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Volatile Solid and Bury Period Influence on Odorous Material Production in Simulating Landfill Treatment

Peng Lu[1,2], Yuanyuan Zhang[3], Linan Xing[1], Ying Wang[4], Hong Lu[5], Dongbei Yue[3], Wei Cheng[2], Jin Liu[2]

[1]Department of Environmental Engineering Technology, Beijing Polytechnic, Beijing, China
[2]Beijing Municipal Institute of City Management, Beijing, China
[3]Department of Environmental Science and Engineering, Tsinghua University, Beijing, China
[4]Chinese Academy of Meteorological Sciences, Beijing, China
[5]China Meteorological Administration, Beijing, China
Email: lupeng531@163.com

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Abstract

Odor pollution in landfill area has attracted more social attention in China. It is very important to control the generation of odor pollutants *in situ*. Analyzing odorous materials production form buried waste, simulated columns of different volatile solid (VS) content and different buried period waste were designed. Gas compounds produced from the columns were collected and analyzed by comprehensive two-dimensional gas chromatography (GC × GC) method. It has remarkable relationship between VS content and concentrations of odorous material. When VS content more than 40%, the total amount of odorous compounds increases remarkably. It can be inferred that reduced VS content of original waste may effective decreasing odorous materials production in landfill area. The old rubbish produced more odorous compounds than that of fresh one in simulated columns.

Keywords

Volatile Solid, Odorous Material, Bury Period

1. Introduction

In China, the amount of MSW reached 242.06 million tons in 2019, 45.6% of which was disposed in landfills (National Bureau of Statistics of China, 2020). Large landfill was a notable emission source of odorous and hazardous substances, which produced both odor nuisance effects and health risks to nearby

residents (Zhang et al., 2021; Du et al., 2023). For the high proportion of perishable organic matter and high moisture content of domestic waste, odor pollution in the process of treatment is a common phenomenon and has attracted more and more social attention.

The content of organic matter in the mixed waste and the landfill time has obvious effects on the occurrence of malodorous substances. Similar research is rarely reported in domestic literature. Volatile solid (VS) has an obvious indication function for the content of perishable organic matter in domestic waste. Through the analysis of the malodorous substances released from rubbish columns with different VS ratios and different landfill time, the characteristics of the generation and release of malodorous substances can be found, providing a basis for the prevention and control of odor pollution in the landfill.

2. Materials and Methods

The domestic waste landfill column of the same specification is made of PVC (Polyvinyl chloride) plastic as shown in **Figure 1**, with a height of 0.8 meters and a diameter of 0.5 meters. At the bottom of the column is a leachate collection system and a vent pipe. Six water distribution pipes are evenly distributed at the top of the column, and the center is an air outlet pipe. The gas outlet pipe is connected to the condensate pipe, condensate collection bottle, acid absorption bottle and wet flowmeter in turn. The five simulated columns are filled with 77.5 kg of original waste which were taken from the working face of the domestic landfill in Daxing District of Beijing city in China and placed on the scale. The initial height of waste in the column is 0.6 m. After different degrees of aerobic pretreatment, the VS gradients of waste materials are 65%, 50%, 40%, 35% and 30% respectively. After two weeks of storage, generated gas and leachate was sampled and analyzed; In addition, the stale waste excavated from the landfill

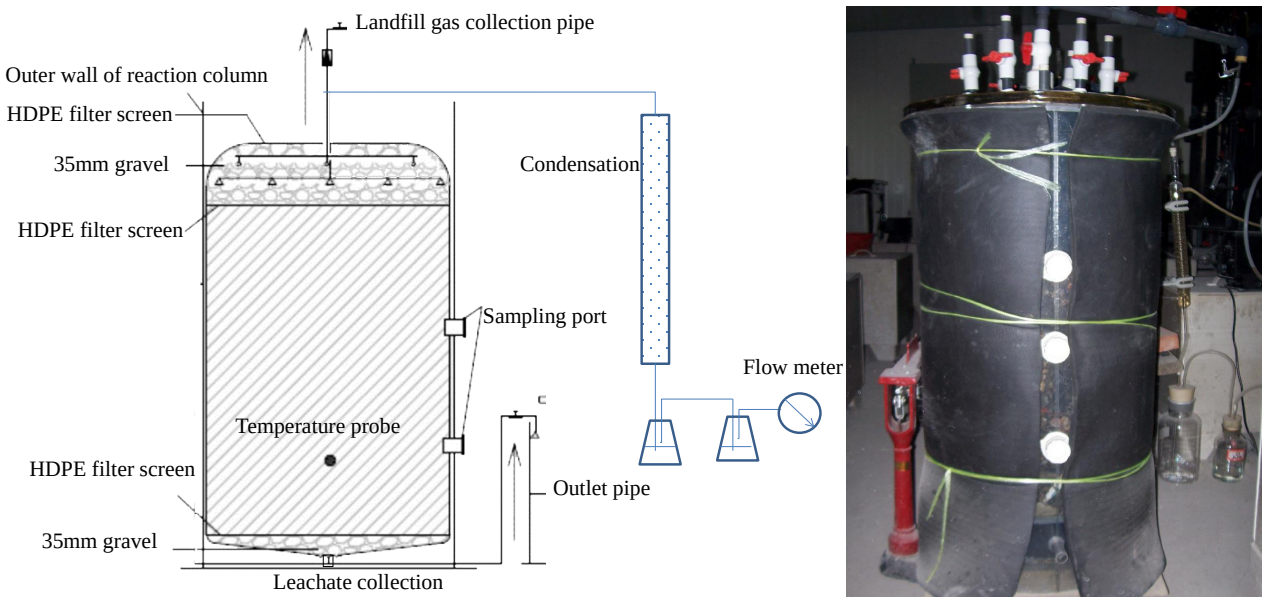

Figure 1. Design layout and photograph of simulated column for landfill treatment of municipal solid waste (MSW).

which buried after 3 years and the fresh waste which buried after 1 year were filled in different columns respectively. After 1 year time experiment, the gas in the column is sampled to analyze the landfill time influence on the malodorous substances production.

The gas samples from domestic waste landfill simulated columns were taken for analysis. Meanwhile, the collected leachate was analyzed. The determination of volatile solids (VS) of the sample is baked to constant weight at 105˚C, and then put into a muffle furnace to burn at 550˚C for 6 h. The percentage of the quality difference (volatile solids) of the sample before and after burning in the muffle furnace to the dehydrated sample (total solids) before burning is the VS content of the sample (Zhang et al., 2008). pH is measured by PHS-25 digital pH meter, Chemical oxygen demand (COD) is determined by potassium dichromate method, biochemical oxygen demand after 5 days (BOD) detected by dilution and inoculation method (GB 7488-87) (Ministry of Ecology and Environment of the People's Republic of China, 2009), total nitrogen (TN) detected by alkaline potassium persulfate digestion-UV spectrophotometric method (GB11894) (Ministry of Ecology and Environment of the People's Republic of China, 2012), Ammonia nitrogen was tested by sodium reagent method.

A 6 L SUMMA canister was used to collect the gas sample that produced in simulated columns. Within 24 hours after sampling, the gaseous compounds are analyzed by comprehensive two-dimensional gas chromatography (Xu et al., 2003). The sample was concentrated into a cold trap which contains two beds of sorbent, i.e. Tenax TA and Carbograph, supported by quartz wool before analysis by gas chromatography coupled with flame ionization detector. The measurement setup consisted of a flow controller, a thermal desorber (Markes International, UK), and a gas chromatograph (GC6890, Agilent, USA) equipped with a flame ionization detector and jet-modulated parts (KT2001, Zeox, USA) for comprehensive two-dimensional gas chromatography (GC × GC) according to the method used by Xu et al. (2003). The sampling and desorption control software (Markes International, UK) and Chem Station (Agilent, USA) installed on a personal computer were used to control the sampling/thermal desorption system and the gas chromatograph, respectively

A 150 mL gaseous sample was extracted from the adsorption tube (Tenax/Carbograph 1TD/Carboxen 1000), and the sampling flow was set at 50 mL/min. The thermal desorption temperature was 300 C. Both cold and hot jets of nitrogen gas were used. The standard mixture, EPA TO-15, was used for calibration. The jet modulated system was operated for 6 s at a cryotrap temperature of 20˚C.

The column set used for GC × GC equipment was composed of an HP-5MS (5% phenyl, 95% dimethylpolysiloxane phase) primary column (30 m × 0.25 mm I.D. × 0.25 mm film thickness) directly coupled to a DB-WAX (polyethylene glycol phase) secondary column (1.05 m × 0.1 mm I.D. × 0.1 mm). The temperature program for the primary column began at 35˚C, holding for 8 min, and then rose to 221˚C at 3˚C/min. For the secondary column, the temperature was

initially held at 30˚C for 8 min, and then raised to 210˚C at 3.3˚C/min and held for 7.5 min. The transfer line column for the system was a 1.5 m deactivated fused silica column with a 0.1 mm I.D. Liquid nitrogen was used to refrigeration the transfer line column and keep 6 seconds of modulation period.

3. Results and Analysis

3.1. Influence of Volatile Solid (VS) Content on Pollutant Production

Through the analysis of collected leachate, it is found that chemical oxygen demand (COD) and biological oxygen demand (BOD) have an obvious trend of increase with the increased of volatile solid (VS), while the change of total nitrogen and ammonium nitrogen (NH_3-N) is not obvious as shown in Table 1.

There are a total of 89 compounds were detected in the gas samples by comprehensive two-dimensional gas chromatography. These materials can be divided into 8 kinds of chemical compounds, including aromatic hydrocarbons, ketones, olefins, aldehydes, cycloalkanes, chlorinated hydrocarbons, straight-chain hydrocarbons, esters and alcohols. There is different concentration change trend with the different VS content treatments. Accumulate the concentration of all quantitatively detected compounds and calculate the total volatile organic compounds (VOCs) concentration. It can be seen that there is a certain relationship between VS content and VOCs release as shown in Figure 2. When VS content exceeds 50%, VOCs release increased sharply.

The VOCs concentration increases in uneven content percentage come from two aspects. Firstly, the analytical methods used cannot identify all VOC substances. Secondly, VOCs concentration increase in uneven content percentage comes from sampling and measurement errors. This kind of error is amplified by the fact that the drawing scale is measured in micrograms per cubic meter. In fact, this change does not affect the final conclusion relative to the experimental gradient.

3.2. Correlation between Volatile Solid (VS) and Total VOCs

The correlation between VS (volatile solid) and the quality of various VOCs were analyzed. There was a significant correlation between VS content and the total amount of ketones (Table 2, $P = 0.05$), but the correlation between VS and total VOC is not obviously.

Table 1. Pollution parameters of leachate from simulated columns.

VS (%)	30	35	40	50	65
COD (mg/m^3)	37,808	23,973	28,082	40,753	105,556
TN (mg/m^3)	3940.4	3564.6	3672.0	3741.6	3820
NH_3-N (mg/m^3)	1352.7	2124.8	2156.7	3725.3	2056
BOD (mg/m^3)	10218.0	6055.8	8187.8	20944.2	91497
pH	8.66	7.45	7.07	6.83	5.42

Table 2. Correlation analyzing of volatile solid (VS) and volatile organic Compounds (VOCs).

Item	Aromatic hydrocarbons	Ketones	Alkene	Aldehydes	Cycloalkanes	Chlorinated hydrocarbons	Linear hydrocarbon	Esters	Alcohols	Total
VS	−0.175	0.869*	0.791	0.729	0.776	0.737	0.632	0.715	0.643	0.792
Aromatic hydrocarbons		0.277	0.408	0.453	0.432	0.493	0.562	0.459	0.514	0.418
Ketones			0.985**	0.947**	0.979**	0.960**	0.811	0.947**	0.895*	0.983**
Alkene				0.986**	0.999**	0.978**	0.848*	0.986**	0.955**	0.998**
Aldehydes					0.989**	0.963**	0.857*	0.993**	0.991**	0.986**
Cycloalkanes						0.982**	0.861*	0.987**	0.962**	0.999**
Chlorinated hydrocarbons							0.918**	0.948**	0.935**	0.986**
Linear hydrocarbon								0.807	0.848*	0.874*
Esters									0.980**	0.980**
Alcohols										0.956**

Note: *Significant correlation at 0.05 level (2tail test), **Significant correlation at 0.01 level (2tail test).

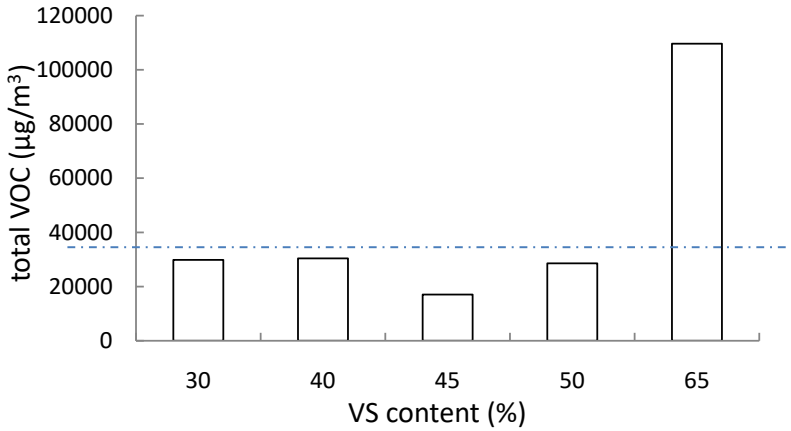

Figure 2. VOCs concentration in different VS content simulated columns (μg/m^3).

It can also be seen from the last column of Table 2, among the eight main compounds constitute the total VOC, ketones, alkene, aldehydes, cycloalkanes, chlorinated hydrocarbons, esters and alcohols are extremely significantly correlated with the total VOC release (P = 0.01), and linear hydrocarbon are significantly correlated with the total VOC release (P = 0.05). Only aromatic hydrocarbons have no relationship to total VOC release.

To detecting which category of compounds that directly related to volatile solid (VS) content, the correlation between the VS values and the concentration of 89 VOCs materials has analyzed as shown in Table 3.

The results showed that VS was positively correlated with o-xylene, 2-hexanone, camphene, cyclohexane and isopropanol (P = 0.05), and the correlation coefficients were 0.863, 0.848, 0.878, 0.881, 0.851 and 0.893, respectively. There was a

Table 3. Materials influenced by VS contents.

Item	Aromatic hydrocarbon		Ketones	Alkene	Linear hydrocarbon		Cycloalkanes	Alcohols
	Oxylene	Styrene	2-hexanone	camphene	2,3-dimethylbutane	2,2,4-Trimethylpentane	Cyclohexane	Isopropanol
Correlation positive and negative	+	−	+	+	+	−	+	+
Correlation significance	*	**	*	*	*	*	*	*
Correlation coefficient value	0.863	−0.940	0.848	0.878	0.881	−0.822	0.851	0.893

Note: *Significantly correlated ($P = 0.05$), **Extremely significant correlation ($P = 0.01$).

significant negative correlation between VS and 2,2,4-trimethylpentane ($P = 0.05$), and the correlation coefficient was −0.822. There was a very significant negative correlation between VS and styrene ($P = 0.01$), and the correlation coefficient was −0.940. Among the above substances, O-xylene, styrene and camphene are substances with obvious bad smell.

The analysis shows that aldehyde, chlorinated hydrocarbon and ester have significant contribution to VOCs release (extremely significant positive correlation, $P = 0.01$). But, there is no significant correlation between VS and specific compounds belonging to these three substances.

3.3. Buried Time Influence on Odoriferous Compounds Production

The landfill simulating experiment was carried out with the rubbish that had been buried in landfilled area for 2 years and that had been buried for 4 years respectively, and the VOCs produced in the simulated columns were compared as shown in **Table 4**. The values of BOD and COD of leachate which produced in simulated column filling fresh rubbish (after 2 years of buried in landfill area) are significantly higher than that filling old rubbish (after 4 years of landfill treatment). The content of total nitrogen and ammonia nitrogen content in different treatment was showing the same regular.

The VOCs substance analysis of the gas released from the landfill column showed that the total VOCs released by the old rubbish was lower than that fresh one as shown in **Table 5**.

It can be seen that the release of other VOCs from the simulated column of old rubbishis relatively large, except for the slightly small release of olefins. The total release amount reached 7.6 times to that of the fresh rubbish simulation column (**Table 5**). Benzenes are recognized as important representative compounds in the study of landfill odor pollution (Davoli et al., 2003; Zou et al., 2003; Dincer, Odabasi, & Muezzinoglu, 2006), the concentrations of benzene homologues in landfill columns at different buried time were compared. The total amount of benzene series is about 4.7 times to that of fresh rubbish simulated column.

Table 4. Parameters of rubbish and leachate in the simulated columns.

Buried time	Rubbish		Leachate				
	VS	Water concentration	COD mg/m^3	TN mg/m^3	NH_3-N mg/m^3	BOD mg/m^3	pH
4 years	15.6%	40.6%	863.3	503.2	21.8	40.4	6.75
2 years	25.3%	50.4%	15555.6	4821.2	3906.6	8593.5	6.26

Table 5. VOCs concentration in simulated columns (μg/m^3).

VOCs type	Old rubbish (buried 4 years)	Fresh rubbish (buried 2 years)	Difference between two treatment Old rubbish/Fresh rubbish
alcohols	292	20.0	14.6
aldehydes	200.1	39.0	5.1
ketones	189.7	34.0	5.6
esters	312.4	18.7	*16.7*
aromatic hydrocarbons	250.8	53.5	4.7
alkanes	357.5	42.6	*8.4*
alkenes	38.8	7.03	5.5
total	1641.0	214.9	*7.6*

Note: show the ratio of total VOCs released in 4 years landfill treatment to that in 2 years of landfill treatment.

The total amount of benzene homologues released from old rubbish is 4.7 times to that in fresh one as shown in **Table 6**. The concentration of other benzene series compounds from old rubbish were higher than that of fresh rubbish except for the 1,2,4-trimethylbenzene (**Table 6**). The eight compounds such as chlorobenzene, biphenyl, benzene, acetyl benzene, benzaldehyde, isopropyl benzene, naphthalene, and phenol have a significant difference between the old and fresh rubbish, with the volume differences of 14.2, 13.1, 12.4, 9.0, 7.0, 5.5, 5.2, and 4.7 times, respectively. Except isopropyl benzene, benzene, benzaldehyde, naphthalene, phenol, acetyl benzene, and chlorobenzene are also the top seven substances released in the old landfill column.

4. Discussions

The VS values of experiment are detected by sampling from different height in the simulated column and repeat for several times to provided representing the real VS in the experiment. The VS gradient is obtained after loading the same waste material in experiment beginning and after aerobic treatments for several months. The VS value is lower than that of compost maturity stage (approximately 30 days) (Luo et al., 2004).

Some researcher founded the concentration of VOCs materials have positive correlation to odorous pollution (Dincer, Odabasi, & Muezzinoglu, 2006). The

Table 6. Aromatic compounds concentration in simulated columns of different rubbish.

Compounds	Old rubbish	Fresh rubbish	Difference between landfill time
benezene	93.14	7.51	12.4
benzaldehyde	53.41	7.65	7.0
naphthalene	19.51	3.76	5.2
Phenol	12.23	2.59	4.7
toluene	10.31	4.25	2.4
acetophenone	7.4	0.82	9.0
chloro-benzene	6.96	0.49	14.2
biphenyl	6.66	0.51	13.1
m,p-xylene	6.61	4.49	1.5
2-methyl-naphthalene	6.55	1.7	3.9
styrene	5.88	5.25	1.1
ethylbenzene	4.42	2.61	1.7
o-xylene	3.91	2.55	1.5
1-ethyl-4-methyl-benzene	3.82	0.91	4.2
1-methyl-Naphthalene	3.42	0.91	3.8
benzene, isoprophyl	2.62	0.48	5.5
1,2,4-trimethylbenzene	1.94	5.59	0.3
1,3,5-trimethylbenzene	1.19	1.12	1.1
propylbenzene	0.78	0.34	2.3
total	250.76	53.53	4.7

Note: show the ratio of total VOCs released in 4 years landfill treatment to that in 2 years of landfill treatment.

experiment filling different buried time rubbish from landfill site show that: the large amount of VOCs compounds released after a period of burry treatment, especially the aromatic odorous compounds. This phenomenon shows that there has an obvious periodicity of the transfer and changing of odorous compounds in municipal solid waste landfill treatment.

Generally, the degradation order of macromolecular organic compounds in nature from fast to slow is nucleic acid, protein, polysaccharide, fat, cellulose, and lignin. Lignin is very difficult to degraded in macromolecular Because lignin is a polymer composed of monomers derived from phenyl propane (Xie & Luo, 2007), Lignin can be considered as a complex phenolic polymer, formed by free radical polymerization (i.e., lignification) of syringyl (S), guaiacyl (G), and p-hydroxyphenyl (H) units. The benzene ring is its characteristic structure, and a large amount of benzene series compounds may come from the degradation of lignin like substances. The landfill experiments of old rubbish and fresh rubbish have also verified this speculation from one side.

Sulfur-containing compounds are representative substances of malodorous materials that emission from landfill area (Kim et al., 2005). Sulfur-containing compounds (excluding hydrogen sulfide) in well managed landfills generally have extremely low concentrations measured on-site, and the total amount generally does not exceed 0.5 μg/m^3 (Saral et al., 2009). It is often not detected during on-site inspections at landfills by the reason of its proportion in the total VOCs is less than 1%. Therefore, although limited by detection methods, sulfur containing compounds and compounds with less than 4 carbon atoms are not included. The overall results can still indicate the generation of odorous substances during the landfill process.

5. Conclusion

The total release of VOCs amount has significant positive correlation to VS; Styrene in benzene series has significant negative correlation to VS that weakening the correlation between the total amount of benzene homologues and VS. The old rubbish produced more odorous compounds than that of fresh one in the experiment inferred that waste buried period also has a significant impact on odorous material production. The large amount of VOCs released after a period of buried treatment in landfill area.

Control VS value below 50% in original rubbish by some pretreatment method before landfill operation might effectively reduce the odorous substances generation in the later stage. In the next experiment, 55% and 60% VS content treatment experiments should be set up to explore the critical points of VOC release. Reasonable pre-treatment to reduce the content of VS in landfill materials, such as composting and screening methods, will be the focus of future research.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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