 40.2% at the end. At the genus level (Fig. S3), the genus *Lactobacillus* has the highest abundance throughout the fermentation period. That is, *Lactobacillus* plays an important role in the acidogenic fermentation of various polysaccharides present in mix fruit wastes, therefore leading to acidification. This result is consistent with a previous study, which revealed the association of effective digester performance with the microbial nexus by investigating the microbial dynamics during the anaerobic digestion following augmentation with

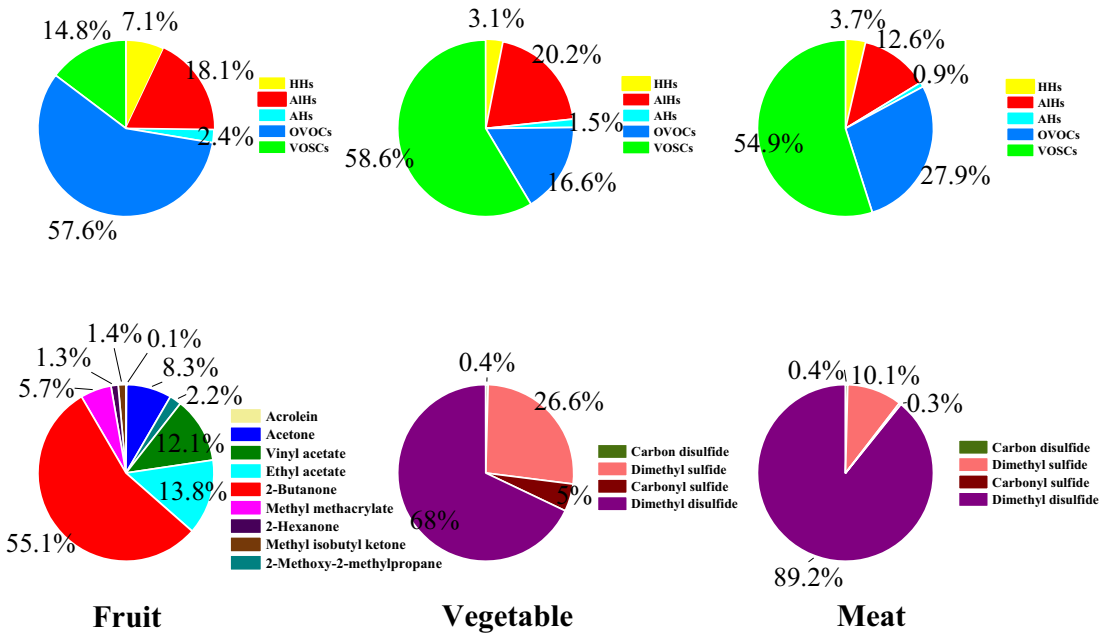

Fig. 2. Percentage of (a) different groups of VOCs and (b) different VOCs of dominant groups emitted from the three types of food wastes.

polysaccharide wastes (Saha et al., 2019). While, the fruit wastes are considered as polysaccharide wastes (Zhao et al., 2018). In addition, it is worth noting that, *Serratia* with a relatively high abundance (21.4%) on day 2, which has long been considered as a pathogen in lichens and earthworm gut, decreased rapidly to 0.6% on day 8 and thereafter (Hussain et al., 2016). In this case, the fermentation of these food wastes may not lead to the propagation of the pathogens.

Different from fruit wastes, the most dominant phylum in vegetable wastes was Proteobacteria (accounting for 16.9%–83.6% of the total microbial community), followed by Bacteroidetes, Firmicutes, Spirochaetae, Synergistetes and Actinobacteria (Fig. 4b). This finding coincided with a previous study, which found that the predominated phyla were Proteobacteria, Bacteroidetes, Actinobacteria, and Firmicutes phyla in swine farm environments (He et al., 2019). In addition, Huang et al. (2014) also found that Proteobacteria and Bacteroidetes presented in the entire decomposition process of fresh fruit and vegetable wastes, and thus were considered them as the common decomposing bacteria for these food wastes (Huang et al., 2014). At the genus level (Fig. S9), the relative abundance of *Lactobacillus* was declined from 13.4% to 6.9% when the decomposing time increased from day 2 to day 8. *Enterobacteriaceae* was the most abundant genus on day 2 (23.2%), and declined to <1% as degradation time prolonging to day 47. Previous study also reported that *Enterobacteriaceae* was mainly associated with the endophytic communities, while *Pseudomonadaceae*, *Sphingomonadaceae* were

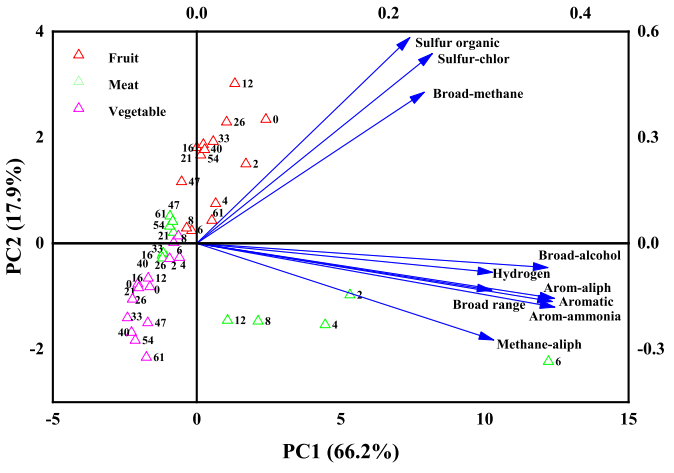

Fig. 3. PCA analysis based on the gases identified by *E*-nose. The sensors were displayed as blue Vectors and the distance between the points represented the relationship between the gases. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

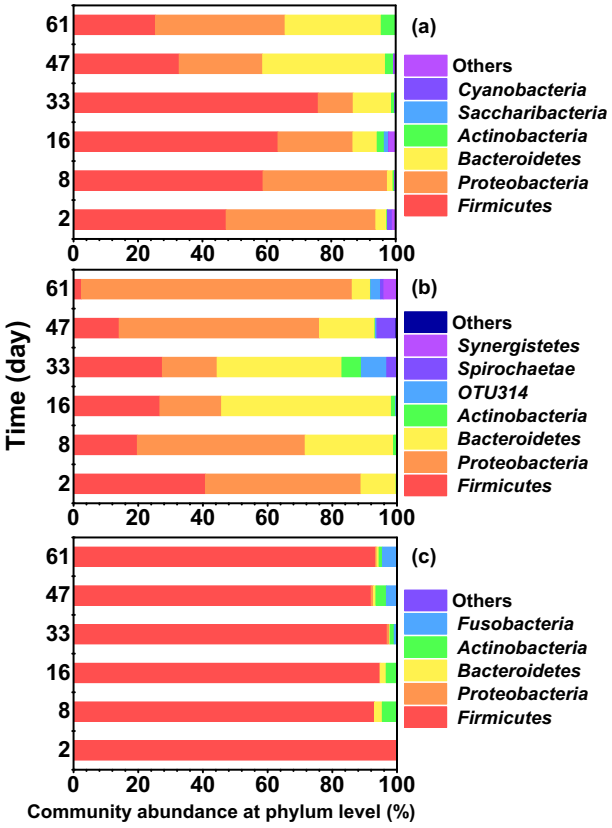

Fig. 4. Abundance of different microbial community structures at the phylum level in (a) fruit, (b) vegetable, and (c) meat wastes.

associated both with the rhizosphere and endosphere bacterial communities (Mitter et al., 2017). In addition, the abundance of *Acinetobacter* was 10.8% on day 2, declined to 0.9% on day 16, but increased to 67.4% on day 61, suggesting that *Acinetobacter* has become the most dominant genus during later period. This might be explained for the high fluxes of VOSCs emitted during vegetable wastes decomposition, considering *Acinetobacter* can utilize dimethyl sulfide as the sulfur source and participate in the thiosulfate oxidization (Luo et al., 2013).

In meat wastes, Firmicutes (accounting for 93.0%–99.9% of the bacterial community) is the predominant phylum during the whole decomposition process (Fig. 4c), suggesting that this population might be well adapted to protein-enriched environment. Previous studies also found that Firmicutes is the most dominant phyla in anaerobic digesters to treat different biomasses including lipid wastes, food wastes and animal carcass (Ferguson et al., 2018; Kim et al., 2017; Wang et al., 2019). In addition, Firmicutes can degrade various substrates such as fats, oils, and grease to produce volatile fatty acids including acetic acid, which is a prime substrate to produce methane for acetoclastic methanogens (Amha et al., 2017). Therefore, the occurrence of Firmicutes such as *Clostridia, Lactobacillales* is expected to represent the beginning of a new decomposition stage. Moreover, the majority of the Firmicutes reads pertains to the class *Clostridia*; while only 8.3% to 30.9% of the Firmicutes sequences were related to the other subdivision like *Bacilli*. At the genus level, the bacterial community shifts from *Pepstreptococcus* and *Peptoniphillus* to *Sporanaerobacter* with prolonging decomposition time (Fig. S10). The result is in accordance with studies showed that *Sporanaerobacter* sp. has the ability to concurrently hydrolyze fats, oil, grease, protein and polysaccharide (Kurade et al., 2019; Zhao et al., 2017), indicating that it is the primary degrader during meat decomposition.

To show the microbial composition, a ternary plot was further constructed based on three main wastes matrix: fruit, meat and vegetable wastes. As Fig. 5 shows, meat wastes harbored a high abundance of *Clostridiales* and *Peptostretococcaceae*, whereas vegetable wastes had a high abundance of *Moraxellacea*. Comparatively, the phylum *Proteobacteria* tends to have the highest relative abundance in vegetable and fruit wastes. The thrived *Proteobacteria* in systems may be linked to low concentration of dissolved organic carbon (Gerrity et al., 2018). Comparatively, only a few families existed in mix meat wastes, while most families could be found in both fruit and vegetable wastes, which further support our previous conclusion that that low diversity of microorganisms in meat wastes.

3.4. Correlations between microbial diversity and VOCs as well as physicochemical parameters

The microbial community structure will change with the different fermentation materials and environmental characteristics (Fitamo et al., 2017). Therefore, the relationship between the microbial community structure and environmental characteristics was studied in this work. Nevertheless, it is very necessary to select the environmental factors with less interaction by performing the environmental factor screening using VIF (variance inflation factor) analysis and remove those that linked to the composition of the microbial community but auto-correlated. By comparing the VIF values before and after screening, we retained those environmental factors with VIF value < 10. As Table S5 shows, methyl methacrylate, 2-butanone and dimethyl sulfide are redundant variables which need to be removed. In total, 11 environmental characteristics including 9 VOCs emitted from the mixed food wastes composting systems, pH and temperature were chosen for canonical correlation analysis.

As Fig. S11 shows, the combination of the two ordination axes explained approximately 31.0% of the total variables. The pH, acetone, 2-hexanone, methyl isobutyl ketone, carbonyl sulfide had positive correlations with the first ordination axis, while the others had major negative correlations. As for the second ordination axis, the major positive

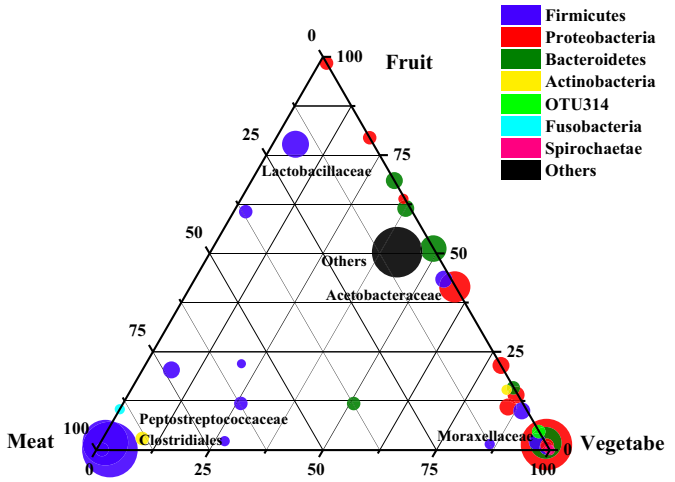

Fig. 5. Ternary plot representing the relative occurrence of individual families (circles), Families enriched in different compartments are colored by taxonomy of the most abundant phylum. The size of the circles is proportional to the mean abundance in the community.

associates were linked with carbonyl sulfide, while the major negative correlations were linked with the ethyl acetate, acetone, n-pentane, dimethyl disulfide. Specifically, vinyl acetate and temperature were correlated with bacterial community changes both in fruit and vegetable wastes initially (days 2–16). Dimethyl disulfide has a greater effect on the distribution of bacterial communities in meat on days 2, 8, 16; while methyl isobutyl ketone and acetone have significant correlation with fermentation of meat on days 33, 47 and 61. Beside, 2-hexanone, pH and carbonyl sulfide were significantly correlated with the late stage (day 33–61) vegetable wastes fermentation. All of the environmental factors examined, pH ($r^2 = 0.560$, $P = .004$), methyl isobutyl ketone ($r^2 = 0.744$, $P = .001$), 2-hexanone ($r^2 = 0.430$, $P = .024$) and acetone ($r^2 = 0.475$, $P = .025$) were the most important factors, shaping the microbial community structure (Table S5). Overall, these results confirmed that the changes of community structure could affect the VOCs released from wastes.

To further evaluate the relationship between VOCs, physicochemical parameters and the specific bacterial genera, a correction heatmap was constructed (Fig. 6). The pH was significantly correlated with *Arcobacter*, *vadinBC27_wastewater-sludge_group*, *Advenella* and *Acinetobacter*, while negatively correlated with *Komagataeibacter*, *Acetobacter*, *Cloacibacterium*, *Lactobacillus*. This suggested that pH played an important role in the evolution of microorganisms during fermentation. Further, significant positive correlations were found between temperature and *Cloacibacterium*, as well as *Lactobacillus*. That is, temperature might affect food wastes decomposition and VOCs emission by influencing *Lactobacillus* activity. Previous study also demonstrated that the emission fluxes of VOSCs were significantly correlated with measured food waste temperature, due to the production of VOSCs species were induced mainly by microbial activities (Wu et al., 2010). *Weissella*, *Leuconostoc* and *Enterobacteriaceae*, which have high abundance in vegetable wastes, showed significant positive correlations with carbon disulfide and dimethyl sulfide; while the dominant *Lactococcus* in meat wastes showed a strong correlation with dimethyl sulfide. Additionally, the predominant genera including *Peptococcus*, *Bacteroides*, *Lactobacillales*, *Peptoniphilus*, *Gallicola* and *Peptostreptococcus* were also significantly positively associated with dimethyl disulfide. Some abundant bacteria in meat wastes including *Sporanaerobacter*, *Coriobacteriaceae, Fusobacterium* and *[Eubacterium]_coprostanoligenes_group* strongly correlated with methyl isobutyl ketone, acetone and methyl methacrylate; while the

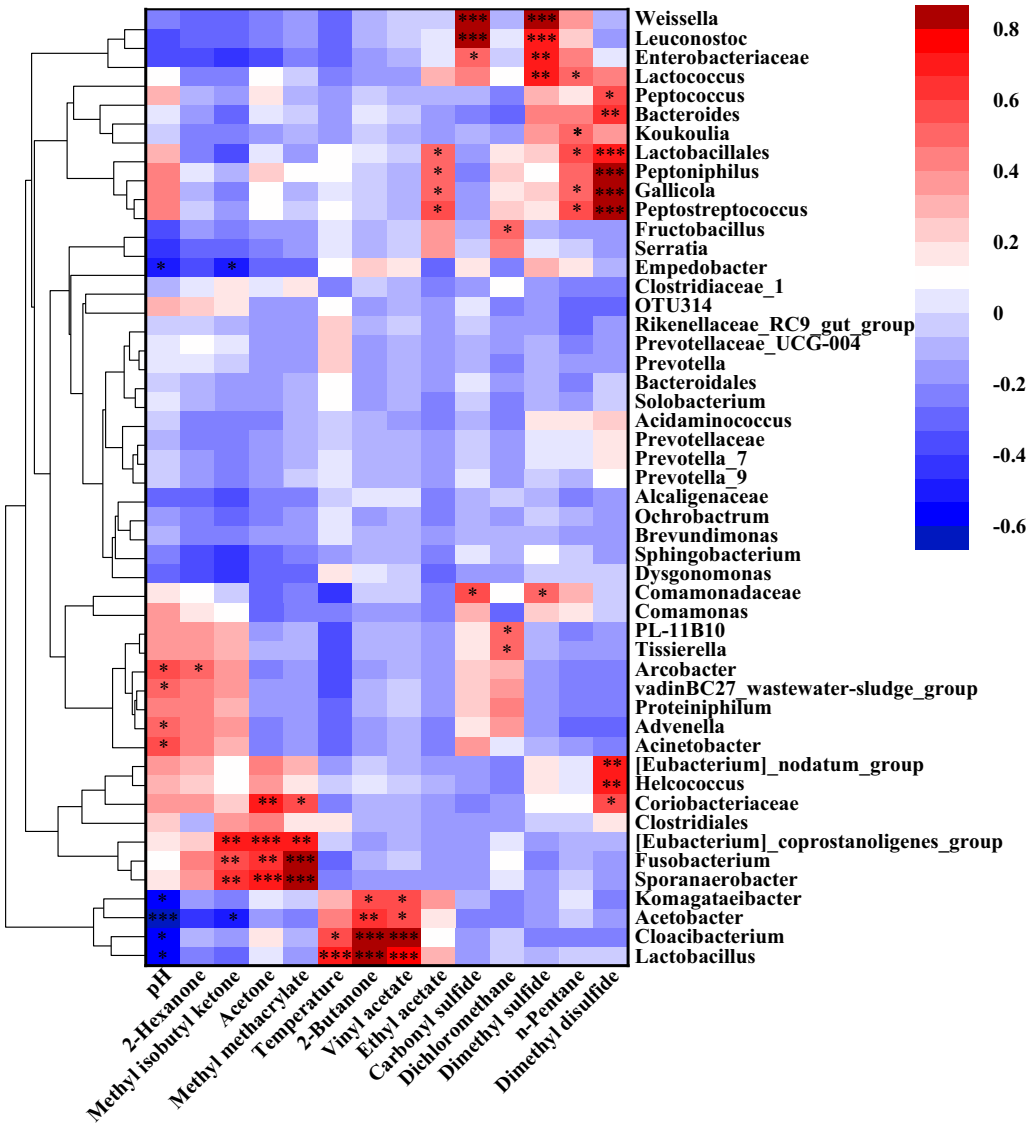

Fig. 6. Pearson correlation heat map of the top fifty genera, physicochemical properties and the contents of VOCs during food wastes decomposition processes. The right side of the legend indicated the color range of different R values. When $0.01 < P < .05$, it is marked with "*", while $0.001 < P \leq .01$ and $P \leq .001$ were marked with "**" and "***", respectively.

emission of 2-butanone and vinyl acetate in fruit wastes were related to *Komagataeibacter*, *Acetobacter*, *Cloacibacteriuma* and *Lactobacillus.*

To sum up, those bacteria mentioned above might be involved in the decomposing processes producing VOCs. However, only limited studies demonstrated the relationship between the emission of VOCs and bacteria. For example, *Comamonas* could oxidize sugars and alcohol to produce acetic acid (Fisgativa et al., 2018) and *Acinetobacter* played an important role in sulfur metabolism in waste biocover soil (Xia et al., 2015). *Bacillus* genus and Actinomycetes of the *Pseudonocardia* genus might involve in decomposing processes to produce VOSCs (Mayrhofer et al., 2006). Therefore, it is essential to further explore the actual interaction between VOCs emission and these microorganisms, providing more information on the control of malodor in food wastes treatment facilities.

4. Conclusions

In this study, the emitted VOCs and the dynamics of bacterial communities in the food wastes were assessed in self-made laboratory reactors to provide information on the correlation between microbial community structure and VOCs profiles. Up to 39, 40 and 38 VOCs were detected with OVOCs, VOSCs and VOSCs as the most abundant species during the decomposition of fruit, vegetable and meat wastes, respectively. Total VOCs emitted from fruit and vegetable wastes degradation were 232.8 and 191.1 $\mu g \cdot kg^{-1} \cdot h^{-1}$ in average, respectively, which were lower than meat wastes ($373.5\ \mu g \cdot kg^{-1} \cdot h^{-1}$). The emission fluxes of these VOCs in vegetable and meat wastes were very low at day 0, peaked at days 2–16 and then decreased sharply; while the peaked time was at days 12–16 for fruit wastes. The microbial community structures changed with incubation time. Fruit wastes had high abundance of *Lactobacillus* throughout decomposition period, while vegetable wastes had an increasing abundance of Acinetobacter with capability of sulfur metabolism. Whereas, the dominant genera were shifted from protein-using *Pepstreptococcus* and *Peptoniphillus* to *Sporanaerobacter* with increasing decomposition time of meat wastes. Furthermore, strong correlations were also found between *Weissella*, *Leuconostoc*, *Enterobacteriaceae*, *Lactococcus, Peptococcus*, *Bacteroides*, *Lactobacillales*, *Peptoniphilus*, *Gallicola*, *Peptostreptococcus* and malodorous VOCs. Overall, these results suggested that there were significant differences and correlation between VOCs emission profiles and bacterial communities among different food wastes decomposition, which will definitely provide useful information for waste management and odor control.

Declaration of competing interests

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.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (41877363, 41425015, 41807405 and U1901210), and the Science and Technology Program of Guangzhou, China (201704020185), Science and Technology Project of Guangdong Province, China (190307184964278 and 190325224778589), and China Postdoctoral Science Foundation (2018M643025).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.137175.

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