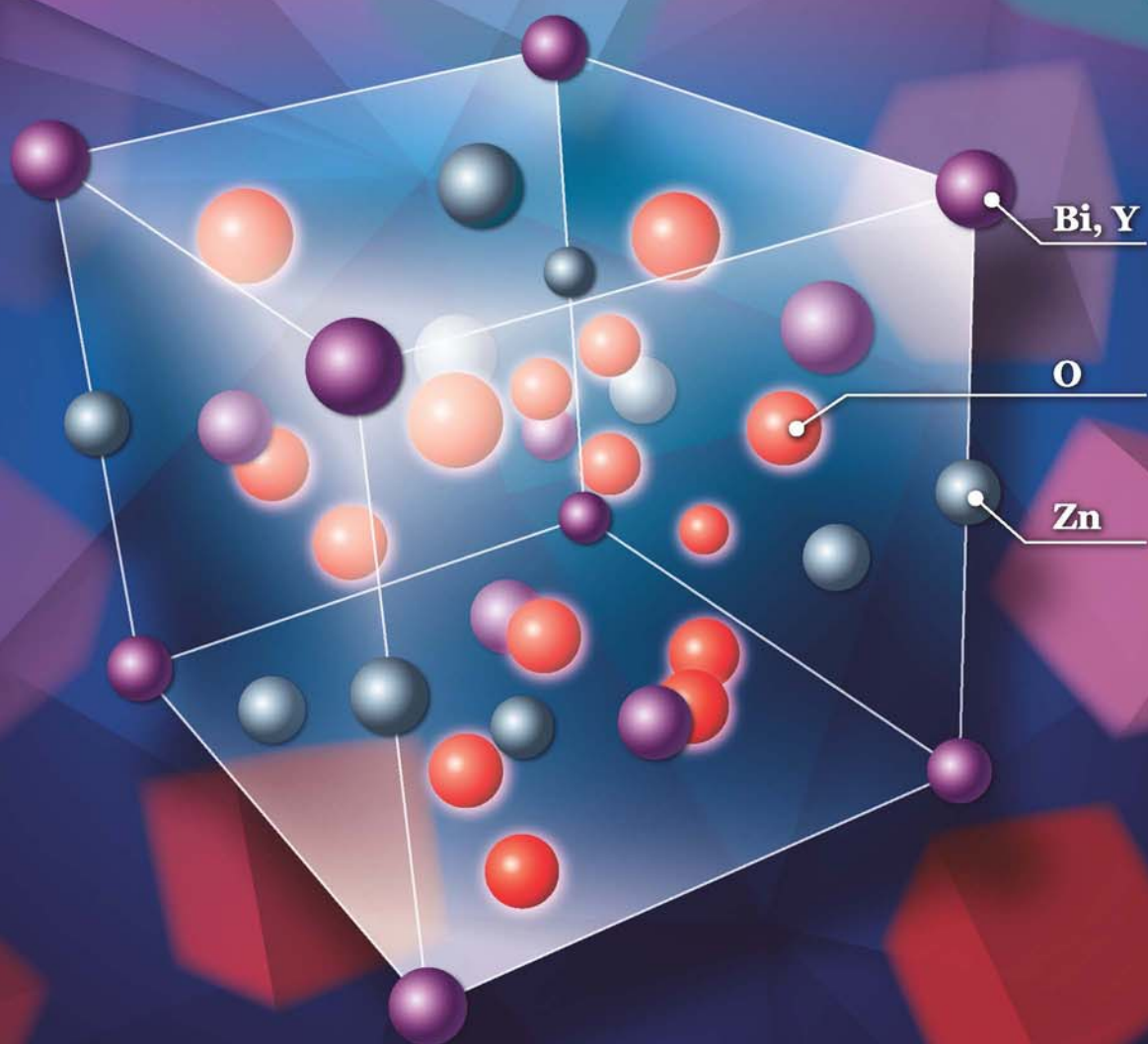


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Characterization of odorous charge and photochemical reactivity of VOC emissions from a full-scale food waste treatment plant in China

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ABSTRACT

Food waste treatment plants (FWTPs) are usually associated with odorous nuisance and health risks, which are partially caused by volatile organic compound (VOC) emissions. This study investigated the VOC emissions from a selected full-scale FWTP in China. The feedstock used in this plant was mainly collected from local restaurants. For a year, the FWTP was closely monitored on specific days in each season. Four major indoor treatment units of the plant, including the storage room, sorting/crushing room, hydrothermal hydrolysis unit, and aerobic fermentation unit, were chosen as the monitoring locations. The highest mean concentration of total VOC emissions was observed in the aerobic fermentation unit at 21,748.2–31,283.3 $\mu\text{g}/\text{m}^3$, followed by the hydrothermal hydrolysis unit at 10,798.1–23,144.4 $\mu\text{g}/\text{m}^3$. The detected VOC families included biogenic compounds (oxygenated compounds, hydrocarbons, terpenes, and organosulfur compounds) and abiogenic compounds (aromatic hydrocarbons and halocarbons). Oxygenated compounds, particularly alcohols, were the most abundant compounds in all samples. With the use of odor index analysis and principal components analysis, the hydrothermal hydrolysis and aerobic fermentation units were clearly distinguished from the pre-treatment units, as characterized by their higher contributions to odorous nuisance. Methanethiol was the dominant odorant in the hydrothermal hydrolysis unit, whereas aldehyde was the dominant odorant in the aerobic fermentation unit. Terpenes, specifically limonene, had the highest level of propylene equivalent concentration during the monitoring periods. This concentration can contribute to the increase in the atmospheric reactivity and ozone formation potential in the surrounding air.

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Introduction

With the accelerating socioeconomic growth and urbanization in China, people's living standards have been improving rapidly. However, such improvement has had the negative effect of the

generation of a large amount of food waste (FW). The current FW generation in China is estimated to have reached 60 million tons per year (Wang et al., 2013). Compared with other bio-wastes, FW is rich in easily biodegradable organic matters and high water content, thus becoming a potential source of environmental pollution.

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Under this scenario, industrial FW treatment has been paid considerable attention in China, where a series of regulations and technical specifications have been issued recently.

However, during FW processing, a mass of volatile organic compounds (VOCs) are inevitably released into the indoor and outdoor environment. These VOC emissions mainly contain hydrocarbons, ketones, esters, alcohols, aldehydes, terpenes, and organic sulfur compounds (Mao et al., 2006; Tsai et al., 2008; Wu and Wang, 2012). It was reported that total non-methane organic compounds and total volatile sulfur compounds derived from FW anaerobic decomposition and aerobic composting accounted for 0.221 and 0.379 mg-C/dry g, respectively, and these values were comparatively higher than those emitted from paper, yard waste, and municipal solid waste decomposition (Staley et al., 2006; Zhang et al., 2013). Most VOC emissions are generally responsible for odor nuisance and health risks due to their relatively low olfactory thresholds and potential toxicity. On the other hand, such compounds also can contribute to the atmospheric OH radical budget and tropospheric ozone production, while VOC oxidation can facilitate the formation of secondary organic aerosols (SOAs), which are the main components of particulate matter 2.5 (PM_{2.5}) (Chen et al., 2013; Ran et al., 2011).

The “Technical code on food waste treatment” has been issued in China recently, and this specification is anticipated to accelerate the development of the domestic FW treatment industry. However, the corresponding technical specifications on risk assessment and pollution control during FW treatment remain unavailable in China. One possible reason for this unavailability is the scarcity of information on the *in-situ* monitoring of pollutant emission, specifically for VOC emissions from a full-scale food waste treatment plant (FWTP). Although VOC emissions from both aerobic and anaerobic processes have already been reported in the literature, most of the references were only related to bench or pilot experiments (Pierucci et al., 2005; Staley et al., 2006; Zhang et al., 2012), and others were only focused on an isolated operating unit, *i.e.*, the composting process (Kumar et al., 2011; Mao et al., 2006; Tsai et al., 2008), rather than the whole treatment process from initial receiving to final treating (Gallego et al., 2012; Nadal et al., 2009). Moreover, despite the growing interest in the atmospheric reactivity of VOC emissions, most papers have primarily referred to such scenarios as atmospheric environment, industrial environment, vehicle sources, and even VOC emissions from waste water treatment and incineration plants (Jia et al., 2009; Ran et al., 2011; Tang et al., 2011; Wei et al., 2014), while few reports have focused on the atmospheric reactivity contributions of VOC emissions derived from FWTPs. Hence, a better understanding of VOC emissions released from industrial FWTPs and their potential contributions to atmospheric pollution is required.

With these findings, this study aims to develop a surveillance program focused on VOC emission in a full-scale FWTP with the core processes of hydrothermal hydrolysis followed by aerobic fermentation. This FWTP was selected for its relatively advanced technology, good management practices, large scale, long period of normal operation, and potential for widespread application in China. In addition, the main odorous charge and atmospheric reactivity of target VOC emissions were estimated based on the *in-situ* monitoring results to provide new insights into air pollutants released from FWTP. Such findings can also contribute to preventive recommendations for VOC abatement during industrial FW treatment to help regulatory bodies.

1. Materials and methods

1.1. Site description

The study was conducted in a full-scale FWTP in China. The FWTP was put into operation in 2004 and has a treatment capacity of 300 ton/day. The feedstock used in this plant was

mainly obtained from local restaurants. The basic compositions of FW were as follows (represented as mean \pm SD): moisture = 74.4 ± 1.7 (% (W/W)); VS = 80.2 ± 6.4 (% (DM (dry matter))); carbohydrate = 23.8 ± 5.2 (% (DM)); protein = 24.8 ± 2.0 (% (DM)); lipid = 25.9 ± 5.1 (% (DM)); and salt content = 4.6 ± 0.6 (% (DM)). After being received and stored for a short period, FW was sorted mechanically and manually to remove the inorganic materials, followed by the crushing treatment. The crushed FW was then pumped to the hydrothermal hydrolysis reactors (treated at 120°C for 80 min), where the macromolecular organic matters were converted into simple organic matters through physical and chemical reactions. After hydrothermal processing, solid–liquid separation was conducted by a simple gravity effect. The solid residue was pumped up to aerobic fermentation reactors for the final protein feed, whereas the liquid fraction was used for biodiesel production (oil phase) and anaerobic digestion (aqueous phase) after oil–water separation. The aerobic fermentation unit adopted forced ventilation mode with an average discharge rate of approximately 100 m³/min. The retention time and fermentation temperature were set as 12–18 hr and 70–80°C, respectively. Meanwhile, exogenous microbes were added to accelerate the aerobic decomposition of thermally hydrolyzed food waste. The process flow chart and plan view are shown in Fig. 1.

1.2. Sampling and analysis

The four three-month basis samples were collected from April 2012 to January 2013 and fairly represented the possible conditions during the seasons of spring, summer, autumn, and winter. According to the treatment process and the potential discharge capacity of VOC emissions, four major treatment units inside the plant were chosen as sampling points. The locations were as follows: (1) storage room; (2) sorting and crushing room; (3) hydrothermal hydrolysis unit; and (4) aerobic fermentation unit (Fig. 1). Monitoring was conducted once or twice a day within the continuous five-day working period. The mean of the monitored data was recorded.

Different sampling strategies were used for fugitive and fixed emission sources. The fugitive emission samples, mainly from treatment units 1 and 2, were collected beside the raw waste storage pool and discharge port of the crushing device with a distance and a height of 0.5 and 1.2 m respectively. For the fixed emission samples in treatment units 3 and 4, sampling was carried out at the open face of the respiration valve of the hydrothermal hydrolysis reactor and the exhaust outlet of the aerobic fermentation reactor, respectively. To assure their representative nature, samples were collected during working hours, and simultaneously to avoid air disturbance caused by anthropic factors. The concentrations of total VOC emissions (TVOC) were first measured on site using a hand-held TVOC analyzer with PID detector (ppbRAE 3000 PGM-7340, ICON Safety Co., Inc., Cerritos, USA) equipped with a gas-discharge lamp (10.6 eV). Subsequently, the air samples were dynamically collected by using self-extracting equipment connected to an 8 L Teflon sampling bag for component analysis of VOC emissions. The speed of the mass flow controller (Buck Libra Pump L-4, A. P. BUCK, Inc., Orlando, USA) was 0.1 L/min and continually sampled for 1 hr. Therefore, approximately 6 L ambient air

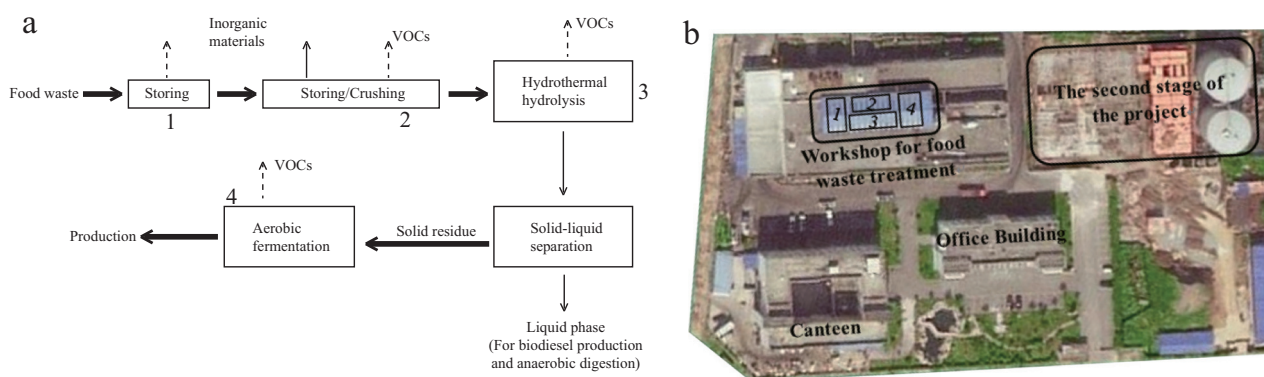


Fig. 1 – Technological process of the food waste treatment plant (a) and its plan view (b). Sampling points: (1) storing room; (2) sorting/Crushing room; (3) hydrothermal hydrolysis unit; (4) aerobic fermentation unit.

was collected. After sampling, the samples were sealed immediately and then sent for analysis within 48 hr.

Analysis was conducted in the State Key Laboratory of Environmental Protection and Odorous Pollution Control located in Tianjin, China, following the USEPA methods TO-14A and TO-15, using a GC–MS system composed of a gas chromatograph (Agilent 7890A, Agilent Technologies, Inc., Wilmington, USA) equipped with a mass-selective detector (Agilent MSD5975C, Agilent Technologies, Inc., Wilmington, USA). This combined system was also used in a previous study to determine the VOC reactivity of ambient air at a suburban site in the North Plain, China (Ran et al., 2011).

The equipment and optimum conditions determined for the performance analysis were as follows: gas chromatograph equipped with a 60 m × 0.32 mm × 1.0 μm DB-5 capillary column with helium as the carrier gas (1.5 mL/min). The injection volume was 50–400 mL, according to the actual concentration. Temperature programming was 35°C for 7 min with an increase to 140°C at 4°C/min followed by bake-out at 250°C for 5 min. The mass spectra were obtained by electronic impact at 70 eV with SIM scanning and full scanning synchronously (scanning time < 1 sec). Data were collected at an m/z range of 15 to 300. Quantitative analysis of all the identified VOC emissions was conducted by the internal standard method, and the results were expressed in $\mu\text{g}/\text{m}^3$. The error of standard matter concentration must be under 10%. Limits of detection (LOD) were below $0.1 \mu\text{g}/\text{m}^3$.

1.3. Data analysis

Data analysis was performed with the SPSS 18.0 statistics software (SPSS Inc., Chicago, USA). A probability lower than 0.05 ($p < 0.05$) was regarded as statistically significant. For the calculation, non-detected chemicals (or those lower than LOD) were denoted by a concentration of zero. Principal component analysis (PCA) was performed using CANOCO software (version 4.0 Biometris, Wageningen, Netherlands).

2. Results and discussion

2.1. Variation of TVOC concentrations on the site

The mean and maximum TVOC concentrations in four sampling points are depicted in Fig. 2 over temporal and

spatial variation. As expected, relatively higher concentrations were found in the core treatment units (points 3 and 4). Hydrothermal hydrolysis treatment enabled the conversion of crushed organic matters to low-molecular-weight organic acids, glucose, and other intermediate materials through physical and chemical reactions. Thus, a large amount of VOC emissions can be dispersed along with the gases discharged from the respiration valve of the hydrothermal hydrolysis reactor while working. The aerobic fermentation unit (point 4) showed the maximum TVOC concentrations among all the monitoring points with the average ranging from 21,000 to 31,000 $\mu\text{g}/\text{m}^3$. These values were 1.4–9 times higher than those of the other treatment units. Although the pollution levels in various studies were significantly variable, the organic matter composting unit was generally considered to be an important source of VOC emissions. Kumar et al. (2011) thoroughly characterized the VOC emissions from green waste compost piles of different ages. More than 100 types of VOC emissions were quantified, and the average VOC emission was 15.23 mg/m^2 min from young composting windrows (3–6 days). Two other laboratory-based studies conducted by Wu et al. (2010) and Zhang et al. (2013) also found that volatile sulfur compounds emitted from aerobic composting of food waste and kitchen waste totaled 409.9 and

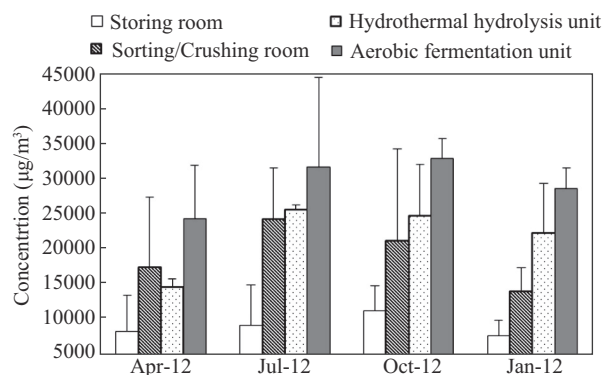


Fig. 2 – The mean and maximum TVOC concentrations in four sampling points. The top of error bar represents the maximum concentration detected during monitoring periods in the respective treatment unit.

379.3 mg/kg, respectively. The lowest TVOC concentrations in this study were observed in the storage room, with mean concentrations of 3000–7000 $\mu\text{g}/\text{m}^3$. In this location, initial FW was transiently stacked for subsequent treatment, such that lower levels of VOC concentrations were expected to be present. The sorting/crushing unit involved rotating agitation of feedstock to remove inorganic fractions (*i.e.*, plastic, bottles, chopsticks, and cardboard), and the remaining materials (organic fraction) were crushed into smaller diameters. Considering the agitation and possible further decomposition of FW, a TVOC concentration higher than that of the storage room can be expected in this location, with mean concentrations ranging from 10,000 to 21,000 $\mu\text{g}/\text{m}^3$.

When studying the temporal trend of organic compounds, a slight increase in TVOC concentrations in the hydrothermal hydrolysis and aerobic fermentation units could be observed during the months of July and October. Combined with the monitoring data in January, the total mean concentration of VOC emissions was observed to be relatively stable in these two locations. Thus, no concrete evidence demonstrates that TVOC production during these two processes was influenced by season or ambient conditions. A possible explanation for this result is that relatively stable operation parameters, such as internal temperature, humidity, air flow rate, and operation cycle, are controlled by the corresponding reactors. Nevertheless, a few exceptions were observed in the month of April when the TVOC emissions in these two units were relatively lower than those of the other monitoring periods, probably due to great differences in the compositions of feedstock. However, inverse results were found in the storage and sorting/crushing units (points 1 and 2), and a relative increase in TVOC concentrations could be observed in July and October. The TVOC growth, possibly derived from the increase in the ambient temperature (34–36°C in July and 27–29°C in October), has been suggested to cause seasonal differences in the VOC emissions in the storage and sorting/crushing units because of the semi-closed workshop and equipment design.

As expected, *in-situ* monitoring results of TVOC were notably variable even in the same unit over 5 continuous days. The maximum concentrations detected during monitoring periods in the respective units were 1.3–1.7 times higher than mean concentrations in the same sampling points (Fig. 2).

2.2. VOC profiles

The compositional patterns of VOC emissions were analyzed using the samples collected in October and January. These samples were characterized by relatively high mean concentrations of TVOC in points 3 and 4 and the lowest mean concentrations in points 1 and 2 (Fig. 2). A total of 65 VOC species were quantified in October, and 57 species in January. The dominant VOC species detected are shown in Table 1 based on their categories. These VOC emissions contained alkanes, alkenes, terpenes, aromatic hydrocarbons, halocarbons, organosulfur compounds, alcohols, aldehydes, and ketones. Similar compositions of VOC species, both derived from aerobic and/or anaerobic degradation of bio-waste at laboratory or field scale, were reported in previous studies (Chiriac et al., 2007; Staley et al., 2006; Zhang et al., 2012). Compared with the VOC emissions from other industrial processes, more VOC categories were detected in this study

compared with those derived from wastewater treatment and a ship painting process, but were slightly less than those emitted from a coking process (Celebi and Vardar, 2008; Jia et al., 2009; Tang et al., 2011). More alkanes were detected in this study (Table 1) compared with previous reports for FW aerobic fermentation (Mao et al., 2006; Tsai et al., 2008; Wu and Wang, 2012). This result implied that thermally hydrolyzed FW could increase alkane species during the subsequent biological processing, but further lab-scale experiments were needed to demonstrate this viewpoint. It is also worth noting that a portion of organosulfur compounds could be generated during aerobic fermentation, which was consistent with the results reported by Wu et al. (2010) in lab experiments. They revealed that organosulfur compound generation should maintain different patterns in terms of odorant species, and peak at days 2 to 4. In the present treatment system at the investigated plant, the retention time of thermally hydrolyzed FW in the aerobic fermentation reactor was only 16–18 hr, but still a considerable portion of organosulfur compounds could be detected. This suggested that thermally hydrolyzed FW was easily turned to anaerobic conditions, and thus a more optimum ventilation mode should be selected during aerobic treatment for thermally hydrolyzed FW.

The abundance of pollutant families in the different sampling points is illustrated in Fig. 3. As summarized, alcohols were the largest category of VOC emissions, accounting for 53%–88% (October) and 42%–70% (January) of TVOC (*w/w*), and were present in all monitoring points. Remarkably, this result is consistent with the conclusion proposed by Kumar et al. (2011), who performed *in-situ* monitoring during the composting process for green waste. They also found that alcohol was dominant among the TVOC emissions (66%–85%, *w/w*). Meanwhile, FW treatment tends to produce a higher percentage of alcohols than MSW anaerobic/aerobic treatment or landfilling (Duan et al., 2014; Staley et al., 2006).

Aside from alcohols, aldehydes (5%–21%, *w/w*) and terpenes (2%–20%, *W/W*) were the dominant components of VOC emissions in October, while terpenes (7%–20%, *w/w*), organosulfurs (5%–16%, *W/W*), and alkanes (5%–11%, *W/W*) were found to have significant percentages in January. The concentration percentage of terpenes emitted from the hydrothermal hydrolysis unit and aerobic fermentation unit in October was slightly higher than in January. Meanwhile, the concentration percentage of organosulfurs emitted in January was higher than in October. This implied that variations in VOC patterns occurred more or less with the changes in the seasons, possibly because the dietary structure of people for each season, such as the content of vegetal wastes or meat, might change accordingly. Other categories of VOC emissions that comprised the remaining fractions exhibited relatively low percentages.

Pearson correlation analysis was performed based on the mean concentrations of detected VOC emissions to identify the characteristics of their generation. To reduce the uncertainty of analysis, only dominant VOC emissions with mean concentration of more than 10% of the total concentration of the individual family were used. Correlation within the same family of VOC was observed, as shown in Table 2. For example, dimethyl disulfide had a significant correlation with carbon disulfide ($r = 0.779$, $p < 0.05$, $n = 8$), as well as with

Table 1 – Dominant VOC emissions found in the different sampling locations and their pollution characteristic values.

Compounds	Abbreviation	Detected spots		Odor threshold ($\mu\text{g}/\text{m}^3$) ^a	OH-rate coefficient ^b ($\times 10^{12} \text{ cm}^3/\text{molecule s}$, 298 K)
		October	January		
<i>Organosulfurs</i>					
Carbon disulfide	CS ₂	1,2,3,4	1,2,3,4	110	2.89
Methanthiol	MM	1,2,3,4	2,3	0.14 ^c	31.6 ^d
Dimethyl sulfide	DMS	1,2,3,4	1,2,3,4	1	4.4 ^d
Dimethyl disulfide	DMDS	1,2,3,4	1,2,3,4	7	198 ^d
<i>Terpenoids</i>					
a-Pinene	a-Pin	1,2,3,4	1,2,3,4	230	52.3
b-Pinene	b-Pin	1,2,3,4	ND	8900	74.3
Limonene	Limo	1,2,3,4	1,2,3,4	1700	164
<i>Alcohols</i>					
Ethanol	Ethano	1,2,3,4	1,2,3,4	2000	3.2
<i>Esters</i>					
Ethyl acetate	EA	1,2,3,4	1,2,3,4	4600	1.82 ^d
<i>Aldehydes</i>					
Acetaldehyde	Acetalde	1,2,3,4	ND	2.7	9.37
Propionaldehyde	Propional	3,4	ND	3.6	15
Butyraldehyde	Butyral	3	ND	15	20
<i>Halocarbons</i>					
Dichloromethane	DCM	1,2,3,4	1,2,3,4	4100	0.11 ^d
Chlorobenzene	CB	1,2,3,4	1,2,3,4	980 ^c	0.94 ^d
Chloromethane	MC	1,2,3,4	1,2,3,4	21100 ^c	0.04 ^d
Trichloromethane	TMA	1,2,3,4	1,2,3,4	500	0.1 ^d
1,2-Dichloroethane	1,2-DE	1,2,3,4	1,2,3,4	24700 ^c	0.26 ^d
Tetrachloroethylene	PCE	1,2,3,4	4	8300	0.17 ^d
1,4-Dichlorobenzene	1,4-DB	1,2,3,4	1,2,3,4	730	0.32 ^d
<i>Ketones</i>					
Acetone	Ato	2,3,4	ND	8600	0.17
<i>Aromatic hydrocarbons</i>					
Benzene	B	1,2,3,4	1,2,3,4	1500	1.22
Toluene	T	1,2,3,4	1,2,3,4	3800	5.63
Ethylbenzene	E	1,2,3,4	1,2,3,4	400	7.0
m-Xylene	m-X	1,2,3,4	1,2,3,4	770	23.1
p-Xylene	p-X	1,2,3,4	1,2,3,4	770	14.3
o-Xylene	o-X	1,2,3,4	1,2,4	770	13.6
1,3,5-Trimethylbenzene	1,3,5-TMB	1,2,3,4	1,2,3,4	10700	56.7
1,2,4-Trimethylbenzene	1,2,4-TMB	1,2,3,4	1,2,3,4	140	32.5
1,2,3-Trimethylbenzene	1,2,3-TMB	1,4	ND	NR	32.7
Naphthalene	Naphth	1,2,3,4	1,2,4	7	23
p-Ethyltoluene	p-ET	1,2,3,4	1,2,3,4	42	11.8
m-Ethyltoluene	m-ET	1,2,3,4	1,2,3,4	42	18.6
o-Ethyltoluene	o-ET	1,2,3,4	1,3,4	42	11.9
<i>Alkane</i>					
Propane	Propan	1,2,3,4	1,2,3,4	NR	1.09
iso-Butane	i-Bu	1,2,3,4	1,2,3,4	NR	2.16
Butane	Bu	1,2,3,4	1,2,3,4	NR	2.36
Pentane	Pentan	1,2,3,4	1,2,3,4	NR	3.8
Octane	Octan	1,2,3,4	2	NR	8.11
<i>Alkene</i>					
Propylene	Propyl	1,2,3,4	1,2,3,4	NR	26.3

NR: not reported; ND: not determined.

^a Source: Gallego et al. (2012).^b Source: Tang et al., 2006.^c Source: Calculated based on ppm values derived from “Measurement of odor threshold by triangle odor bag method” (Nagata, 2003).^d Source: Atkinson (1986).

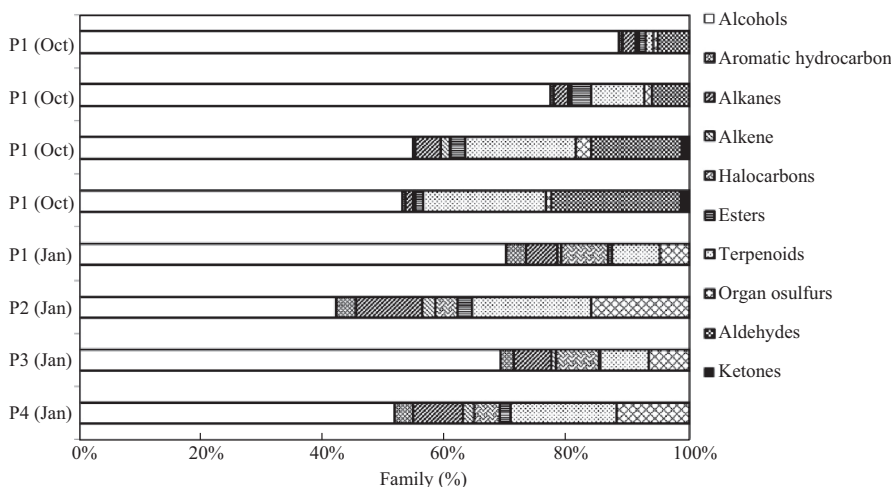


Fig. 3 – Contribution of VOC groups in the emission profile of different treatment units. P1–P4 represents sampling points 1–4.

methanethiol and dimethyl sulfide ($r = 0.850$ and 0.835 , $p < 0.01$, $n = 8$). Likewise, *a*-Pinene had a significant correlation with limonene ($r = 0.990$, $p < 0.01$, $n = 8$). Xylene had a significant correlation with toluene and ethylbenzene ($r = 0.986$ and 0.897 , $p < 0.01$, $n = 8$).

From these results, in fact, two different trends of dominant VOC emissions from FW treatment process can be observed. First, all biogenic compounds (oxygenated compounds, organosulfurs, alkanes, alkene, and terpenes) were statistically correlated with each other because these compounds are usually considered to be by-products of easily degradable organic matters (Zhang et al., 2012). Meanwhile, the emission of abiogenic VOC species, such as halogenides and benzene, which were usually inherent in waste, exhibited low correlation with other groups. However, a few exceptions were observed for toluene, ethylbenzene, and xylene. These compounds also exhibited a statistical correlation with some of the biogenic compounds, although their concentrations were relatively low during the whole process (36.9 – $120.3 \mu\text{g}/\text{m}^3$ and 20.5 – $47.6 \mu\text{g}/\text{m}^3$ in October and January, respectively). In the previous research conducted by Staley et al. (2006); Komilis et al. (2004), toluene, ethylbenzene, and xylene were also detected during FW anaerobic or aerobic decomposition. However, these aromatic pollutants were usually considered to be directly volatilized from waste mass (Powell et al., 2005), and the higher the fat content present, the more aromatics will be released (Heikes et al., 1995). Hence, for this FWTP, it is difficult to deduce which factor, between stripping and decomposition from lipid and lignin, mainly affects aromatic hydrocarbons emission.

2.3. Odorous charge on the site

To estimate the olfactory nuisances of VOC emissions, a theoretical calculation was performed based on the concentrations and odor thresholds of individual VOC detected on the site following the method of Fang et al. (2012); Nagata (2003). It should be noted that the odor thresholds of odorant chemicals vary widely, which means that a high concentration of a VOC does not always have strong odor impacts on

people who have been exposed to this odor. Furthermore, the theoretical calculation of odor concentration only refers to the compounds for which the odor threshold can be determined. Other compounds for which odor threshold values were not reported cannot be used to determine their odorous strength using this method. The odor index (OI) can be calculated according to Eq. (1) and (2).

$$OI_i = C_i/T_i \quad (1)$$

$$TOI = \sum_{i=1}^k OI_i \quad (2)$$

where, C_i is the analytical concentration of compound i , T_i is the odor threshold concentration of compound i , and k is the number of compounds in each VOC groups. Therefore, TOI represents the total sum of individual OI.

To develop a representative odorant characteristic for FWTP, the concentrations of detected VOC emissions were averaged for the two-season data (October and January). The targeted VOC emissions and their odor thresholds are listed in Table 1. The calculated results of OI and TOI are shown in Fig. 4. Esters, halocarbons, and ketones were removed intentionally because their theoretical OI were very low (less than 1). As shown in Fig. 4, organosulfurs and aldehydes were the main substances contributing to odor, specifically methanethiol, dimethyl sulfide, and acetaldehyde, which accounted for more than 95% of TOI derived from VOC emissions. To investigate the olfactory nuisance related to solid waste treatment, organosulfurs were consistently considered the main contributors, although the different processes would exhibit differences in the compositions of organosulfurs (Kim et al., 2005; Wu et al., 2010; Zhang et al., 2013). Total odorant strength derived from the different units in the plant was the highest in the hydrothermal hydrolysis unit, followed by the aerobic fermentation unit, sorting/crushing unit, and storage unit (Fig. 4). This order was a little different from that of TVOC emission (Fig. 1). One probable explanation is that more compounds with low odor thresholds were present in the outflow of the hydrothermal hydrolysis unit. Organosulfurs were dominant in olfactory nuisance in the hydrothermal hydrolysis and sorting/crushing units, whereas

Table 2 – Pearson correlation coefficients of the concentrations of the detected VOC emissions ($n = 8$).

	CS ₂	MM	DMS	DMDS	a-Pin	b-Pin	EA	CB	DCM	1,2-DE	Ethano	B	T	E	X	Propan	i-Bu	Pentan	Propyl	Acetalde	Ato
CS ₂	1.000	0.974	0.730	0.779	0.829	0.806	0.819	-0.276	-0.309	-0.105	0.695	0.304	0.727	0.741	0.730	-0.441	0.940	0.976	0.994	0.696	0.838
MM		1.000	0.699	0.850	0.877	0.876	0.920	-0.347	-0.479	-0.305	0.827	0.146	0.824	0.863	0.801	-0.507	0.975	0.994	0.989	0.774	0.876
DMS			1.000	0.835	0.552	0.509	0.490	0.364	-0.371	0.004	0.314	0.360	0.544	0.503	0.553	-0.208	0.614	0.694	0.721	0.385	0.527
DMDS				1.000	0.713	0.736	0.833	-0.012	-0.601	-0.422	0.697	0.021	0.767	0.809	0.726	-0.408	0.806	0.830	0.807	0.609	0.675
a-Pin					1.000	0.990	0.829	-0.341	-0.520	-0.279	0.840	0.206	0.938	0.918	0.927	-0.134	0.779	0.844	0.839	0.975	0.997
b-Pin						1.000	0.880	-0.381	-0.547	-0.347	0.884	0.153	0.962	0.948	0.946	-0.178	0.787	0.836	0.822	0.976	0.982
EA							1.000	-0.475	-0.584	-0.538	0.942	-0.080	0.868	0.933	0.821	-0.558	0.919	0.898	0.859	0.790	0.814
CB								1.000	0.250	0.497	-0.587	0.387	-0.215	-0.388	-0.140	0.472	-0.438	-0.359	-0.318	-0.411	-0.374
DCM									1.000	0.840	-0.695	0.493	-0.612	-0.732	-0.488	0.450	-0.487	-0.474	-0.398	-0.560	-0.489
1,2-DE										1.000	-0.680	0.836	-0.379	-0.595	-0.225	0.544	-0.397	-0.314	-0.208	-0.366	-0.253
Ethano											1.000	-0.270	0.869	0.961	0.794	-0.492	0.837	0.812	0.750	0.862	0.832
B												1.000	0.130	0.282	0.282	0.404	-0.008	0.104	0.209	0.933	0.917
T													1.000	0.955	0.986	-0.178	0.732	0.777	0.752	0.919	0.898
E														1.000	0.897	-0.355	0.820	0.834	0.788	0.919	0.908
X															1.000	-0.090	0.689	0.747	0.738	0.911	0.908
Propan																1.000	-0.665	-0.552	-0.504	-0.064	-0.140
i-Bu																	1.000	0.986	0.968	0.679	0.783
Pentan																		1.000	0.993	0.735	0.847
Propyl																			1.000	0.716	0.845
Acetalde																				1.000	0.971
Ato																					1.000

For abbreviations of the pollutants refer to Table 1. The bold values represent statistically significant correlation ($p < 0.01$, 2-tailed).

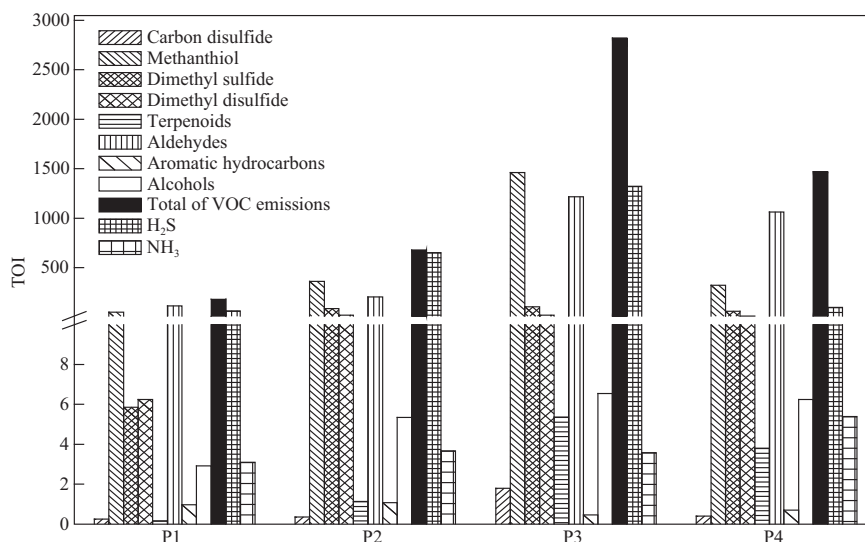


Fig. 4 – Odor contributions of VOC species, H₂S and NH₃.

aldehydes were found to rank first in terms of total odor strength in the aerobic fermentation unit.

Along with VOC emissions, H₂S and NH₃ also contributed to odorous nuisance problems during FW treatment. These two pollutants were designated as offensive odorants that must be controlled in industrial plants (GB 14554-93). The OI of H₂S and NH₃ were estimated using the same method as Eq. (1) and the calculated results are also shown in Fig. 4. The highest OI of H₂S was detected in the hydrothermal hydrolysis unit, followed by the sorting/crushing unit, and this order differed from TOI of VOC emissions. Especially in the sorting/crushing unit, H₂S was found to be the most abundant odorant, approximately accounting for 48.8% of TOI. However, the odorant strength of H₂S had an obvious abatement in the aerobic fermentation unit, possibly because of a better aerobic condition than in the other units. Compared with H₂S, the OI of NH₃, ranging from 3.0 to 5.4, was relatively low throughout the whole treatment system.

PCA analysis enables the evaluation of the inter-relationships between the cases and the variables (Fang et al., 2012; Li et al., 2014). In this study, different treatment units were selected as cases, whereas theoretical OI values of different types of odorants were selected as variables. Three principal components were extracted, while the first two factors had high loading of almost all the odorants except halocarbons, which accounted for approximately 98.4% of the total variance (84.8% and 13.6% for PC1 and PC2 with standard deviation of 1.33 and 0.73, respectively). Fig. 5 shows a bi-plot graph for PC1 and PC2. Point 1 was found to cluster closely with point 2, indicating similar characteristics of odorant emission. The iconic odorants in these locations were aromatics, despite the fact that their OI value was relatively low. One concern for this study was the finding of organosulfurs, H₂S and esters in point 3. The presence of these compounds may have been derived from the hydrothermal and relatively anaerobic environment. Positive signals from PC2 were obtained for NH₃, aldehydes, terpenes, alcohols and

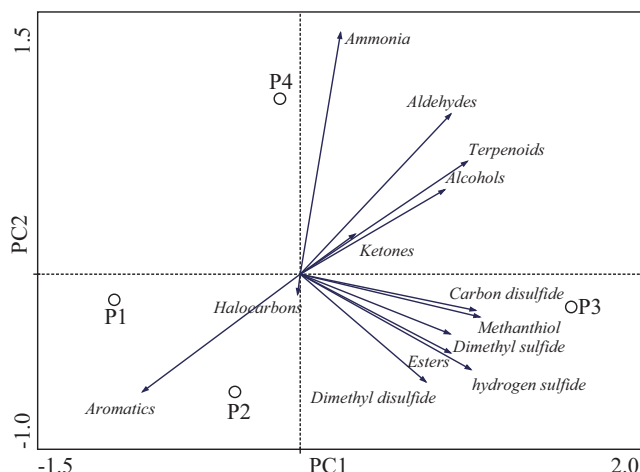


Fig. 5 – Principal component analysis (PCA) bi-plot graph, depicting the inter-relationships between odorants and treatment units.

ketones near point 4, whereas inverse signals were found for organosulfurs and H₂S. These results possibly resulted from a better oxidation environment, which might have increased the oxygenated compounds (except esters) and terpenes while eliminating the sulfur compounds. Both PC1 and PC2 had low loading signals of halocarbons, which were the main abiogenic compounds inherent in waste. Consequently, PC1 and PC2 represented two possible reaction types corresponding to hydrothermal-hydrolysis and aerobic fermentation, which occurred at points 3 and 4, respectively. Thus, from the perspective of odor control, the hydrothermal hydrolysis unit should be the primary control target, followed by the aerobic fermentation unit.

2.4. Contributions of atmospheric reactivity from VOC emissions

In this study, propylene-equivalent (Propy-Equiv) concentration was used to weight the atmospheric reactivity (OH-reactivity-based) of VOC species derived from the FW treatment processes. This method has typically been used to supply a simple estimation of the relative contributions of VOC being oxidized to alter the chemistry of atmosphere in the absence of necessary direct measuring instruments (Duan et al., 2008; Ran et al., 2011; Tang et al., 2007). The calculation equation can be expressed as:

$$\text{Propy-Equiv concentration} = C_J \frac{K_{\text{OH}}(J)}{K_{\text{OH}}(\text{C}_3\text{H}_6)} \quad (3)$$

$$\text{Accumulated Propy-Equiv concentration} = \sum_{i=1}^k \text{Propy-Equiv}(C_i) \quad (4)$$

where, C_J is the quantified concentration of species J expressed in ppbC, and K_{OH} is the OH-reaction rate of individual VOC species obtained primarily from Atkinson (1986) and Tang et al. (2006). Accumulated Propy-Equiv concentration represents the sum of individual Propy-Equiv concentrations.

The results were calculated from the two seasonally averaged concentrations (October and January) of detected VOC emissions to develop a representative level of atmospheric activity. The total Propy-Equiv concentration of certain VOC species was derived by summing up the calculated results of the four sampling points. A total of 39 types of VOC emissions were extracted for calculation of their Propy-Equiv concentrations (Table 1). The top ten substances (in order of their ranking based on the results) were limonene > b-Pinene > a-Pinene > ethanol > acetaldehyde > dimethyl disulfide > propylene > propionaldehyde > pentane > methanethiol. The Propy-Equiv concentrations of all these substances ranged from 1×10^2 to 5×10^3 ppbC levels, except for limonene, which reached the 5×10^4 ppbC level. The sum of these substances accounted for approximately 99% of the accumulated Propy-Equiv concentration. This order was very different from that of odor emissions, and terpene compounds tend to have high contributions to atmospheric chemical reactivity during FW treatment.

Compared with other scenarios, the Propy-Equiv concentrations of VOC emissions from FWTP in this study was one order of magnitude higher than that of wastewater treatment and coking plants in previous investigations (Fig. 6). This result can be explained further by the fact that a large amount of biogenic VOC species, specifically terpenes, oxygen compounds, and organic sulfur compounds, can be generated during the FW treatment process. These compounds usually possess a high level of chemical reactivity.

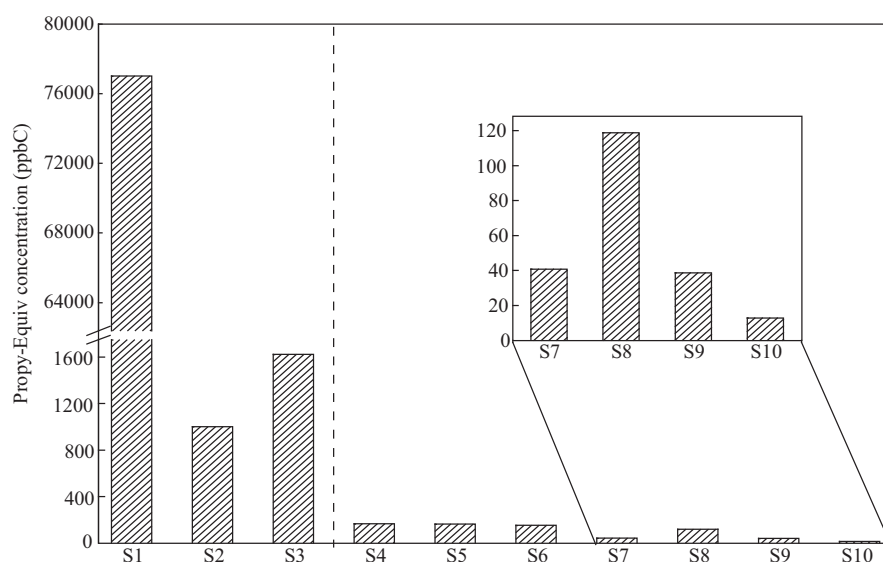


Fig. 6 – Accumulated Propy-Equiv concentrations of VOC emissions in various scenarios. S1–S3 represents industrial scenarios: S1, FW treatment process in this study; S2, waste water treatment process (Tang et al., 2011); S3, coking process, calculated based on the data from Jia et al. (2009). S4–S10 represents normal ambient condition in urban, suburban and rural site in China: S4, urban site, Beijing city in summer of 2008 (Duan et al., 2008); S5, urban site, Tianjing city in summer of 2009 (Lu et al., 2011); S6, urban site, Guangzhou city in spring of 2005 (Tang et al., 2007); S7, urban site, Shanghai city in 2007–2010 (Cai et al., 2010); S8, suburban site, Panyu, Guangzhou city in spring of 2005 (Tang et al., 2007); S9, rural site, Guangzhou city in spring of 2005 (Tang et al., 2007); S10, remote site, Jianfeng mountain, Hainan province in spring of 2005 (Tang et al., 2007).

To receive FW rapidly, FWTPs were usually established in suburban sites not too far from the urban center. Compared with normal ambient chemical reactivity in urban or suburban sites, VOC emissions derived from FWTP can have higher Propy-Equiv concentrations by approximately two orders of magnitude (Fig. 6). Thus, from the perspective of photochemical reactivity, FWTPs are expected to become a significant source to increase the atmospheric reactivity and the potential of ozone formation in a city region.

3. Conclusions

The characteristics of VOC emissions from a full-scale FWTP in China were monitored and investigated on specific days in each season (spring, summer, autumn, and winter) for one year. Several VOC emissions in different families (alkanes, alkene, terpenes, aromatic hydrocarbons, halocarbons, organosulfur compounds, alcohols, aldehydes, and ketones) were quantified with the following mean TVOC concentrations: storage room, 3045.6–7077.5 $\mu\text{g}/\text{m}^3$; sorting/crushing unit, 10,206.3–21,616.3 $\mu\text{g}/\text{m}^3$; hydrothermal hydrolysis unit, 10,798.1–23,144.4 $\mu\text{g}/\text{m}^3$; and aerobic fermentation unit, 21,748.2–31,283.3 $\mu\text{g}/\text{m}^3$. Oxygenated compounds, particularly ethanol, were observed to be the most abundant compounds in all samples. Terpenes, organosulfurs, aldehydes, and alkanes also contributed significant percentages of TVOC, although their order showed slight seasonal differences.

This study highlighted the odorous charge and photochemical reactivity of VOC emissions derived from FWTP. Some compounds (e.g., organosulfurs and oxygenated compounds) were not only major contributors to odors but also had high level of atmospheric reactivity. The highest OI was observed in the hydrothermal hydrolysis unit, followed by the aerobic fermentation unit, with the top contributors being methanethiol and acetaldehyde, respectively. Additionally, H_2S also contributed significantly to odorous problems during FW processing, with OI arranging from 100 to 1323, and it was found to be the most abundant odorant in the sorting/crushing unit. The most important substances from this plant in terms of atmospheric reactivity were terpenes, which comprised a possible primary source for atmospheric VOC increment and thus can impact the ozone formation potential in ambient air.

The *in-situ* monitoring data can provide useful information to support preventive measures and recommendations for VOC abatement in FWTPs, especially for different operational processes. Regular monitoring and selection of suitable collection and treatment systems can reduce or minimize potential environmental risks. Likewise, workers in the critical treatment unit of FWTPs are suggested to wear the necessary protective equipment (e.g., filter masks, gloves) to minimize contact, inhalation, and exposure to toxic VOC emissions.

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