



Odorous gas emissions from sewage sludge composting windrows affected by the turning operation and associated health risks

Zhenhan Duan^{a,b,*}, Wenjing Lu^b, Muhammad Farooq Mustafa^{b,c}, Jianwei Du^a, Yong Wen^a

^a South China Institute of Environmental Science (SCIES), Ministry of Ecology and Environment (MEE), 510655, Guangdong, PR China

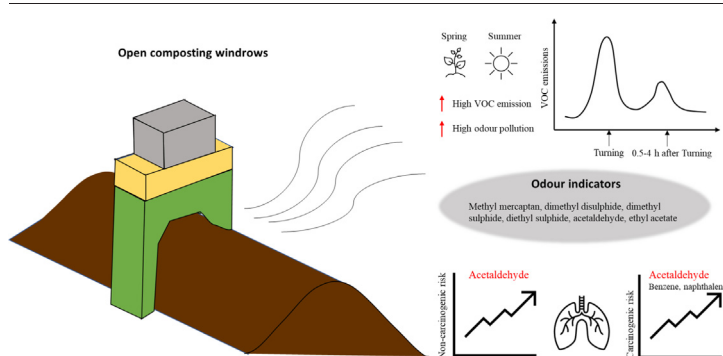
^b School of Environment, Tsinghua University, 10084 Beijing, PR China

^c Department of Environmental Design, Health and Nutritional Sciences, Allama Iqbal Open University, Islamabad, Pakistan

HIGHLIGHTS

- The highest odorous gas emissions occurred while turning composting windrows.
- Raw material mixing ratios and temperature affect odorous gas emissions.
- Sulphur and oxygenated compounds are major odour contributors.
- Acetaldehyde poses unacceptable health risks to on-site workers.

GRAPHICAL ABSTRACT



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ABSTRACT

The treatment and disposal of sewage sludge (SL) has long been a challenging task in China. Open windrow composting, coupled with mechanical turning, is preferred in small cities and rural areas, due to low costs and ease of operation. However, the emission of odorous volatile organic compounds (VOCs) from open composting windrows, as well as related health risks, has aroused strong protests from surrounding populations. This study investigated VOC emissions (including hydrogen sulphide) from five open SL composting windrows at a single site, before, during and after turning operations, and across different seasons. As expected, the highest VOC concentration ($6676 \mu\text{g m}^{-3}$) was measured while turning the windrows, whilst an additional emission peak was observed at all windrows at different times after turning, which was determined by the raw material mixing ratio (SL: woodchips), as well as ambient and windrow temperatures. In general, higher VOCs emissions and odour concentrations were measured in summer, and odour pollution was mainly caused by sulphur and oxygenated compounds, due to their high odour activity values (OAVs). Methyl mercaptan, dimethyl disulphide, dimethyl sulphide, diethyl sulphide, acetaldehyde and ethyl acetate were identified as the odour pollution indicators for the composting facility. The results from a health risk assessment showed that acetaldehyde was the most hazardous compound, with both non-carcinogenic and carcinogenic risks exceeding acceptable levels. The carcinogenic risks of benzene and naphthalene were also above acceptable levels; however, their risks were insignificant at the studied site due to the low concentrations.

1. Introduction

The growing amount of sewage sludge (SL) in China is generating increasing concerns, due to its adverse impacts on both the environment and human health (Yang et al., 2015; Zhu et al., 2016). In 2019, 39 million

* Corresponding author at: South China Institute of Environmental Science, No. 7, West Street, Tianhe District, Guangdong Province, PR China.
E-mail address: duanzhenhan@scies.org (Z. Duan).

tons of SL (80% water content) were produced (Wei et al., 2020). Historically, most of the generated sludge was not safely treated and disposed, with more than 80% improperly dumped (Yang et al., 2015). Under recent policy changes, the SL treatment and disposal market has grown rapidly, and aerobic composting, followed by land application, has become one of the most promising techniques currently available, due to the possibility of utilising sludge compost as an alternative soil amendment (Chen et al., 2012; Yildiz et al., 2016). In fact, aerobic composting is recommended by the Chinese Agricultural Ministry as the preferred SL disposal method and has been applied in various regions (Cao et al., 2011; Duan et al., 2017; Feng et al., 2015; Zhou et al., 2018). In small cities and rural areas, open windrow composting systems, coupled with mechanical turning, are often preferred, due to their low cost and minimal technical requirements. However, odour pollution and health risks caused by open windrow composting facilities have aroused strong protests from adjacent communities.

Gaseous emissions during the composting process can be divided into two groups: 1) inorganic compound (IC) emissions, which mainly include carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), hydrogen sulphide (H₂S) and ammonia (NH₃); 2) volatile organic compound (VOC) emissions, including organic sulphur compounds, oxygenated compounds (refer to O containing VOCs such as alcohols, esters and ethers, aldehydes, ketones, etc.), aliphatic hydrocarbons (alkanes and alkenes), aromatics, halogenated compounds, etc. (Dhamodharan et al., 2019; Smet et al., 1999; Zhu et al., 2016; Zigmontiene and Zuokaite, 2010). In this paper, we mainly target VOC emissions from SL composting windrows. However, H₂S is also included in this report, due to its important odour contribution. These compounds are termed collectively as VOCs.

VOC emissions from SL composting windrows are originated from two processes: 1) biodegradation of organic matter in the SL, which produces biogenic-originated compounds, 2) direct volatilisation of compounds – often xenobiotics – contained in SL (and the bulking agents) (Kissel et al., 1992; Zigmontiene and Zuokaite, 2010). Many studies investigated VOC emissions during the composting processes of SL under various conditions and evaluated the associated odour pollutions. Shen et al. (2012) measured the production rate of VOCs during SL composting to be 1.09 g C kg DM⁻¹, with peak production (444 mg kg DM⁻¹ d⁻¹) observed in the mesophilic phase, while VOC emissions were about two times lower than the production rate. However, high VOC emission rates were reported by other researchers (0.82–3.9 g kg DM⁻¹) (Cadena et al., 2009; Maulini-Duran et al., 2013), probably due to the different raw materials, composting techniques as well as the applied measurement methods, and several studies have found that more VOCs were emitted from SL composting windrows during the mesophilic stage (Komilis et al., 2004; Pagans et al., 2006; Shen et al., 2012). Terpenes, organic sulphur compounds, ketones and carboxylic acids were found to be the most abundant VOCs in the gaseous emissions, while NH₃ and H₂S usually dominated the total mass flowrate (Fisher et al., 2019; González et al., 2019; Maulini-Duran et al., 2013; Zhu et al., 2016). Among these compounds, NH₃, H₂S, methanethiol, dimethyl sulphide (DMS), dimethyl disulphide (DMDS), limonene and pinenes were frequently identified as key odour contributors due to their high concentrations and/or low odour thresholds (Blazy et al., 2015; Hort et al., 2009; Van Durme et al., 1992; Zhu et al., 2016), while others such as aromatics have gained attention as a result of their potential hazardous health effects (Durmugoglu et al., 2010; Mustafa et al., 2017; Zhu et al., 2016).

VOCs production and emissions from an open windrow composting system are affected by many factors, such as oxygen (O₂) content inside the windrow, turning frequency, inherited concentration of VOCs in raw sludge, adsorption, dissolution and the retention of VOCs in free air space (He et al., 2018, 2010). Gutiérrez et al. (2017) found that the maximum O₂ consumption rate or organic matter degradation during composting were directly related to the maximum odour emission rate, and He et al. (2018) revealed that high O₂ levels together with a high degree of biostability can decrease the emission rate of specified odorous VOCs from SL composting. The turning operation in open composting windrows, as a simple way of aeration, can provide O₂ for bacteria, mix composting

materials, enhance water reduction and control windrow temperatures, which could significantly affect VOCs emission from SL composting windrows (Nian et al., 2013; Zhou et al., 2018). In addition, during the turning process, large quantities of VOCs are released into ambient air, which can lead to serious odour pollution (Defoer and Van Langenhove, 2002). It is therefore essential to understand how VOC emission changes during the turning process of SL composting windrows, and to assess potential odour pollution issues and health risks. However, most previous studies focused on VOC and odour emissions throughout the entire composting process, and there was scant focus in the literature on VOC emission changes from open SL composting windrows as affected by the turning operation, with even fewer studies have evaluated the associated health impacts (Defoer and Van Langenhove, 2002; Zhao et al., 2015).

This paper aims to demonstrate the effect of turning operations on VOC emissions from open SL composting windrows and to evaluate associated odour pollution and health risks. A series of field campaigns was conducted at an SL composting facility in southeast China, and the VOC emissions from different composting windrows were measured before, during and after turning. Olfactometric and theoretical odour concentrations were measured and calculated for selected gas samples, and the most important odorous contributors were identified for each windrow. In addition, the carcinogenic and non-carcinogenic health risks caused by exposure to hazardous compounds were assessed for on-site workers. The results of this study will deepen understanding on VOC emission characteristics from SL composting windrows as affected by the turning operation and provide guidance on odour pollution and health risk control at SL composting facilities.

2. Materials and methods

2.1. Site description

Field sampling was conducted at an SL composting facility located in southeast China with a treatment capacity of 100 t/day. Dewatered SL (water content ~80%) was collected from a municipal wastewater treatment plant and transported to the composting facility for final disposal. The SL was first mixed with woodchips (crushed garden waste, 2–3 cm in length and 3 mm thick) in different ratios and then piled into large windrows measuring 100 m in length, 1.8 m in width and 1 m high (compositions of raw SL and woodchips are shown in Table 1). The composting process took place in covered sheds, to protect the windrows from rainfall, while the surroundings were open to the atmosphere (see Fig. S1 in the Supplementary Information (SI)). At optimal material mixing ratios, the temperature inside the windrows would increase quickly to above 40 °C within one or two days, and this temperature was maintained for 10–20 days while organic matter rapidly degraded. During this period, the windrows were turned daily with a compost turner to provide O₂ for

Table 1
Compositions of raw sewage sludge and woodchips.

Item ^a	Sewage sludge	Woodchips
Water (%)	82.9 ± 3.0	20.1 ± 6.7
VS (%TS)	59.6 ± 4.5	96.6 ± 2.0
C (%TS)	27.2 ± 0.3	37.3 ± 2.2
H (%TS)	4.8 ± 0.1	5.0 ± 0.2
N (%TS)	4.4 ± 0.1	1.6 ± 0.3
P (%TS)	1.26	0.06
K (%TS)	0.65	0.73
Cd (mg/kg)	17.2	Not detected
Cr (mg/kg)	56.2	27.9
Cu (mg/kg)	162	26.5
Ni (mg/kg)	27.7	21.7
Pb (mg/kg)	14.7	535
Zn (mg/kg)	629	93.5

^a The water content and VS were determined by weight method using a drying oven and a muffle furnace. Elements of C, H and N were analysed using an Elemental Analyser CE440 (Exeter Analytical, Inc., USA).

microbiological activities, and exhaust air was emitted directly into ambient air. The overall composting process lasted for approximately 30 days in summer and 50 days in winter until the windrow temperature dropped below 30 °C. The materials were then screened to recycle woodchips, and the SL raw compost was sent to the post-treatment unit for further maturation before it can be used for landscaping or as fertiliser (Fig. S2). During composting, changes of major gas concentrations (CH₄, CO₂ and O₂) within the windrows were regularly recorded using a biogas analyser (Geotech Biogas 5000, Shanghai Zhonglin Co., China) by inserting the sampling probe into the windrows to 10–20 cm below surface, and NH₃ concentration was measured in the same way with a portable multi-gas analyser (Dräger X-am 7000, Drägerwerk AG & Co., Germany) (Figs. S3–S8). The windrow temperatures were measured using a portable temperature detector (Sanywun, Shanghai, China). Three measurements were conducted each time at the beginning, middle and end of the windrows, and temperatures at different depth (20 cm, 40 cm, 60 cm and 80 cm) were recorded at each location. The average temperature was then calculated as the windrow temperature.

2.2. VOCs sample collection

Three sampling campaigns were conducted during winter, summer and spring across two years (average ambient temperatures between 2–8, 27–35 and 14–22 °C, respectively). Five windrows with different raw material mixing ratios were selected for gas sample collection (Table 2). All the samples were collected when the windrows had entered thermophilic stages for about one week (Figs. S3–S8), each with an average windrow temperature of above 45 °C, except for Pile B (Figs. S4, S6–S8). At Pile B, the composting process was not successfully initiated, due to high SL content, and it remained in the mesophilic stage throughout the entire composting cycle. The windrows were turned by a compost turner on all sampling days.

In the first two sampling campaigns, four samples were collected at different times, before and after turning (Table 2). Gas sampling was conducted using the “lung method,” i.e. an 8 L bioriented polyester bag (Sinodour co., Tianjin, China) was placed in an air-tight container with the opening connected to the lid using ¼ in. Stainless steel screws. A vacuum pump was connected to the container. When the pump started to work, the air inside the container was removed, creating a pressure difference between the container and the atmosphere, and ambient air was drawn into the sampling bag through a stainless-steel sampling tube coated with polytetrafluoroethylene (PTFE). The sampling flowrate was around 10 L min⁻¹, and each sampling took 40–50 s to fill the bags. Before sampling, the bags were flushed twice to remove any background concentrations. The sampling tube was placed at 5 cm above the top of the windrow, and it was moved several metres along the windrow during sampling to ensure proper mixing of the air. In addition, two background samples were collected outside the composting area (at factory boundary) during the first campaign for comparison purposes. The samples were stored in dark boxes and transported to the lab for chemical analysis within 24 h.

In the third campaign, a portable gas analyser was used (see Section 2.3), which allowed for on-site monitoring of odorous gas emissions during windrow turning. The sampling was repeated over three days, and four samples were taken on each of the first two days, i.e. during turning, 0.5 h, 2 h and 5 h after turning (Table 2). An additional sample was taken on the third day before turning for comparison.

2.3. Chemical analysis

VOC samples collected in the first two campaigns were analysed according to Compendium Method TO-15 (USEPA, 1999). Pre-concentration of the samples was conducted through a three-stage cold trap concentrator (Entech 7100, USA). The concentrated samples were then injected into a stationary gas chromatograph (GC, Agilent 7890A, USA)-mass spectrometry (MS, Agilent 5975C, USA) system for qualitative and quantitative analysis. A DB-5 ms column (60 m × 0.32 mm × 1.0 µm) was used with the following temperature setup: initial temperature 35 °C held for 10 min, 35 to 140 °C at 4 °C min⁻¹, 140 to 250 °C at 15 °C min⁻¹, 250 °C held at 5 min. Helium (99.999%) carrier gas flow in the analytical column was maintained at 1.5 mL min⁻¹. Electron impact spectra were obtained with electron energy of 70 eV, and mass spectral data were acquired over a mass range of 15–300 amu. The MS was operated in synchronous Selected ion monitoring (Sim)/Scan mode for qualitative and quantitative analysis. Identification of the VOCs was based on matching their retention times with standards and ratios between the quantifier and qualifier ions. The identified compounds were then quantified, using the internal standard method. Calibration of the GC–MS was conducted by using several standard mixtures in different concentrations (including certificated TO-15 and PAMS standard mixtures (Restek Corporation, Bellefonte, PA, USA), Sulphide Standard Gas, Trimethylamine standard gas, Low-grade fatty aldehyde standard gas and Terpene Standard Gas). The TO-14A Internal standard mixture was used for quantifying the samples. With the applied analytical method, up to 120 compounds can be quantified with method detection limits between 0.1 and 1 ppb. A list of the quantified compounds in this study – divided into seven chemical groups – can be found in Table S1.

In the third sampling campaign, a portable GC–MS (HAPSITE® ER, Inficon, East Syracuse, USA) was used, which enabled simultaneous on-site sampling and chemical analysis. Air samples (100 mL) were collected via a hand-held sampling probe, pre-concentrated in a concentrator and then injected into a non-polar column (100% polydimethylsiloxane, 15 m × 0.25 mm × 1.0 µm) for separation. The temperature program for the GC column was as follows: hold at 60 °C for 7 min, ramp up to 150 °C at 20 °C min⁻¹, then ramp up to 180 °C at 10 °C min⁻¹ and hold for 30 s. The mass spectrometer was operated in full scan mode within a mass range of 41–300 amu, and electron energy was 70 eV. Overall analysis time for one sample was 15 min. VOCs were identified by comparing the mass spectra with those of the US NIST (National Institute of Standards and Technology) 98 Library. The GC–MS was calibrated by using the TO-15 standard solution (Restek Corporation, Bellefonte, PA, USA). Fifty-four compounds were quantified with method detection limits ranging between 1 and 10 ppb (see Table S1).

Table 2
Sampling windrows and sample collection times.

Campaign	Season	Windrow	Material mixing ratio (SL: woodchip in wet weight)	Sample collection time ^a
#1	Winter	Pile A	3:1	0.5 h BT, 0.5 h AT, 4 h AT, 8 h AT
#1	Winter	Pile B	5:1	0.5 h BT, 0.5 h AT, 4 h AT, 8 h AT
#2	Summer	Pile C	3:1	0.5 h BT, 0.5 h AT, 4 h AT, 8 h AT
#2	Summer	Pile D	4:1	0.5 h BT, 0.5 h AT, 4 h AT, 8 h AT
#3	Spring	Pile E	3:1	DT, 0.5 h AT, 2 h AT, 5 h AT

^a BT: before turning, AT: after turning, DT: during turning.

2.4. Olfactometric odour concentration measurement and odour activity value (OAV) calculation

Odour concentrations of the samples collected in the first two sampling campaigns were determined by using the Triangle Odour Bag Method, according to the Chinese standard GB/T 14675–93. Briefly, the sample was diluted into different ratios in one of three odour-free bags, and six panellists were required to determine which bag was odorous. The dilution and identification procedure was repeated multiple times until the panellist gave a wrong answer, and the geometric mean of the dilution factors of the last correct answer was calculated as the individual threshold. The odour concentration of the sample was then determined as the geometric mean of individual thresholds, excluding the maximum and minimum individual thresholds. A more detailed description of the method can be found in Lu et al. (2015).

The odour contribution of individual compounds in one sample can be evaluated by using the odour activity value (OAV), which is defined as the ratio between the chemical concentration of one compound and its odour threshold (OT), as expressed in the following (Capelli et al., 2013; Wu et al., 2017):

$$OAV_i = \frac{CA_i}{OT_i} \quad (1)$$

$$SOAV = \sum_{i=1}^n OAV_i \quad (2)$$

where OAV_i is the odour activity value of compound i in one air sample ($\text{ou}_E \text{ m}^{-3}$), $SOAV$ is the odour activity value of the entire sample ($\text{ou}_E \text{ m}^{-3}$), CA_i is the chemical concentration of compound i in air ($\mu\text{g m}^{-3}$) and OT_i is the odour threshold of compound i ($\mu\text{g ou}_E^{-1}$). The odour threshold values are obtained from previous studies (Lu et al., 2015; Wu et al., 2017) and are summarised in Table S2.

The OAVs of samples collected in the third sampling campaign were not calculated, as most of the important odorants (e.g. sulphur, oxygenated compounds and terpenes) could not be detected by the portable GC–MS.

2.5. Health risk assessment

Inhalation is the most significant route of human exposure to airborne chemicals. Hence, the USEPA specific exposure methodology was used to assess chronic non-carcinogenic and carcinogenic effects (USEPA, 2009). The time-weighted exposure concentrations were calculated by using the following equation:

$$EC_i = \frac{CA_i \times ET \times EF \times ED}{AT \times 365 \text{ days/year} \times 24 \text{ hours/day}} \quad (3)$$

where EC_i is the exposure concentration of compound i ($\mu\text{g m}^{-3}$), CA_i is the chemical concentration of compound i in air ($\mu\text{g m}^{-3}$), ET is the daily exposure time (4 h d^{-1} for on-site workers), EF is exposure frequency (335 d y^{-1} , considering 30 days of annual vacation), ED is exposure duration (which is assumed to be 20 years) and AT is the averaging time, i.e. 20 years for non-carcinogenic effects and 70 years for carcinogenic effects (USEPA, 2011; Wu et al., 2018).

The non-carcinogenic and carcinogenic risks were calculated by:

$$R = \sum_{i=1}^n R_i = \sum_{i=1}^n (EC_i \times IUR_i) \quad (4)$$

$$HI = \sum_{i=1}^n HI_i = \sum_{i=1}^n \left(\frac{EC_i}{RfC_i \times 1000 \mu\text{g/mg}} \right) \quad (5)$$

where R is the cumulative cancer risk of all targeted compounds, HI is the cumulative hazard index for all targeted compounds, HI_i and R_i are the corresponding hazard index and cancer risk of compound i , EC_i is the calculated exposure concentration of compound i ($\mu\text{g m}^{-3}$), RfC_i is the

reference concentration of compound i (mg m^{-3}) and IUR_i is the Inhalation Unit Risk ($\mu\text{g m}^{-3}$)⁻¹ of compound i . RfC and IUR values for each compound were obtained from the IRIS (Integrated Risk Information System) and RAIS (Risk Assessment Information Systems) databases. $HI_i > 1$ and $R_i > 1 \times 10^{-6}$ are considered unacceptable risks.

Average VOC concentrations measured before and after turning (0.5 h BT, 5 h AT (for pile E) and 8 h AT) at all five windrows were used for the calculation of CA_i , as these concentrations were close to the background concentrations measured at the factory boundary and were judged suitable to represent long-term exposure for on-site workers. Concentrations measured during turning (DT), 0.5 h and 4 h after turning were significantly higher than the background concentrations and were considered as short-term exposures. Hence, they were not included in the calculation of CA_i . Instead, these concentrations were compared to the limit values in the Chinese standard *Occupational exposure limits for hazardous agents in the work place – Part 1: Chemical hazardous agents* (GBZ 2.1–2019), to assess if they exceeded the acceptable ranges of short-term exposure limits.

3. Results and discussion

3.1. VOC emissions before, during and after the turning operation, and affecting factors

Total VOC concentrations varied significantly between samples. The highest concentration ($6676 \mu\text{g m}^{-3}$) was measured during turning at Pile E, while the lowest concentration ($308 \mu\text{g m}^{-3}$) appeared at 0.5 h before turning at Pile B, which was slightly higher than the background concentrations measured outside the composting area (251 and $269 \mu\text{g m}^{-3}$) (Fig. 1). It should be noted that the VOC concentrations in the third campaign were measured by the portable GC–MS, which detected much less compounds than the stationary GC–MS used in the other two campaigns (Table S1). Hence, the actual VOC emissions from Pile E might be higher than the reported values. However, the general trend shown in Fig. 1 still strongly suggested that highest VOC emissions would appear during windrow turning, and higher VOC emissions were found in spring and summer when the ambient temperatures were higher.

The change in VOC concentrations before and after turning differed between windrows with different raw material mixing ratios (SL: woodchip in wet weight). At Piles A, C and E, each with a material mixing ratio of 3:1, an emission peak was observed at 0.5 h after turning (0.5 h AT), while at Piles B and D, with higher material mixing ratios (5:1 and 4:1), this emission peak appeared at 4 h after turning (4 h AT) (Fig. 1). The production and emission of VOCs at a composting windrow is governed by the O_2 level as well as microbial activities (Shen et al., 2012). The material mixing ratio affects VOC emissions from SL composting windrows by restricting O_2 content as well as (gaseous) mass transfer inside the windrows. Piles A, C and E had higher porosity than the other windrows (based on our visual inspection, these windrows were less compacted and the materials were visually more porous), thereby allowing more O_2 to enter the windrow after turning – and thus boosting the aerobic decomposition of organic matter and the production of VOCs (Zhang et al., 2013). Also, the high gas permeability and good ventilation conditions in these windrows mean that fewer VOCs were accumulated inside the windrow before the next turning operation took place (Gutiérrez et al., 2017; Zhu et al., 2016). This could explain the emission peak at 0.5 h after turning in Piles A, C and E, in that VOCs produced after turning were quickly released from the windrows. In contrast, Piles B and D were more compacted, and so gas convection from the windrows into ambient air was limited. Most of the produced VOCs would have been trapped inside the windrow and released slowly into the atmosphere (Zhou et al., 2018), thus leading to a delayed emission peak after turning.

Temperatures can also affect the production and emission of VOCs. High ambient temperatures could help maintain a windrow's internal temperature as a result of less heat exchange between the windrow surface and the atmosphere (Zhou et al., 2018), while high windrow temperatures in the thermophilic composting stage prompted the microbial degradation of organic matter and thus producing more VOCs as intermediates (such

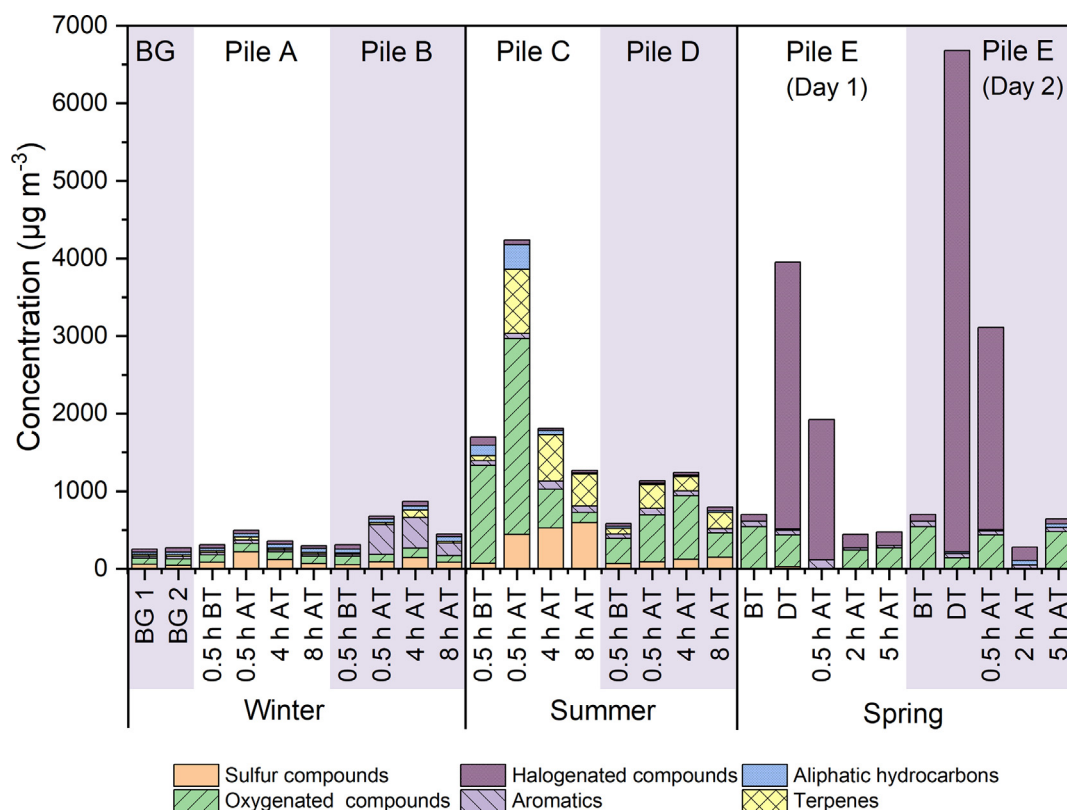


Fig. 1. Total VOC concentrations measured from the three sampling campaigns. BG: background concentration, BT: before turning, DT: during turning operation, AT: after turning. Note: the BT concentration at Pile E was measured on Day 3, before turning, and does not reflect the actual concentrations before turning on Day 1 and Day 2. It is included in this figure to provide a reference value for the first two days.

as ketones, alcohols, acids, etc.). High temperatures could also accelerate the volatilisation of inherent VOCs in the raw material and increase emissions, which could explain the high VOC emissions in spring and summer

when both ambient and windrow temperatures were higher (Fig. 2). It should be mentioned that the windrow temperatures at Pile B were below 30°C during the entire composting period (Fig. 2 and Fig. S6), indicating

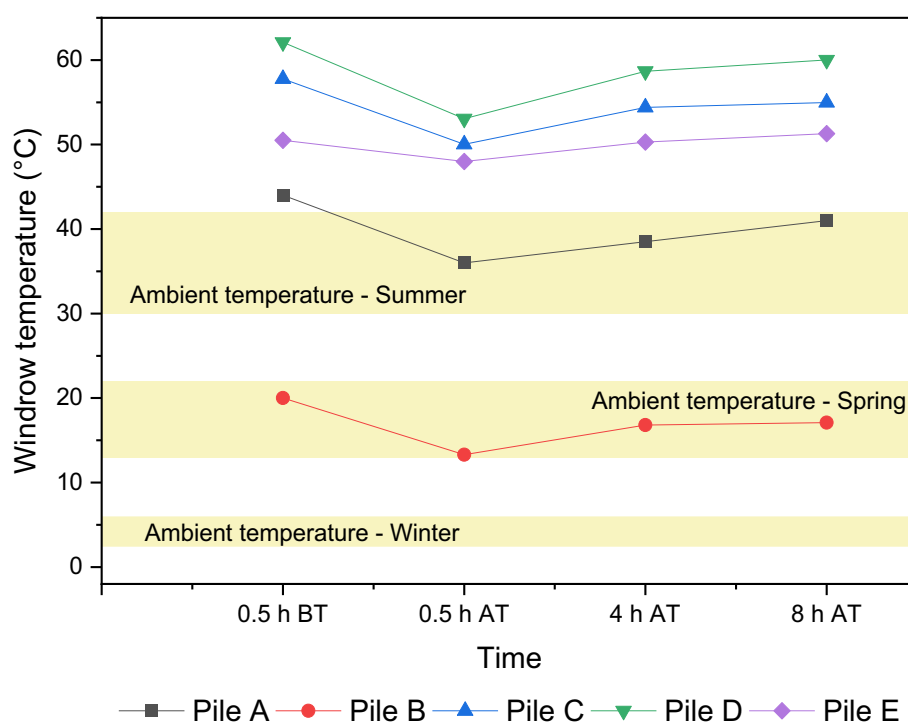


Fig. 2. Ambient and windrow temperatures before and after turning on the measurement days. BT: before turning, AT: after turning.

that this windrow was in the mesophilic stage, during which more VOCs were likely to have been produced (Kumar et al., 2011; Shen et al., 2012). The anaerobic condition formed inside this windrow due to high SL content (as indicated by the high CH_4 concentration in Fig. S6) also contributed to the relatively high VOC emissions from Pile B.

In total, 91 compounds were identified in the samples, including six sulphur compounds, 10 oxygenated compounds, 18 aromatics, three terpenes, 30 aliphatic hydrocarbons and 25 halogenated compounds. The concentration of individual compounds measured at different windrows are reported in Tables S3–S5, while cumulative concentrations of each chemical group are summarised in Table S6. Oxygenated compounds dominated the total VOC concentrations in Piles A, C and D, followed by sulphur compounds and terpenes, while noticeable high concentrations of aromatics and halogenated compounds were measured at Pile B and Pile E. Oxygenated compounds are typical intermediates of incomplete aerobic degradation (Smet et al., 1999), and their increased concentrations at all windrows after turning were a result of increased O_2 content inside them. Sulphur compounds are normally produced under anaerobic conditions, due to insufficient aeration (Chen et al., 2011; Homans and Fischer, 1992), and their presence in all samples indicated that anaerobic zones were developed in the windrows both before and after turning. The high concentration of sulphur compounds in summer was likely a result of rapid O_2 consumption, due to accelerated microbial activities under high temperatures. Abundant terpenes were only measured during summer, which were probably released from the degradation of woodchips in warm temperatures (Van Durme et al., 1992). Halogenated compounds and most aromatics are xenobiotic and are inherent in raw materials, which were released from the composting windrows through direct volatilisation (Komilis et al., 2004; Zhang et al., 2012). Aromatics can also be produced during the anaerobic degradation of lignin (Wilber and Murray, 1990); however, this amount was considered to be small due to the aerobic conditions in the composting windrows as well as the slow degradation rate of lignin (mostly in woodchips). Hence, the emissions of halogenated compounds and aromatics are mainly affected by the raw material mixing ratio and windrow temperatures. The high concentrations of aromatics and halogenated compounds at Piles B and E probably indicated a high content of these compounds in the raw sludge. Table S7 lists the most concentrated VOCs at each windrow, and the

compounds that were measured in most samples with high concentrations were identified as being the most concentrated VOCs at the composting facility, i.e. ethanol, dimethyl disulphide, dimethyl sulphide, dichloromethane, α -pinene, β -pinene, limonene, toluene and acetone.

3.2. Odour concentration and major odour contributors

Fig. 3 shows the odour concentrations measured by olfactometric tests, and the odour activity values (OAVs) calculated from chemical concentrations for the first two campaigns. Higher odour concentrations were generally measured in summer, which corresponded well with the high VOC concentrations reported in Section 3.1, although no statistical correlation was found between odour concentration and total VOC concentrations (data not shown). The highest odour concentration measured at Pile A, before turning (winter), was probably caused by high NH_3 emissions from the windrow (NH_3 concentration was 226 ppm at 20 cm below the windrow surface, see Fig. S3). However, NH_3 concentration in the gas samples was not measured, due to limitations of the applied analytical method. The change of odour concentration before and after turning also differed in winter and summer. In winter, odour concentration at the surface of the composting windrows decreased over time after turning, while the opposite was observed in summer (Fig. 3). This could be explained by the temperature difference between the composting windrow and ambient air in the two seasons. In winter, the large temperature difference between the windrow surface and ambient air enhanced convective gas flow from the composting windrow and accelerated the dispersion of odorous VOCs in ambient air (Fig. 2). Consequently, odour concentration decreased quickly after turning, until reaching background concentration levels. In summer, gas convection from the composting windrows and dispersion into ambient air, driven by temperature differences, was less pronounced, thus leading to the accumulation of odour over time after turning. It could also be expected that the highest odour concentration would appear during the turning of windrows, as already found by Defoer and Van Langenhove (2002). However, the lack of gas samples during turning does not support a solid conclusion from this study.

The change in SOAV before and after turning was inconsistent with the olfactometric results and should not be used to assess odour pollution issues

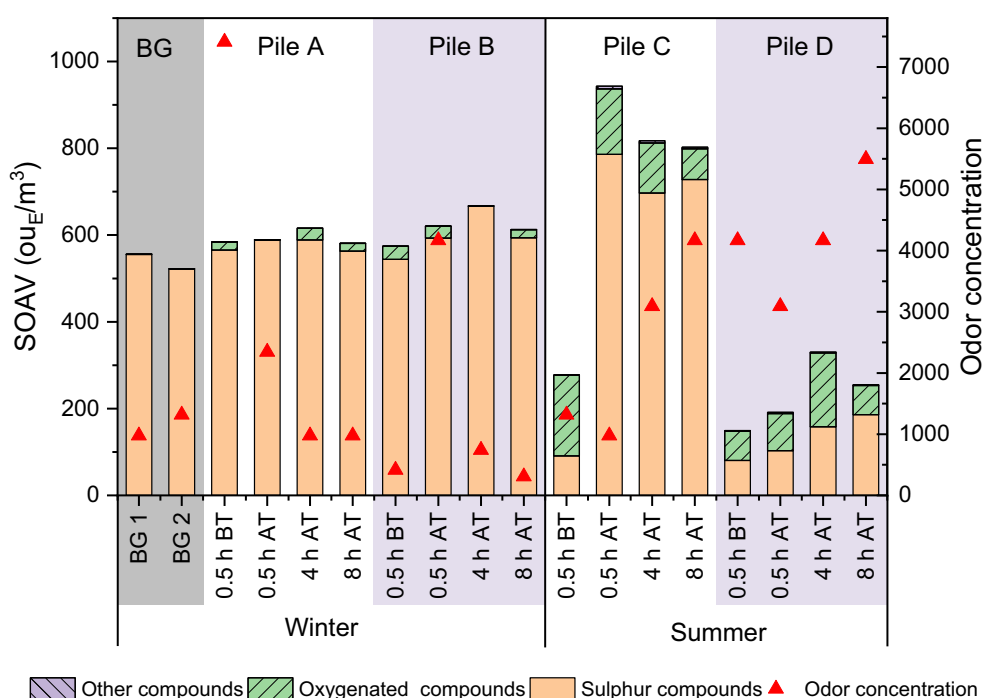


Fig. 3. Measured odour concentration and calculated odour activity values (SOAVs) for each sample from the first two sampling campaigns. BG: background concentration, BT: before turning, AT: after turning. Odour concentration at 4 h AT from Pile A is missing, due to technical problems during analysis.

(Fig. 3). This discrepancy might attribute to several reasons: 1) the chemical analysis by GC–MS only covered limited numbers of compounds, and some important odorous gases such as NH_3 – which often appeared in high concentrations – and hexanol (OT value 0.28 ppb) cannot be detected by the applied analytical method, so they were not included in the SOAVs calculation. However, their contributions to the odour concentration might be significant. 2) The OT values used in calculating OAVs were obtained from different studies (USEPA, 1992; Wu et al., 2017; Yoshio and Nagata, 2003). As different OT values were reported in these studies even for one compound (probably due to the use of different olfactometric measurement methods), the choice of database in the calculation can also affect the calculated SOAVs. 3) There might be synergy and mask effects between different odorous compounds, which cannot be estimated by the SOAVs (Kim, 2011; Ruth, 1986). Thus, the simple sum up of OAV of individual compounds does not reflect the real odour concentration sensed by humans (olfactometric results). Similar poor correlation of SOAV and chemical concentrations had also been found in other studies (Blazy et al., 2015; González et al., 2019). Nevertheless, the calculation of OAVs for individual compounds enables the identification of major odour contributors at each windrow as affected by the turning operation (Capelli et al., 2013). As evident in Fig. 3, sulphur compounds constituted the most important odorous group in almost all samples, and they were more dominant in winter, while oxygenated compounds contributed a larger portion to the total odour concentrations in summer. The major odour contributors were recognised for each composting windrow by calculating the average OAV of each compound in gas samples collected from the same windrow, the results for which are presented in Table 3. It seems that the most odorous compounds were highly consistent at different windrows, and all the sulphur compounds, except for carbon disulphide, were found to be the most important odorants. These compounds were produced from bacterial sulphate reduction as well as decomposition of sulphur-containing organic matter under anaerobic conditions (He et al., 2018; Zhang et al., 2016). Since only limited aeration was provided to the windrows through turning, anaerobic environments were easily developed within the windrows (as also indicated by the presence of CH_4 , see Figs. S5, S7–S8), which favoured the production of volatile sulphur compounds. Given the low odour thresholds of these compounds (Table S2), volatile sulphur compounds became the most important odour contributors at the composting facility, as was also found in many other studies (Blazy et al., 2015; Fisher et al., 2019; He et al., 2018; Hort et al., 2009; Zhu et al., 2016).

Compounds with an OAV > 1 were compiled for all gas samples, and those measured in 80% of cases were identified as the odour pollution indicators for the studied composting facility, i.e. methyl mercaptan, dimethyl disulphide, dimethyl sulphide, diethyl sulphide, acetaldehyde and ethyl acetate. Among them, methyl mercaptan, dimethyl sulphide and dimethyl disulphide are regulated odorous compounds in the Chinese standard *Emission standards for odour pollutants* (GB 14554–1993). Although the concentrations of these three compounds were below the regulated limits in this study, odour concentration significantly exceeded the limit values at the composting facility, thereby indicating the potential of odour pollution affecting adjacent communities, especially during warm seasons. Hence, odour control measures must be established at the composting facility,

such as sealing the composting area for gas emission control and using forced aeration at the composting windrows (to reduce the production of sulphur compounds).

3.3. Health risks

Peak VOC concentrations measured while turning the composting windrows were compared to the occupational exposure limits (OELs) in the Chinese standard GBZ 2.1–2019. None of the measured concentrations exceeded the maximum allowable concentrations (MACs, defined as concentrations that should never be exceeded at any time or any workplace on a workday). However, the ambient concentration of NH_3 during windrow turning, albeit not continuously measured, sometimes exceeded the permissible concentration – short-term limits (PC-STEL, defined as concentrations allowable for short-term exposures (<15 min) at a workplace under the condition that the average exposure concentration does not exceed the time-weighted average permissible concentration (8 h per day, 40 h per week)), indicating that exposure to ambient air during turning for more than 15 min might lead to irritation to the eyes and the upper respiratory tract.

Nineteen compounds were selected for the calculation of non-carcinogenic risks, based on their hazardous characteristics. The calculated risks are shown in Table 4. Only one compound, acetaldehyde, showed unacceptable risks to humans, with a hazard index (HI) of 3.13 – mainly due to its high concentration in the summer samples (Table S4), while the rest all showed minor or negligible risks and were not even considered as “likely concerns” (HI < 0.5) (Mustafa et al., 2017). Previous studies have identified acetaldehyde as one of the main odorants in some waste management facilities (Fang et al., 2012; Lim et al., 2018); however, its health risk has rarely been documented. Long-term exposure to acetaldehyde might lead to changes in the nasal mucosa and trachea, growth retardation, slight anaemia and increased kidney weight (USEPA, 2000). The high HI calculated in this study suggests that there is an elevated risk for on-site workers of contracting a chronic disease, due to exposure to acetaldehyde. Nevertheless, the high concentrations of acetaldehyde were only detected in several samples (mostly in summer), thus suggesting that the adverse health effects caused by this compound might vary seasonally. The cumulative HI of the selected VOCs also exceeded 1, thus indicating elevated non-carcinogenic risks for on-site workers at the composting facility.

Five compounds were included in the carcinogenic risk calculation (Table 5). The R values ranged from 1.39E-09 to 1.77E-05, with three compounds (acetaldehyde, benzene and naphthalene) exceeding the acceptable carcinogenic risk level ($R = 1\text{E-}06$). Acetaldehyde is classified in group B2 by the USEPA as a probable human carcinogen, as it might increase the risk of nasal tumours and laryngeal tumours (USEPA, 2000). The carcinogenic risk of acetaldehyde was 1.77E-05, and according to Sexton et al. (2007), this can be labelled as a “probable risk” (R between 1.0E-05 and 1.0E-04). Benzene and naphthalene have often been recognised as health indicators at waste management facilities, due to their potential risks (Durmugoglu et al., 2010; Moolla et al., 2013; Zhu et al., 2016), and they have also been found to pose both non-carcinogenic and carcinogenic risks in previous studies (Liu et al., 2016; Mustafa et al., 2017;

Table 3
Major odour contributors in each windrow, and their corresponding odour activity values (OAV > 1).

NO.	Pile A		Pile B		Pile C		Pile D	
	Compound	OAV	Compound	OAV	Compound	OAV	Compound	OAV
1	Diethyl sulphide	523	Diethyl sulphide	526	Dimethyl sulphide	292	Dimethyl sulphide	101
2	Dimethyl sulphide	36	Dimethyl sulphide	55	Methyl mercaptan	265	Ethyl acetate	86
3	Ethyl acetate	16	Ethyl acetate	19	Ethyl acetate	104	Methyl mercaptan	29
4	Methyl mercaptan	10	Methyl mercaptan	10	Acetaldehyde	27	Acetaldehyde	12
5	Dimethyl disulphide	8	Dimethyl disulphide	4	Dimethyl disulphide	13	Dimethyl disulphide	3
6			Hydrogen sulphide	4	Hydrogen sulphide	7		
7					α -pinene	2		

Table 4

Non-carcinogenic risks of selected VOCs through inhalation at the composting facility.

Compound	EC ($\mu\text{g m}^{-3}$)	RfC (mg m^{-3})	HI
Acetaldehyde	28.16	0.009 ^a	3.13E+00
Acetone	20.09	30.9 ^b	6.50E-04
Ethyl acetate	1.65	0.07 ^b	2.35E-02
hydrogen sulphide	0.30	0.002 ^a	1.48E-01
Carbon disulphide	0.70	0.7 ^a	1.01E-03
Benzene	0.59	0.03 ^a	1.98E-02
Toluene	3.74	5 ^a	7.49E-04
Ethylbenzene	0.80	1 ^a	8.05E-04
m-xylene	0.50	0.1 ^a	5.03E-03
p-xylene	0.96	0.1 ^a	9.62E-03
o-xylene	0.51	0.1 ^a	5.06E-03
Naphthalene	1.00	0.003 ^a	3.34E-01
Pentane	1.48	1 ^b	1.48E-03
Propylene	0.93	3 ^b	3.09E-04
methyl chloride	0.49	0.09 ^a	5.41E-03
Ethyl Chloride	7.67	10 ^a	7.67E-04
Dichloromethane	4.16	0.6 ^a	6.93E-03
Dichlorodifluoromethane	0.66	0.1 ^b	6.62E-03
Trichloromonofluoromethane	0.18	0.7 ^b	2.60E-04
Cumulative HI			3.70E+00

^a Obtained from the IRIS (Integrated Risk Information System) database.

^b Obtained from the RAIS (Risk Assessment Information Systems) database.

Wu et al., 2018). In this study, benzene and naphthalene are classified as “possible risks,” since their R values were between 1.0E-06 and 1.0E-05 (Sexton et al., 2007). Hence, they do not pose significant health risks to workers at the composting facility. The R values of ethylbenzene and methyl chloride were 1–3 orders of magnitude below the regulated acceptable level, indicating that their risks to workers at the composting facility were minimal.

It should be noted that the calculated risks were likely conservative, as the peak concentrations during turning were not included in the exposure concentration calculation, and the actual time-averaged exposure concentrations might be higher. In addition, if several windrows are turned at the same time (which is often the case), ambient VOCs concentrations will be higher than those measured in this study, thus leading to increased odour pollution and health risks. To reduce these health risks, protective measures should be established for on-site workers, such as installing air filters in the pressurised cab of the turning machine, wearing masks and protective suits, etc. In addition, as the high exposure concentrations were mostly measured in summer, shortening working hours in this season could also reduce the potential health risks.

4. Conclusions

Aerobic composting is becoming increasingly important as a sewage sludge (SL) disposal technology in China. This study investigated odorous volatile organic compound (VOC) emissions from open SL composting windrows and explored how the emissions were affected by the turning operation. Several field campaigns were conducted at an SL composting facility in southeast China, and gas samples were collected from different windrows before, during and after windrow turning. It was found that the

Table 5

Carcinogenic risks of selected VOCs through inhalation at the composting facility.

Compound	EC ($\mu\text{g m}^{-3}$)	IUR ($\mu\text{g m}^{-3}$) ⁻¹	R
Acetaldehyde	8.05	2.20E-06 ^a	1.77E-05
Benzene	0.17	7.80E-06 ^a	1.32E-06
Ethylbenzene	0.23	2.50E-06 ^b	5.75E-07
Naphthalene	0.29	3.40E-05 ^b	9.75E-06
methyl chloride	0.14	1.00E-08 ^b	1.39E-09
Cumulative R			2.94E-05

^a Obtained from the IRIS (Integrated Risk Information System) database.

^b Obtained from the RAIS (Risk Assessment Information Systems) database.

highest VOC emissions appeared during the turning of windrows, and the high emission would continue for several hours after turning. The raw material mixing ratio (SL: woodchips in this study) could affect the change in VOC emissions from individual windrows before and after turning. At windrows with higher raw material mixing ratios, a delayed VOC emission peak would appear several hours after turning, due to VOC accumulation inside the windrows. In addition, higher VOC emissions were measured in spring and summer, when the ambient and windrow temperatures were higher. The chemical composition of emitted VOCs also varied between different windrows. Oxygenated compounds dominated VOC emissions from windrows with low material mixing ratios, followed by sulphur compounds and terpenes, while aromatics and halogenated compounds contributed more to total VOC concentrations at windrows with higher SL contents. Ethanol, dimethyl disulphide, dimethyl sulphide, dichloromethane, α -pinene, β -pinene, limonene, toluene and acetone were identified as the most concentrated compounds at the composting facility.

Odour concentrations were generally higher in summer under high temperatures, and it was expected that the highest odour pollution would occur during windrow turning. However, odour concentration during turning was not measured, due to a lack of gas samples. Based on the calculated odour activity values (OAVs), sulphur and oxygenated compounds were found to contribute most to total odour concentration, and methyl mercaptan, dimethyl disulphide, dimethyl sulphide, diethyl sulphide, acetaldehyde and ethyl acetate were selected as the odour pollution indicators for the composting facility. Regarding health risks, acetaldehyde was found to pose both non-carcinogenic and carcinogenic risks to the workers, with HI and R values exceeded acceptable levels, while benzene and naphthalene were identified as “possible risks” but were not considered to pose significant carcinogenic risks to humans. To control odour pollution and reduce the health risks for on-site workers, it is suggested that exhaust gas collection systems should be installed at the composting facility, and protective measures as well as an adjusted working schedule should be established.

CRediT authorship contribution statement

Zhenhan Duan: Investigation, Methodology, Data curation, Writing – original draft. **Wenjing Lu:** Supervision, Writing – review & editing. **Muhammad Farooq Mustafa:** Investigation, Methodology, Data curation. **Jianwei Du:** Supervision, Writing – review & editing. **Yong Wen:** Supervision, Writing – review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

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