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**Proceedings of the  
2<sup>nd</sup> Chinese-Austrian Workshop  
on Environmental Odour:  
Odour Emission - Dispersion -  
Impact Assessment - Abatement**

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The general objective is to promote and extend the use of statistical and mathematical methods in veterinary epidemiology. Special emphasis is given on methods and results. Monographs, paper collections or conference proceedings will be published in German as well as in English in the Austrian Contributions to Veterinary Epidemiology if judged consistently with these general aims. All contributions will be refereed.

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## Preface

Environmental odour is perceived as major nuisance by rural as well as by urban populations. The sources of odorous substances are manifold. In urban areas restaurants, small manufacturing businesses and other sources can be found inside residential areas. In the suburbs we can expect waste water treatment plants, landfill sites and other infra structures as major causing sources. These problems are often aggravated by the accelerated growth of cities. In rural spaces, livestock farming and the spreading of manure on the fields is blamed for severe odour nuisance. As a matter of fact, environmental odours are considered to be a common cause of public complaints by residents to local authorities, regional or national environmental agencies.

In the 1<sup>st</sup> Chinese-Austrian Workshop on Environmental Odour held in Tianjin, China in February 2015 this environmental issue was addressed to compile experiences in this field. Participants from several universities as well as state agencies took part. The goal of this workshop was to establish cooperations in this field. One of the major results of this workshop was a bilateral joint project between the University of Science and Technology Beijing and Austrian partner organisations. The proceedings of this workshop were published in Volume 8 of the *Austrian Contributions to Veterinary Epidemiology*, a journal, which offers open access to all papers.

In February 2016, the 2<sup>nd</sup> Chinese-Austrian Workshop on Environmental Odour was held in Shanghai, as one of the hotspots of environmental odour research in China. The workshop was hosted by Prof. Dr. Pinjing He, Institute of Waste Treatment & Reclamation at the College of Environmental Science and Engineering, Tongji University in Shanghai. The workshop included the following topics (1) Characterisation of odour sources by emission factors and emission models, (2) Monitoring of odour emission in diverse environments, (3) Equipment and methods of odour measurement, and (4) Assessment of the relevant stimuli concentration and the odour impact criteria. Prof. Dr. Pinjing He and his team deserve gratitude for the successful organisation on site and his Austrian counterpart Prof. Dr. Günther Schauburger from the University of Veterinary Medicine Vienna for the initiation and organisation of the meeting. Both are the guest editors for this issue.

The workshop as well as Volume 9 of the *Austrian Contributions to Veterinary Epidemiology* was partly funded by Eurasia-Pacific Uninet as a network, which aims at establishing contacts and scientific partnerships between Austrian universities and member institutions in East Asia, Central Asia, South Asia and the Pacific region.

In the light of the current air quality crisis confronting China and the world, I am confident that such bilateral efforts are an incentive for finding future solutions by cooperation of our two countries.

Vienna, February 2017

Prof. Dr. Wolf-Dieter Rausch  
President of the Eurasia-Pacific Uninet



## Determination of the hedonic odour tone in China and the behavior curve of ammonia

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The hedonic tone of 73 panellists is tested with reference to the Guideline VDI 3882 with two standard odorants: vanillin(5 g/L) and guaiacol (5 µg/L). For guaiacol, about 77% of the panellists judged the smell to be unpleasant with the mean value -1.3. For vanillin, about 81% of the panellists observed a pleasant tone with the mean value of 1.2. Based on the data analysis, the domestic screening criteria for the panel of the hedonic tone test (not the individual hedonic tone) is obtained as follows:

vanillin: from +1.1 ~ +2.4

guaiacol from -1.6 ~ -0.4.

Ammonia is presented as an example in this paper. The odour concentration of the original sample was 1713, and it was diluted to 5 concentration gradients which differ by a factor of approximately 3. The result shows that the hedonic tone at different dilution levels all belong to the category of unpleasant. Moreover, the hedonic tone was approximately 0 with an odorant concentration of 26 ppm. In addition, the behaviour curves of the hedonic tone and odour concentrations are plotted. The trend of the behaviour curve demonstrates a multiple nonlinear relationship between the hedonic tone and the odour concentration, and the regression equation is  $Y = 3.26 - 3.26 X + 0.42 X^2$  ( $R^2=0.99$ ). The model can reflect the changing tendency of the hedonic tone at different concentrations, which will be helpful for the prediction of the hedonic tone.

### 1. Introduction

Environmental odour is an important subcategory of perceived air pollution, which is discussed under the common heading of environmental stress together with noise, heat and light (Sucker et al.,2008). With the progress of society, people seem to have become less tolerant to environmental odour (Capelli et al.,2013), and it has become a major environmental problem among neighborhood communities, local municipalities, state agencies and national governments around the world (Carmo et al.,2010). There are great differences in the odour substances and characteristics of different sources. At present, the odour concentration, intensity, odour quality are the most important indicators for the odour impact evaluation, among which the odour concentration is the most widely used (Nicell et al.,2009). However, odour concentration refers to the dilution times of odour samples with clean air to the olfactory threshold, and it has nothing to do with the characteristics of the odour. Therefore, odour concentrations cannot reflect the discrepancy in the effects of different odours.

The hedonic tone is a measurement which can truly reflect the person's "pleasant - unpleasant" psychology. It is contingent on the nature of the odorous substance or mixture of the substances, on the odorous concentration and hence on the perceived intensity of the odour, and on the mental condition of the individual person. Experience

has shown that the hedonic tone can reflect the injury and the psychological influence better than odour concentration (Both et al., 2004). A study on the odour classification was carried out in 2004, it is considered that the hedonic tone is an important dimension of classification for the people with diverse cultural background (Chrea C et al., 2004). Besides, the hedonic tone has been found to affect annoyance and the number of highly annoyed persons (Steunenbergh, 1998), and also to strongly influence associations between odour exposure and symptom reporting (Sucker et al., 2009). A large scale of surveys was executed from 1999 to 2001, the results showed that only the unpleasant and neutral odour would cause the annoyance and complaint, and the pleasant smell almost won't induce annoying reactions (Sucker et al., 2008). In Germany, a standard of hedonic tone determination (VDI 3882, 1994) was published in 1994. It proposed a nine-point scale with values ranging from "-4 - extremely unpleasant" through "0 - neither pleasant nor unpleasant" ("neutral") to "+4 - extremely pleasant". Furthermore, the standard proposed the qualified criterion of the panel as follows:

vanillin: from +1.9 ~ +2.9

guaiacol: from -0.8 ~ -2.0

Currently, the evaluation and management of odour pollution in China is mainly based on the odour concentration, yet the studies and applications on the hedonic tone have not been carried out. In this paper, the panel selection criterion is proposed through the test of two odorous substances referred to VDI3882. Furthermore, ammonia is presented as an example to reveal the relationship between the hedonic tone and the odour concentration, and a mathematical model of the two factors is established.

## 2. Material and methods

### 2.1 Method for panel selection

VDI 3882 stipulates the panel selection criterion: vanillin is from +2.9 to +1.9, while guaiacol is from -0.8 to -2.0. However, the psychological feelings for a certain odorant may be different between China and Western countries because of the discrepancy of the cultural background and living habit. So it is necessary to propose a panel selection criterion which will be conform to the actual condition of China.

73 members aged from 18 to 45 participated in the panel screening, including 36 males and 37 females. They are all tested with the reference materials: vanillin (5 g/L, dipropylene glycol), guaiacol (5 µl/L, distilled water). The fresh samples have been prepared each day and have been presented in 500 mL wide-neck bottles with 45 mm standard ground stoppers. Each bottle contained 200 ml of the solution. The assessor was asked to evaluate the hedonic tone one by one. The fixed sequence is as follows:

1. vanillin

2. guaiacol.

The results of the evaluation are given according to the 9 point hedonic scale as shown in Table1.

Before the experiment, the members have been told that there is no "right" or "wrong" answer in the test. They should respond on behalf of their individual feelings. Once the members got the bottle, they should open the stopper immediately, sniff two or three times and then quickly replace the stopper. The decision according to the 9 point hedonic scale should be given as spontaneously as possible, without too much reflection. The evaluation process can be carried out only once.



*Table 1: 9 point hedonic scale for odour.*

Hedonic Tone	Verbal description
-4	extremely unpleasant
-3	moderate unpleasant
-2	unpleasant
-1	slightly unpleasant
0	neutral
1	slightly pleasant
2	pleasant
3	moderate pleasant
4	extremely pleasant

## 2.2 Procedure for the hedonic odour tone evaluation

The panel should consist of at least 15 persons to account for the inter-individual differences which may occur when determining the hedonic odour tone. The odour samples to be presented to the panel members consist of several dilution levels according to the method of the triangle odour bag. The lower limit corresponds to the panel threshold. The entire range of concentrations to be presented to the panel members is checked for toxicity, and all possible risks of injury to the panel members must be reliably excluded. In the case of low odorant concentrations, a smaller number of dilution levels may be presented.

The presentation of odour samples is carried out at random. With this method, the dilution levels are presented in any desired sequence for each series of the measurements. Adaptation effects must be minimized by ensuring that an above-threshold odour sample should not be presented for longer than 15 seconds, with an allowance of an additional decision time of 5 seconds. The minimum break between any two stimuli should be at least 1 minute. In order to avoid the phenomenon of guessing by the panellists, additional samples of pure air was offered in a stochastic order.

It is necessary to ask the panel members whether he or she smelled the odour or not. If the answer is "Yes", the hedonic tone of the perceived concentration should be evaluated according to the 9 point hedonic scale. If the answer is "No", then a symbol of " - " will be recorded.

*Table 2: Hedonic tone of vanillin and guaiacol.*

Hedonic Tone	Vanillin	Guaiacol
+4	1	3
+3	11	6
+2	21	16
+1	26	31
0	3	15
-1	7	0
-2	4	1
-3	0	0
-4	0	1
Average Value	1.3	-1.2

### 3. Results and discussion

#### 3.1 The criteria of the panel selection

The hedonic tones of vanillin and guaiacol are shown in Table 2. The proportion of the hedonic tones of the two standard materials are presented in Figure 1 and Figure 2, respectively.

For vanillin, nearly 59 (81%) members record a pleasant tone, while about 11 (15%) people feel unpleasant, and 3 (4%) persons consider it neither pleasant nor unpleasant. The results are mainly concentrated in the range of +1 ~+3 with an average value of 1.3, which is lower than the criterion presented in VDI 3882. It means that the psychological impact of vanillin for the Chinese is not as strong as for Germans.

The major proportion of the panel judged guaiacol as unpleasant (56 (77%) persons). About 15 (21%) people, which occupy the second largest rate, feel neither pleasant nor unpleasant, only 2 (2%) members feel pleasant. The results are mainly concentrated in the range of 0 ~-2, with the average value of -1.2, which belongs to the category “slightly unpleasant”.

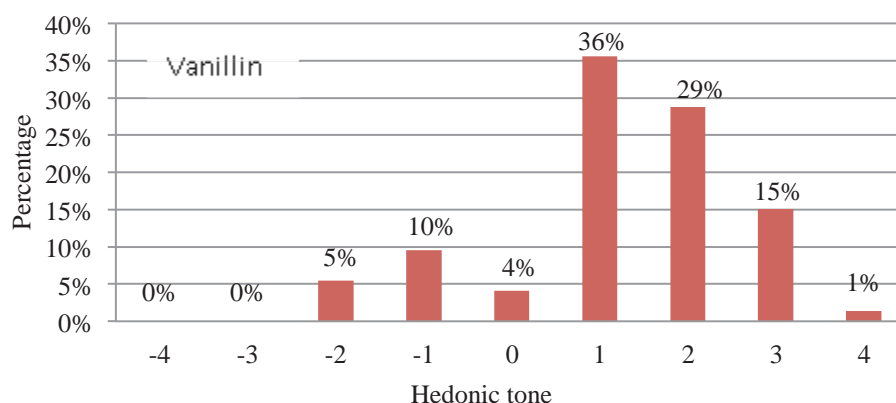


Figure 1. The hedonic tone percentage of vanillin.

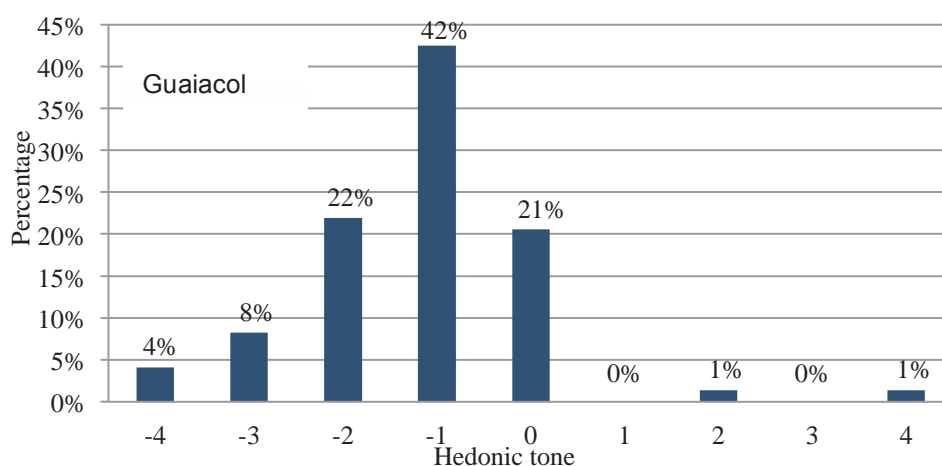


Figure 2. The hedonic tone percentage of guaiacol.

The criterion for panel selection is not the panel member's sensitivity, but the mean value of the whole panel results. The values of the hedonic tone given by the panel

members are subject to the normal distribution, the range of values under different confidence levels can be determined by the formula:

$$[\bar{X} - \frac{\sigma}{\sqrt{n}} Z_{\frac{\alpha}{2}}, \bar{X} + \frac{\sigma}{\sqrt{n}} Z_{\frac{\alpha}{2}}] \quad (1)$$

Where

$\bar{X}$  - mean value,

$\sigma$  - standard deviation,  $n$  quantity of sample

$Z_{\frac{\alpha}{2}}$  - a coefficient that can be obtained by the normal distribution table

From the perspective of statistics, the higher the confidence level, the larger the probability that the sample falls into the interval, and the wider the corresponding interval. However, for the panel selection, the confidence interval is not as wide as possible. It is necessary to exclude the extreme values in order to improve the panel selection conditions. In this study, the hedonic tone samples of vanillin are chosen as {1, 2, 3}, while those of guaiacol are {0, -1, -2}. Through the comprehensive analysis of the range corresponding to different confidence levels and the distribution characteristics of the hedonic tone determined by the panellists, the panel selection criteria are finally put forward as follows:

vanillin: +1.1 ~ +2.4 ,

guaiacol: -1.6 ~ -0.4.

### 3.2 Determination of the typical odorant hedonic tone

Ammonia ( $\text{NH}_3$ ) is presented as a typical odorant. The original odour concentration of the sample is 1713 ou/m<sup>3</sup> (under the assumption that 1 ppm correspond to 1 ou/m<sup>3</sup>), and it is diluted in steps of three by 0, 3, 10, 30, 100. The hedonic tone at different odour concentrations is determined by 18 qualified panel members.

The results are shown in Table 3. Generally, the hedonic tones are all negative, which means that  $\text{NH}_3$  belongs to the range of unpleasant substances. At the concentration index of 3.23, the corresponding hedonic tone is the maximum (-2.89). As the dilution factor increases, the degree of aversion gradually mitigates, until the concentration index declines to 1.23, at which point the average value of the hedonic tone decreases to the minimum of -0.08.

Fig. 3 demonstrates the percentage of the hedonic tone at different concentrations. For the concentration level of  $Z_1$ , the level “-3” ranks the first, which accounts for 39%. The levels “-2” and “-4” occupy the second largest proportion with 28% each, and the level “-1” constitutes only 5%. As the dilution factor increases, the proportion of “serious disgust” is gradually reduced, while the percentage of “slight disgust” increases. The odour concentration of  $Z_5$  is the lowest with the concentration index of 1.23. At this point, about 78% of the panellists feel neither pleasant nor unpleasant, which makes up the largest rate, and only 4% persons feel unpleasant.

### 3.3 Relationship between odour concentration and hedonic tone

A behaviour curve of the hedonic tone in the above-threshold concentration range can provide useful information for the extent to which abatement measurements are necessary. Assuming that the downstream waste gas cleaning process doesn't change the composition of the crude gas with respect of its constituents, the behaviour curve

may serve to determine what odorant concentration of cleaned gas is permissible in order to obtain a gas emission with an acceptable hedonic tone.

Table. 3 Hedonic odour tone evaluation for  $\text{NH}_3$ .

Panel member	Blank sample		Concentration steps	Z <sub>1</sub>	Z <sub>2</sub>	Z <sub>3</sub>	Z <sub>4</sub>	Z <sub>5</sub>
			lgZ <sub>k</sub>	3.23	2.76	2.23	1.76	1.23
1	0	-	Hedonic tone	-4	-2	-2	0	0
2	-	-		-3	-2	-1	-1	0
3	0	0		-2	-1	-1	0	0
4	-			-3	-1	-2	-1	-
5	1	1		-3	-1	0	-	2
6	-	-		-4	-2	-1	0	-
7	0	0		-2	-1	-1	0	-
8	-	-		-3	-3	-2	-1	-
9	0	0		-2	-2	-2	-1	0
10	0	0		-4	-2	-1	-2	0
11		0		-2	-2	-2	0	0
12	0	0		-3	-2	-2	-2	-1
13	-	0		-2	-1	0	-2	-1
14	-	-		-4	-2	-4	-3	-1
15	0	1		-1	-1	-1	0	-
16	-	-		-3	-2	-2	-1	-
17	-1	0		-4	-4	-4	-2	0
18	-	-		-3	-2	-2	-1	0
Frequency	+4		0.00	0.00	0.00	0.00	0.00	
	+3		0.00	0.00	0.00	0.00	0.00	
	+2		0.00	0.00	0.00	0.00	0.00	
	+1		0.00	0.00	0.00	0.00	0.00	
	0		0.00	0.00	0.11	0.39	0.78	
	-1		0.06	0.06	0.33	0.33	0.17	
	-2		0.28	0.56	0.44	0.22	0.06	
	-3		0.39	0.33	0.00	0.06	0.00	
-4		0.28	0.06	0.11	0.00	0.00		
Average value			-2.89	-2.40	-1.96	-1.21	-0.08	

The behaviour curve of the hedonic tone as a function of odour concentration index is shown in Fig.4. A significant decrease in the hedonic tone by concentration was found. When the odour concentration index is 1.42 (odour concentration approximately 26 ou/m<sup>3</sup>), the corresponding hedonic tone is -0.5. If the concentration index is less than 1.42, people will feel neither pleasant nor unpleasant, which means that the odour will have no influence on persons. In order to prevent from odour pollution of  $\text{NH}_3$ , the odour concentration should be below 26 ou/m<sup>3</sup>.

To fit the hedonic tone and the odour concentration index by the data analysis software of Origin, the following regression equation is obtained:

$$Y = 3.26 - 3.26 X + 0.42 X^2 \quad (2)$$

with the hedonic tone Y and the odour concentration index X.

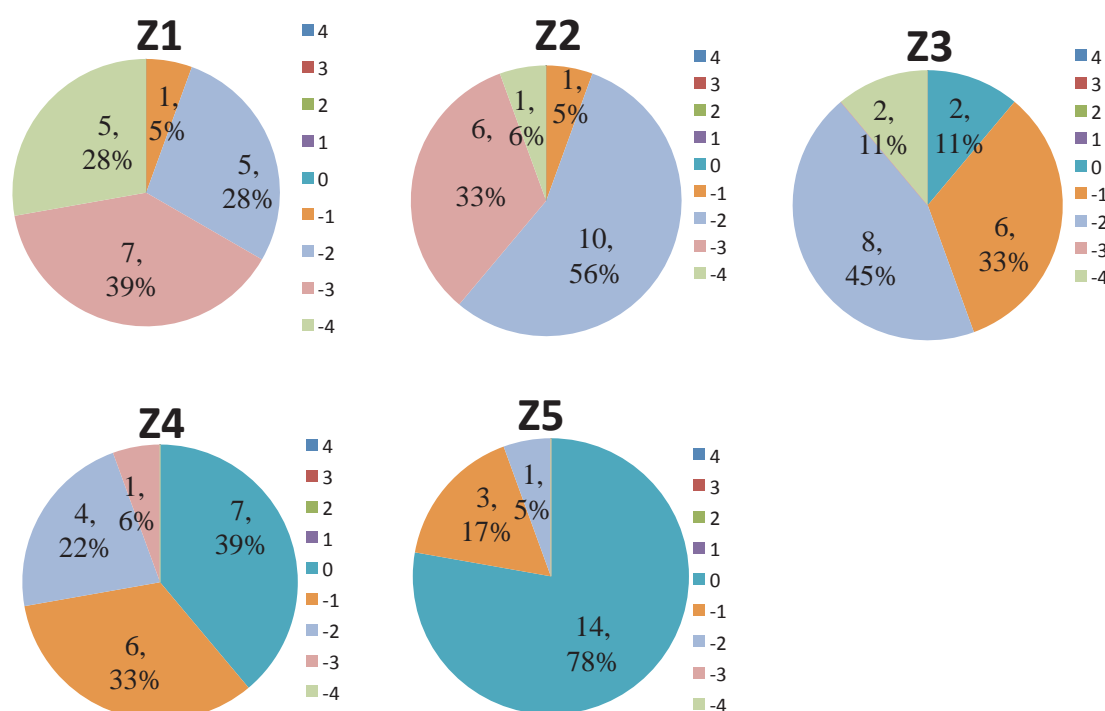


Figure 3: The percentage of hedonic tones at different concentrations.

The correlation coefficient is 0.99, which means a strong correlation between the two factors. The model indicates that there is a multiple nonlinear relationship between the hedonic tone and the odour concentration

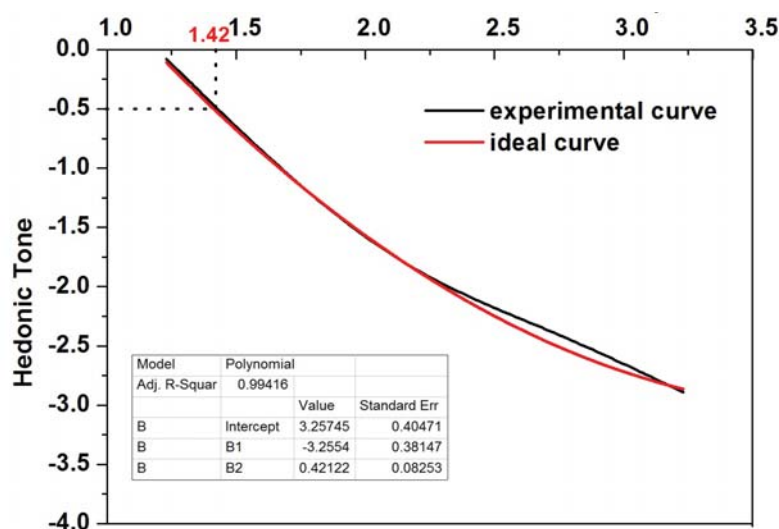


Figure 4: Relationship between the odour concentration index and the perceived hedonic tone of  $\text{NH}_3$ .

#### 4. Conclusion

The hedonic tone is a measure which can quantify the subjective feeling of individuals (pleasant or unpleasant). In this study, the characteristics of the Chinese hedonic tone are analyzed with the two reference materials according to the method provided by VDI 3882, and a panel selection criterion is proposed. Furthermore, the hedonic tone of  $\text{NH}_3$  is determined to study the relationship between hedonic tone and odour concentration. The conclusions drawn from the research are as follows:

- (1) For vanillin, 81% of the members feel pleasant with the average value of 1.3 which is lower than the criterion of 1.9 ~ 2.9 presented in VDI 3882. For guaiacol, 77% of the members feel unpleasant with the average value of -1.2 which conforms to the criterion of -2.0 ~ -0.8 presented in VDI 3882.
- (2) The criterion of panel selection for China is put forward as follows: vanillin: +1.1 ~ +2.4, guaiacol: -1.6 ~ -0.4. The selection of the panel is not based on the individual sensitivity of the olfactory sense, but on the average value of the whole group.
- (3) The hedonic tone of  $\text{NH}_3$  belongs to the category of unpleasantness, and the degree of disgust gradually increases with the odour concentration. If the odour concentration is below 26  $\text{ou}/\text{m}^3$ , the humans won't be disturbed by the smell of  $\text{NH}_3$  as it is neither pleasant nor unpleasant.
- (4) There is a multiple nonlinear relationship between the hedonic tone  $Y$  and the odour concentration  $X$  according to  $Y = 3.26 - 3.26 X + 0.42 X^2$  with a correlation coefficient of 0.99.

#### Acknowledgments

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## Conversion of the chemical concentration into odour concentration: evaluation of the key parameters

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Continuous odour measurements are seldom realised, mainly because of their high costs. They are therefore often substituted by chemical concentration measurements of odorous substances. Then a conversion of the chemical concentrations  $C$  ( $\text{mg m}^{-3}$ ) into odour concentrations  $OC$  ( $\text{ou}_E \text{ m}^{-3}$ ) is necessary. Several methods are in use for the conversion: the chemical concentration  $C$ , the sum of odour activity value SOAV, the sum of odour intensity SOI, the equivalent odour concentration EOC, and the sum of odour activity factor SOAF. These conversion methods are evaluated by comparison with the olfactometric measurements of odorous mixtures. The results indicate that the SOI and EOC methods deliver reliable values for odorous mixtures composed of seven substances, and the accuracy is much better than for the first two methods, since SOI and EOC methods use not only the odour threshold concentration but also the slope of the Weber-Fechner law to include the sensitivity of the odour perception of the individual substances. On the other hand, the SOAF method shows a good precision on the prediction of odour concentrations of real odorous air samples in a waste disposal plant. The reason lies in the fact that the SOAF method includes the interaction effects in the complex odorous air samples.

### 1. Introduction

For environmental odour, it is difficult to realise continuous odour measurements of ambient concentrations and gaseous emissions of odour sources. This is mainly because the olfactometric measurements can only be done discontinuously and usually inside an odourless laboratory, and the costs of the olfactometric measurements are high. Thus, in many cases, odour measurements are substituted by concentration measurements of the odorous compounds.

Using concentration measurements of the odorous compounds instead of olfactometric measurements, a conversion of the chemical concentrations  $C$  ( $\text{mg m}^{-3}$ ) into odour concentrations  $C_{OD}$  ( $\text{ou}_E \text{ m}^{-3}$ ) and odour intensities  $OI$  is necessary. For this conversion, several concepts are in use. The simplest approach is the direct use of the concentration of a single substance (e.g.  $\text{H}_2\text{S}$ ) or a group of substances (e.g. VOC concentrations) as a surrogate of the odour concentration using a regression analysis. The second concept is called “Odour activity value” (OAV) and the “Sum of odour activity values” (SOAV). It is based on the normalisation of the concentration  $C$  by the odour threshold concentration  $C_{OT}$  ( $\text{mg m}^{-3}$ ). If more than one substance is involved, then the sum of the individual OAVs is used (SOAV).

For a more sophisticated conversion of the concentrations of odorous substances into odour concentrations, we use not only the odour threshold concentration but also the slope of the relationship between odour concentration and odour intensity. Using these parameters, two conversion methods are possible: the “Sum of the odour intensity” (SOI) and the concept of the “Equivalent odour concentration” (EOC) by Wu et al. (2016).

On the other hand, considering the potential odour interaction effects among the odour compounds in complex odorous mixtures, the “Sum of odour activity factor” (SOAF)-method was developed to assess the binary odour interaction effects. An odour activity value coefficient is proposed to evaluate the type and the level of binary interaction effects based on the measurement of OAV variation in a research on odorous gases in a waste disposal plant.

For the above five methods of converting the chemical concentrations of single substances into the odour concentrations and odour intensities of odorous mixtures, several key parameters are involved. Generally, the key parameters include the chemical concentration, the odour threshold and the sensitivity of the odour perception of the individual substances, as well as the odour interaction effects in the odorous mixtures. The topic of this paper is to compare these conversion methods as well as to explore the key parameters.

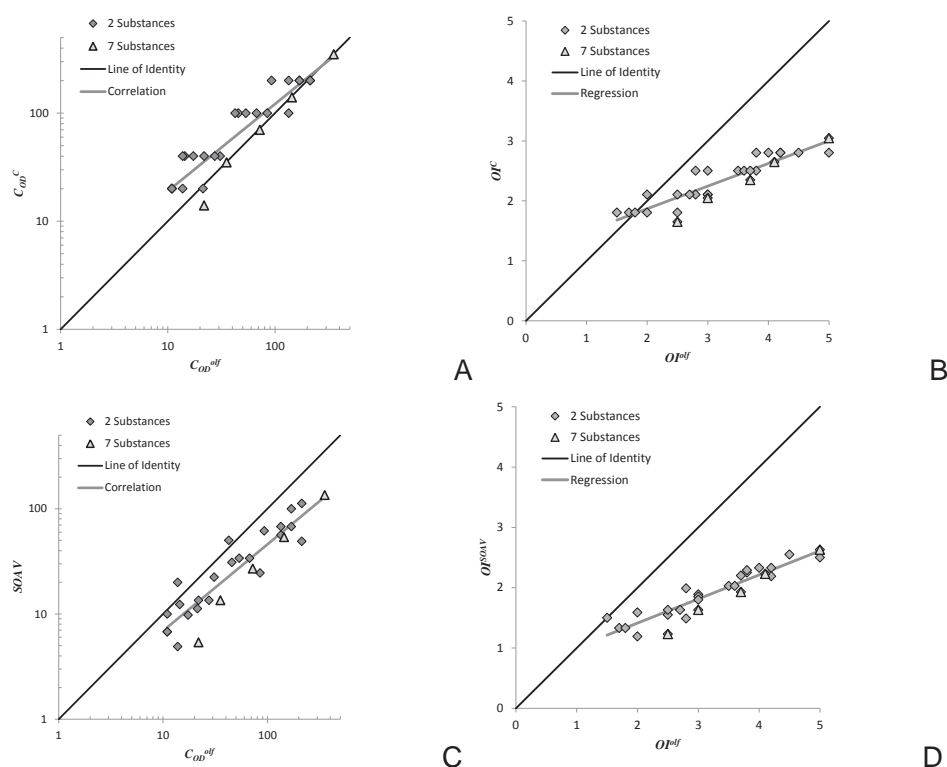


Figure 1: Comparison of the converted odour concentrations  $C_{OD}^C$  (A) and SOAV (C) with  $C_{OD}^{olf}$  ( $ou_E m^{-3}$ ) and the converted odour intensities  $OI^C$  (B) and  $OI^{SOAV}$  (D) with the  $OI^{olf}$  for the 23 binary mixtures and the 5 mixtures of all seven substances.



## 2. Material and methods

The conversion of the chemical concentration of single substances to the odour concentrations and odour intensities of an odorous mixture using the five methods is the central topic of this paper. The ability of the first four conversion methods (C, SOAV, SOI, and EOC) to produce reliable odour concentrations is investigated here by comparing them with olfactometric odour concentration measurements; also the odour intensities will be compared. The experiments were conducted on 24 binary odorous mixtures and 5 odorous mixtures of seven substances which comprise seven typical odorous compounds: Butyl acetate, Benzene, Ethyl acetate, Toluene, m-Xylene, o-Xylene and  $\alpha$ -Pinene. The four conversion methods were described in detail by Wu et al. (2016).

The ability of the last conversion method (SOAF) to produce reliable odour concentrations is investigated by comparing its conversion values with the olfactometric odour concentration measurements and the conversion results of the SOAV method. The experiments were conducted on 16 odorous air samples which were collected at four sampling sites in a waste disposal plant during a year. These measurements and conversion details of the SOAF method were described in detail by Wu et al. (2015).

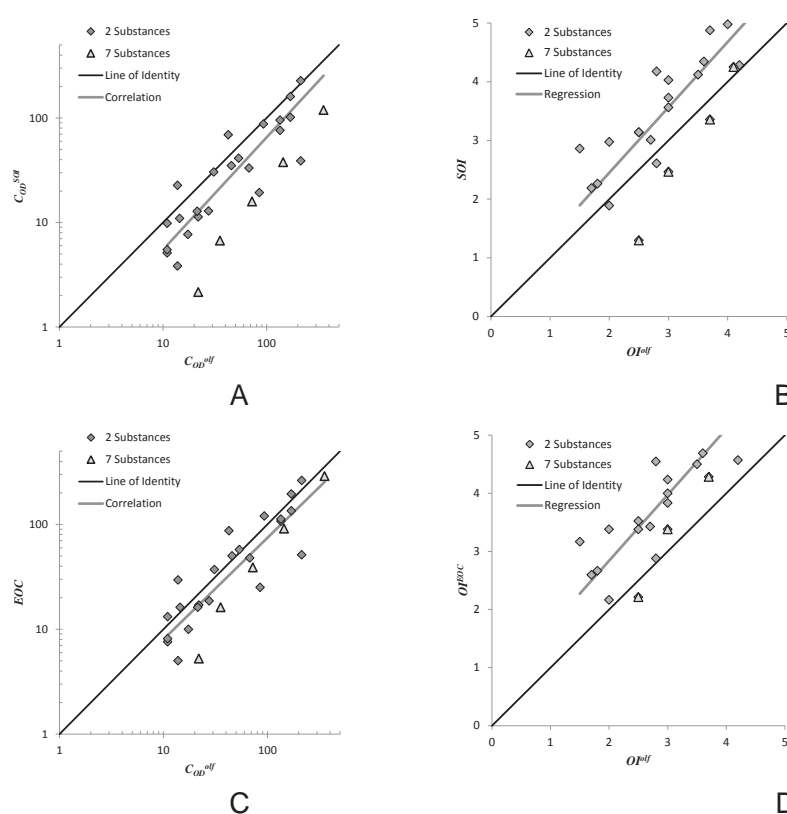


Figure 2: Comparison of the converted odour concentrations  $C_{OD}^{SOI}$  (A) and  $EOC$  (B) with  $C_{OD}^{olf}$  ( $ou_E m^{-3}$ ) and the converted odour intensities  $SOI$  (C) and  $OI^{EOC}$  (D) with the  $OI^{olf}$  for the 23 binary mixtures and the 5 mixtures of all seven substances.

## 3. Results and discussion

The converted odour concentrations and odour intensities from the first two methods (C method and SOAV method) show the weakest quality (Fig. 1). These conversion

methods will not provide odour intensities which are close to those measured by the olfactometer. Instead, the odour intensities are severely under-estimated.

The odour concentration, calculated by the SOI (Fig. 2A), shows a good correspondence with the line of identity with a slope of 0.9471. The converted odour concentration underestimates the measured odour concentration by about 37%. This under-estimation is even more pronounced for the mixtures of the seven substances. The regression line for the odour intensity shows a good agreement with the line of identity (Fig. 2B). The slope of the linear regression is 1.12 which results in an overestimation of about 0.5 grades for a high odour intensity of grade 5.

The equivalent odour concentration  $EOC$  shows a slope of 0.9688 which is close to the line of identity with a weak underestimation of about 13% (Figure C). The regression line of the resulting odour intensities  $OF^{EOC}$  lies parallel to the line of identity with a slope of 1.14 and an overestimation of about 0.6 grades of the 5 grade intensity scale (Fig. 2D).

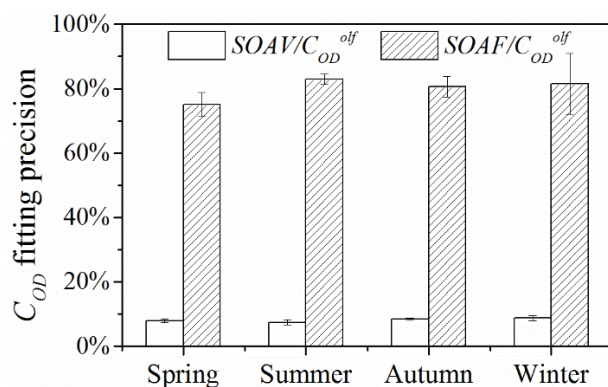


Figure 3: Comparison of the converted odour concentrations SOAF and SOAV with  $C_{OD}^{olf}$  for the gaseous samples collected in a waste disposal plant.

The last two conversion methods, SOI and EOC, yield the best results. The regression lines for the odour intensity show a good agreement with the line of identity. It could be shown that conversion methods which use not only the odour threshold concentration but also the slope of the Weber Fechner law to include the sensitivity of the odour perception of the individual substances deliver more reliable values.

On the other hand, Fig. 3 reflects the comparison of mean SOAV, SOAF and  $C_{OD}$  of odour samples collected at four sites in each season. On average, SOAF matches  $80.0\% \pm 5.7\%$  of  $C_{OD}$ . This is 10 times higher than SOAV, which means that the SOAF method could be a promising method to convert chemical concentrations of compounds in complex gaseous mixtures into odour concentrations. The reason is that odour interaction effects among the compounds are taken into consideration.

#### 4. Conclusion

The first four concepts (C, SOAV, SOI and EOC) are based on the working hypothesis that the mixture of odorous substances behaves additively, which is only a rough estimate. This means that no odour interaction effects between the substances is taken into consideration. Then the calculated odour concentration  $C_{OD}^{theo}$ , calculated by the SOAV, the SOI or the EOC methods, and the odour concentration measured by an olfactometer  $C_{OD}^{olf}$  lie on the line of identity, namely  $C_{OD}^{olf} = C_{OD}^{theo}$ . The SOI and EOC

conversion methods deliver reliable values, since these methods use not only the odour threshold concentration but also the slope of the Weber Fechner law to include the sensitivity of the odour perception of the individual substances. For complex odorous mixtures such as gaseous emissions in a waste disposal plant, the interaction effects between various compounds can be complicated. The odour activity value coefficient was determined to evaluate the type and the level of binary interaction effects based on the measurement of the OAV variation.

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## Odour assessment methods: appropriate uses to obtain the most accurate results

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This paper presents a brief summary of the different methods for assessing odours and discusses their advantages and disadvantages. Odour assessment methods are divided into two categories: assessments at potential sources and ambient assessments at affected areas, which are usually residential areas located close to potential odour sources. Despite there being several available methodologies, the emphasis is on the following methods: source odour testing with dynamic olfactometry evaluations and dispersion modelling analysis to predict off-site odour concentrations; ambient sampling with dynamic olfactometry analysis, monitoring of ambient odour concentrations using the Nasal Ranger and the Scentroid SM100. In addition to the brief summaries of each available methodology, seven case studies are also introduced to compare their results when different techniques are used.

### 1. Introduction

What is the purpose of odour assessments and why are odour assessments performed? For many facilities, odour can be problematic and may cause complaints from adjacent residential areas. When complaints occur, an investigation may be triggered in the same area, which may eventually lead to an odour assessment which could verify an odour complaint. Odour complaint data can be a very good indicator for odour discharges in the area, but on the other hand, complaint records may not necessarily demonstrate the full adverse effects due to several factors. These factors may include: residents not realizing they may be able to complain, residents tired of complaining and ceasing to do so, and persons not being at their homes when an odour episode occurs. However, odour complaints are very important and useful, especially when they end up being substantiated and are validated.

Odour assessments may also be performed as proactive measures to determine current odour emissions or rank potential odour sources in a facility or area to determine the short or long term odour exposure in residential areas. In addition, when a facility is planning to expand, odour assessments can be performed to estimate expected impacts on the surrounding areas. They can also be performed to determine compliance with odour legislation, to comply with conditions outlined in operating permits and in environmental compliance certificates. It is also important that when a new facility is planned, that background odour be established beforehand with these assessments.

Odour assessments are associated to different types of operations, which may include: any type of industrial operations, wastewater treatment plants, refineries, automotive facilities or landfill/compost operations, as well as any agriculture operations.

Different assessments may be required and will depend on several factors, such as, the type of operation performed, the specific country and the purpose of the

assessment. Most importantly, the type of assessment can depend on the country, province or state's odour regulations specific to those regions.

There is no standard method for odour assessment but odour assessment can include or be a combination of several approaches. These include:

- Source odour and/or specific compound testing with analysis and dispersion modelling to predict off-site odour concentrations. This is the most common method for assessing odours at the source. This method is introduced later in this paper in greater detail with some case study examples.
- Ambient sampling with odour panel evaluations. This is a very common approach used in Ontario, Canada to assess odours in residential and complaint areas. It is an inexpensive method which will be introduced in greater detail with case study examples.
- Odour monitoring using portable instruments such as the Nasal Ranger or Scentroid SM100. This is a very inexpensive method for measurement and can be used as a screen tool. Results are based on one - person measurement. Both instruments tend to underestimate or overestimate real odour concentrations due to several factors such as: very short measurement time (peak, or no peak), sensitivity of the person involved in monitoring. In the United States (USA) however, the Nasal Ranger is commonly used by inspectors for regulatory purposes. This method is introduced in this paper in detail with some case studies listed.
- Ambient monitoring using grit or plume method. In this method, observations are made by a panellist who performs odour observations for odour intensity following grit or plume methods. The grit method is commonly used in Europe. However, it is a very expensive method and requires a large data base and therefore months of observations. Most data are based on a one-person observation and depend on his or her sensitivity. The data can be very easily different if different persons with different sensitivities towards odour perform this type of assessment
- Continuous odour monitoring using electronic noses- a very expensive method and difficult to use. They are not applicable for all types of sources. Also sensors need to be changed frequently. In addition, a calibration is required before their use.
- Continuous specific compound or a group of compounds monitoring – this can be applied to source monitoring or ambient monitoring. Continuous monitors typically measure concentrations for individual odorants or group of compounds every few seconds and record the data as a one-minute average value. Examples: monitoring systems for hydrogen sulphide and reduced sulphur compounds. Some of these systems are easy to operate but most of them are complex and installation and operation require extensive technical expertise. There are also some limitations such as the detection limits for the instrument - some odours may be detected by a human nose but not detected by an instrument. There are also possibilities that problems may occur with interferences and a false response of the instrument.
- Specific compound sampling followed by specific analysis. For different measured compounds a different collection media is used. Sampling is inexpensive but the analytical cost may be expensive depending on the compound. Limited to detection limits of the analytical instrument used. Some

- odours may be detected by the human nose but are below the analytical detection level.
- Community odour surveys: usually performed by screened and trained independent observers downwind from the potential source, namely the facility in question. The number of observers will depend on how large the facility is, the budget for the project, terrain and other factors. Community odour surveys can be an effective alternative or supplement to source testing for odour, particularly in cases where there are a number of potential odour sources that can affect a community, where sources are difficult to sample, or when sources are expected to vary with meteorological conditions. Usually observations are made periodically over an extended time frame. A large data set is required to determine the odour levels at specific locations covering a range of weather conditions. They require a long period of observations; therefore, they can be very expensive. There are some limitations to community odour surveys, such as odour adaption and fatigue. If community odour surveys are performed by community members or from a facility, it may lead to adaptation to particular odours, and would therefore tend to underreport odour occurrence. To avoid this limitation, independent outside observers should be used. In most cases, due to the unavailability or for safety reasons, observations are not performed during early morning or late night hours when odours could be at their worst. Therefore, overall, if performed outside these hours, they may underestimate the actual situation in the affected area.
  - Resident observations and questionnaires for residents. A simple logging of odour observations by an individual such as the intensity of the odour, character, duration, and pleasantness. Date and time of the odour episode is usually also recorded, as well as environmental conditions at the time of the odour episode. This method is not expensive and is easy to use; just requires a short training and screening for residents. It may be helpful for initiating an investigation of potential odour or potential odour sources in the area. Limitations: based on the sensitivity of one individual.

## **2. Selected methods for odour assessment**

In this section, we introduce in greater detail some of the methods for odour assessments such as odour testing at the sources with olfactometry analysis, ambient sampling with dynamic olfactometry analysis and the use of two available on the market field olfactometers: the Nasal Ranger and Scentroid SM100.

### **2.1 Odour Testing at the Sources, Dynamic Olfactometry Evaluation and Dispersion Modelling Analysis**

The general approach for odour testing at the source is to first select the potential source at the facility, then collect odour samples, perform odour analysis using dynamic olfactometry and then use dispersion modelling to predict off-site odour concentrations at sensitive receptors. These predicted-by-model odour concentrations could be verified by actual ambient odour sampling at sensitive receptors on the days of the odour testing at the sources. Several factors should be considered before or during assessments and these include:

- Careful selection of all potential odour sources in the plant including point, area and fugitive sources. All sources should be included in the assessment,



otherwise the predicted-by-model odour concentrations may be underestimated when compared to the actual measured ambient odour concentrations.

- Determination of any odour background before any odour testing commences – it is important to determine any other potential sources in the area which may contribute to the off- site odour from the ‘facility in question’- otherwise the facility may be blamed for someone else’s odour releases.
- Methodology used for the collection of samples from sources. It is especially important for point sources which are humid or/and at high temperature, and also for any area or fugitive sources
- Number of collected samples which should be representative to the actual process of the assessed facility.
- Determine if the process is continuous or batch. In case of a batch process, adequate sampling time should be established.
- Odour analysis techniques.
- Dispersion modelling analysis - type of model used for the estimation of off-site odour concentrations.

There are different methods for odour sampling and they vary by jurisdiction. In Europe, most countries use the methods outlined by VDI 3880 (2011) and/or European Standard EN 13725 (2003). The EN13725 standard is now under revision but was designed more for odour analysis, not sampling. Sampling in Australia and New Zealand follows methods outlined by the Standards Association of Australia (2001) “Stationary Source Emissions. Part 3: Determination of Odour Concentration by Dynamic Olfactometry”. Compliance sampling for odour in Ontario, Canada follows the Ministry of Environment and Climate Change (MOECC) Ontario Source Testing Code, Method ON-6” Determination of Odour Emissions from Stationary Sources”<sup>4</sup>).

Different sampling techniques will apply to different types of odour sources. Types of sources:

- Point Source - a single, identifiable source of air pollutant or odorant emissions. Point sources are characterized as being either elevated or at ground-level. Point sources have a defined exhaust diameter. Examples include stacks and vents.
- Area Sources - two-dimensional source of diffuse air pollutant emissions. The dimensions of these sources are either known or can be estimated. Examples include tanks, such as preliminary, aeration tanks at wastewater treatment plants, tailing ponds. See subcategories of the area sources in the text below.
- Fugitive Sources - any open doors, windows, and trucks waiting to unload or load odorous material.

At a point source during testing, odour samples are usually dynamically diluted with nitrogen. For this purpose, a dynamic dilution sampler is usually used to collect samples. After collection of samples at the source, the samples are evaluated for odour detection threshold values (ODTV), which together with volumetric flow rates measured at the source, result in the determination of odour emission rates from the source. For some point sources where expected odour is low, a lung sampling method may be used for collection of the samples. Lung samplers contain a pump which creates a vacuum inside a sealed container (a vacuum chamber) which draws a source sample into the sample bag.

The sampling time depends on the jurisdiction and the nature of the emission source. For example, in Europe a common sample period is 30 minutes for continuous



sources. In Ontario, Canada, it is 10 minutes, while in other jurisdictions, a grab sample is common. The sampling time may be adjusted for a batch process. The sampling equipment should be odourless. Storage times between sample collection and evaluation by an odour panel should also be minimized. Criteria for the maximum allowed storage time are outlined in some standard test methods and generally range from six hours (New Jersey) to 30 hours (most European countries, Australia and New Zealand). However, it is expected that some changes for the time will be made with revision of the European EN13725 standard.

When it comes to the estimation of odour emission rates from area or fugitive sources it becomes more complicated. There are some challenges mostly attributed to the difficulty in accurately measuring emissions from these potential odour sources. Area sources are categorized as sources that are open tanks, such as primary or aeration tanks. Within area sources, three different subcategories are determined:

- Active surface sources, *i.e.*, sources that have a noticeable air flow (aeration tanks)
- Passive surface sources: *i.e.*, sources without outward air flow (primary tanks)
- Partially active sources and partially passive sources. These can include complex tanks where the aeration occurs only a few hours a day, which is when the complex tank becomes an active surface source.

Fugitive sources, on the other hand, can be categorized as a truck loading or unloading area, open doors, windows, leaks. Both area and fugitive odour sources are difficult to reliably measure. Therefore, a careful selection of the method is important for a proper assessment.

There are five methods commonly used to predict odour emissions from area sources:

- (1) Flux Chamber Method - where nitrogen is used as a sweep gas and a sample is collected at the outlet of the chamber. Usually three samples are collected into a container, which is in most cases a Tedlar bag. These samples are analysed by dynamic olfactometry with a screened panellist to determine the odour concentration. The nitrogen flow rate is used together with the odour concentration to determine the odour emission rate. This method is very frequently used, but the sampling method does not represent the actual conditions on site and therefore tends to underestimate the emission rate.
- (2) Portable Wind Tunnel Method – where a portable wind tunnel is used as a replacement of the flux chamber. In this method, the odour samples are taken under different flow rates simulating different wind speeds, which affect odour transfer from a liquid to the gas phase. The odour emission rates are then used for different wind speeds. This approach is much costlier than the flux chamber method due to the instrumentation and set-up procedures, but is more representative of the actual emission rate.
- (3) A Back Calculation with an Air Dispersion Model Method - For this method, the ambient odour concentrations are measured at several downwind locations, which are later used in conjunction with a dispersion model to back-calculate odour emission rates. However, that approach requires the collection of a large number of samples at different downwind locations from the area source. Also for this approach, each area source should be separated. In some cases, it is not possible to separate area sources for the specific wind directions.
- (4) Static Hood Method - This method is commonly used for active sources such as biofilters and aeration tanks. In this method, a static hood is placed over the

emitting surface. The hood isolates a part of the emitting surface and therefore channels the flow into the hood outlet which is in the shape of a stack. Samples are collected at the port installed on the stack

- (5) Mass Transfer Method – Recently, a new method for estimation of the emission rates from the water surfaces was developed which is based on the principle of mass transfer from liquid to gas phase.

For fugitive sources, a back-up calculation method is commonly used for estimation of emissions from these sources.

When estimating fugitive emissions, the following steps are usually required:

Step 1 - Collection of ambient samples within the cavity of the building or structure attached to the fugitive source. Usually more than one sampling location is chosen within the cavity region and at the near wake region. The cavity dimensions are calculated before sampling.

Step 2 - Evaluation of the collected samples using dynamic olfactometry.

Step 3 - Calculations of the cavity concentration. The model is used to calculate the concentration within the cavity. In order to calculate the cavity concentration, the fugitive emission has to be modelled as a point source with certain parameters.

Step 4 - Calculation of the dilution factor

Finally, the odour emission rates are calculated based on the formula

$$\text{Odour Emission Rate (ou/s)} = \text{Dispersion Factor (m}^3\text{/s)} \times \text{Ambient Odour Concentration (ou/m}^3\text{)}$$

In order to predict off-site odour concentrations at selected sensitive receptors, the measured or estimated emission rates are used in the dispersion modelling analysis. Odour models can be classified according to their working principles. The following are categories of models: Gaussian plume models, Gaussian puff models, Lagrangian particle models, Computational Fluid Dynamics models (CFD), In Eulerian models. In Ontario, Canada only two models are approved for regulatory purposes: AERMOD and CAPUFF. In order to run dispersion models for odour assessments, several inputs are necessary such as: emission and source parameters, including nearby buildings, meteorological data, terrain data, and land use characteristics.

Advantages and disadvantages of the source odour sampling method:

- The most common reasonably not expensive method for assessing odour at the sources. If specific compounds are measured at each source, it may be expensive. It may be expensive when CALPUFF model is run.
- Possible ranking of the odour sources which may help in developing methodology for controlling individual odour sources which may contribute the most, therefore in some cases it may be a money savings for the companies struggling with odour issues.
- Easy to monitor emissions for any changes from year to year, which may be beneficial for any changes in the process, expansions.

However, experience in sampling and in designing the source sampling program is essential. The method requires an experienced professional for selecting all potential sources of odour and a careful selection of the methodology for assessing emissions from these sources, especially area or fugitive sources or hot, humid point sources.

The method provides full true odour emissions at each selected source and off site odour impacts when all potential odour sources are included and the right methodology

is chosen. Choosing an appropriate procedure for each source is essential, otherwise a significant underestimation of odour emissions may result.

## 2.2 Case Study 1- Point Sources: Different Methods Used for Collection of the Samples from Hot, Humid Sources as well as Inlet to the Biofilter

For each study, at each location or source three odour samples were collected using a dynamic dilution technique and at the same time three samples were also collected using Lung sampler, meaning samples were not diluted with nitrogen on site during the testing.

The results for odour detection threshold values obtained when the two methods were used (dilution versus no dilution) for the two separate sources are presented in Table 1. The results are presented for hot, humid sources as well as two locations of the Biofilter Inlet.

The odour detection values were determined using dynamic olfactometry and screened panellists according to the European standard EN 13725:2003 and MOECC ON-6 Method.

*Table 1: Summary of the Results.*

Sampling Description/Location	Odour Detection Threshold Values (OU)		Factor
	Dynamic Dilution Method	Lung Sampler Method	
Hot Source-Location 1	34,294	2,075	16
Hot Source-Location 2	1,588	124	13
Humid Source- Location 1	9600	1100	9
Humid Source-Location 2	7200	850	8
Biofilter Inlet- Location 1	13272	3090	4
Biofilter Inlet- Location 2	15400	2435	6

Based on test results, the odour losses at the hot/humid sources were significant, and were recorded by the lung sampler method as thirteen to sixteen times lower for hot sources for the undiluted samples and between 8 to 9 times lower for humid sources for undiluted samples. Also for samples collected at the inlet of the biofilter, the odour losses were significant and were 4 to 6 times lower for undiluted samples with the lung sampler method.

**Conclusion from Case Study 1:** The lung sampling method for collection of samples especially from hot or humid sources underestimates the odour emissions.

## 2.3 Case Study 2- Area Sources - Different Methods Used for Estimation of Odour Emissions from Area Sources

Three different methods were used for estimating odour emissions from area sources located at one of the wastewater treatment plants in Canada. These estimated emissions were used in dispersion modelling to predict off-site odour concentrations at sensitive receptors. The emission rates were established for two different process conditions. These predicted-by-model odour concentrations were compared to actual measured ambient concentrations at three sensitive receptors.

**Method 1 - Flux Chamber Method.** At each area source, two sampling locations were chosen for the flux chamber sampling. At each sampling location, three odour samples were collected for odour panel evaluations. Odour emission rates were calculated

based on the nitrogen flow rate and geometric mean of odour concentrations from all collected samples.

**Method 2** - Wind Tunnel Method. At the same locations for the area sources, three odour samples were collected for odour panel evaluations. Odour emission rates were calculated based on the air flow rate and the geometric mean of odour concentrations from all collected samples.

**Method 3** - Mass Transfer Method. For this approach, odour samples were collected using the Flux Chamber that acted as a capture hood. It was assumed that the transfer of gases between the water and air was directed by turbulent and molecular transport processes which can be characterized by diffusion coefficients.

*Table 2: Summary of the Results.*

Location/Condition		Measured Ambient Odour Concentration OU	Predicted Ambient Odour Concentration OU Method 1 Flux Chamber	Predicted Ambient Odour Concentration OU Method 2 Wind Tunnel	Predicted Ambient Odour Concentration OU Method 3 Mass Transfer
Sensitive Receptor Condition 1	1-	13	2	12	14
Sensitive Receptor Condition 2	1-	68	7	14	54
Sensitive Receptor Condition 1	2-	10	1	6	14
Sensitive Receptor Condition 2	2-	69	7	43	63
Sensitive Receptor Condition 1	3-	55	5	30	45
Sensitive Receptor Condition 2	3-	138	14	64	73

The AERMOD model was used for prediction of off-site odour concentrations at selected sensitive receptors.

In addition to sample collection at the source, several ambient locations were chosen for ambient odour sampling. Samples were collected for different process conditions. At each location, three samples were collected using the Lung Sampler. All ambient samples were evaluated the same way as actual samples collected at the source for odour concentration.

The table below presents the predicted-by-model off-site odour concentrations using three different methods versus measured ambient odour concentrations.

Based on this study, it was found that when a new method for estimating emissions from the area sources (Mass Transfer Method) was used, the predicted-by-model off-site odour concentrations at three sensitive receptors were within the range of measured concentrations or slightly higher than those predicted by the AERMOD

model. The values were within a factor of two, which is a factor commonly used for this model. When the Flux Chamber Method was used, the predicted-by-model off-site odour concentrations were significantly below the measured ambient concentrations. In addition, the Wind Tunnel Method gave results within the measured ambient levels. Therefore, when the Flux Chamber Method was used for sampling, the odour emissions were significantly lower compared to Mass Transfer Method or Wind Tunnel Method.

**Conclusions from Case Study 2** In conclusion, it is very important to consider the right methodology to be used for the estimation of the odour emission rates from any area or fugitive sources.

## 2.4 Case Study 3 - Fugitive Sources- Back Up Calculation Method to Estimate Fugitive Odour Emissions

The study was performed at a Waste Facility where two main areas were considered as potential odour sources. At each area, the odour emissions were discharged from either the exhaust stacks located on the roof when the exhaust fans were turned on or from the receiving doors when the exhaust fans were turned off and the doors were open. Theoretically, the emission rates from either the exhaust stacks or receiving doors should be the same. For the condition when the exhaust fans were turned on, samples were collected at the stacks (fan on), and a procedure for the collection of samples from point sources was applied. The geometric mean of odour detection threshold values from the three samples collected was multiplied by the measured volumetric flowrate to determine the odour emission rates. This was done separately for the two areas.

The second condition was when the exhaust fans were turned off, therefore it was estimated that the odour emissions came from the receiving doors only. For this scenario, in order to estimate the sampling location from the doors, before even sampling started, a cavity of the region was calculated based on building dimensions. Two sampling locations were chosen within the cavity and three samples were taken concurrently at approximately 1.8 m above the ground. Based on the analysis in the laboratory using a dynamic olfactometer, an ambient odour concentration within the cavity was calculated. Later on the model was used to back-up calculate the odour emission rates from these receiving doors. Table 3 present the results for the study.

Based on this study, the odour emission rates estimated using back-up calculation are very much in line with those estimated using a conventional method for estimating odour from point sources.

**Conclusions from Case Study 3** The methodology used for assessing emissions from fugitive sources was accurate.

*Table 3: Comparison of Odour Emission Rates Estimated Using Two Methods.*

Process	Odour Emission Rate ou/s (m <sup>3</sup> basis)	Ratio Odour Emission Rates Stacks to that Estimated from Doors
Area 1- Stack- Fan On	175060	1.1
Area 1- Fugitive- Doors	158450	
Area 2-Stack- Fan On	949412	0.98
Area 2- Fugitive- Doors	964190	

## 2.5 Case Study 4

This study was based on odour assessments at an Organic Waste Facility in Canada. At this facility, samples were collected at point sources (stacks) as well as at three main fugitive sources which were the open doors at the receiving area, trucks waiting to unload and an opening at a material storage. The odour emissions from stacks were estimated using a conventional extractive method. Odour emissions from fugitive sources were estimated using the Back-Up Method. All collected samples were evaluated using dynamic olfactometry.

In addition to sampling inside the facility, ambient odour samples were collected at the most impacted sensitive receptors. A dispersion model (AERMOD) was used to predict off-site odour concentrations for ambient measurement sensitive receptors.

This facility was assessed during different seasons.

All predicted off-site concentrations and measured off-site were compared during different seasons.

The paired comparison of the modelled and monitored odour concentrations is tabulated in Table 4.

*Table 4: Summary of the Results - Measured Odour Concentrations Versus Predicted.*

Sampling Season	Measured Ambient Odour	Predicted by Model Odour
	Concentration OU	Concentration OU
Season 1	24	22
Season 2	37	31
Season 3	11	18
Season 4	23	29

As shown in Table 4, in two seasons, (Season 1, Season 2) the AERMOD model slightly under-predicted odour concentrations when estimated fugitive odour emission rates were plotted into the model. Overall, the combined analyses are very much in line with the often quoted “factor-of-two” accuracy for AERMOD.

**Conclusions from Case Study 4** Estimation of the fugitive odours from receiving doors, storage area and trucks was very close to the actual odour emissions, therefore the back-up method used for the estimation of fugitive odour sources was very accurate.

## 2.6 Ambient Odour Assessments using Ambient Sampling and Dynamic Olfactometry Analysis

When it comes to ambient odour assessments there are different techniques and they may vary from jurisdiction to jurisdiction. In Ontario, Canada, the most popular method for assessing ambient odour is ambient odour testing and dynamic olfactometry analysis.

Ambient sampling for odour assessment is typically conducted using the lung sampling technique with the sample collection done in ambient air rather than within a source such as a stack.

This method is one of the most common methods to assess ambient odour, provided the odour concentration is sufficient to give panelist responses at the lowest dilution levels of the olfactometer. If the odour concentration is too low for a sample to be



evaluated by olfactometry, then the odour panelists can evaluate the sample directly from the sample bag.

The lung sampling procedures for ambient odour monitoring are similar to sampling at the sources using the lung method with exception that the sampling probe is located about 1.5 m above ground level but specific heights may be selected based on the nature of the monitoring program. Sampling periods depend on the jurisdiction (e.g., 10 minutes in Ontario) but can vary depending on the nature of the upwind source and the meteorology.

## 2.7 Case Study 5 - Measured Odour Emissions and Dispersion Modelling Analysis versus Ambient Sampling and Olfactometry Analysis

In this study, the odour emissions were measured and used in the dispersion modelling analysis to predict off-site odour concentrations. At the same time, a lung sampler was used for ambient assessments of odours at sensitive receptors, downwind from the tested facility. Three samples were collected at each source as well as three samples at the sensitive receptors. In addition, an upwind location was selected and tested for any background odour. The results for ambient odour concentrations are taken as geometric mean from three collected samples at that location.

Table 5 presents the results.

*Table 5: Summary of the Results - Measured Odour Concentrations Versus Predicted by the AERMOD Model.*

Location	Predicted by AERMOD Model Off- Site Odour Concentration OU	Measured Ambient Odour Concentration OU
Sensitive Receptor 1	41	80
Sensitive Receptor 2	46	72
Sensitive Receptor 3	15	18
Sensitive Receptor 4	82	126

Based on these results, the predicted by-model off-site odour concentrations are slightly lower than measured. However, they are in line with the model error of factor 2.

## 2.8 Ambient Odour Monitoring Using Field Olfactometers

A field olfactometer such as the Nasal Ranger or Scentroid SM100 directly determines the odour concentration in the ambient air without the requirement to collect a sample in a container.

The field olfactometer, which is used by one person at a time, draws ambient air into the instrument. The diluted sample is presented to the odour observer via a face mask and the observer indicates whether an odour can be detected at each dilution. The results from the Nasal Ranger are used to calculate the detection to threshold (D/T) which is the number of dilutions needed to make the odour ambient air non detectable.

All field olfactometers (Nasal Ranger and SM100) are based on individual one person readings for one-minute maximums whereas ambient sampling for laboratory olfactometry generally occurs for a longer time. Due to this short time frame, accurate results cannot be guaranteed, therefore it should be considered a screening tool only, because the method is expected to be less accurate than any other method and mainly depends on the sensitivity of the person performing readings at a short time. As one observer operates the instrument, results depend on operator sensitivity. In addition,

the observer likely breathes odorant before using the field olfactometer, increasing the opportunity for odour fatigue.

Limitations of the field olfactometers include:

- Provide peak instantaneous odours, whereas ambient sampling provides odour concentrations averaged (10 minutes, 30 minutes.)
- Readings can easily accommodate needs and can be performed only when odours are detected and at their peak. Whereas the odour sampling time averages from 10 minutes to 30 minutes.
- Use of masks require the use to alter their breathing process and pattern, which may affect results. Typically results tend to be lower.
- Readings are based on one - person sensitivity which can highly vary from person to person.
- Fatigue can occur and quite quickly with the person performing the readings.
- Not capable to perform any readings over a long period of time.
- Data are based on the day and the time of the readings and do not cover all hours or meteorological conditions.

In addition, the Nasal Ranger has attached carbon filters which may not be capable of filtering some odorants sufficiently (Bokowa 2008.) On the other hand, Scentroid SM100 uses separate air tanks which are filtered, but the tanks have only a sufficient volume of clean air which only lasts for 30 minutes at a time, and which may not be sufficient enough to perform ambient readings without any interruption.

## 2.9 Case Study 6 - Odour Concentrations Obtained by Ambient Sampling and Dynamic Olfactometry Analysis versus Nasal Ranger Readings

This study is based on the results obtained by two methods: the Nasal Ranger and ambient sampling with dynamic olfactometry analysis. At each location, Nasal Ranger readings were taken to determine the detection to threshold (D/T). At the same location, three odour samples were taken for odour evaluations. Table 6 presents the results.

As shown in Table 6, the results obtained with using the collection of bag samples with evaluation by dynamic olfactometry and screened panellists are by a factor of two or more higher than the results obtained with the Nasal Ranger instrument.

**Conclusion from Case Study 6** It was expected that the Nasal Ranger measurements would be higher due to the possibility of catching odour peaks over a shorter period of time. However, the Nasal Ranger data are much lower than data obtained by ambient sampling.

*Table 6: Comparison of Ambient Odour Levels Using Ambient Sampling versus Nasal Ranger Readings.*

Location	Odour Concentration using Ambient Sampling OU	Nasal Ranger D/T
Location 1	9	2
Location 2	16	2
Location 3	12	4
Location 4	29	15
Location 5	91	30



### 2.10 Case Study 7 - Odour Concentrations Obtained by Ambient Sampling and Dynamic Olfactometry Analysis versus Scentroid SM100 Monitoring

Several studies were performed involving the SM100 unit. The first study (Bokowa, 2012) compared odour concentrations in the collected bags obtained by evaluation of the sample using dynamic olfactometry and eight panellists to readings obtained by the SM100 with attached port, not a mask, as designed by the manufacturer. All of the studies presented in that paper were studies when the samples were already collected and dynamic olfactometry was applied. In addition, the readings were taken in the bag using the SM100. In real conditions the SM100 is designed to be used on site, not on collected samples, and secondly with attached mask, not a port, as studied in 2012. Therefore, caution should be applied when interpreting studies from 2012. Table 7 below shows some of the results from 2012.

*Table 7: Comparison of Odour Concentrations in Collected Bags Using the Dynamic Olfactometry and SM100 with Attached Port.*

Description	ODTV SM100 (using port) OU	ODTV using Odour Panel OU
Sample 1	94	116
Sample 2	164	108
Sample 3	721	515
Sample 4	164	201
Sample 5	3750	2363
Sample 6	3000	1955
Sample 7	3330	2048

Based on these results, the odour detection threshold values obtained by the SM100 with attached port for samples collected at the sources which were high in odour (above 1900 OU) were much higher than obtained by standard dynamic olfactometry analysis with a variation of results between 24% to 38%. For other samples (lower than 1900) the SM100 readings were in line with dynamic olfactometry results but with the variation that some of the results were higher and some lower than the ODTV obtained by dynamic olfactometry. All studies were performed in the laboratory, not on site and SM100 readings were performed with attached port, not mask. Therefore, all presented data cannot be used to compare these two methodologies for assessing ambient odour.

The second study was performed on-site during the collection of samples. All collected samples for ambient sampling were evaluated by eight panellists using dynamic olfactometry. At the same time SM100 readings were performed on site. One reading was performed using SM100 with attached port and the second reading with attached mask. Table 8 summarizes the results.

As shown in Table 8, the results obtained by using the collection of bag samples with evaluation by dynamic olfactometry and screened panellists are either higher or lower than the results obtained with the SM100 instrument when ports are attached which may indicate that SM100 might catch a peak or not during short readings (1 minute) versus 10-minute sampling time using a Lung sampler. When the mask is attached, the

SM100 results are much lower than obtained when the port was attached. However, the ports cannot be used during ambient monitoring due to the interference with readings. Therefore they can be used only in the laboratory.

*Table 8: Comparison of Odour Concentrations Using Dynamic Olfactometry and SM100 with Attached Mask*

Description	Odour Concentration using Ambient Sampling	Scentroid SM100 Using Ports	Scentroid SM100 Using Mask
	OU	OU	OU
Location 1	450	319	164
Location 2	255	328	94
Location 3	32	55	22
Location 4	44	55	24
Location 5	81	109	62

### 3. Conclusions

There are different methods for assessing odours, however they will depend on several factors. A careful selection of the methodology used should be considered before any assessments are performed. Considerations should be made that involve assessing the location of the jurisdiction, the appropriate standards needed to be met, the amount of time available for assessment, whether it be days, months or years, and the financial budgets allowed for the project. Careful thought should be placed on appropriate methods also relating to the type of sources tested (point, fugitive and area sources) and the difficulties in being able to assess them. Lastly, any investigation should attempt to yield the most accurate results and therefore a variety of assessment methods can and should be used, so that underestimation does not occur.

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## Emission characteristics of odorous compounds during short-term pre-aeration of municipal solid waste prior to landfilling

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An important advantage of using a short-term pre-aeration of municipal solid waste (MSW) with high organic fractions prior to landfilling is that it intensively decomposes a part of organic matter, especially easily biodegradable fractions in waste, and thus regulates the subsequent anaerobic degradation of MSW under landfill conditions, meanwhile conserving the bio-methane potential as much as possible. However, this organic matter degraded during the pre-aeration process is converted by aerobic microorganisms into high concentrations of organic leachate and exhausted gases (e.g.  $\text{NH}_3$  and volatile organic compounds), which are also potential pollution sources to the surrounding environment.

In this study, the emission characteristics of leachate and odorous gases generated during a short-term pre-aeration process of MSW are investigated in a laboratory reactor. It is found that MSW, pre-treated aerobically for 7 days, results in ~28.1% of weight loss and ~30.3% of capacity loss, respectively. Due to the initial hydrolysis, acidification and leaching of organic solids, TOC concentrations in leachate generated during the pre-aeration process increase rapidly at the beginning, and then are maintained at relatively stable levels, averaging from 16,700 to 26,000 mg/L. A certain amount of VFAs, mainly butyric acid (50-75%) and acetic acid (20-50%), is detected in leachate, with the concentration range of 1,000-18,000 mg/L, which can increase the odorous risk from leachate storage and treatment operation.  $\text{NH}_3$  emission in the exhaust gases is highly consistent with the degradation of proteins and temperature evolution, with the maximum release capacity of ~37.6 mg/(kg DM.h) occurring at day 4. Compared to the initial untreated waste, the release strength of halocarbons and aromatic hydrocarbons from the pre-aerated waste is obviously decreased, whereas higher concentrations of organosulfurs, such as ethanethiol and dimethyl disulfide, and alkanes are observed (~10 times higher). This finding suggests that more attention should be paid to control the odorous charge during the subsequent storage, transportation and landfilling operation of the pre-aerated waste.

### 1. Introduction

Landfilling is the most prevalent method of municipal solid waste (MSW) disposal in the world, especially in developing countries. In order to minimize adverse impacts to the surrounding environment and improve the performance of MSW landfilling, multistage landfill modes are of increasing interest in recent landfill practice (Clarke et al., 2015; Cossu et al., 2015; Xu et al., 2014). Among these modes, a short-term pre-aeration of MSW prior to landfilling shows its advantages that allows, on the one hand, an abatement of landfill pollutant emissions and accelerated stabilization of MSW, and, on

the other hand, the conservation of a certain amount of biogas producible under anaerobic operation (Gerassimidou et al., 2013; Ni et al., 2016; Salati et al., 2013; Scaglia et al., 2013). Considering economic, environmental and social benefits, this short-term biological pre-aeration followed by anaerobic landfilling is effective to meet the recent requirements of MSW management in developing countries.

The operation of short-term pre-aeration may be used as a composting-like process, and a part of organic matter in waste, especially easily biodegradable fractions, can intensively be decomposed by aerobic microbes. During this process, it cannot be avoided to generate high concentrations of organic leachate and exhaust gases, such as  $\text{NH}_3$  and volatile organic compounds (VOCs) (Kumar et al., 2011). It is reported that total non-methane organic compounds and total volatile sulfur compounds derived from food waste aerobic composting account for  $\sim 0.379$  mg-C/dry g, and this value is comparatively higher than those emitted from paper, yard waste, and MSW decomposition (Staley et al., 2006; Zhang et al., 2013). MSW generated in developing countries is typically characterized by high moisture and high organic content, in which food waste is dominant, usually accounting for more than 50% by weight (Norbu et al., 2005; Zheng et al., 2014). These special characteristics of MSW can strengthen the pollution or health risk potential to the surrounding environment and people during the pre-aeration process, to some extent.

In this study, the emission characteristics of odorous charge, containing leachate and exhaust gases, generated during a short-term pre-aeration process of MSW, are investigated in a laboratory reactor. Based on the daily monitoring, the results obtained here can contribute to preventive recommendations for pollutant abatement during aerobic pre-treatment to help regulatory bodies.

## **2. Materials and methods**

### **2.1 Substrate and experimental equipment**

Based on the typical MSW compositions in Beijing city (Sun et al., 2014), synthetic MSW is used in the present study to minimize variability and guarantee the comparability between the reactors. A detailed breakdown of the initial waste composition was shown in a previous report (Ni et al., 2016). The initial water content and biodegradable VS content of the synthetic MSW are 55.7% (w/w) and 48.7%TS, respectively. The MSW is manually shredded and well mixed with a uniform size of 20–40 mm.

Two laboratory-scale Plexiglas columns are used as pre-aeration reactors in the present research. Each reactor has an internal diameter of 240 mm and a height of 1,100 mm. To channel air into the waste body, a vertical stainless steel pipe with side perforations is installed at the centre of the reactors. The ventilation rate is regulated by a LZB-12 flowmeter. A thermo-regulated insulation system is designed to cover all the reactor lateral surfaces and to maintain a constant temperature at  $35 \pm 1^\circ\text{C}$ . The waste mass temperature is monitored using three Pt100 temperature sensors installed in the side of each reactor (upper, middle and lower layers, respectively). Additionally, a 10-cm thick gravel layer is placed at the bottom of the reactors for leachate discharge.

### **2.2 Methodology**

Before pre-aeration,  $\sim 16$  kg MSW is filled into each reactor, resulting in a density of  $\sim 0.5$  t/m<sup>3</sup>. While loading, approximately 800 g of sludge compost (5% inoculation rate) is mixed in the waste to enhance the aerobic degradation process. The airflow rate is

fixed at 0.5 L/min/kg waste at room temperature in both reactors, with a frequency of 10 min every 20 min, to maintain a favourable aerobic condition. The pre-aeration time lasts for 7 days, which is considered a reasonable and effective duration to regulate the subsequent performance of landfilling operation (Ni et al., 2016). Additionally, no leachate recirculation is used during the pre-aeration process.

### 2.3 Analytical methods

The physical and chemical characteristics of the substrate before and after aerobic pre-treatment are measured according to the methods reported by Ni et al. (2016). Leachate samples from the pre-aeration process are tested for the following analytical parameters daily, according to APHA methods (2005): pH, TOC, TN,  $\text{NH}_4^+\text{-N}$ , and VFA. The  $\text{CO}_2$  content emitted on this stage is measured by a GC-2014 gas chromatographer equipped with a thermal conductivity detector. Exhaust air is bubbled through a 250 ml 2 N  $\text{H}_3\text{BO}_4$  solution to capture  $\text{NH}_3$ , and this solution is tested every 12 h to determine the amount of  $\text{NH}_3$  stripping. All the analysis on solid and liquid samples is conducted in duplicate.

VOCs emitted from the waste mass at the beginning and the end of the pre-aeration process is grabbed using summa canister and analyzed according to the TO-15 method, using a GC–MS system. This method was also used in a previous study to determine the VOC emissions from the actual landfill site in Beijing city (Duan et al., 2014).

## 3. Results and discussion

### 3.1 Evolution of temperature and $\text{CO}_2$ concentration during the pre-aeration process

The temporal evolution of the temperature and  $\text{CO}_2$  content in the free space of the reactors before ventilation is shown in Fig. 1. Pre-aeration of 7 days enables the waste mass to undergo the complete stage of temperature rise ( $29.8 < T < 45^\circ\text{C}$ ) and thermophilic stage ( $T > 45^\circ\text{C}$ ), with the maximum temperature of  $58.5^\circ\text{C}$ . It was reported that the thermophilic temperature is beneficial for the decomposition of easily degradable components in waste, while a mesophilic condition can accelerate the degradation of lignocelluloses (Ni et al., 2016). Based on this reason, it is advisable to interrupt the pre-aeration process before the waste mass temperature entering into the cooling stage to give way to anaerobic treatment, with the purpose of conserving the fraction of lignocelluloses in the waste as much as possible for the subsequent methane production. This is also assumed to support the reason why a pre-aeration of 7 days is used in this study. At the end of the pre-aeration of 7 days, the temperature of the pile mass decreases to less than  $50^\circ\text{C}$ . Liu et al. (2009) reported that the pile temperature, if it remains above  $55^\circ\text{C}$  for 24 h, will result in an almost complete destruction of human and animal pathogens. In this study, the pile average temperature remains above  $55^\circ\text{C}$  around 3 days, it infers that the post-sorted waste is free from pathogens after this short-term pre-aeration.

The  $\text{CO}_2$  concentration can indicate the degradation efficiency of organics by aerobic microorganisms under the given ventilation modes, to some extent. Coinciding with the temperature evolution, the  $\text{CO}_2$  concentration in the free space of the reactors before ventilation gradually increases from 130 mL/L and peaks at 730 mL/L at day 4, then decreases rapidly. At the end of the experiment, the  $\text{CO}_2$  level slightly increases and remains at a high level ( $> 470$  mL/L), possibly implying that mesophilic organisms are

activated again, especially fungi and actinomycetes, which can consume lignocelluloses in waste as carbon source further. Finally, the pre-aeration of 7 days results in ~28.1% of weight loss of substrates and ~30.3% of capacity loss, respectively.

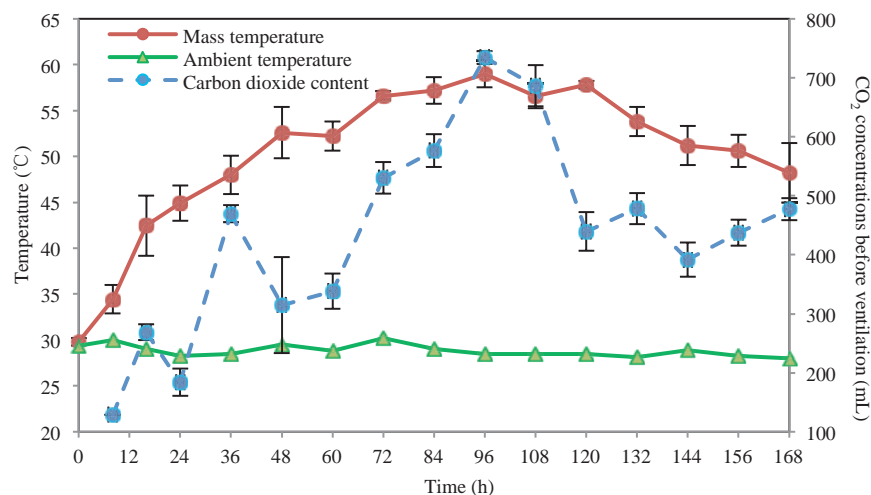


Figure 1: Temporal evolution of temperature and CO<sub>2</sub> concentration before ventilation.

### 3.2 NH<sub>3</sub> and VOC emissions during the pre-aeration process

NH<sub>3</sub> emissions from the exhaust gas are of utmost concern for the odorous nuisance during the pre-aeration of MSW. Coincident with the temperature increase, NH<sub>3</sub> generation from the degradation of proteins is evidently accelerated. It gradually increases from almost 0 mg/(kg DM·h) and peaks at ~37.6 mg/(kg DM·h) at the end of day 4, and then decreases rapidly due to consumption of substrates. This generation rate of NH<sub>3</sub> is also impacted by the pH of the system. In this study, it is observed that the pH values of leachate generated daily gradually increase from 4.7 to 7.5 during the same period. This finding is in agreement with Charles et al. (2007) who also found that the high protease exoenzyme activities during the thermophilic stage and high pH were responsible for the high NH<sub>3</sub> volatilization.

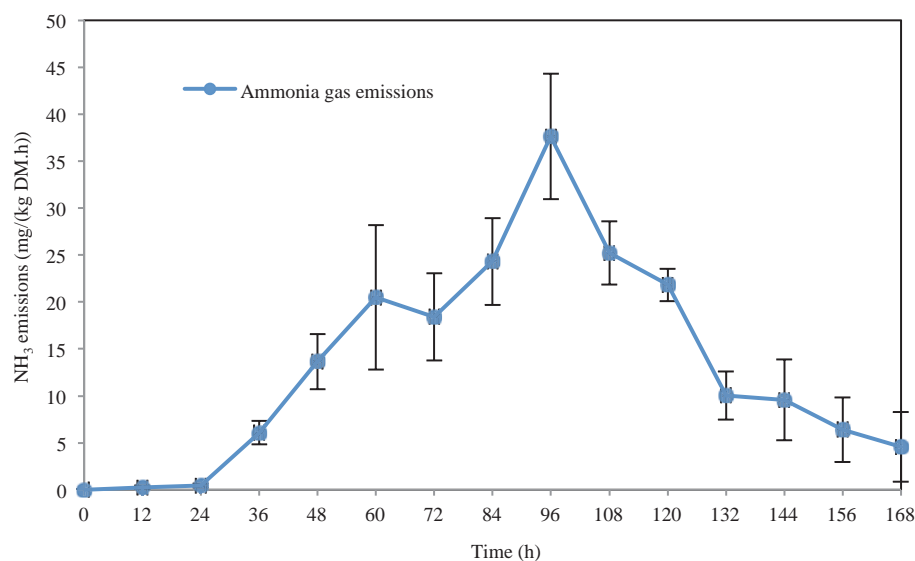


Figure 2: Ammonia emission during short-term pre-aeration of MSW.



During pre-aeration, a mass of VOCs is inevitably generated and emitted to the surrounding environment. Most VOC emissions are generally responsible for odorous nuisance and health risks due to their relatively low olfactory thresholds and potential toxicity. In this study, the VOC emissions detected contain mainly alkanes, halocarbons, aromatic hydrocarbons, organic sulfur compounds and oxygenated compounds (Fig. 3). For the initial untreated waste, ethanol is the largest category of VOC emissions, accounting for 36.1% (w/w) of the total VOCs tested, followed by esters, sulfur compounds, halocarbons, alkanes and aromatic hydrocarbons. To investigate the olfactory nuisance related to the solid waste treatment, organosulfurs are consistently considered to be the main contributors, although aerobic processes are sometimes applied (Ni et al., 2015). Four representative organosulfurs are identified in this study, mainly containing methanliol, dimethyl sulfide, carbon disulfide and dimethyl disulfide, and their concentrations are in the range of 0.003~0.3 mg/m<sup>3</sup>.

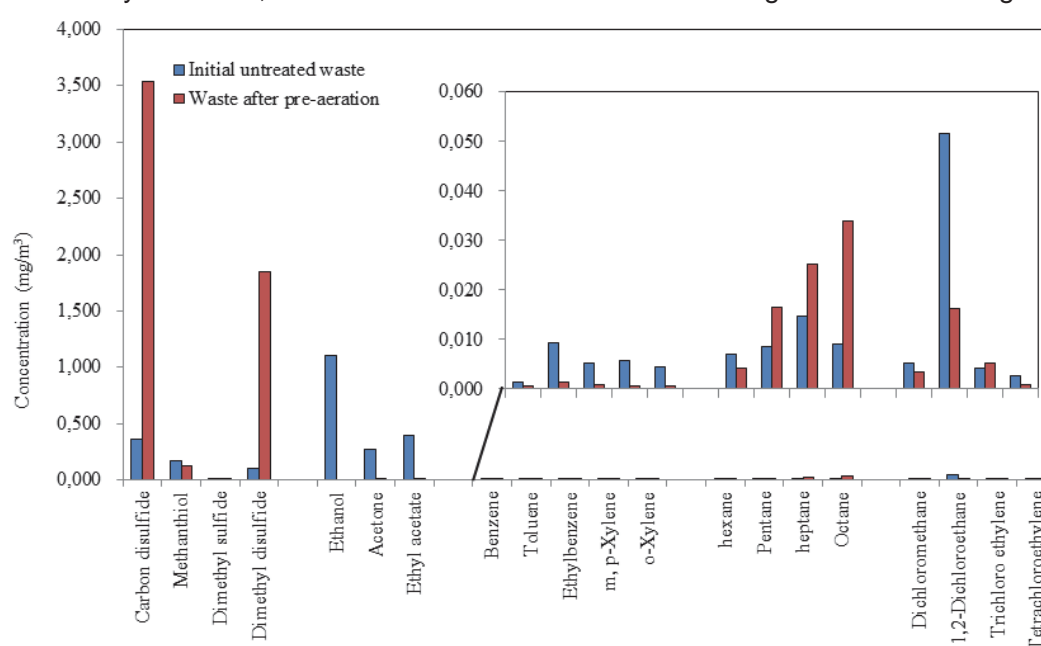


Figure 3: Dominant VOC emissions detected from waste mass before and after pre-aeration.

Compared with the VOCs emitted from the untreated waste, higher emission concentrations of alkanes and organosulfurs are detected from the waste after pre-aeration (~10 times higher), but less halocarbons and aromatic hydrocarbons are detected (Fig. 3). The decomposition of organic sulfur, mainly from the proteins contained in waste, under aerobic conditions, result in mercaptanes (Organic group -SH), and dimethyl sulphide, tending to be generated under conditions of anoxic circumstances, predominates (Fang et al., 2015); meanwhile, alkanes are more likely generated from decomposition of lipids in waste. These two kinds of VOCs are commonly considered as biogenic compounds, which are generated due to biodecomposition during the short-term pre-aeration process. On the contrary, halocarbons and aromatic hydrocarbons are usually considered as abiogenic compounds in waste (Powell et al., 2005) rather than as intermediate products of waste biodegradation, thus pre-aeration is beneficial for eliminating their contents through air stripping. Due to the low odour threshold value of organosulfurs, the above findings suggest that, from the perspective of odour control, more attention should be paid to

the subsequent storage, transportation and landfilling operations of the pre-aerated waste.

### 3.3 NH<sub>3</sub> and VOC emissions during the pre-aeration process

Due to the initial hydrolysis by aerobic organisms and leaching of organic solids, TOC concentrations in leachate generated during the pre-aeration process increase rapidly at the beginning, and then are maintained at relatively stable levels (~20,000-26,000 mg/L) (Fig. 4). A certain amount of VFAs is also detected in leachate generated during the pre-aeration process. Some species of VFAs, such as butyric acid and valeric acid, possess the pungent smell, which is, thus, an important source of odour emission (Fang et al., 2015). In this study, the concentrations of VFAs in leachate increase evidently from the initial value ~1,000 mg/L to 18,000 mg/L at the end of the pre-aeration (Fig. 4). The rapid increase of VFA concentrations, occurring at the end of day 3, may be attributed to the accelerated decomposition of proteins in waste, which is consistent with the increase of NH<sub>3</sub> emissions (Fig. 2); meanwhile, the settlement of pile mass can increase the difficulty of the oxygen transmission. This results inevitably in the locally anaerobic condition of the system, which can enhance the yield of VFAs. Among the VFAs detected, butyric acid (50-75%) is dominant, followed by acetic acid. The presence of these VFAs can aggravate the odorous risks from the leachate storage and treatment units.

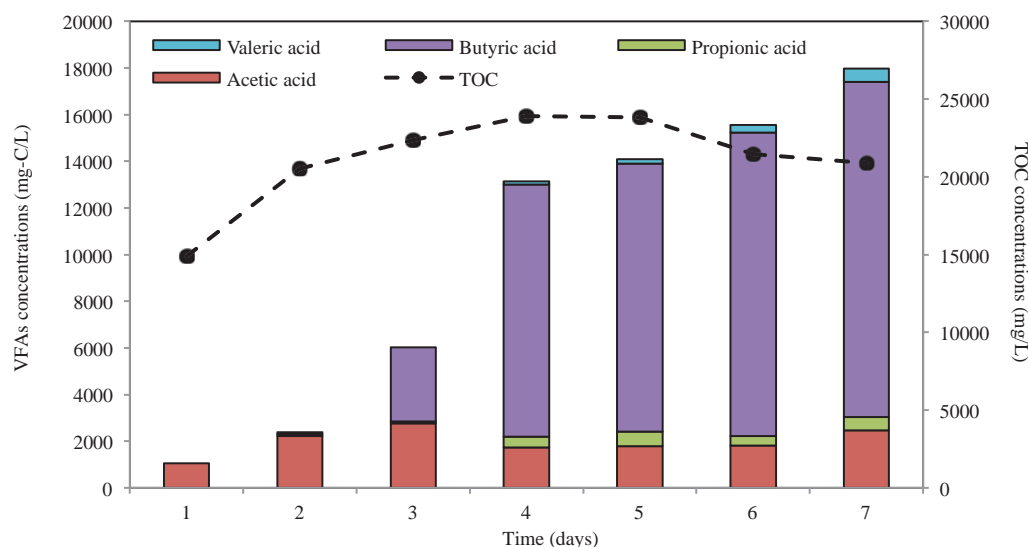


Figure 4: Evolution of VFAs in leachate during the pre-aeration process.

## 4. Conclusions

Short-term pre-aeration is effective in reducing the amount and volume of waste prior to its disposal. However, intensive pre-aeration could generate high concentrations of leachate and odorous gases, which make it a potential source of odorous nuisance. During the pre-aeration process, the NH<sub>3</sub> emission in exhaust gases is highly consistent with the temperature evolution and the degradation of proteins, with the maximum value detected in this study of ~37.6 mg/(kg DM.h). Compared to the initial waste, the release strength of halocarbons and aromatic hydrocarbons from the pre-aerated waste is decreased, whereas higher concentrations of organosulfurs and alkanes are observed, suggesting that more attention should be paid to the subsequent

storage, transportation and landfilling operations of the pre-aerated waste. Additionally, the presence of VFAs in leachate, mainly butyric acid and acetic acid, aggravate the odorous risks from the leachate storage and treatment units.

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## Endogenous mitigation of H<sub>2</sub>S inside of landfills

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Vast quantities of H<sub>2</sub>S emitted from landfill sites require urgent disposal. The current study focuses on source control and examines the migration and conversion behaviour of sulfur compounds in two lab-scale simulated landfills with different operation modes. It aims to explore the possible strategies and mechanisms for H<sub>2</sub>S endogenous mitigation inside of landfills during decomposition. It is found that the strength of H<sub>2</sub>S emissions from the landfill sites depends on the municipal solid waste (MSW) degradation speed and vertical distribution of sulfide. Leachate recirculation can shorten both the H<sub>2</sub>S influence period and pollution risk to the surrounding environment. H<sub>2</sub>S endogenous mitigation may be achieved by chemical oxidation, biological oxidation, adsorption and/or precipitation in different stages. Migration and conversion mainly affects H<sub>2</sub>S release behaviour during the initial stabilization phase in the landfill. Microbial activities related to sulfur, nitrogen and iron can further promote H<sub>2</sub>S endogenous mitigation during the high reduction phase. Thus, H<sub>2</sub>S endogenous mitigation can be effectively enhanced via the control of the aforementioned processes.

### 1. Introduction

Landfills are widely used around the world because of their low cost and easy management (Long et al., 2008; Lei et al., 2007). However, one of the most common public complaints associated with landfills regards odour pollution, originating from the degradation of organics containing sulfur (OCS). Hydrogen sulfide (H<sub>2</sub>S), with an extremely low odour threshold (around 0.5 ppb) and high toxicity (Firer et al., 2008), is the most noticeable OCS and has gained increasing attention in recent years. It has been reported that H<sub>2</sub>S concentrations range from hundreds of micrograms to thousands of milligrams per cubic meter in different MSW landfill sites (Kim et al., 2005; Ding et al., 2012). The emission of H<sub>2</sub>S into the atmosphere reduces the air quality, but also affects public health, causing headaches, pulmonary edemas and neurotoxicity in humans (Olson, 2011; Schiffman et al., 2002).

Currently, H<sub>2</sub>S emission control in landfills mainly focuses on altering the traditional cover materials using filter materials (He et al., 2012; Bergersen and Haarstad, 2014; Xu et al., 2010). For example, Plaza et al. (2007) demonstrated that sandy soil infused with lime and fine concrete had H<sub>2</sub>S removal efficiencies of greater than 99%, while clayey and sandy soils had average removal efficiencies of 65% and 30%, respectively. Sungthong and Reinhart (2011) proved that an autotrophic denitrification landfill biocover is an effective alternative for controlling H<sub>2</sub>S emissions. Moreover, chemical (Xu et al., 2014) and microbial (He et al., 2012) H<sub>2</sub>S control mechanisms have also been investigated in the biocover soil of landfills. However, these techniques are all end control strategies, focusing on the stage of odour being emitted from the landfill.

H<sub>2</sub>S endogenous mitigating behavior inside landfills has not been studied, to our best knowledge.

Landfills are complex artificial habitats, which include various physical and biochemical processes (Zacharof and Butler, 2004). Before H<sub>2</sub>S is formed and released out of the cover layer, OCS in landfills experience complicated migration and conversion processes during decomposition. Moreover, the behaviour of compounds related to the final emission of H<sub>2</sub>S in refuse (e.g., sulfate, sulfide, nitrate and ferrous) differ with different operation modes. Unfortunately, current research related to H<sub>2</sub>S mitigation from the cover layers has failed to explain the endogenous behavior of H<sub>2</sub>S inside the landfill, because biocover is just a H<sub>2</sub>S sink, rather than an original source. Therefore, for controlling H<sub>2</sub>S emissions, it may be more helpful to evaluate the specific migration and conversion behavior of sulfur compounds inside of landfills during decomposition. In this study, we investigate whether H<sub>2</sub>S can be mitigated endogenously within the landfill, via source control. The H<sub>2</sub>S endogenous mitigating behavior inside landfills is examined by tracking the specific migration and conversion behavior of sulfur compounds in two lab-scale simulated landfills with different operation modes. The purpose of the present study is to find a controllable strategy for H<sub>2</sub>S endogenous mitigation during MSW decomposition in landfills.

## 2. Materials and methods

### 2.1 Experimental set-up

Two simulated bioreactor landfill sets, constructed with watertight polypropylene, are established in the study (one is a conventional landfill (CL) with single pass leaching; another is set as a recirculated bioreactor landfill (RL), with recirculated leachate.). The complete configurations of the two landfills are shown in Fig. 1.

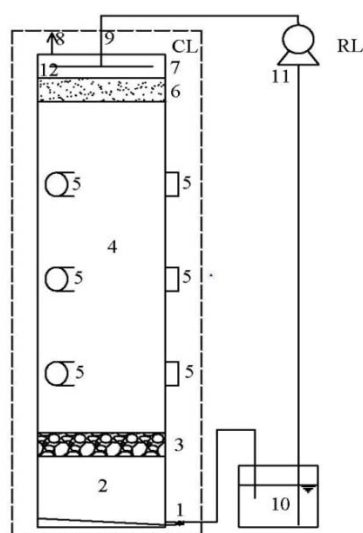


Figure 1: Scheme of the simulated landfill systems (1. Leachate outlet; 2. Leachate collection tank; 3. Gravel layer; 4. Landfill site; 5. Sampling port; 6. Sandy layer; 7. Headspace; 8. Gas outlet; 9. Vent-port; 10. Leachate collection tank; 11. Peristaltic pump; 12. Leachate distribution system).

Each landfill has a diameter of 0.5 m and a height of 2.0 m, with a 100-mm-thick layer of headspace, a 1600-mm-thick layer of landfill site and a 300-mm-thick layer of

leachate collection from top to bottom. Each landfill site is equipped with 12 ports: two inlet/outlet ports in the top lid for gas exporting or leachate recycling (only for the RL); one port at the bottom of the landfill for leachate drainage and sampling; the remaining nine ports on the side wall of the landfills for refuse and gas sampling.

At the bottom of each landfill site, a 100-mm-thick layer of gravel (including the upper layer with the smaller size of gravel (less than 5 cm) and the lower layer with the bigger size of gravel (10~15 cm)) is placed to simulate a leachate collection system, also preventing clogging of the leachate withdrawal outlets. Above this layer, the MSW is loaded in 1450-mm layers and is compacted using a shovel and a sledgehammer. The MSW layer is divided into three layers: shallow, middle and deep, and each layer has three sampling ports, the angle of which between the three sampling ports is 120°. Then a 50-mm-thick layer of sand is placed on top of the MSW to simulate intermediate cover and an upper drainage layer, and to provide even distribution of the recirculated leachate. Headspace on top of each landfill creates a leachate distribution system. Finally, the two landfills are sealed using rubber gaskets and silica gel to ensure an anaerobic environment, and are then operated at room temperature.

## 2.2 Characteristics of MSW

The MSW used in this experiment was collected from the refuse collection station in Hangzhou, Zhejiang, China. Visual inspection of the refuse showed the presence of a variety of food waste, paper, plastic, textile, timber, orthod, glass, metal and mixture. The average wet density of the refuse compacted in the landfills is 880 kg m<sup>-3</sup>. Larger particles of the collected refuse were shredded into approximately 2 cm, and the refuse was thoroughly mixed prior to loading the landfills. The physical composition of MSW used in this experiment is shown in Table 1 (by wet weight, w/w).

*Table 1: Components of experimental MSW sample*

Component	Food waste	Paper	Plastic	Textile	Timber	Orthod	Glass	Metal	Mixture
w/w, %	66.76	9.16	3.17	0.50	1.61	1.75	0.38	0.22	16.45
Moisture content, %	74.56	59.71	43.80	56.61	62.78	21.33	1.54	1.21	59.90

## 2.3 Operation of simulated bioreactor landfills

After loading, the moisture content of the refuse is adjusted to 75% by adding tap water to the bioreactor landfill (Benson et al., 2007). The leachate is collected and stored in a leachate collection tank. The leachate of the CL is discarded without further treatment, while leachate of the RL is continuously recirculated using peristaltic pumps with adjusted flow rates.

## 2.4 Sampling and analytical methods

Refuse is sampled periodically from the refuse sample ports at the side of the landfills, when approximately 100 g refuse samples are collected from each refuse sampling port. In each layer, refuse samples from each of three ports are mixed as a sample (~300 g). Meanwhile, leachate is collected from the leachate outlet ports (~100 mL). To maintain equilibrium of leachate volume in the RL before recirculation, the same volume of tap water (~100 mL) is added back into the leachate after sampling. Gas



samples are also monitored periodically from the gas outlet port in the top lid and the nine ports in the sides of the landfills.

Refuse samples are analyzed for pH, moisture content, dissolved organic carbon (DOC), sulfate ( $\text{SO}_4^{2-}$ ), sulfide ( $\text{H}_2\text{S}$ ,  $\text{HS}^-$ ,  $\text{S}^{2-}$ ), ferrous ( $\text{Fe}^{2+}$ ), nitrate ( $\text{NO}_3^-$ ) and nitrite ( $\text{NO}_2^-$ ) after samples being passed through a  $0.22\ \mu\text{m}$  filter. Distilled water is used as the extraction solution (solid-liquid ratio of 1:10) for the analysis of pH, DOC and  $\text{SO}_4^{2-}$ . The pH is determined using a pH meter (SevenEasy, Mettler-Toledo, Switzerland). The DOC is determined using a TOC analyzer (TOC-V CPN, Shimadzu, Japan). The  $\text{SO}_4^{2-}$  is determined using an ion chromatograph equipped with a Metrosep A Supp 5 column (150 mm long, 4.0 mm i.d.) and a conductivity detector (882 Compact IC plus; Metrohm, Herisau, Switzerland) (Fang et al., 2015). Sulfide concentrations are determined by the method described by Qiu et al. (1992). The ferrous concentrations are determined by the o-phenanthroline method after extraction with  $1\ \text{mol L}^{-1}$  HCl (solid-to-liquid ratio of 1:100) (Li et al., 2010).  $2\ \text{mol L}^{-1}$  KCl solution is used as the extraction solution (solid-liquid ratio of 1:10) for the analysis of  $\text{NO}_3^-$  and  $\text{NO}_2^-$ .  $\text{NO}_3^-$  and  $\text{NO}_2^-$  are determined by ultraviolet spectrophotometry and N-(1-naphthyl)-ethylenediamine dihydrochloride, respectively (APHA, 1999). The moisture content in MSW is determined using standard methods (APHA, 1999). Leachate samples are analyzed for volume (V), volatile fatty acids (VFA), chemical oxygen demand (COD) and sulfide. The VFA are measured using the acidified ethylene glycol colorimetric method (Shen et al., 2004). The COD is measured using a DR2800 spectrophotometer (HACH Company, Loveland, CO, USA). Gas samples are collected for  $\text{H}_2\text{S}$  analysis. The  $\text{H}_2\text{S}$  in the gas samples is analyzed using a gas chromatograph equipped with a flame photometric detector (GC 7890A; Agilent Technologies, Santa Clara, CA, USA) (Fang et al., 2015).

All analyses are carried out in triplicate to ensure the validity of the results, and all the results of the chemical analyses are calculated on a dry-weight basis.

### 3. Results and discussion

#### 3.1 Release behaviour of $\text{H}_2\text{S}$ from the landfill

During the degradation of OCS under anaerobic conditions in landfills,  $\text{H}_2\text{S}$  is produced and released (Mescia et al., 2011). As shown in Fig. 2, both simulated landfills promptly release  $\text{H}_2\text{S}$  after start-up. In the first 135 days,  $\text{H}_2\text{S}$  concentration released out of the CL cover layer remains at a low level, with the highest concentration less than  $10\ \text{mg m}^{-3}$ . However, after this initial period, the  $\text{H}_2\text{S}$  concentration fluctuates greatly, and the maximum  $\text{H}_2\text{S}$  concentration is  $19.4\ \text{mg m}^{-3}$ , which is higher than that specified in the World Health Organization air quality guidelines for  $\text{H}_2\text{S}$  ( $0.15\ \text{mg m}^{-3}$ ) (World Health Organization, 2000). However, the release behaviour of  $\text{H}_2\text{S}$  from the RL completely differs from the CL. During the first 135 days, the  $\text{H}_2\text{S}$  concentration is significantly higher in the RL than in the CL, and the highest  $\text{H}_2\text{S}$  concentration in the RL is  $24.1\ \text{mg m}^{-3}$ . After 135 days, the  $\text{H}_2\text{S}$  concentration in the RL decreases gradually to less than  $5.6\ \text{mg m}^{-3}$  for the remainder of the experiment.

The release of  $\text{H}_2\text{S}$  is mainly induced by the decomposition of organics in landfills. Correspondingly, a clear rising trend in VFA and COD concentrations in the leachate is observed (Fig. 3) after start-up. It indicates that the landfill experiences a fast degradation and acidification process (Long et al., 2010; Li et al., 2011). In this period, a large amount of easily degradable sulfur-containing substrates found in the refuse is hydrolyzed and/or decomposed into inorganic sulfur, coupled with the degradation of

organics in MSW. Then,  $H_2S$  is emitted after physical, chemical and/or biological equilibration in solid and liquid phases. After 135 days, the fast decomposition process promoted by leachate recirculation in the RL attenuates, and the corresponding  $H_2S$  release also declines.

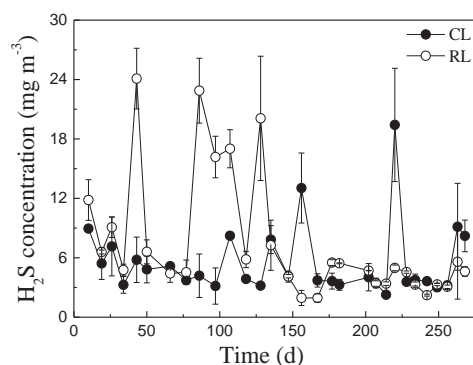


Figure 2: Release behaviour of  $H_2S$  from the cover layer of the two simulated landfill sites.

In contrast, the  $H_2S$  release in the CL gradually experiences the strongest period (e.g., day 220) and then sharply declines. It is mainly ascribed to the slower degradation rate in the CL without leachate recirculation, compared with the RL. This shows that  $H_2S$  emission at landfill sites depends on MSW degradation speed. From the viewpoint of the high concentration emission which means high toxicity, the influence caused by  $H_2S$  from RL is shorter than from CL. In other words, landfills with leachate recirculation can thus shorten the  $H_2S$  influence period and pollution risk to the surrounding environment.

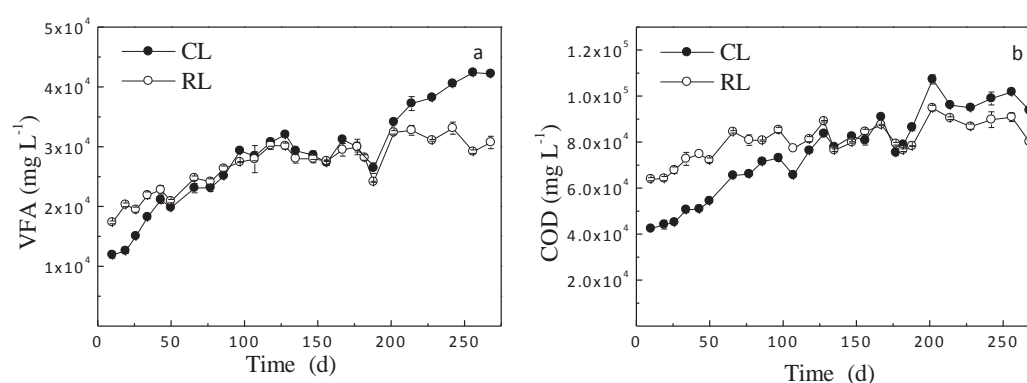


Figure 3: Changes of VFA (a) and COD (b) in leachate from the two simulated landfill sites.

Inside the landfill,  $H_2S$  emissions should experience a long migration process, in which various kinds of physical, chemical and/or biological reactions, equilibriums and transformations occur in the complex solid-liquid-gas phases (Yongsiri et al., 2003). As shown in Fig. 4, the  $H_2S$  distribution differs with depth in the landfill. In the shallow layer, the  $H_2S$  concentration distribution is consistent with its final releasing characteristics from the cover layer. Namely, the  $H_2S$  concentration in the shallow layer is much greater in the RL than in the CL before day 135, but is the opposite in the following period. For example, on day 43, the  $H_2S$  concentration in the shallow layer of

the RL increased to its highest concentration of  $16.4 \text{ mg m}^{-3}$ , which coincided with the highest concentration of  $\text{H}_2\text{S}$  emitted from the landfill. Subsequently, after a sharp fall,  $\text{H}_2\text{S}$  concentration increased again on day 86. However, no such corresponding phenomena can be observed when comparing the finally emitted  $\text{H}_2\text{S}$  concentration with concentrations in the middle and deep layers. This indicates that the final release of  $\text{H}_2\text{S}$  from the landfill is mainly influenced by its distribution in the shallow landfill layer, due to positive vertical migration. Therefore, to effectively reduce  $\text{H}_2\text{S}$  emission in landfills, research should focus on the shallow and cover layers.

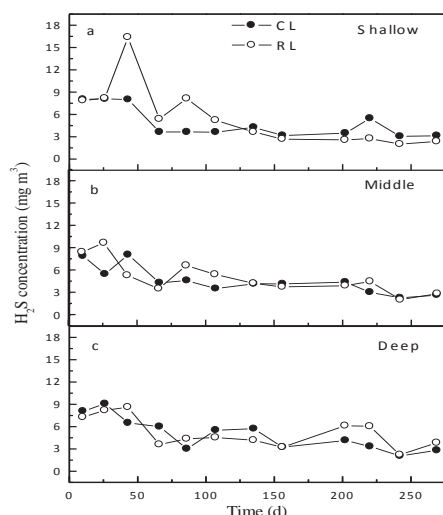


Figure 4: Release behavior of  $\text{H}_2\text{S}$  in the shallow (a), middle (b) and deep (c) layers of the two simulated landfill sites.

### 3.2 Migration and conversion of sulfur compounds in landfills

Sulfur in refuse includes inorganic, such as sulfide, elemental sulfur, sulfate, and thiosulfate, and organic fractions. Among them, sulfide is the reduction product of high-valence sulfur and a potential  $\text{H}_2\text{S}$  source (Yongsiri et al., 2003). As shown in Fig. 5, the sulfide content is generally higher in the CL than in the RL, and differs among layers in both landfills. In the shallow layer, the sulfide content remains at a low level in both simulated landfills, at not more than  $20 \text{ mg kg}^{-1}$ , and no temporal obvious changes are seen. This results suggest that a lot of sulfide in the refuse has been transformed to  $\text{H}_2\text{S}$  and emitted, as mentioned above. In the middle layer, the sulfide content is obviously higher in the CL than in RL. For instance, the highest content observed in the CL was on day 223, at  $75.9 \text{ mg kg}^{-1}$ , which is three times the maximum value in the RL. In the deep layer. Differences between the CL and RL can only be found after day 135, in which CL showed a greater increase than RL, and the highest sulfide content of  $115.2 \text{ mg kg}^{-1}$  was observed in the CL. This result further indicates that the landfill with leachate recirculation has lower odour pollution risk than the one without, because of the differences in the transformation from sulfide to  $\text{H}_2\text{S}$ .

In landfills, anaerobic microbe-like sulfate-reducing bacteria (SRB) can use sulfur compounds (mainly sulfate) as electron acceptors to produce sulfide that is finally released as  $\text{H}_2\text{S}$  (Zhang et al., 2008). As shown in Fig. 6, the CL and RL show similar changing trends in sulfate content. Before deposition in the landfill, the initial sulfate content, mainly an inorganic fraction, in the raw MSW is  $3198 \text{ mg kg}^{-1}$ . With the rapid

degradation of refuse, the sulfate content increases sharply because of hydrolysis. After day 135, sulfate can either be converted into other sulfur forms or assimilated to form OCS, which is accompanied by an increase in sulfide content.

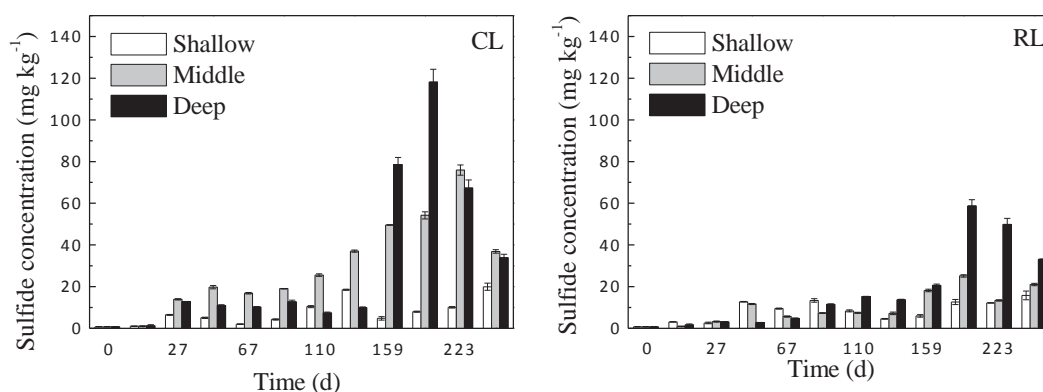


Figure 5: Changes in sulfide content in each layer of the simulated CL and RL.

The above differences in sulfide in both landfills can further be verified by their releasing amounts by leachate (Fig. 7). For example, the accumulated release amount of sulfide is higher from the RL than from the CL after day 135, while the opposite trend is observed for sulfate. The release responses of sulfide and sulfate in leachate correspond to the release behavior of  $H_2S$  and sulfur compounds in refuse. Because of the negative vertical migration of leachate, more sulfides are released into leachate in the RL, leading to less  $H_2S$  emitted from the landfill. Moreover, there are more sulfates reduced to other forms of sulfur or assimilated to form organic sulfur compounds, which will pose a risk of producing  $H_2S$  at a lower level.

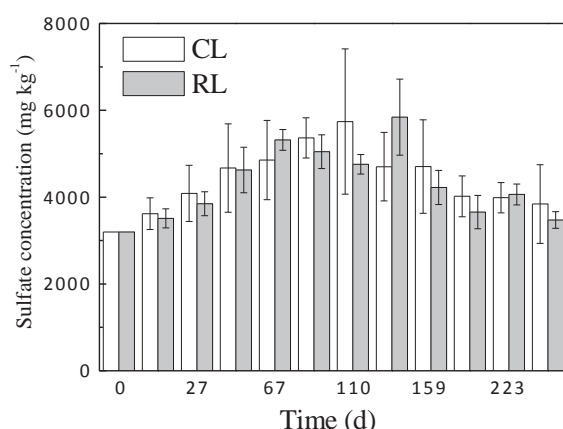


Figure 6: Changes in sulfate content in each layer of the simulated CL and RL.

### 3.3 Possible $H_2S$ endogenous mitigation behavior in landfills

As mentioned above, the release of  $H_2S$  is closely related to sulfide content. The migration and conversion of sulfide in landfills is a complicated process influenced by many factors. Stepwise linear regression analysis shows that the possible  $H_2S$  endogenous mitigation behaviour differs with operation modes (Table 2). With the leachate recirculated in the RL,  $H_2S$  release is weakened because of the higher moisture content and pH of refuse in the RL than in the CL. Additionally, iron and

nitrogen including nitrate and nitrite are also favorable for H<sub>2</sub>S endogenous mitigation inside the landfill.

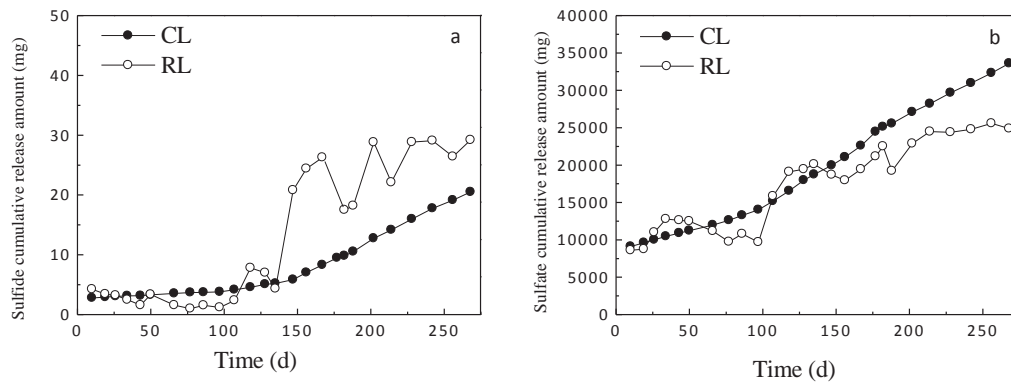


Figure 7: Accumulated amount of sulfide (a) and sulfate (b) leached out from simulated landfills.

Table 2: Stepwise linear regression analysis of sulfide and sulfate, ferrous, nitrate, nitrite, pH, DOC, moisture content in each layer of the simulated landfill (y, x<sub>1</sub>, x<sub>2</sub>, x<sub>3</sub>, x<sub>4</sub>, x<sub>5</sub> represent sulfide concentration, ferrous concentration, nitrite concentration, pH value, moisture content, nitrate concentration, respectively).

		Regression equation	R <sup>2</sup>	F	Sig.
CL	Shallow	y=0.012x <sub>1</sub> -7.726	0.660	19.437	0.001
	Middle	y=0.044x <sub>1</sub> -32.496	0.731	27.129	0.000
	Deep	y=0.089x <sub>1</sub> +0.525x <sub>2</sub> -155.872	0.764	14.57	0.002
RL	Shallow	y=14.914x <sub>3</sub> -85.068x <sub>4</sub> -21.426	0.819	20.306	0.000
	Middle	y=10.850x <sub>3</sub> -0.035x <sub>5</sub> -32.756	0.732	12.284	0.003
	Deep	y=0.026x <sub>1</sub> -0.041x <sub>5</sub> +5.324	0.883	33.946	0.000

A possible H<sub>2</sub>S endogenous mitigation behavior in landfills is described in Fig. 8. H<sub>2</sub>S endogenous mitigation may be achieved by chemical oxidation, biological oxidation, adsorption and/or precipitation inside the landfill via the following stages.

#### (1) Initial stabilization stage

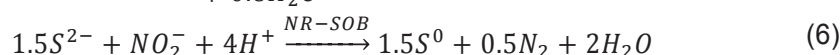
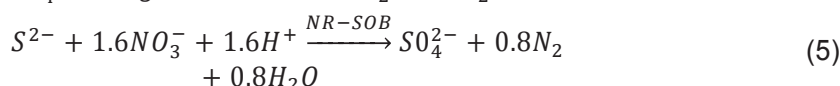
After the landfill start-up (Fig. 8-a), residual oxygen from MSW is still available. The low-concentration background sulfide undergoes a chemical oxidation process (Eq. 1). However, in this period, a large amount of easily degradable OCS in refuse is hydrolyzed and/or decomposed, and the inorganic sulfurs including sulfate and sulfide are released. Then, H<sub>2</sub>S is emitted after physical, chemical and/or biological equilibration in solid and liquid phases. The H<sub>2</sub>S can be absorbed into the liquid film and/or adsorbed by refuse, and then metabolizes by microorganisms present in the refuse. This biodegradation requires continuous adsorption of H<sub>2</sub>S into the liquid film and/or adsorbed by refuse. H<sub>2</sub>S is highly soluble in water, but is affected by the pH level. Between a pH value of 5 and 9, the fraction of H<sub>2</sub>S available decreases as the pH increases because of dissociation into HS<sup>-</sup> (Rumsey and Aneja, 2014) (Eqs. 2 and 3). Thus the lower the pH, the more H<sub>2</sub>S is available to be transferred from the landfills into the atmosphere.





## (2) High reducing phase

When landfills gradually come into a high reducing phase (Fig. 8-b), anaerobic microbe-like SRB can use sulfur compounds (mainly sulfate) as electron acceptors to produce sulfide (Eq. 4). Moreover, microorganism-like nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB) can use nitrate or nitrite to oxidize sulfide under anaerobic conditions (Zhang et al., 2008) (Eqs. 5 and 6). At this stage, the inhibition of  $H_2S$  production seen in the RL may have been caused by these autotrophic sulfur-oxidizing bacteria. Moreover, the presence of nitrate and nitrite may also have inhibited the SRB activities and then attenuated  $H_2S$  production in refuse (Kumaraswamy et al., 2011).



In addition to nitrogen, iron with alterable valence is another common element in landfills. It has also been reported that iron oxides can efficiently remove  $H_2S$  (Bergersen and Haarstad, 2008). In landfill, Fe(II) can remove dissolved sulfide via the formation of ferrous sulfide precipitation, while Fe(III) can oxidize sulfide to elemental sulfur while itself being reduced to Fe(II) (Nielsen et al., 2005) (Eqs. 7 and 8). The increasing Fe(II) content in each layer of a landfill provides a favorable condition for  $H_2S$  mitigation. With the help of leachate recirculation, iron is captured by landfilled refuse because of the adsorption and precipitation reactions (Kjeldsen et al., 2002), especially in the shallow layer where a large amount of iron is available. Obviously, iron in landfills represents a good opportunity for mitigation of  $H_2S$  release. Moreover, the higher moisture content caused by leachate recirculation stimulates microbial activity and provides better contact opportunity between substrates, nutrients, and microorganisms (Long et al., 2009).



## 4. Conclusions

The landfill with leachate recirculation can obviously shorten the  $H_2S$  influence period and pollution risk to the surrounding environment, which is attributed to the MSW degradation speed and vertical distribution of sulfide. The  $H_2S$  endogenous mitigation behavior may be influenced by many environmental factors, such as moisture content, pH, iron, nitrate and nitrite. Migration, like adsorption/desorption, and conversion (i.e. chemical oxidation/reduction) mainly affect the  $H_2S$  release behavior in the initial stabilization phase of a landfill. Microbial activities related to sulfur, nitrogen and iron can further promote  $H_2S$  endogenous mitigation in the high reducing phase.



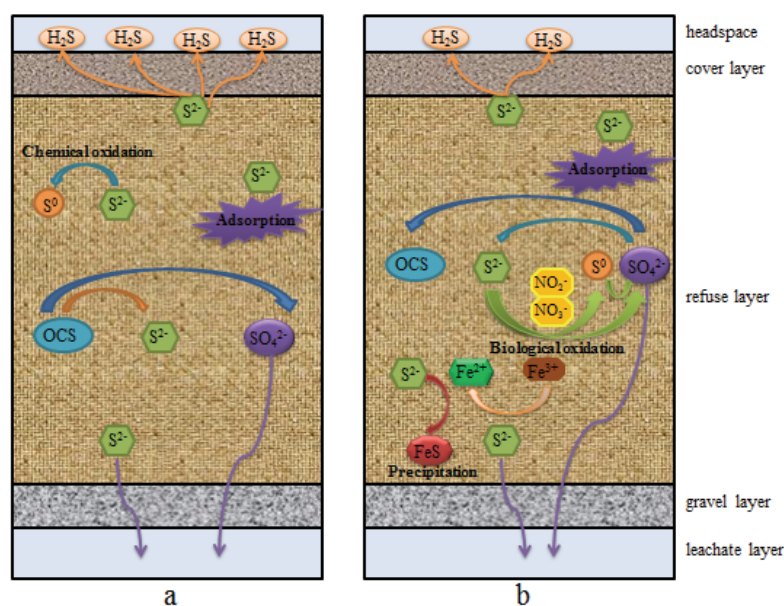


Figure 8: Schematic diagram of possible  $\text{H}_2\text{S}$  endogenous mitigation behavior in landfills.

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# A rapid and green determination of ammonia in indoor air

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Sodium hypochlorite-salicylic acid spectrophotography method, Nessler's reagent spectrophotography method, ion selective electrode method and ion chromatography method are the common methods for detecting ammonia content of indoor air. This study compares the advantages and disadvantages of these methods in experiments. The sodium hypochlorite-salicylic acid spectrophotometry method has good correlation when the ammonia concentration is  $0.5\sim 5\text{ }\mu\text{g}/10\text{mL}$ , the result of the uncertainty evaluation is  $0.792\pm 0.132\text{ mg/L}$ , which is more close to the standard value. Moreover, the pre-treatment of this method is rapid and green. This study confirms the sodium hypochlorite-salicylic acid spectrophotography method as a rapid and green method for the determination of ammonia in indoor air.

## 1. Introduction

Generally, there are various sources of indoor ammonia pollution, including interior drainage pipes, indoor decoration materials and concrete admixtures etc. (Yang et al., 2007; Wei et al., 2015). Ammonia in sewage pools or septic tanks escapes into the indoor air through the water channel. A lot of furniture with additives and whitening agent use ammonia. Common concrete admixture contains some ammonia stuff that increases indoor ammonia concentration. Sodium hypochlorite-salicylic acid spectrophotometry (GB/T18883-2002), Nessler's reagent spectrophotometry (GB50325-2010), ion selective electrode (HJ534-2009) and ion chromatography (GB/T18204.25-2000) are the common environmental monitoring methods of ammonia in indoor air. There are also the acetylacetone spectrophotometry method (Liu et al., 2010), the ninhydrin spectrophotometry method (Li et al., 2008), the field measurement method (Wang et al., 2014) and other methods. A previous study has demonstrated that ion selective electrode method and ion chromatography are simple and with high selectivity (Deng et al., 2006). But these methods require high accuracy apparatus and high cost. Nessler's reagent spectrophotography method (Li et al., 2007; Mei et al., 2000) is simple and convenient, but its sensitivity is lower. As the standard limit value of the ammonia concentration in indoor air is  $0.2\text{ mg/m}^3$ , the daily detection of the concentration of ammonia in indoor air by Nessler's reagent spectrophotography method is not suitable (Hu et al., 2000). On the other hand, the sodium hypochlorite-salicylic acid spectrophotography method has a higher sensitivity, a lower detection limit and a better correlation than Nessler's reagent spectrophotography.

## 2. Methods

A preliminary experiment is carried out to determine the dilution ratio according to the comparison of the depth of the colour of air samples with that of the ammonia standard series samples. That is different from the traditional pre-treatment. In this way, experimental reagents are reduced and testing time is saved since repeated dilution is

avoided. Finally, standard curves are drawn according to the national standard methods, as shown in Figure 1.

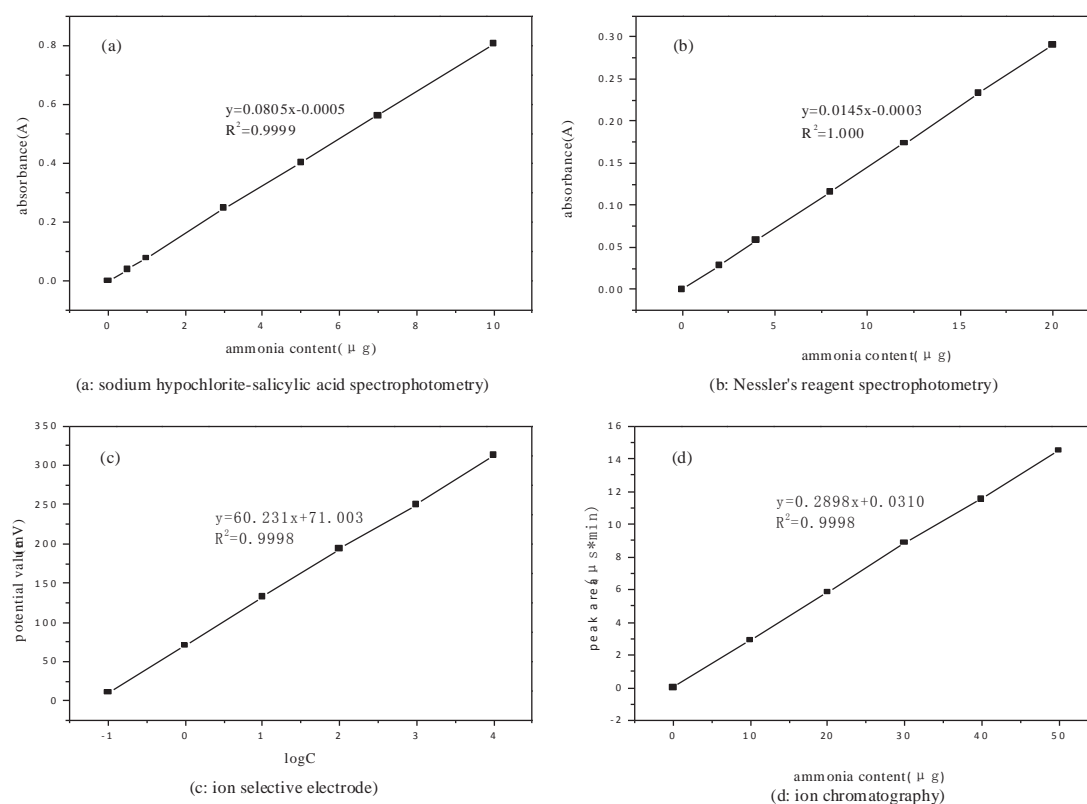


Figure 1: The standard curves of the four methods.

### 3. Results

The methods are tested by the same standard sample of ammonia which is used in the CNAS qualification determination, and the concentration of ammonia in the standard sample is  $0.796 \pm 0.038$  mg/L. The determination results are shown in Table 1.

Table 1: Results of the determination of ammonia by the four methods.

No.	Sodium hypochlorite-salicylic acid - spectrophotography	Nessler's reagent spectrophotography	Ion selective electrode	Ion chromatography
1	0.792	0.792	0.795	0.797
2	0.801	0.806	0.796	0.796
3	0.796	0.778	0.796	0.795
4	0.796	0.792	0.796	0.796
Average	0.796	0.792	0.796	0.796

The ion selective electrode method and the ion chromatography method have merits of simple operation, high sensitivity and good reproducibility. However, ion chromatography needs expensive instruments and high cost (Xiong et al., 2005). Table 1 shows that the four methods have no significant difference within the scope of quality control. The measurement of ammonia in indoor air in the laboratory is just state-of-the-

As shown in Figure 2, for 10 mL absorption liquid containing 1  $\mu\text{g}$  ammonia, the absorbance is 0.0781 for the sodium hypochlorite-salicylic acid spectrophotography method, and the absorbance is only 0.0146 for Nessler's reagent spectrophotography method. This indicates that the first method is more sensitive than the second one. Nessler's reagent (Li et al., 2007) contains large amounts of mercury salt. its toxicity and potential of secondary pollution is larger. The sodium hypochlorite-salicylic acid spectrophotography method has a good correlation when the ammonia content is  $0.5 \sim 5 \mu\text{g}/10 \text{ mL}$  in Fig. 2.

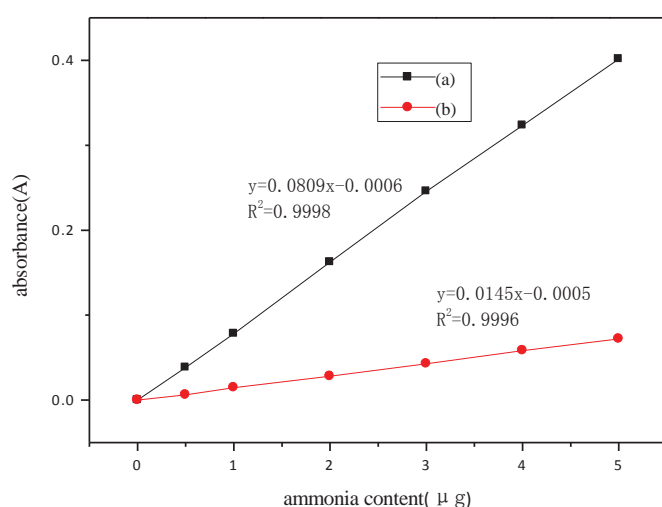


Figure 2: The standard curves of two kinds of spectrophotography methods.

The analysis of uncertainty showed that the result of the sodium hypochlorite-salicylic acid spectrophotography method is more close to the standard value.

The determination of ammonia in indoor air is a routine laboratory testing procedure. Sodium hypochlorite-salicylic acid spectrophotography method, Nessler's reagent

spectrophotography method, ion selective electrode and ion chromatography method have their own advantages and disadvantages.

The analysis of linear correlation and uncertainty showed that Nessler's reagent spectrophotography method is simple and convenient, but is not suitable for the determination of ammonia in indoor air since the standard limit value of the concentration of ammonia in indoor air is  $0.2 \text{ mg/m}^3$ . The sodium hypochlorite-salicylic acid spectrophotography method has a good linear correlation when the ammonia content is  $0.5 \sim 5 \text{ } \mu\text{g}/10 \text{ mL}$ . Its pre-treatment is rapid and green. This study confirmed sodium hypochlorite-salicylic acid spectrophotography method as a rapid and green method for the determination of ammonia in indoor air.

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## Odour dispersion modelling with Lagrangian and Gaussian models

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The Lagrange particle diffusion model LASAT and the Gaussian Austrian Odour Dispersion Model AODM are used to calculate direction-dependent separation distances around livestock farms. As the method has been published already in several papers, the focus is here on the differences of the results due to model physics. With both models, the same peak-to-mean algorithm is used to calculate short-term peak concentrations relevant for odour dispersion. For both models, the same emission and meteorological data, but model-specific atmospheric stability classes have been used. The estimate of atmospheric stability is obtained from three-axis ultrasonic anemometers using the standard deviations of the three wind components and the Obukhov stability parameter. Separation distances are determined for two odour impact criteria, namely exceedence probabilities of 3 and 8 %, each in combination with the odour threshold of 1 ou<sub>E</sub>/m<sup>3</sup>. The results will be presented and discussed for two sites with very different topographical surroundings.

### 1. Introduction

The calculation of separation distances between odour sources and residential areas is an appropriate method to reduce annoyance. Separation distances can be obtained from dispersion models. We use a Lagrangian and a Gaussian model to calculate direction-dependent separation distances. As dispersion models usually calculate concentrations over some integration time (half an hour or an hour), the authors developed a mechanism to account for short-term concentrations relevant for the perception of the human nose. This so-called peak-to-mean algorithm is explained in detail in (Piringer et al., 2015; Piringer et al., 2016) and applied with the Lagrangian dispersion model LASAT (Janicke Consulting, 2013) as well as the Gaussian Austrian Odour Dispersion Model AODM (Schaubberger et al., 2002).

LASAT and AODM calculations have been conducted at two Austrian sites of very different surroundings, namely Kittsee east of Vienna and Weißbach in the Saalach valley in the county of Salzburg. At both sites, meteorological input data are provided by three-axis ultrasonic anemometers operated over the period of at least one year. Separation distances at both sites are determined for Austrian odour impact criteria allowing an exceedence of the odour threshold by 3 % (high level of protection) or 8 % (lower level of protection). Material and methods are outlined in Section 2, the results and a brief discussion are presented in Section 3. Section 4 contains a summary of the findings.

## 2. Material and methods

A description of the dispersion models and the peak-to-mean approach is in the latest version given in Piringer et al. (2016), so only a brief summary is given here to maintain the status of a stand-alone paper. The dispersion model LASAT (Janicke Consulting, 2013) simulates the dispersion and the transport of a representative sample of tracer particles utilizing a random walk process (Lagrangian simulation). It computes the transport of passive trace substances in the lower atmosphere (up to heights of about 2000 m) on a local and regional scale (up to distances of about 150 km). LASAT is usually run with the Klug-Manier stability scheme (TA Luft, 2002). The Austrian odour dispersion model (AODM, Piringer et al., 2007; Piringer et al., 2013; Schauburger et al., 2000; Schauburger et al., 2013; Schauburger et al., 2002) estimates mean ambient concentrations by the Austrian regulatory dispersion model (Österreichisches Normeninstitut, 1996; Kolb, 1981) and transforms these to instantaneous values depending on the stability of the atmosphere. The model uses a traditional discrete stability classification scheme with dispersion parameters developed by Reuter (1970). The peak-to-mean approach used to transform the half-hourly model concentrations into short-term peak values as well as the scheme to transform the Obukhov stability parameter  $OSP [m^{-1}]$  to atmospheric stability classes depending on the local roughness length  $z_0 [m]$  are described in full detail in Piringer et al. (2016).



Figure 1: Map of Austria indicating the sites of investigation.

The Obukhov stability parameter and the standard deviations of the three wind components are derived from at least a one-year time series of ultrasonic anemometer measurements. Sonic anemometers measure the along-path velocity components from the travel time of acoustic waves between transducers separated about 10-20 cm. In addition to the three-dimensional wind vector, the sound velocity is derived, from which the so-called “sonic temperature” is calculated. The measurement of sonic temperature fluctuations is necessary to calculate the sensible heat flux. Other quantities which are derived from sonic measurements are the means, standard deviations, and co-variances of the wind components and the momentum flux, the Obukhov stability

parameter, and the friction velocity. Sonic anemometers usually sample at 10 Hz, and the data are usually stored as averages over 10 minutes or half an hour.

The investigation has been carried out at two very different sites (Fig. 1) where a one year continuous data set of half-hour ultrasonic anemometer measurements is available. One is Kittsee, east of Vienna near Bratislava. The site is located at 17.070° E and 48.109 N at 136 m asl. It is within flat terrain, mainly farmland. Weissbach near Lofer (12.789 E, 47.498 N, at 678 m asl.) is situated in the Saalach valley in the county of Salzburg which at the site stretches from SE to NW. The valley with approx. only 1 km in width is relatively narrow, flanked by steep slopes with heights of several hundred meters.

Table 1: Source data for dispersion calculations.

Stack height	[m]	8.0
Stack diameter	[m]	2.7
Outlet air velocity	[ms <sup>-1</sup> ]	3.0
Volume flow rate	[m <sup>3</sup> h <sup>-1</sup> ]	60,000
Temperature	[°C]	20
Odour emission rate	[ou <sub>ES</sub> <sup>-1</sup> ]	5,200

For all model runs, the same source data are used (Table 1 and Piringer et al., 2016). These data are typical for a pig fattening unit with about 750 animals with a mean life mass of 70 kg. For the model runs, the source data are assumed constant over the year.

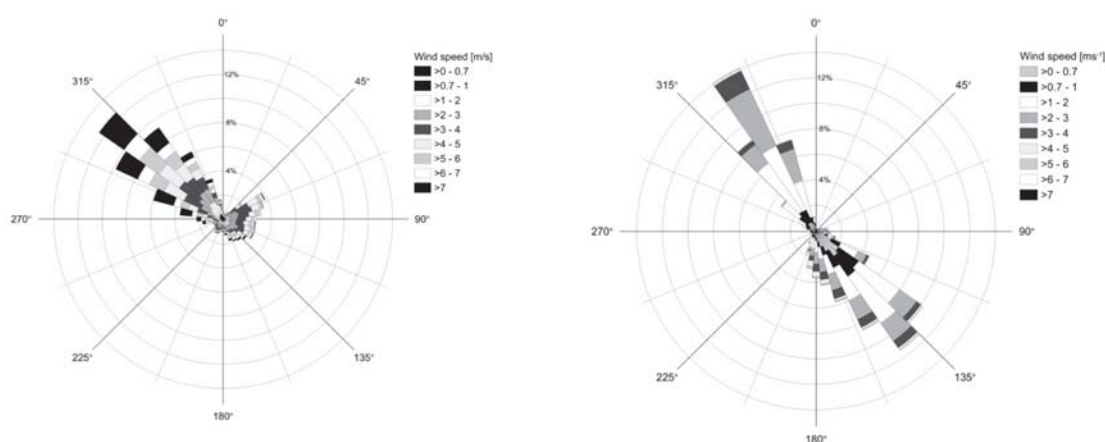


Figure 2: Wind roses at (left) Kittsee (03.03.2006 – 31.05.2007) and (right) Weissbach (01.09.2010 – 31.08.2011); black-white coding denotes wind speed.

### 3. Results and discussion

Kittsee can experience high wind speeds, mainly from northwesterly directions, often associated with frontal of systems and storms (Fig. 2). The secondary maximum of wind directions is from Northeast, in contrast to a lot of other meteorological stations in the area. This is explained by a topographical deflection of the regional flow in the area caused by the southernmost tip of the Carpathian mountains in the region of Bratislava North of the site. These wind directions show on average lower wind speeds as they are mainly observed in anti-cyclonic conditions.

Due to the topographical situation at Weissbach, the wind is channelled along the valley axis. Up-valley flow is from NW, down-valley flow from SE (Fig. 2b). The down-valley flow from SE shows a larger fraction of weaker winds, but also slightly more stronger winds than the up-valley flow. The strong southerly winds might also be associated with Foehn events.

The frequency of stability classes is at both sites determined from the OSP with a roughness length of 0.02 m at Kittsee and 0.5 m at Weissbach derived from on-site wind measurements. Neutral conditions are most abundant at Kittsee (Fig. 3, left panel), with 40 % for class 4 (AODM) and over 50 % for class III/1 (LASAT). This is due to the high wind speeds and/or cloudy conditions. The large differences for classes 4 and 5 compared to III/2 and III/1 is due to the different stability concepts of the Reuter and Klug-Manier schemes. Reuter's class 5 has no appropriate counterpart in the LASAT scheme.

From Fig. 3 (right panel), the frequency distribution is roughly 30:30:40 for unstable-neutral-stable conditions at Weissbach, especially for the AODM scheme. The LASAT scheme calculates slightly more stable and less unstable conditions.

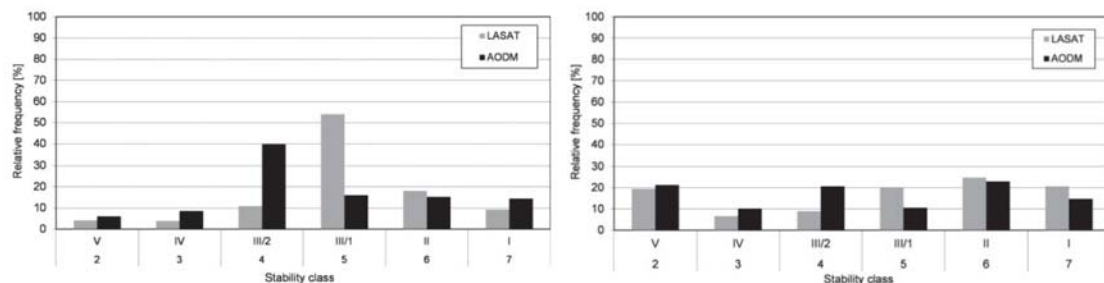


Figure 3: Relative frequency [%] of stability classes at (left) Kittsee and (right) Weissbach.

The different meteorological conditions displayed in Figs. 2 and 3 lead to remarkable differences in the dependence of the peak-to-mean factors with distance between the two sites (Figs. 4 and 5). The peak-to-mean ratios, however, start at similar values near the source. Their decrease with distance is much more pronounced at Weissbach, compared to Kittsee. In unstable conditions at Kittsee, peak-to-mean ratios at 100 m are between 5 and 8 for AODM (Fig. 4, left panel) or 4 and 5 for LASAT (Fig. 4, right panel). At Weissbach, they show a value of about 2 (Fig. 5, both panels). At Kittsee (Fig. 4), the attenuation curves in unstable conditions deliver values above 1 several 100 m downwind. At Weissbach (Fig. 5), all peak-to-mean curves reach the value of 1 about 200 m downwind, unstable conditions being most relevant here. For neutral and stable conditions at Kittsee, the peak-to-mean ratios reach the value of 1 even further downwind than in unstable conditions, so that stable conditions are most important at large distances.

The differences in the attenuation curves between Kittsee and Weissbach are due to the fact that Kittsee shows much higher wind speeds, but far less turbulence than Weissbach. The increased turbulence due to topography at Weissbach leads to a quicker decrease of peak-to-mean ratios with distance. This is explained in detail in Piringer et al. (2016) comparing the conditions at Weissbach to another flatland site in Austria.

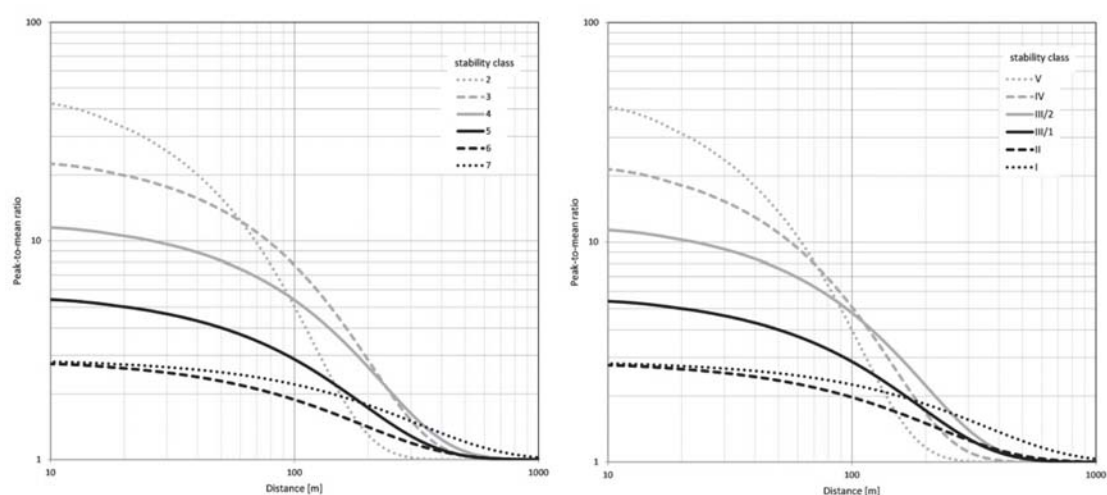


Figure 4: Peak-to-mean factors at Kittsee derived from ultrasonic anemometer data (OSP) for AODM (left) and for LASAT (right).

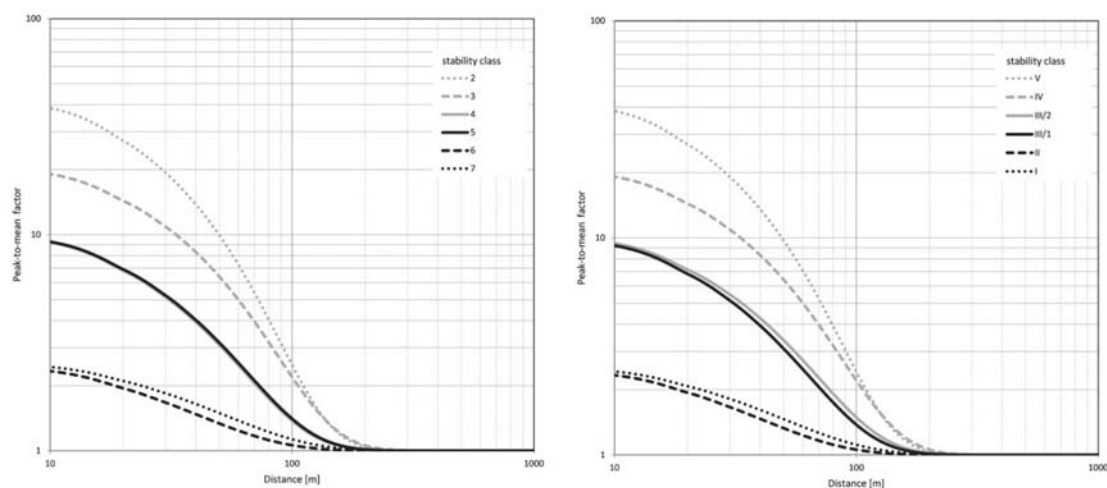


Figure 5: Peak-to-mean factors at Weissbach derived from ultrasonic anemometer data (OSP) for AODM (left) and for LASAT (right).

Direction-dependent separation distances are calculated for two odour impact criteria used in Austria:  $1 \text{ ou}_E/\text{m}^3$  and 3 % exceedence probability, representative for recreation areas (high odour protection),  $1 \text{ ou}_E/\text{m}^3$  and 8 % exceedence probability, representative for residential areas mixed with commercial activity (lower odour protection). Following Piringer et al. (2016), the separation distances are shown as isolines in the coming figures, encompassing the area of exceedence of the given thresholds. The larger the area, the more unfavourable is the odour impact criterion. In each of the figures showing the separation distances, AODM results (left) are compared to LASAT results (right) for the same scenario.

At Kittsee, using AODM, maximum separation distances for an exceedence probability of 3 % are about 350 m towards SE and about 250 m towards W; with 8 %, these distances are about 250 and 150 m, respectively (Fig. 6, left). The calculation with LASAT leads to a further increase of separation distances for an exceedence probability of 3 % by about 100 m (Fig. 6, right) compared to AODM. For 8 %, however,



there is only a slight increase towards SE to about 320 m, whereas a decrease to about 100 m is observed towards W, compared to the AODM results.

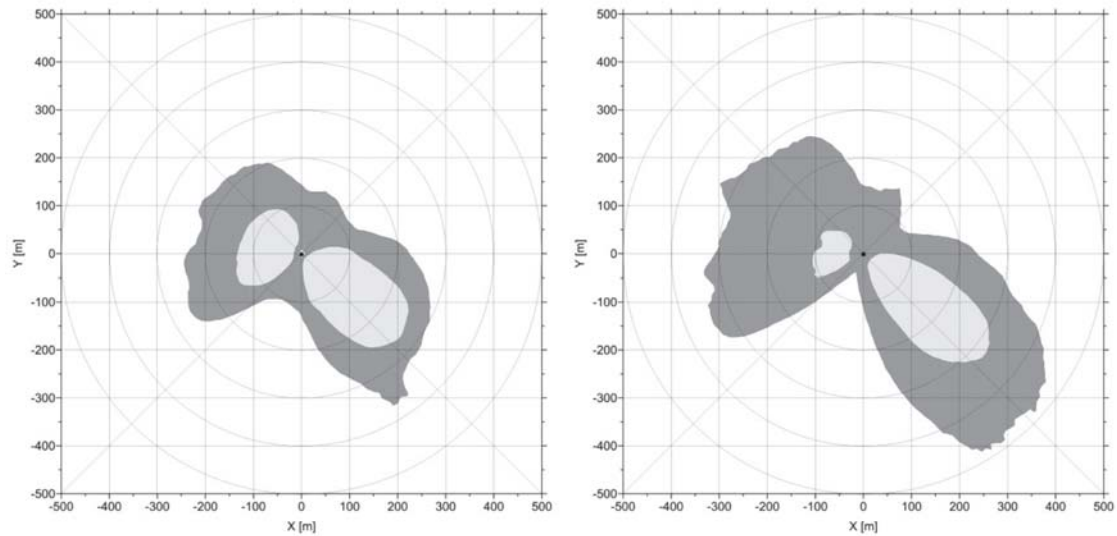


Figure 6: Direction-dependent separation distances [m] with (left) AODM and (right) LASAT for 1 ouE/m<sup>3</sup> and 3 % (dark grey) and 8 % (light grey) exceedance probability with peak-to-mean factors derived from ultrasonic anemometer measurements at Kittsee.

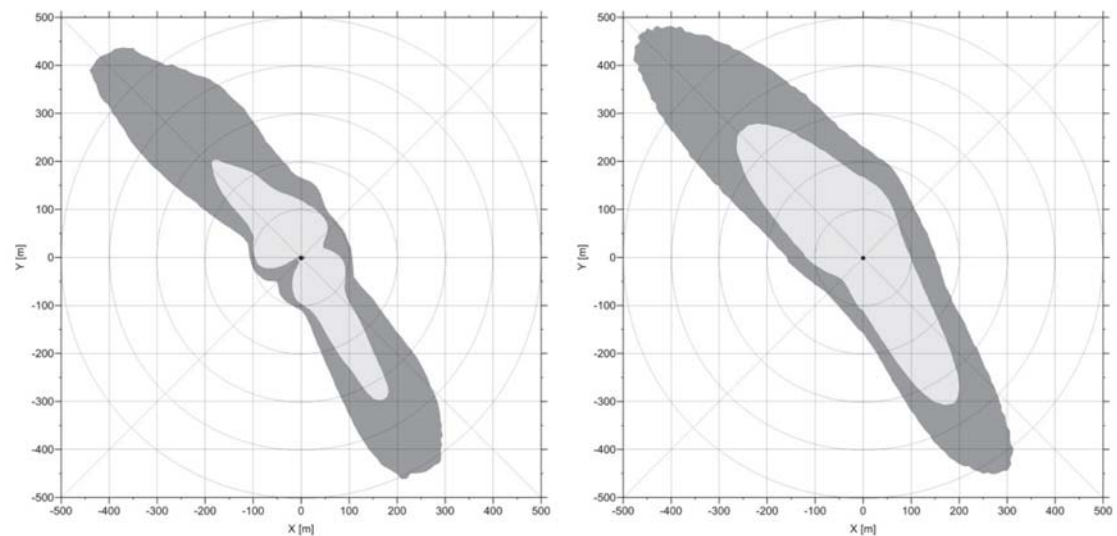


Figure 7: Direction-dependent separation distances [m] with (left) AODM and (right) LASAT for 1 ouE/m<sup>3</sup> and 3 % (dark grey) and 8 % (light grey) exceedance probability with peak-to-mean factors derived from ultrasonic anemometer measurements at Weissbach.

At Weissbach, using AODM, maximum separation distances for an exceedance probability of 3 % are about 600 m towards NW and about 500 m towards SE; with 8 %, these distances are about 300 m in both directions (Fig. 7, left). The calculation with LASAT leads again to an increase of separation distances for an exceedance probability of 3 % (Fig. 7, right) compared to AODM. For 8 %, however, maximum separation distances calculated with LASAT only slightly increase to about 400 m in both directions, compared to the AODM results. The area of exceedances is generally larger for LASAT than for AODM.



Looking at Figs. 6 and 7, considerable differences in separation distances between the chosen sites and also systematic differences between the two models used are observed. Looking first on the differences caused by the sites, these are certainly due to the different meteorological conditions, in the first place. We selected two topographically very different sites, a flatland and a narrow valley site, for the calculation of separation distances (Fig. 1). Kittsee can experience high wind speeds, especially with winds from NW (Fig. 2a); half-hour average wind speeds up to  $14 \text{ ms}^{-1}$  are observed. In contrast, wind speeds of up to only  $7 \text{ ms}^{-1}$  occur at Weissbach in the Saalach valley, where the flow is strongly channelled along the valley axis (Fig. 2b). The near-surface atmosphere, apparently due to the topographical conditions, is much more turbulent at Weissbach than at Kittsee (Piringer et al., 2016; Piringer et al., 2015). The overall effect on the peak-to-mean factors is a more rapid decrease with distance at Weissbach compared to Kittsee (compare Figs. 4 and 5).

Another important meteorological parameter for the calculation of separation distances is atmospheric stability which is also entirely different between the two sites. Atmospheric stability is here determined from three-axis ultrasonic anemometer measurements via the Obukhov stability parameter (Section 2, see Piringer et al. (2016) for details). When transformed into stability classes, the local roughness length is also considered, and the stability schemes associated with the models (the Reuter (1970) scheme with AODM, the Klug-Manier scheme (TA-Luft, 2002) with LASAT) use different class widths and limits for this transformation. The distribution of stability classes is different between the schemes as well as between the sites (Fig. 3).

Atmospheric stability exerts an influence on the separation distances. As the peak-to-mean factors are stability-dependent, a separate curve is obtained for each stability class for their decrease with distance (Figs. 4 and 5). The differences between Kittsee and Weissbach are very pronounced, for all stability classes. Kittsee, compared to Weissbach, is thus characterized by higher wind speeds, lower turbulence and larger peak-to-mean factors. Nevertheless, the separation distances along the valley axis at Weissbach are considerably larger than the maximum separation distances at Kittsee, especially with AODM (Figs. 6 and 7).

The solution to this problem is seen in the fact that the along-valley wind directions at Weissbach are often associated with stable conditions, whereas the main wind direction sector at Kittsee is mainly combined with neutral atmospheric stability. The channelling of the flow in combination with frequent stable conditions causes higher odour concentrations and leads to the enhanced separation distances at Weissbach. This enhanced frequency apparently compensates for the peak-to-mean factors which are not relevant at Weissbach for large separation distances. At Kittsee, separation distances south-east of the source, calculated with LASAT, are with almost 500 m for an exceedence probability of 3 % comparable to those at Weissbach along the valley (compare Figs. 6 and 7, right panels). The combination of a high occurrence of wind directions and frequent neutral conditions is also prone for large separation distances. A more detailed discussion is available from Piringer et al. (2016).

As already discussed in Piringer et al. (2016), a reason for generally larger separation distances with LASAT compared to AODM is probably due to the different model physics: whereas AODM, a Gaussian model, produces a stationary concentration field for a half-hour time step, LASAT calculates concentrations as long as the trajectories stay within the model domain, thus likely increasing residence times and also separation distances. Because of this more realistic assumption we conclude that,

even in flat terrain, the use of a Lagrangian model is to be preferred over a Gaussian model.

#### 4. Conclusions

In this paper, direction-dependent separation distances are discussed derived for Austrian odour impact criteria to avoid odour annoyance at two sites, calculated with two models, the Gaussian Austrian Odour Dispersion Model AODM and the Lagrangian particle diffusion model LASAT. Short-term peak odour concentrations have been calculated with a stability-dependent peak-to-mean algorithm (Section 2). The same emission (Table 1) and meteorological data have been used for both models. The estimate of atmospheric stability is obtained from three-axis ultrasonic anemometers using the standard deviations of the three wind components and the Obukhov stability parameter. The results are demonstrated at the Austrian villages Kittsee and Weissbach (Fig. 1) which are very different with respect to their topographical surroundings (Section 2) and thus the resulting on-site meteorological conditions (Section 3). The causes for the differences in separation distances between the two sites were analysed.

The shape of the contour lines of separation distances is primarily determined by the distribution of wind directions; the elongation in the main wind directions is in addition caused by wind speed (at Kittsee) and the frequency distribution of stability classes with wind direction (at Weissbach); see Figs. 6 and 7. Weissbach is a very specific site as its location in the narrow Saalach valley leads to a strong channelling of the flow and the development of a valley wind system. Both contribute to higher odour concentrations and thus extended separation distances along the valley axis despite to the lower wind speeds (Fig. 2) and the smaller peak-to-mean factors at larger distances (Figs. 4 and 5) compared to Kittsee. The combination of a channelled flow with an enhanced occurrence of stable conditions leads to larger separation distances than larger peak-to-mean factors. Atmospheric stability in combination with frequent wind directions can be very important for large separation distances, outweighing the values of the peak-to-mean factors. In the vicinity of livestock farms, even slight reductions can become important, and they will increase with higher exceedence probabilities, often valid in agricultural areas. This underlines the importance of the peak-to-mean concept and the use of on-site meteorological information for odour impact analyses.

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## Odour control versus potential pollution and risks

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Odours can cause hazards to the environment and human health. The increasing public odour complaints and expectations, and stricter environmental regulations have made odour control important all over the world. There are various technologies for odour control, mainly divided into physical, chemical, and biological treatment. For a sustainable solution of odour problems, there is more to consider besides the performance of the technologies to remove the target pollutants. This article focuses on the potential pollution and the environment-health risks that may be created during the odour control processes, including activated carbon adsorption, chemical scrubber, thermal treatment, biological treatment, UV photodegradation, and non-thermal plasma. Accordingly, the solutions to decrease the pollution and risks are discussed.

### 1. Odour control

Odours can be caused by a variety of harmful substances, such as H<sub>2</sub>S, NH<sub>3</sub>, and volatile organic compounds (VOCs), which lead to a threat to the environment and human health. There are significantly increasing public complaints due to odorous emissions from municipal activities, such as waste water treatment plants, or industrial activities (Schlegelmilch et al., 2005b, Lebrero et al., 2011). As a result, the regulations related to air emissions have become stricter in many countries in recent years. For instance, in China, there was a project called the 12<sup>th</sup> Five Year Plan (2011–2015), which clearly aimed to improve the quality of water and air (NPC, 2011).

A variety of technologies have been developed for odour control. According to the nature of each technology, they are mainly classified into physical, chemical, and biological technologies (Schlegelmilch et al., 2005b). To select an appropriate odour control technology, it is necessary to consider the nature and concentration of odorants, the level of efficiency required, the type of odour sources (active or passive), the air flow rates to be treated, site characteristics including operation and maintenance capabilities, etc. (Lebrero et al., 2011, Burgess et al., 2001). Moreover, there should be more to consider besides the factors mentioned above (especially not only the performance of the technologies to remove the target pollutants). The potential pollution and environment-health risks that may be induced during odour control processes should not be neglected. To the authors' knowledge, there are few works that focus on the potential pollution and the environment-health risks of each technology for odour control. In this study, we review the potential problems of pollution and risks of odour control processes, including activated carbon adsorption, chemical scrubber, thermal treatment, biological treatment, UV photodegradation, and non-thermal plasma. Moreover, the according solutions are discussed.

## 2. Potential problems and solutions

### 2.1 Activated carbon adsorption

#### 2.1.1 Potential problems

Porous carbonaceous materials, such as activated carbon, are widely used in the removal of pollutants present in liquid and gaseous effluents due to their excellent adsorbent properties and large surface areas. They do not destroy the pollutants, but just concentrate them (Kittrell et al., 1991).

However, once exhausted, activated carbon can become hazardous residues. The Environmental Protection Agency (EPA, USA) defines that solid wastes generated during the treatment, storage, or disposal of a listed hazardous waste or their mixture with a listed hazardous waste are also hazardous waste. Therefore, activated carbon wastes from the treatment of contaminated air that contains listed hazardous wastes should be managed as hazardous waste. Spent activated carbon is usually incinerated or disposed in landfills, resulting in risks of pollution (Salvador et al., 2015a). The disposal of hazardous waste in landfills is less and less accepted due to the growing concerns about the effect of pollutants on the environment and more stringent environmental standards (Yuen and Hameed, 2009, Zanella et al., 2014). Adsorbates from granular activated carbon can be released into the atmosphere in reactive, oxidized and partially oxidized states, aggravating the air pollution (Nath and Bhakhar, 2011). The current Anniston Chemical Agent Disposal Facility operating permit states that used carbon will be evaluated for chemical agent contamination if it has been exposed to agent concentrations of  $\geq 0.0001 \text{ mg/m}^3$ . Once the carbon is determined to be a hazardous waste, it should be sent to a permitted hazardous waste treatment, storage, and disposal facility (NRC, 2009).

In addition to the disposal, the spent activated carbon can be regenerated, which attempts to remove pollutants retained on the activated carbon's surface to restore adsorptive capacity without modifying porosity or causing adsorbent mass losses. However, the removal process will result in either water or air streams polluted with the contaminants adsorbed by the activated carbon, and the liquid or air stream can be a disposal problem. The desorbed contaminants must be treated or reclaimed (Kittrell et al., 1991).

Regeneration methods include three major groups: thermal, chemical and microbiological regeneration (Salvador et al., 2015a, Salvador et al., 2015b). Thermal regeneration is the most widely used industrial-scale method (Salvador et al., 2015a). Thermal regeneration is typically processed at 800–1000 °C, which involves high energy consumption (Yuen and Hameed, 2009). Román et al. (2013) analysed the gas released during the thermal regeneration of spent activated carbon, and found that there were H<sub>2</sub>, CO, CH<sub>4</sub> and CO<sub>2</sub> in the gas. CH<sub>4</sub> and CO<sub>2</sub> are both greenhouse gases, which could enhance global warming (Sommerfeld et al., 1993), while CO is poisonous. Regeneration with liquid water implies that the fluid becomes contaminated by the same pollutants, subsequent purification of the liquid should be carried out (Salvador et al., 2015b).

Complete regeneration of the spent activated carbon using microbial processes is difficult to achieve, because microbial degradation products will be adsorbed and the pores will be blocked by the decay products of microbial cells (Nath and Bhakhar, 2011, Kim et al., 2014). Moreover, its ecological impacts should be further investigated.

### 2.1.2 Solutions

To make the saturated activated carbon re-usable and to avoid the contamination that is caused by its disposal, the regeneration of spent activated carbon is an attractive option, even though problems can arise during regeneration (Salvador et al., 2015a, Salvador et al., 2015b).

Regarding the high energy consumption of thermal regeneration, microwave energy can be a potential method and powerful replacement (Yuen and Hameed, 2009).

Adsorption can be used as an enrichment step prior to thermal gas treatment (Rafson, 1998), and it can be combined with thermal treatment or other treatments for the subsequent treatment of CH<sub>4</sub> and CO.

## 2.2 Chemical scrubber

### 2.2.1 Potential problems

Chemical scrubbers are the most commonly employed abatement techniques in waste water treatment plants (Gabriel and Deshusses, 2004). Chemical scrubbers are implemented in packed towers, using NaClO, NaOH, KMnO<sub>4</sub>, H<sub>2</sub>O<sub>2</sub>, or H<sub>2</sub>SO<sub>4</sub> as reagents. Considerable amounts of chemicals are required, and reaction products must be removed or treated. Chemicals used in chemical scrubbers for off-gas treatment might also generate residual odours as well as odorous reaction products such as aldehydes. The hazardous nature of the employed chemicals and the generated by-products represents a serious challenge to its supremacy in a world increasingly devoted to sustainable development (Schlegelmilch et al., 2005b, Charron et al., 2004, Gabriel and Deshusses, 2004, Jeavons et al., 2000, Metcalfeddy et al., 2003).

The use of chemical scrubbers can lead to climate change, freshwater eutrophication, photochemical oxidant formation, human toxicity and ecotoxicity, mainly due to the use of large amounts of chemicals. The use of chemicals in chemical scrubbers can account for approximately 80% of its total impact on climate change, with NaClO presenting the largest contribution. The chemicals added in chemical scrubbers (especially NaClO) can induce ecotoxicity and human toxicity. Moreover, hypochlorite has the tendency to oxidise and chlorinate the compounds at the same time. Furthermore, chlorine gas (Cl<sub>2</sub>) is also generated (Schlegelmilch et al., 2005b). Even though NaClO is both relatively inexpensive and easy to handle, the significant amount required for odorant oxidation can play a key role in the impacts associated with the use of chemicals (Alfonsín et al., 2015). In addition, surface water criteria to protect aquatic life (IDAPA 58.01.02.250) prescribes that pH should be within the range of 6.5 to 9.0 (EPA, 2016). However, the pH value of discharged water from the chemical scrubber is usually out of that range (Gabriel and Deshusses, 2004). Therefore, the use of chemicals in chemical scrubber could lead to negative burdens (Estrada et al., 2011, Alfonsín et al., 2013).

### 2.2.2 Solutions

As NaClO plays a key role in the impacts associated with the use of chemicals (Alfonsín et al., 2015), alternative chemicals, such as ferric and ferrous ions chelated with EDTA, Fe<sub>2</sub>O<sub>3</sub> and FeO, and H<sub>2</sub>O<sub>2</sub>, can be potential alternatives (Shareefdeen et al., 2005). The investigations on alternative chemicals, which are more environmentally friendly and cost-effective, could be conducted.

The chlorine gas formed in the chemical scrubber could be absorbed by NaOH in the same or another scrubber (Drust and Deacon, 1995).



The pH of the discharged water from the chemical scrubber should be regulated by adding acid or alkali (Drust and Deacon, 1995). If there are multiple stages, acidic waste water could be used to neutralize the alkali one, thus reducing the consumption of chemicals.

## **2.3 Thermal treatment**

### **2.3.1 Potential problems**

Thermal treatment can be basically applied to any exhaust air. Thermal treatment includes catalytic and non-catalytic techniques. In a non-catalytic thermal treatment process, VOC-laden air is thermally treated (oxidized/decomposed) at temperatures of about 730-850 °C (Chuah and Warahena, 2009). Since the concentration of VOCs is often low, it usually requires the addition of natural gas or a pre-concentration, such as by adsorption. However, this method produces secondary emissions like nitrous and sulphur oxides (Schlegelmilch et al., 2005b). In presence of NO<sub>x</sub>, photochemical oxidation can happen (Alfonsín et al., 2013).

If the waste gas contains chlorine, the production of chlorine during the thermal treatment process is not desired, since it can create an air pollution problem. However, even the most highly active oxidation catalysts can oxidize the HCl product, and the product is chlorine rather than HCl (Kittrell et al., 1991).

Little attention has been paid to the production of dioxin-like compounds (DLCs), which have been classified by the World Health Organization (WHO) as one of the most persistent toxic chemical substances in the environment (WHO, 2002). The main risk of dioxins for human health is that they can alter the development of many cells and can cause illnesses, such as cancer, disruption of the endocrine system, or reproductive and development problems. The main sources of DLCs are the human interaction, especially combustion-related processes. PCDD/Fs are formed during the reactions between organic compounds and chlorine at high temperature (Dopico and Gómez, 2015). Several investigations have proven that DLCs can be generated during the thermal waste gas treatment (Li et al., 2012, Teng et al., 2015).

Therefore, some researchers indicate that incineration technologies are not an attractive odour abatement alternative due to their high investment and operation costs and the potential production of toxic dioxins and furans (Delhoménie and Heitz, 2005).

### **2.3.2 Solutions**

To reduce the additional fuel, catalytic process can be an alternative, because they can be operated at lower temperatures (Zhang et al., 2016). Besides, for non-catalytic processes, using advanced systems with heat recovery (recuperative thermal oxidisers, regenerative thermal oxidisers) can reduce the energy consumption significantly (Greco and Jain, 2016).

NO<sub>x</sub> can be controlled by selective catalytic reduction (SCR), using NH<sub>3</sub>. V<sub>2</sub>O<sub>5</sub>/TiO<sub>2</sub>-based catalysts (Wielgosiński, 2011).

The selectivity of catalysts for oxidation of halogenated hydrocarbons is very important, because appropriate catalysts can convert the chlorine content to HCl rather than to Cl<sub>2</sub> or other chlorine-containing products (Kittrell et al., 1991).

The proper selection of raw materials to avoid the addition of chlorine into the process is an effective method to reduce the formation of dioxins. Extensive research and field measurements have indicated that commercial NO<sub>x</sub> control catalysts also reduce the PCDD/F emission (Wielgosiński, 2011, Dopico and Gómez, 2015). Another way to

prevent the regeneration of DLCs is the application of the accelerated cooling of the off-gas (UNEP, 2006).

## **2.4 Biological treatment**

### **2.4.1 Potential problems**

Biological treatment is regarded as an environmentally friendly and cost-effective waste air treatment method. It simply uses microbes to consume the pollutants from contaminated air streams (EPA, 2003). However, biological treatment systems may also be a source of pollution.

Biological processes may produce high concentrations of bioaerosol, which are a potential health risk. Some investigations have been conducted on the sanitary consequences for workers. The results indicate an increase in respiratory, gastrointestinal, and skin symptoms compared with a control group (Schlegelmilch et al., 2005a). Ottengraf and Konings (1991) reported that there was an extra contamination due to the filtration process at low CFU-numbers of the inlet gas, therefore, biofilters may serve as a source of emission of microorganisms. Wang et al. (2009a) measured bioaerosol concentration of exhaust gas from the biofilter to investigate the ecological safety of the biological process. The result shows that the microorganism emission from the biofilter is as high as  $1.38 \cdot 10^3$  CFU/m<sup>3</sup>. Schlegelmilch et al. (2005a) found that the number of mesophilic bacteria determined in the outlet of the biofilter is about two times higher than that in the inlet, and microbial analysis revealed that the species of microorganisms at the inlet are different from that at the outlet of the treatment system. All these investigations have proven that biological treatment systems can cause secondary emissions of bioaerosol.

Besides the emissions of bioaerosol, biological treatment systems can also produce pollutants in the waste water. Sulphurous and nitrogenous organic or inorganic compounds are oxidized to sulphuric and nitric acid, which can cause acidification of water (Hartikainen and Ruuskanen, 1996). In addition, N-compounds discharged in the waste water, including free ammonia, ammonium, nitrite and nitrate, can result in water eutrophication (Alfonsín et al., 2013).

### **2.4.2 Solutions**

Ozone is a biocide, which can control the bioaerosol emissions from the biological treatment system and the growth of excess biofilm in the bioreactor. UV photodegradation process produces ozone. Therefore, an integrated UV-biofiltration system is investigated, and proved to be a promising option to effectively reduce the bioaerosol emissions from biological treatment system (Wang et al., 2008, Wang et al., 2009b, Moussavi and Mohseni, 2007, Wang et al., 2009a).

The emitted concentration of bioaerosol is considerably affected by the structure of the packing material. Finely structured, homogeneous packing materials would emit lower CFU-numbers of bacteria than more inhomogeneous materials. Therefore, the selection of packing materials and optimization of its structure could be an approach to control the bioaerosol emissions (Ottengraf and Konings, 1991).

The rate of the bioaerosol emissions process is assumed to be proportional to the gas velocity. There is a clear decrease trend of the outlet concentration as the velocity increased. Thus bioaerosol emissions could also be reduced by controlling the gas velocity (Ottengraf and Konings, 1991).

Regarding the acidification and eutrophication caused by the formation of sulphuric acid, nitric acid and other N-compounds, the waste water from the biological treatment

system should be treated (neutralized or biological treated) before it is discharged into the aquatic environment.

## **2.5 Thermal treatment**

### **2.5.1 Potential problems**

UV photodegradation, using UV irradiation to rapidly oxidize the pollutants, is capable of oxidizing a wide range of contaminants (Koh et al., 2004).

A few studies indicate that some products, which are more toxic compared to their parent contaminants, can be generated during UV photodegradation (Koh and Thiemann, 2001, Cantavenera et al., 2007, Jeong et al., 2005, Zhang et al., 2006). Wang et al. (2009a) used UV photodegradation to treat chlorobenzene and found that both the acute toxicity and the genotoxicity of the photodegradation products increased in the exhaust gas after UV photodegradation. The exhaust gas containing these toxic products shows potential health risks. Cantavenera et al. (2007) indicated that genotoxic intermediates can be formed during the photocatalytic degradation of paraquat.

UV lamps (185- and 254-nm UV light) can produce a high concentration of ozone (up to 50 mg/m<sup>3</sup>). Moreover, it is reported that 0.2 mg/m<sup>3</sup> of ozone might have a harmful effect on human health. What's more, the half-life of ozone in air is approximately 16 h (20 °C, 1 atm). As a result, health risks for workers are to be expected if the ozone processes self-decay in the gas phase (Moussavi and Mohseni, 2007, Wang et al., 2008, Wang et al., 2009a).

The possible production of toxic contaminants should be carefully taken into consideration when a UV photodegradation process is going to be applied.

### **2.5.2 Solutions**

The toxic products, such as chlorophenol and ozone, produced by the UV photodegradation, can be removed by subsequent biofiltration. A biological treatment system shows an apparent detoxification effect on UV photodegradation. The combined UV-biofiltration process can be a promising option to make UV photodegradation ecologically safer (Wang et al., 2009a, Wang et al., 2008, Wang et al., 2009b). However, a biological system as an ozone deletion process should be used carefully, because ozone might inhibit or even kill microorganisms in the biological system (Moussavi and Mohseni, 2007).

## **2.6 Non-thermal plasma**

### **2.6.1 Potential problems**

Non-thermal plasma provides a highly chemical reactive environment (e<sup>-</sup>, O<sup>\*</sup>, HO<sub>2</sub><sup>\*</sup>, OH<sup>\*</sup>, N<sub>2</sub><sup>\*</sup>, O<sub>3</sub>, etc.) to decompose the VOCs at ambient conditions (Sultana et al., 2015, Vandenbroucke et al., 2011).

Non-thermal plasma can result in the formation of excess ozone, which is a hazardous substance to human health (Pekárek, 2003). Moreover, the industrial implementation of non-thermal plasma for the abatement of VOCs is impaired by three main bottlenecks such as poor product selectivity, formation of undesired by-products (O<sub>3</sub>, NO<sub>x</sub>, other VOCs and aerosols) that often increase the overall toxicity of the treated gas stream (Sultana et al., 2015).

### **2.6.2 Solutions**

To decrease the undesired by-products, the combination of non-thermal plasma with catalysts/sorbents, i.e., plasma-catalysis, has been remarkably investigated during the

last decade (Sultana et al., 2015). In such a hybrid system, the catalyst can be integrated either inside (IPC-Inside Plasma Catalysis) (Kim et al., 2006) or downstream (PPC-Post Plasma Catalysis) (Vandenbroucke et al., 2011, Durme et al., 2008) of the discharge region. Mok and Kim (2011) tested a  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> packed dielectric barrier discharge reactor for the treatment of toluene in a sequential approach. The result shows that ozone is the only by-product besides CO and CO<sub>2</sub>.

As far as ozone removal is concerned, the combination with a biological treatment system can be an option. Wei et al. (2013) combined non-thermal plasma and a biological process to treat dimethyl sulfide, and found that ozone from non-thermal plasma makes the microbial community in the biotrickling more active for dimethyl sulfide removal.

### 3. Conclusions

The increasing public odour complaints and expectations and stricter environmental regulations have motivated the development of odour control technologies. Besides the performance of removing target contaminants of each technology, the potential pollutions and risks should be noticed. This work aims to point out the potential pollution and environment-health risks of the odour control processes, which one might not be aware of. Moreover, some solutions to the problems were discussed.

The contaminants adsorbed in activated carbon can be released into the atmosphere, and spent activated carbon can become hazardous wastes. Moreover, during the regeneration of the spent activated carbon, greenhouse gas and water pollution can be produced. The chemicals used in the chemical scrubber can lead to climate change, freshwater eutrophication, photochemical oxidant formation, human toxicity, and ecotoxicity. The thermal treatment consumes a lot of energy, produces secondary emissions such as nitrous and sulphur oxides, and causes photochemical reaction. It also may produce undesired by-products, such as Cl<sub>2</sub> and DLCs. The main potential problems of biological treatment include the emissions of bioaerosol and the acidification and eutrophication of water. The high toxicity by-product formed during the UV photodegradation should be considered carefully. The formation of O<sub>3</sub>, NO<sub>x</sub>, other VOCs, and aerosols during the non-thermal plasma should be paid attention to.

Process control and optimization are very important to solve these potential problems. Moreover, combinations of different technologies can be an attractive approach to solve the potential problems and improve the efficiency of odour control.

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