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Research Paper

# Quantification of – And determining factors affecting – Methane emissions from composting plants

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ABSTRACT

Methane is a potent greenhouse gas contributing to climate change. Reliable data for methane emissions from the waste management sector are paramount in terms of providing national methane budgets and developing climate mitigation efforts. This study quantified total methane emissions and characterised temporal as well as operational emission patterns at five commercial composting plants in Denmark. Methane emissions were measured over a one-year period, using the tracer gas dispersion method. The results show that methane emission rates ranged from 8.0 ± 0.1 to 42.5 ± 1.5 kg $CH_4$ $h^{-1}$ and were significantly affected by factors including the type of feedstock and composting technology, treated feedstock mass, operational patterns and season. The results indicate that the highest methane emission factors were obtained at the combined anaerobic digestion and open windrow composting plant (4.51–5.21 kg $CH_4$ $Mg^{-1}$ wet garden/park waste (GPW) and food waste), followed by open windrow plants co-composting GPW, sewage sludge and straw (3.49–3.76 kg $CH_4$ $Mg^{-1}$ wet feedstock). The lowest methane emission factors were found at open windrow composting plants treating GPW (1.56–3.24 kg $CH_4$ $Mg^{-1}$ wet feedstock). Emissions tended to be higher when measurements were performed during working hours, in comparison to when they were measured after the plant closed for the day. At one plant, emissions were measured monthly over one year, and emissions were about 50% higher in spring and summer in comparison to autumn and winter.

## 1. Introduction

Bio-waste is defined as biodegradable garden and park waste, food and kitchen waste from households, restaurants, caterers and retail premises and comparable waste from food-processing plants (EC – European Commission, 2008). It accounts for a large proportion of solid waste occurring in municipal solid waste (MSW), and it constitutes about 34% of MSW in Europe (EEA, 2020). In 2017, about 43% of bio-waste was collected separately within the EU-28, amounting to 71 million tons, 64% of which was composted (EEA, 2020). The remaining collected bio-waste was treated by anaerobic digestion in combination with (30%) or without (10%) composting (EEA, 2020). These figures suggest that about 94 million tons of bio-waste was collected, mixed with other waste, and then incinerated or landfilled. Therefore, predictions suggest that an additional 60 million tons (assuming a sorting efficiency of 80%) will be realised in the future, when the separate collection of bio-waste will be mandatory in the EU. Consequently, a continuous increase in manufactured compost is expected, which may give rise to an increase in the numbers of composting plants.

In Denmark, the bio-waste generation rate has steadily risen from 67 kg $capita^{-1}$ $year^{-1}$ in 1994 to 268 kg $capita^{-1}$ $year^{-1}$ in 2019, mainly due to an increase in garden waste and the separate collection of household organic waste (Danish EPA, 2020). Moreover, bio-waste also makes up the predominant fraction of household waste (49%) (Edjabou et al., 2015). Additionally, bio-waste consisting of garden and park waste (GPW) remains one of the largest contributors (about 25% from 2008 to 2016) to MSW collected at recycling centres in Denmark (Edjabou et al., 2019), where bio-waste landfilling was banned in 1997. A more recent national resource strategy urges the diversion of bio-waste away from incineration. In this regard, newly revised Waste Framework Directives set legally binding targets for all EU countries, including the implementation of a separate collection system for bio-waste by 2023. To achieve this target, Danish municipalities are introducing bio-waste source-sorting, resulting in increasing amounts of collected bio-waste as well as considerable amounts of sludge from wastewater treatment plants, which is also composted.

In 2017, 150 Danish composting facilities treated only garden and park waste (GPW), nine facilities treated food waste mixed with GPW or

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other organic waste and 10 facilities treated GPW mixed with sludge and/or "other organic waste" (Nielsen et al., 2022). Of these facilities, 92% exclusively operated windrow composting, i.e., whereby the material is laid out in triangular piles with access to natural air only. Composting is a simple process in Denmark, thereby implying that temperature, moisture and aeration are neither consistently controlled nor regulated (Nielsen et al., 2022).

Composting stabilises heterogeneous organic material in aerobic conditions to generate a "humus-like product," which is traditionally applied as a soil conditioner and fertiliser. The application of compost on agricultural land potentially substitutes for the use of conventional mineral phosphorous and nitrogen fertilisers and the use of peat to improve soil texture. Recently, innovative technology has been employed to apply compost in bio-covers in order to mitigate greenhouse gas (GHG) emissions from landfills (Duan et al., 2022; Scheutz et al., 2022, 2014, 2011).

During composting, a large fraction of the degradable organic carbon in waste material is converted into carbon dioxide. Even though windrows are occasionally turned to support aeration, anaerobic pockets in the compost pile are inevitable and can then produce and emit methane (Andersen et al., 2010a; Boldrin et al., 2009). The trapped methane is generally vented through narrow pores in the top of the windrow pile, due to a chimney effect caused by temperature development in the centre of the material (Andersen et al., 2010a). Moreover, compost turning substantially increases methane emissions (Jensen et al., 2017; Hrad et al., 2014), as methane trapped inside the material is released. This generation and emission pattern prevents the use of local- and small-scale gas emission measurement techniques, including closed and open, static or dynamic chambers (Andersen et al., 2010a). For example, one of the main limitations of these chambers is that they cannot measure total emissions from a composting plant, due to high temporal and spatial variations in gas emissions (Sánchez et al., 2015; Andersen et al., 2010a). In addition, many small-scale measurement strategies are intrusive and affect forces driving the emissions during the measurement campaign. Previous studies applying static surface flux chambers to measure gas transportation from the composted material into the air have shown that this method significantly underestimates emissions (Andersen et al., 2010b). It is therefore questionable if emission rates obtained using small-scale measurement techniques are representative of actual emissions produced by full-scale composting plants.

In the last decade, ground-based remote-sensing techniques have been applied to alleviate the abovementioned limitations. An interesting measurement technique is the tracer gas dispersion method (TDM), combining the controlled release of tracer gas at the source with mobile plume concentration analysis. The method has been shown to provide accurate and robust results (Delre et al., 2018; Fredenslund et al., 2019; Feitz et al., 2018). Andersen et al. (2010a), for instance, applied TDM by using Fourier transform infra-red (FTIR) absorption spectroscopy to quantify methane and nitrous oxide emission rates from a windrow composting facility and measured temporal changes in compost pore gas composition and temperature. Furthermore, Aghdam et al. (2018) and Jensen et al. (2017) applied TDM by using cavity ring down spectroscopy (CRDS) to quantify methane emissions from composting plants in Denmark and Germany, respectively. Additionally, Hrad et al. (2014) applied the inverse dispersion technique in conjunction with open-path tuneable-diode-laser-spectroscopy (OP- TDLS) and meteorological measurements to determine methane emissions resulting from composting bio-waste in windrows.

Although bio-waste generation follows seasonal patterns (Edjabou et al., 2019), none of these studies investigated the influence of temporal variations on methane emissions. In a recent study, Sintana and Whendee (2019) considered total methane emissions from February to September, applying a micrometeorological mass-balance method to estimate total methane emissions from a full-scale composting plant. Although this study covered a longer period, the method requires high statistical and programming skills to compute total emissions; moreover, its reliability highly depends on the choice of statistical model. Additionally, previously published studies on compost methane emissions have tended to focus primarily on developing and testing measurement methodologies and applying these at individual composting facilities. To date, reported studies have covered neither temporal variations nor the influence of weather conditions on methane emissions from composting plants. For these reasons, several studies have pointed out the need to estimate greenhouse gas (GHG) emissions from composting plants, taking temporal changes into consideration and including changes in feedstock composition as well as operational conditions (Cerda et al., 2018; Sánchez et al., 2015; Nordahl et al., 2023). Accurate methane emission data from composting is paramount for (i) national emission inventory reporting, (ii) the validation of existing GHG emission models, (iii) documenting solutions and initiatives to decrease methane emissions and (iv) improving databases used for the life cycle assessment of composting plants. Overall, very few data on methane emissions from composting plants treating garden waste and food waste are available. A recent review, however, included methane emission data from 10 field studies focusing on composting food waste and five focusing on garden waste (Nordahl et al., 2023). In only two of these 15 field studies were emissions measured using non-intrusive measurement techniques.

The overall objective of this study was to quantify methane emissions as well as the impact of temporal variations and weather conditions on methane emissions from full-scale operational composting plants in Denmark. Methane emissions were quantified at five plants, by applying the TDM. In addition, methane emissions measured during normal working hours and when on-site activities had stopped (after the plant closed for the day) were compared. At one unit, methane emissions were measured over a whole year. Finally, historical methane emission data from previous campaigns performed at eight Danish composting facilities, using the same measurement method (TDM), were compiled and included in this study, in order to provide a solid overview.

## 2. Materials and methods

### 2.1. Overview of the selected composting facilities

Table 1 provides an overview of the five selected full-scale commercial composting plants. Their characteristics suggest that they represent composting plants in Denmark as described in Petersen and Hansen (2003). The feedstock for these plants comprised garden and park waste (GPW), garden and park waste and food waste (GPW and FW) as well as garden and park waste mixed with sewage sludge and straw (GPW and SS). The feedstock originated from MSW, including households, commercial units and industrial facilities. Furthermore, feedstock mass processed annually at the individual plants ranged from 22,000 to 111,000 tons of wet waste. Windrow sizes varied between 400 $m^3$ and 25,200 $m^3$, and treatment time ranged from 4 to 15 months (Table 1). During the composting process, windrows were turned between six times per month and every second month (Table 1). The highest turning frequency was applied at the unit co-composting GPW and SS. The numbers of windrows differed among the plants, ranging from eight to 25 (Table 1). Windrow numbers remained relatively constant throughout the year, because plants continuously receive feedstock throughout this period. These plants operated between 8:00 and 16:00, although working hours would extend occasionally.

### 2.2 Composting technologies

Two composting technologies were investigated, namely 1) open windrows (A, B, C and E) and 2) integrated anaerobic digestion and open windrow composting (D) (Table 1). Incoming GPW is usually shredded to reduce particle size, then sieved and screened to remove any plastic and other inorganic contaminants, before placing it in composting windrows. At maturity, the compost is often sieved and screened to remove inorganic contaminants (DEPA, 1999). Food waste and sewage

**Table 1**
Overview of the selected composting plants.

| Plant | Type of plant | Type of feedstock | Yearly mass of feedstock (Mg yr$^{-1}$) | Turning frequency[d] (times per month) | Composting time (months) | Number of windrows | Windrow layout Length (m) | Wide (m) | Height (m) |
|---|---|---|---|---|---|---|---|---|---|
| A | Composting | GPW[a] | 24,000 | 1-2[e] | 10–15 | 15–22 | 50 | 4–8 | 2–4 |
| B | Composting | GPW[a] | 27,000 | 1-2[e] | 10–15 | 8–12 | 50 | 4–8 | 2–4 |
| C | Composting | GPW[a] | 111,000 | <1[f] | 10–13 | 10–15 | 90 | 40 | 7 |
| D | AD and composting | GPW + FW[b] | 76,000 | <1[f] | 10–13 | 10–20 | 40–30 | 8 | 2 |
| E | Composting | GPW + SS[c] | 48,370 | 4–6[g] | 4–7 | 15–25 | 160–280 | 5–8 | 1.5—2.5 |

[a] Garden/park waste.
[b] Garden/park waste and food waste. AD is anaerobic digestion.
[c] Garden/park waste and sewage sludge (co-composted with straw).
[d] A front-end loader is used in A, B, C and D, whereas a windrow turner is used in E.
[e] One to two times per month.
[f] < 1 refers to every second to third month.
[g] Four to six times per month. For confidentiality reasons, the exact locations and names of the plants are undisclosed.

sludge are often mixed mechanically with pre-treated GPW to improve the porosity of the feedstock – and thus aeration – and to ensure the uniform distribution of moisture content. The windrows are naturally aerated, temperature is measured but not controlled, moisture is regulated by watering the windrows in respect to weather conditions and aeration is assisted by turning the windrows. Turning frequency and turning technology depends on the type of feedstock and the size of windrow. For this study, a front-end loader (Krogmann et al., 2010) was used at composting plants A, B, C and D, whereas a windrow turner (Krogmann et al., 2010) was employed at plant E. When a front-end loader turns the windrow, it generally moves the material from its original place to a new place, which was previously occupied by another windrow. The rotation system enables the optimal use of the area while maintaining the same number of windrows throughout the year. In contrast, the windrow turner may turn the windrows without moving them from their original location.

The second technology is integrated anaerobic digestion with composting, which occurs in a batch reactor. This type of composting plant processes primary FW, which is received in a hall, where it is then sieved in a rotation drum to remove any physical contaminants (>10 cm, e.g., bottles, packages). Next, the pre-treated feedstock is mixed with GPW in the waste processing module, which is sprinkled with percolate to encourage hydrolysis and, thereafter, methane production. When the potential of methane drops, forced aeration is provided for the module and the percolate is pumped into the gas-producing reactor. This process is completely enclosed, so air and percolation circulate between the module and the gas reactor. The temperature inside the waste-processing module is above 70 °C, thus helping to sanitise the compost. Forced aeration reduces humidity and initiates the composting process. At the end of this process, the material is discharged and stored outdoors in aerated composting piles for final treatment. The compost is screened and sieved to remove inorganic contaminants (<20 mm), in accordance with Danish regulations (DEPA, 1999).

### 2.3 Description of the measurement method

Total methane emissions were quantified using a mobile TDM combining the controlled release of tracer gas from a waste treatment facility with concentration measurements downwind of the facility, facilitated by a mobile high-resolution analytical instrument (Mønster et al., 2014; 2015). Downwind concentrations were measured by driving along transects across the plume with a cavity ring down spectrometer (CRDS) from Picarro (model G2203). The spectrometer is a fast and highly sensitive gas analyser capable of detecting methane and acetylene concentrations every second and down to ppb level (Yoshida et al., 2014; Mønster et al., 2014). GPS (R330™ GNSS) was connected to the instrument to log the measured concentrations at their geographical location.

The TDM is based on the principle that a tracer gas released from a source (composting plants for this study) disperses into the atmosphere and follows the same trends as methane emitted from the same area. Since the ratio of their concentrations remains constant throughout atmospheric dispersion, the methane emission rate can be calculated via the following expression, when the tracer gas release rate is known:

$$E_{CH_4} = Q_{tr} \bullet \frac{\int_{plumestart}^{plumeend} \left(C_{CH_4} - C_{CH_4 baseline}\right) dx}{\int_{plumestart}^{plumeend} \left(C_{tr} - C_{trbaseline}\right) dx} \bullet \frac{MW_{CH_4}}{MW_{tr}} (1).$$

where $E_{CH_4}$ is methane emission in mass per time, $Q_{tr}$ is the tracer released in mass per time, $C_{CH_4}$ and $C_{tr}$ are the measured downwind mole fractions (ppb), $C_{CH_4 baseline}$ and $C_{trbaseline}$ are baseline mole fractions of methane and the tracer gas (ppb) and $MW_{CH_4}$ and $MW_{tr}$ are the molar weights of methane and tracer gas, respectively (Andersen et al., 2010a). The method is described in detail in Mønster et al. (2014). Recent studies testing the TDM in controlled field release tests have shown that method uncertainty is no higher than ± 20% (Fredenslund et al., 2019; Delre et al., 2018; Mønster et al., 2014).

In this study, the chosen tracer gas was acetylene ($C_2H_2$), due to its long atmospheric lifetime. Two to three $C_2H_2$ gas bottles were released at the composting plants, and the flow was controlled with calibrated flow meters (Sho-rate, Brooks Instrument, Holland), thus permitting the release of the same concentration from each bottle during each measurement event. The total average release rates ranged from 1.11 to 1.73 kg h$^{-1}$ (Table 2). The $C_2H_2$ bottles were placed in line, perpendicular to the wind direction, in areas where the highest methane concentrations were recorded during an initial on-site screening session. Placement is important to simulate the emission in the best possible manner, while release rates allow us to attain a good signal-to-noise ratio (Delre et al., 2018). Measurements were carried out following best practice guidelines (Scheutz and Kjeldsen, 2019).

A portable tower-mounted weather station (Kestrel 5500 Weather Meter) was positioned downwind of the composting plant in open terrain, to record meteorological data such as wind direction and speed, temperature, atmospheric pressure and general weather conditions. Average temperatures and atmospheric pressure are provided in Table 2. In addition, data provided by the Danish Meteorological Institute (DMI, 2020) were also used.

### 2.4 Overview of the performed measurements

In total, 45 measurement campaigns were carried out, 23 of which were performed during working hours, and 22 when operations shut down for the day (Table 2 and Table S1 in the SI). Two to five measurement campaigns were conducted at each facility, except for plant A, where 26 campaigns were performed from March 2019 to March 2020, to investigate methane emission dynamics. A measurement campaign

**Table 2**
Overview of the measurement campaigns (further information is provided in Table S1 in the Supplementary Information (SI)).

| Plant[a] | Type of plant | Operational activities[b] | Number of measurement campaigns | Average temperature (°C) | Average atmospheric pressure (mbar) | Tracer release points[c] | Total tracer gas ($C_2H_2$) release rate (kg $h^{-1}$)[d] |
|---|---|---|---|---|---|---|---|
| A | Composting | Working hours | 15 | 11 | 1014 | 3 | 1.50 |
| | | Closing hours | 14 | 13 | 1010 | 3 | 1.61 |
| B | Composting | Working hours | 1 | 14 | 1007 | 2 | 1.11 |
| | | Closing hours | 3 | 9 | 1008 | 2 | 1.14 |
| C | Composting | Working hours | 3 | 12 | 1009 | 3 | 1.57 |
| | | Closing hours | 3 | 17 | 1009 | 3 | 1.47 |
| D | AD and composting | Working hours | 3 | 14 | 1022 | 3 | 1.62 |
| | | Closing hours | 1 | 18 | 1007 | 3 | 1.60 |
| E | Composting | Working hours | 1 | 21 | 1014 | 2 | 1.73 |
| | | Closing hours | 1 | 6 | 1013 | 2 | 1.17 |

[a] Composting plants were categorised alphabetically to protect their identity.
[b] Methane emissions were measured during and after working hours (closed hours).
[c] The number of release points equals the number of bottles of tracer release gas (acetylene gas).
[d] The total gas release rate was the sum of the release rates from individual release bottles.

consisted of on-site screening (to locate the main emission sources for optimal position of tracer release), off-site screening (to rule out interfering methane sources) and a tracer release (1–3 h) during which time the methane and tracer plumes were traversed multiple times. Measurement campaigns were carried out following best practice as described by Scheutz and Kjeldsen (2019).

### 2.5 Data processing and statistical analyses

Methane emission rates were obtained by plotting methane and tracer gas (acetylene) concentrations above background levels as a function of time in OriginPro (2019). Plume transects of both methane and acetylene were integrated by applying Eq.1, using the integration build function in OriginPro (2019). The computed plume ratio between methane and acetylene was multiplied by the acetylene release rate to obtain the methane emission rate. The best fit between methane and tracer gas indicated the mixing level between both gases and was applied as one of three quality parameters (Delre et al., 2018). This was determined using $R^2$ (R squared), which is the coefficient of determination for the linear regression between the two gases. $R^2$ should be higher than 0.8 for proper plume mixing.

Statistical analyses were performed in statistical programming software R (R Core Team, 2021), using packages such as *tidyverse* (Wickham et al., 2019). Emission rates between working and closing hours were compared by means of the "data analysis using bootstrap-coupled estimates" (dabestr) package. This statistical method has the advantage of allowing the user to accurately compare unpaired groups, and it can effectively deal with outliers and heavy tail distribution (Ho et al., 2018). Bootstrapping linear regression and correlation tests (Casella et al., 2006; Greenwell, 2017; Štrumbelj and Kononenko, 2014) were applied to determine factors affecting methane emissions. In this regard, the influence of atmospheric parameters (atmospheric pressure, temperature, relative air humidity, precipitation, sunshine hours and drought index) and feedstock mass on measured variations, in terms of total methane emission rates at composting plant A, were investigated.

Additionally, multiple linear regression (Casella et al., 2006; Greenwell, 2017; Štrumbelj and Kononenko, 2014) was applied to determine the overall factors affecting methane emissions. Robust stepwise regression was adopted to select the best model, as it effectively handles outliers (Agostinelli, 2002; Yohai et al., 2023). To validate the results obtained from the stepwise regression, the Random Forests model was employed to classify the predictor factors based on their importance. This model offers several advantages, such as its robustness and the ability to handle both categorical variables (e.g., plants, activities on-site, feedstock) and continuous variables (e.g., temperature, pressure) (Liaw and Wiener, 2002).

Finally, the R package "car" was utilised to test the linear model's assumptions, including normality, independence of errors, linearity and homoscedasticity (Fox and Weisberg, 2019).

The observations used for statistical analysis in this study come from the data collected during measurement campaigns and represent the individual plume traverses (Table 3, Table S1).

## 3 Results and discussion

### 3.1. Methane emission rates from composting plants

Fig. 1 (A1) shows an example of a screening session conducted on site and in the vicinity of the composting plant (plant A), as well as the acetylene and methane gas plumes measured downwind of the plant (A2). The outline of the composting plant is marked in blue. For this example, measurements were performed on March 26, 2019. The methane background concentration was 1.93 ppm. During the on-site screening, the highest recorded concentration was 8.23 ppm. The results of the screening in Fig. 1 (A1) highlight the principal sources of methane at the plant, thereby determining where to place the acetylene gas bottles (B1, B2 and B3). In addition, screening conducted upwind of the plant ensured the absence of any interfering methane sources in the vicinity of the units. This demonstrates that the approach is capable of quantifying – in real time – methane emission sources both at a facility and in the surrounding area, thus overcoming the limitations of identifying either (i) the exact methane emission point on a pile or (ii) covering emission variations between piles at large-scale commercial composting facilities. The correlation between the methane and tracer gas plumes was always higher than 0.8, and statistically significant, thereby suggesting a good simulation between the methane emission and the released tracer gas.

Table 3 presents a summary of measured average total methane emission rates. Detailed information about individual campaigns and measured emission rates can be found in Table S1 in the SI. The highest average methane emissions were measured at plants D (42.5 ± 1.5 kg $h^{-1}$) and C (40.9 ± 1.8 kg $h^{-1}$), whereas the lowest were measured at plants A (8.0 ± 0.1 kg $h^{-1}$) and B (9.1 ± 0.5 kg $h^{-1}$). Ostensibly, the lowest emissions were measured at composting plants processing solely GPW (A, B and C), and the highest were seen at the composting plants processing GPW mixed with either food waste or sewage sludge (D and E). The higher emission measured at plant C, in comparison to plants A and B, could be due to the higher amount of waste treated at plant C and the low turning frequency for the windrows (Table 1). Interestingly, the highest methane emissions were measured at the plant combining anaerobic digestion and composting, which processed GPW mixed with food waste (Table 3). Factors influencing emissions are discussed in the following sections.

**Table 3**
Summary of the measured methane emissions and total numbers of measurement campaigns and plume traverses.

| Plant | Type of plant | Type of feedstock | Number of measurement campaigns | Number of plume traverses | Methane emission rate ($kg\ CH_4\ h^{-1}$) | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | Mean | SD[d] | SE[e] | Median | Mean[f] |
| A | Composting | GPW[a] | 29 | 657 | 8.0 | 3.1 | 0.1 | 8.0 | 7.28 |
| B | Composting | GPW[a] | 4 | 59 | 9.1 | 4.1 | 0.5 | 8.0 | 9.67 |
| C | Composting | GPW[a] | 6 | 158 | 40.9 | 22.5 | 1.8 | 33.5 | 34.0 |
| D | AD and composting | GPW + FW[b] | 4 | 117 | 42.5 | 15.9 | 1.5 | 42.1 | 41.9 |
| E | Composting | GPW + SS[c] | 2 | 21 | 22.8 | 15.7 | 3.4 | 22.8 | 19.2 |

[a] Garden/park waste.
[b] Garden/park waste and food waste. AD is anaerobic digestion.
[c] Garden/park waste and sewage sludge (co-composted with straw).
[d] Standard deviation of the mean.
[e] Standard error of the mean.
[f] Weighted mean emission rates computed using emission rates measured during opening and closing hours, assuming eight working hours and 16 closing hours (Table S2 in SI).

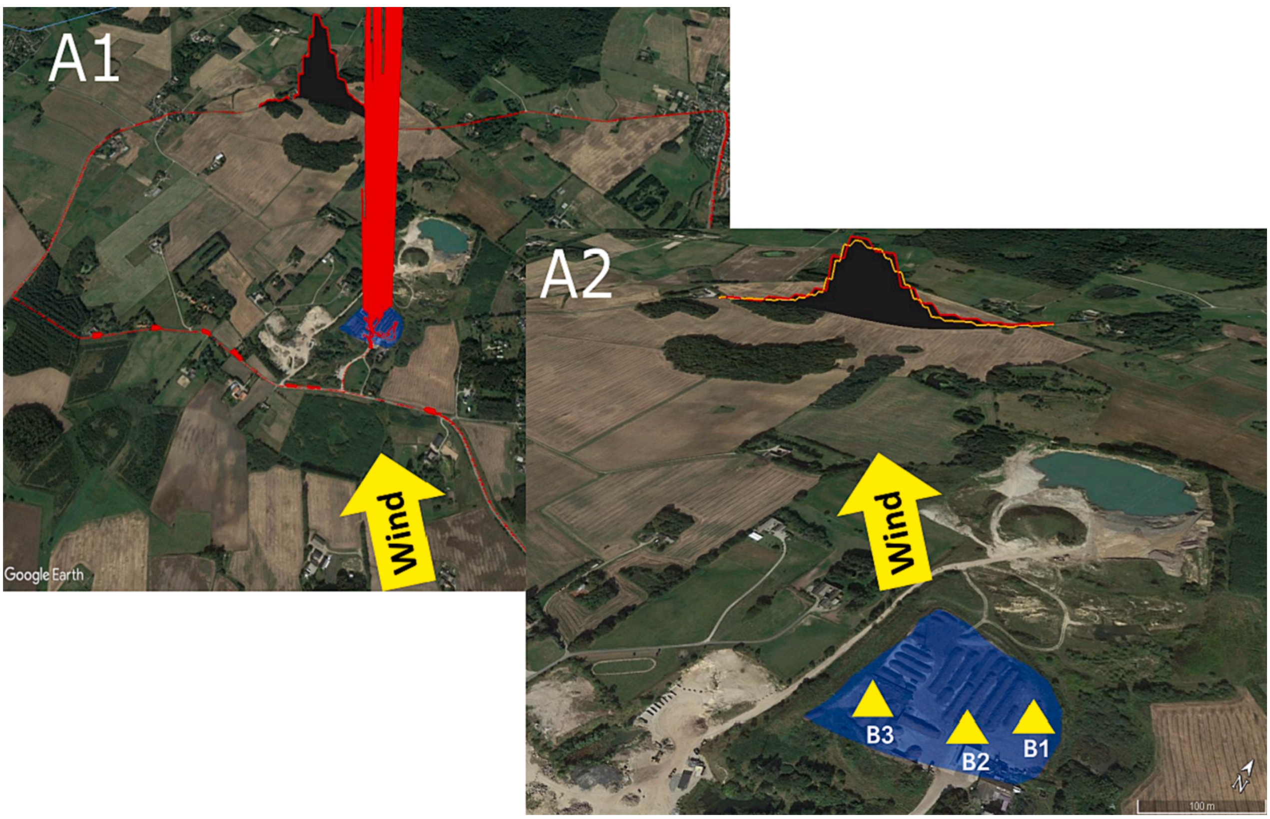


**Fig. 1.** On-site and off-site methane screening of composting plant A, with examples of on-site screening (A1) and downwind plumes showing methane (red) and acetylene (yellow) concentrations above background (A2). The triangles (yellow B1-B3) mark the three release points of the tracer gas ($C_2H_2$) at the composting plant. Map: Google Earth, US Dept. of State Geographer, GeoBasis-DE/BKG. Measurements were performed on March 26, 2019.

### *3.2. Influence of activities at the composting facilities*

Fig. 2 compares total methane emission rates measured at the five composting facilities (A-E) during working hours and thereafter (closing hours). At four of the five plants, the average methane emission rate measured during working hours was higher (a factor of 1.5 to 4.8) than during closing hours. The difference in methane emission rates between opening and closing hours was statistically significant for plants A, B, C and E (Table S3 in the SI). The higher emissions during working hours were most likely due to operational activities at the facilities, where materials were shredded and sieved and the windrows were turned. Especially during windrow turning, enhanced emissions can be expected, as gases accumulating inside the pore spaces of the compost material are released, thus increasing methane emissions (Jensen et al., 2017). The two sites (C and E) with the highest differences in emissions (factors of 2.4 and 4.8) were also those with the most activities (in- and outgoing material, movement of material, shredding of material, etc.). One exception was plant D, where emissions were measured during opening and closing hours, respectively, and were similar (no significant difference, Table S3 in the SI). The possible explanation is that plant D – as the only one of the five – applied a combination of anaerobic digestion and composting processes. It was not possible to distinguish methane emissions from these two treatment processes; however, Jensen et al. (2017) estimated that methane emitted from a similar facility was primarily the result of anaerobic treatment, culminating in insignificant differences between opening and closing hours for total emissions across the whole facility. Anaerobic digestion is a continuous process, regardless of windrow turning events.

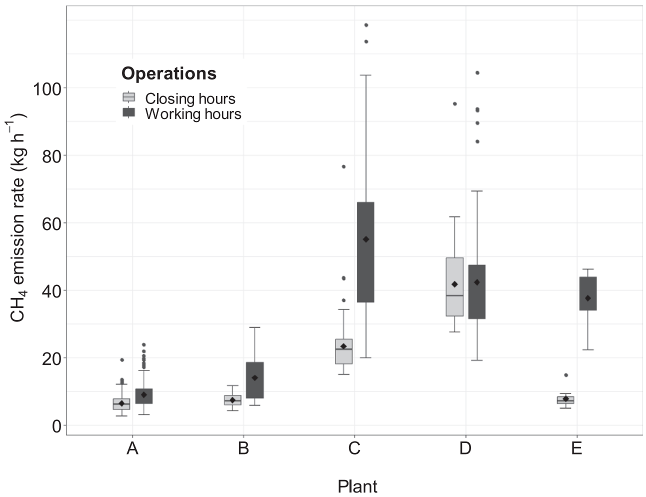


**Fig. 2.** Comparison of methane emission rates measured during working hours (light grey) and after the working day has finished (closing hours in dark grey). Table S3 in the SI provides specific data.

The best model ($R^2$ = 0.8) resulting from the multiple linear regression and robust stepwise regression (Table S6) indicated that the type of composting plant, on-site activities, temperature and atmospheric pressure significantly affect methane emissions (Figs. S3-S7 and Table S6). Furthermore, the Random Forests model (% Var explained = 53) suggested that the most important factor was the type of composting plant (Fig. S8). For this study, differences between composting plants included yearly mass and type of feedstock treated, as well as treatment technology (Table 1). The second most important factor was on-site activities (closing and opening hours), followed by temperature and pressure (Fig. S8). The influence of environmental factors and on-site activities is discussed in section 3.3, while the influence of feedstock (mass and type) and treatment technology is discussed in section 3.4.

### 3.3 Daily, monthly and seasonal variations in methane emission rates

Fig. 3 shows total methane emission rates measured over a whole year at composting plant A. All methane plume traverses performed during each month are presented in the figure, displaying measurements performed during opening and closing hours, respectively.

In six of the 11 measurement campaigns, emissions were measured during both opening and closing hours. For five of the six campaigns, emissions were significantly higher during opening hours (Fig. 3 and Table S4 in the SI). An exception in this regard was the July campaign, where the difference in methane emissions between working hours and closing hours was not statistically significant (Fig. 3 and Table S4 in SI). During daytime hours for the July measurement campaign, smoke and heat were observed at one of the windrows, probably due to the extraordinarily high temperatures in the compost pile approaching ignition (Brown et al., 2008). To prevent fire, the pile was opened after working hours to actively supply oxygen and release heat, most likely resulting in higher methane emissions. As the measurement was carried out after opening the pile during closing hours (Table S1 in the SI), this most likely explains the higher methane emission rate determined at this time of day (Fig. 3).

The results reveal significant differences in methane emission rates between months as well as seasons (Fig. 3, Table S4 and S5 in the SI). Furthermore, they show that average methane emission rates increased markedly from January to April, following which they increased more slowly until August, before starting to decrease steeply. Consequently, the average methane emissions were lower in the September to March period and higher in the April to August period than the yearly average (Fig. 3). For the summer months (May to August), the average methane emission rates were 10.2 and 7.3 kg $h^{-1}$ for working hours and closing hours, respectively, compared to 6.1 and 4.6 $h^{-1}$, respectively, during winter (November to February). These figures suggest that methane emission rates increased by a factor of 1.6 between these two periods, regardless of activities at the plant.

Analysis of a Spearman's rank correlation between environmental parameters and methane emission rates revealed that average monthly methane emission rates were positively correlated with atmospheric temperature (r = 0.55) (Fig. S1 in the SI). Nevertheless, there was a weak

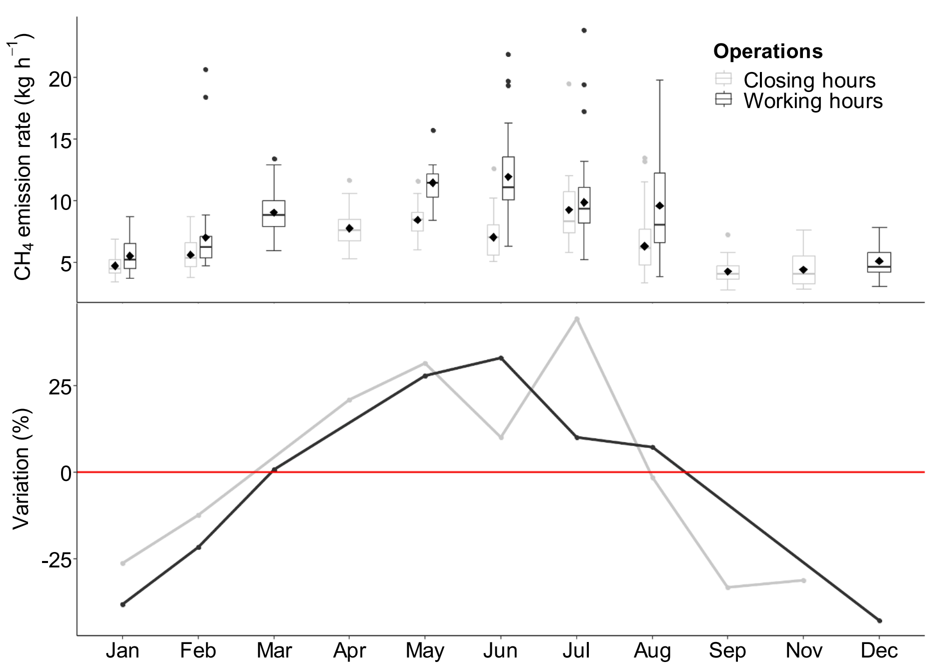


**Fig. 3.** Methane emission rates in kg $h^{-1}$ (a) and emission variations in percentage (b) of monthly methane emissions measured at composting plant A during working and closing hours, respectively.

correlation between the methane emission rate and atmospheric pressure (r = 0.27) and precipitation (r = 0.09), as well as the monthly mass of GPW received (r = 0.01) (Fig. S1 in the SI). These results suggest that methane emissions from composting plants may be driven by atmospheric temperature, thereby corroborating the findings of Husted (1994). To the best knowledge of the authors, our study is the first attempt to correlate these external atmospheric factors with methane emission rates. On the contrary, the weak correlation between monthly received feedstock mass (GPW) and measured methane emission rates can be explained by the fact that received GPW is usually stored for one to five months prior to processing (Krogmann et al., 2010). It is therefore evident that compost plants have windrows with feedstock received on different months of the year. Moreover, compost production takes several months, and as a result, material at different treatment phases (from fresh to mature) will be present at the plant, thus potentially levelling out the effect of variations in the received input material on emissions.

Overall, these results suggest that monthly and seasonal variations, as well as activities at the facility, significantly affect the methane emission rate. Consequently, data extrapolation, using only emission rates measured in either September-March or April-August, leads to under- or overestimating yearly emission rates, respectively. Similar emissions measured solely during opening hours would overestimate annual average emissions and vice versa. To attain reliable methane estimates, measurements should be at a minimum conducted during both periods of the year; moreover, conducting measurements during each season would provide more robust and accurate estimates.

### 3.4 Methane emission factors

Fig. 4 compares average whole-plant methane emission rates with the yearly wet mass of treated feedstock. Previously published methane emission rates from Danish composting facilities were included in the dataset (the whole dataset is provided in Tables S7 and S8 in the SI). Uncertainty relating to the linear relationship (red line) is presented by a 95% confidence interval region in grey. The slope was 0.0005, indicating that an increase of 0.5 kg of total methane emissions for every one-ton increase in feedstock wet mass can be expected. The r-squared value (0.86, $p < 0.001$) suggests a significant linear relationship between the yearly wet mass of treated feedstock and average total methane emission rates.

Owing to these results, methane emission factors were calculated based on measured average annual methane emission rates and reported annual processed wet feedstock amounts (Table S7 in the SI). Fig. 5 presents average methane emission factors (A, B, C, D and E refer to the current study) as a function of composting technologies and the type of feedstock. The results indicate that composting combined with anaerobic digestion generated the highest average emission factors (average of 4.86, ranging from 4.51 to 5.21 kg $Mg^{-1}$ GPW and FW). Conversely, the open windrow composting of GPW yielded the lowest average emission factors (average of 2.65, ranging from 1.56 to 3.24 kg $Mg^{-1}$ waste) (Fig. 5). Moreover, it is apparent that composting facilities can be grouped into three categories based on feedstock and technology, namely (i) open windrows treating primarily GPW (average of 2.65, ranging from 1.56 to 3.24 kg $Mg^{-1}$), (ii) open windrows co-composting GPW and SS (average of 3.63, ranging from 3.49 – 3.76 kg $Mg^{-1}$) and (iii) combined anaerobic digestion and open windrows composting GPW and FW (average of 4.86, ranging from 4.51 – 5.21 kg $Mg^{-1}$).

Until 2020, the national emission inventory for Denmark applied the following methane emission factors for composting waste: GPW (4.20 kg $Mg^{-1}$), FW (4.00 kg $Mg^{-1}$), sewage sludge (0.41 kg $Mg^{-1}$) and the home composting of FW (5.63 kg $Mg^{-1}$) (Table S9 in the SI). In 2021, the values were revised for GPW (3.19 kg $Mg^{-1}$), sewage sludge (0.22 kg $Mg^{-1}$) and home composting (4.2 kg $Mg^{-1}$) (Table S9 in the SI). For FW composting, the emission factors suggested by the IPCC were adopted (4 kg $Mg^{-1}$) (IPCC, 2006; IPCC, 2019). Emission factors for combined anaerobic digestion and open windrow composting are not provided. Even though the emission factor for GPW used in the Danish national inventory was reduced to 3.19 kg $Mg^{-1}$ in 2021, it is still higher (a factor of 1.2) than the average emission factor measured in this study (2.65 kg $Mg^{-1}$) (Fig. 5). The average emission factor for co-composting of GPW with FW or SS is 4.36 kg $Mg^{-1}$ (Fig. 5) and thus comparable to the default value of 4.0 kg $Mg^{-1}$ used by the IPPC.

Table S7 in the SI summarises methane emission factors for composting plants reported in the literature. Estimates of methane emission factors are 0.05 – 0.49 and 0.50 – 1.00 kg $Mg^{-1}$ wet GPW treated, respectively, for Austria and Germany (Amlinger et al., 2008). Similarly, Hrad et al. (2014) suggest 0.11 – 0.94 kg $Mg^{-1}$ wet GPW treated for Austria. Their study was based on a full-scale composting plant treating 100,000 Mg annually, and methane emissions were measured by combining OP-TDLS plume measurements with inverse dispersion modelling. Moreover, Sintana and Whendee (2019) found methane emission factors ranging between 1.4–2.2 kg $Mg^{-1}$ wet waste treated (mean and median), based on a literature review, and estimated an average emission factor of 1.7 ± 0.3 kg $Mg^{-1}$ wet waste treated. Types of waste treated by composting included GPW and manure, mostly from

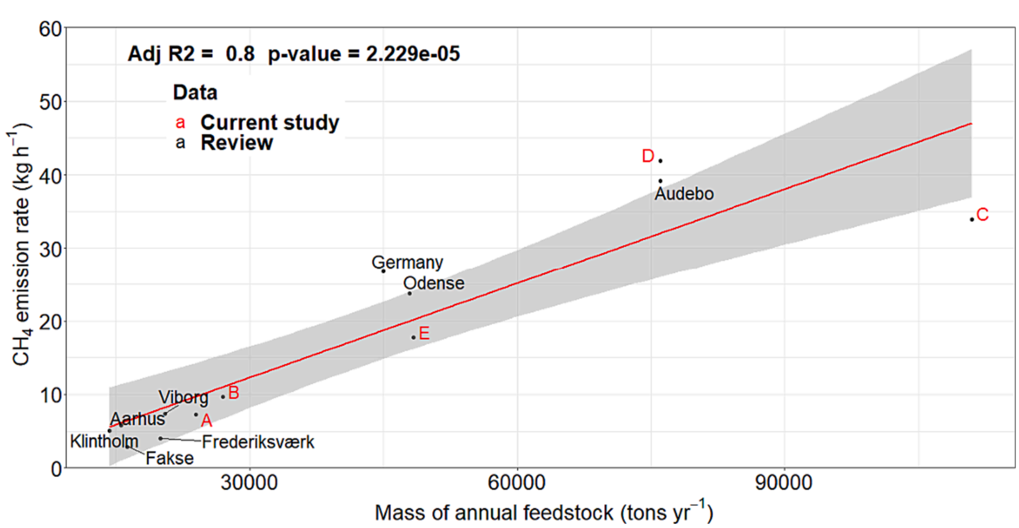


**Fig. 4.** Comparison of total methane emission rates based on the current study (plants A, B, C, D and E) (red) and literature review data (Table S7 and S8 in the SI) (black) and the linear line of best fit (red) with the 95% confidence interval region (grey).

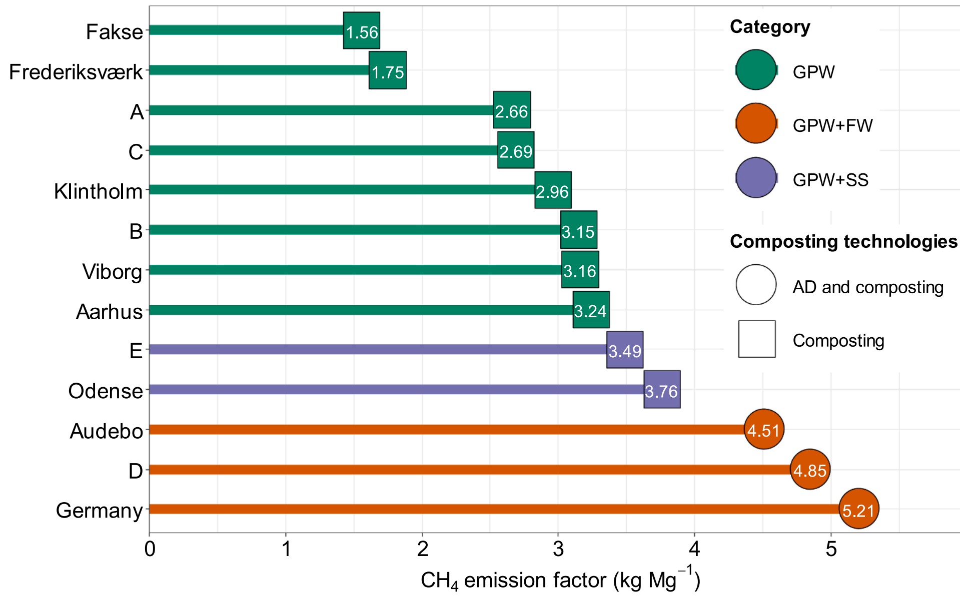


**Fig. 5.** Comparison of methane emission factors (kg $Mg^{-1}$) as a function of feedstock and the type of composting technology. Measured emission rates were normalised to the wet mass of feedstock. Data consisted of the current study (plants A, B, C, D and E) and published studies conducted in Denmark (Tables S7-S8 in the SI).

cattle, goats, horses and chickens. These emission factors were lower than those found by the present study, as well as the default values from IPCC (4.0 kg $Mg^{-1}$). Conversely, Phong (2018) estimated the emission factor for composting facilities by combining the tunnel method and gas measurements at a composting plant processing GPW and food waste at 3.60 – 4.06 kg $Mg^{-1}$ wet food waste treated, 4.41 kg $Mg^{-1}$ wet GPW treated and 10.25 kg $Mg^{-1}$ wet anaerobic digestion materials. These emission factors were higher than estimates in both the current study and the default value from IPCC, thereby implying that estimates should reflect local conditions and the feedstock. Seemingly, very few to no previously published studies consider temporal variations and the influence of activities at a composting facility, which could explain – at least partly – the large discrepancy between emission factors. Our study showed that lower emission factors were obtained when measurements were taken during closing hours, whereas higher emission factors were seen during working hours (Table S3 in the SI). This demonstrates the possible errors that can occur if operational factors are disregarded.

## 4 Conclusion and perspectives

The mobile tracer gas dispersion method (TDM) was successfully applied to quantify total methane emissions at five Danish composting plants. The results show that composting garden/park waste, food waste and sewage sludge generate methane emissions and thus contribute to climate change. The study provides a comprehensive dataset, including methane emission rates measured during working hours and after the end of the working day (closing hours), the influence of temporal variations on emission rates and emission factors.

Methane emission rates ranged between 42.5 ± 1.5 and 8.0 ± 0.1 kg $CH_4$ $h^{-1}$, with the lowest measured at composting plants processing solely garden/park waste, whilst the highest was seen at units processing garden/park waste mixed with either food waste or sewage sludge. Operational activities influenced plant emissions significantly, except for combined anaerobic digestion and composting. At the four windrow composting facilities, average methane emission rates measured during working hours were factors of 1.5 to 4.8 higher than during closing hours. The results revealed significant differences in methane emission rates between months as well as seasons. For the summer months (May to August), average methane emission rates measured at a windrow composting plant (A) were 10.5 kg $CH_4$ $h^{-1}$ compared to 6.1 kg $CH_4$ $h^{-1}$ during winter (November to February) and in working hours. Methane emission rates had a high positive correlation with average monthly atmospheric temperature. The statistical analyses showed that type of composting plant, on-site activities, temperature, and atmospheric pressure significantly affect methane emissions. To obtain emission factors, methane emission rates were normalised to feedstock waste mass (wet weight). Measurements from 13 treatment plants resulted in average methane emission factors from open windrows treating primarily garden/park waste of 2.65 kg $Mg^{-1}$, from open windrows co-composting garden/park waste mixed with sewage sludge of 3.63 kg $Mg^{-1}$ and from combined anaerobic digestion and open windrows composting garden/park waste and food waste of 4.86 kg $Mg^{-1}$.

The findings suggest that methane emissions from composting are site-specific and thus require local monitoring. In this regard, TDM makes it possible to quantify and compare methane emissions accurately between composting plants and to identify controlling factors and document mitigation actions. Due to temporal emission variations (daily, monthly and seasonal), measurements should be performed during each season of the year (at a minimum during summer and winter) to obtain robust and accurate methane emission rates. Moreover, the impact of operational activities on the facility should be accounted for by computing a weighted mean for total annual emissions. To comprehend and describe the patterns that cause emission dynamics as a result of composting, a long-term measurement campaign should be carried out. Finally, it may be possible to reduce methane emissions by changing plant operations such as windrow geometry, windrow turning frequency and technology, pre/post-treatment, material storage time and conditions.

## 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.

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2023.09.010.

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