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Do compost-based landfill biocover systems designed for methane oxidation emit nitrous oxide in significant amounts?

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ABSTRACT

Landfills constitute a significant source of methane (CH_4), thereby contributing to climate change. CH_4 emissions from old and smaller landfills can be mitigated by compost-based biocover systems designed for optimal microbial CH_4 oxidation. It is well-known that the strong greenhouse gas nitrous oxide (N_2O) is generated during the composting process, which potentially could continue after incorporating compost into the biocover system. Field studies were performed at three full-scale biocover systems established at Danish landfills and included surface screenings, surface flux measurements and gas composition analysis. To assess if N_2O generated in the biofilter-compost material would hamper the climate benefit from CH_4 oxidised in a biofilter, CH_4 removed was compared to N_2O generated, with both calculated in CO_2-eq. Two assessments were performed. The first considered individual measurement locations on the biocover, whereas the second considered the overall performance of the biocover. By comparing CH_4 oxidation rates to the emitted N_2O, both approaches showed that there is no risk that N_2O emission will negatively affect the CH_4 mitigation efficiency of compost-based biocover systems established at landfills. The ratio of N_2O emitted to CH_4 oxidized (both in unit kg CO_2-eq per day) was less than 2.3% for both approaches, and in most cases below 1%.

1. Introduction

Decomposition of organic matter deposited in landfills produces landfill gas (LFG), which normally contains 50–60 %v/v of CH_4 and 40–50 %v/v of carbon dioxide (CO_2). CH_4 is an important greenhouse gas with a global warming potential (GWP) 27 times higher than CO_2 over a 100-year period (Foster et al., 2021). Very few studies to date have measured nitrous oxide (N_2O) content in LFG, even though it is of interest because N_2O is a very strong greenhouse gas with a GWP 273 times higher than CO_2 over a 100-year period (Forster et al., 2021). Landfill simulation studies on mixed waste have found very limited amounts of N_2O under anaerobic conditions (Fricko et al., 2022), whilst another study on N_2O production during in situ aeration in a closed landfill site found concentrations lower than 50 ppm under anaerobic conditions prior to the start of aeration (Nag et al., 2016). Most studies conclude that N_2O emissions from landfills are due to N_2O-forming processes in landfill cover soils and not due to its presence in LFG (Rinne et al., 2005, Lee et al., 2023).

In many landfills, LFG is collected and utilised for electricity and/or heat production, or it is flared to reduce CH_4 emissions. However, for older landfills, CH_4 concentrations in LFG decrease gradually over time, and the gas generation rate is often too low for gas utilisation to be economically feasible. In such cases, engineered mitigation systems based on optimised microbial CH_4 oxidation (so-called 'biocover systems') offer an alternative, cost-effective strategy (Huber-Humer et al., 2008; Kjeldsen and Scheutz, 2019). Biocover systems typically consist of a highly porous gas distribution layer covered by a layer supporting CH_4 oxidation, where often organic-rich materials with high CH_4 oxidation capacities are used, such as compost produced from garden waste (Humer-Huber et al., 2008, Kjeldsen and Scheutz, 2019).

Maintaining complete aerobic conditions during the windrow composting of garden waste is challenging, and often emissions of CH_4 and N_2O are observed from such facilities (Edjabou & Scheutz, 2023; Andersen et al., 2010a; 2010b). To the best of our knowledge, it is unknown if N_2O is emitted from landfill biocover systems when mature compost is used as a support material for microbial CH_4 oxidation. N_2O production in biocovers may be affected by several factors such as contents of oxygen, CH_4, NH_3, pH, moisture, etc. High biocover CH_4 oxidation activity may result in oxygen limitation leading to N_2O formation (Lienhart et al., 2024). Such conditions could potentially cause

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significant N_2O emissions, which may counteract the mitigation impact of reducing landfill CH_4 emissions through enhanced microbial CH_4 oxidation. This is because N_2O is a strong greenhouse gas with a GWP about 10 times higher than CH_4 (Foster et al., 2021), so if N_2O is produced in significant quantities in compost-based biocovers, the N_2O emission and its contributions to climate change should be deducted from the climate change mitigation gained by oxidising landfill CH_4.

This study aims to assess N_2O emissions from biocover systems where mature compost is used as CH_4 oxidation material, and to compare N_2O emissions to the mitigation effect from reducing CH_4 emissions. The objective was met by taking N_2O surface emission measurements at three full-scale biocover systems established as part of the Danish Biocover Initiative (Scheutz and Kjeldsen, 2023). The CH_4 mitigation effect of the three biocover systems has previously been published (Duan et al., 2022, Kissas et al., 2023), but this study contains additional measuring campaigns targeting N_2O.

2. Material and methods

2.1. Site descriptions

Table 1 provides an overview of the three established biocover systems at Glatved, Uggeløse and Skellingsted landfills. The biocover systems at Glatved and Uggeløse are both active systems where the LFG is extracted from gas extraction wells and loaded to a gas distribution layer, while the system at Skellingsted is passive, where the generated pressure inside the landfilled waste as a result of LFG generation, feeds the gas to the biocover system. More details are given in Duan et al. (2021; 2022) for Glatved landfill, in Duan et al. (2022) for Uggeløse landfill and in Kissas et al. (2023) for Skellingsted landfill. Table S1 in the Supplementary Material (SM) describes the landfills/landfill cells where the biocover systems are established in respect to age of waste, size of landfill/landfill cell, and waste composition. Table S1 is based on information given in Duan et al. (2022), Kissas et al. (2023) and Kjeldsen and Fischer (1995). For Skellingsted and Uggeløse, the waste is of a mixed nature (Table S1), while the landfill cell at Glatved, which loads CH_4 to the biocover, contains shredder waste, mainly from automotive shredding. It has previous been shown that this type of waste also produces LFG (Aghdam et al., 2017). At all three sites, biofilters/biowindows were constructed with a 30-cm-thick gas distribution layer consisting of virgin gravel overlain by 80 cm of locally produced mature compost material made from garden waste. The three biocover systems are all made as part of the Danish EPA's "Biocover Initiative" (Miljøstyrelsen, 2024). The compost used in the three biocovers comply with the guidelines set up in the "Biocover Initiative": If compost made from garden waste is used, it must be produced by composting in windrows with a duration af more than six months, with adequate supply of oxygen by frequent turning the windrow. Before use, the compost is sieved through a 15 mm sieve and the larger fraction not passing through the sieve is discarded (Miljøstyrelsen, 2018).

The field investigations reported in this study were carried out in June 2022 for the Uggeløse landfill, in the period May to June 2020 for the Glatved landfill and in the period March to May 2022 for the Skellingsted landfill. Table 1 gives details on the investigated landfill biofilters/biowindows, the number of campaigns and the number of flux chamber measurements in each campaign.

2.2. CH_4 loading rates for the active biocover systems

For the two biocover systems at Glatved and Uggeløse, which were actively loaded with LFG, LFG composition at the biofilter inlets was measured using a Biogas 5000 portable gas analyser (Geotech, Warwickshire, UK), whereas gas flow rates (in m^3/h) were read from flow meters installed as part of the biocover systems.

2.3. Surface CH_4 screening

To establish the basis for a representative placement of flux chambers (see next section), surface CH_4 concentration screenings were carried out at the investigated biocover systems using a Laser One Portable CH_4 Leak Detector (Huberg, Italy). More details of the surface screening activities are given in Duan et al. (2022) and Kissas et al. (2023).

2.4. Surface flux measurements

Surface fluxes of CO_2, CH_4 and N_2O were measured using static, cylindrical flux chambers (0.57 m ID or 0.30 m ID). At Glatved, a grid was established on the biofilter and the area divided into 350 small squares, each one of 4 m^2. Based on the surface CH_4 screening of each square, five squares were selected (G1-G5), representing areas with high, medium and low CH_4 loading rates, respectively. At Uggeløse, surface fluxes were measured at 17 locations. The placement of chambers was based on the initial surface CH_4 screening. At Skellingsted landfill (Biocover 8), 12 flux chambers were placed, thereby creating a uniform grid. A typical setup for measuring surface fluxes is showed in Fig. S1 in SM.

Gas concentrations of CH_4, CO_2 and N_2O were measured using a photoacoustic Multi Gas Monitor INNOVA 1512i (LumaSense Technologies, Denmark). The flux was measured by connecting the INNOVA sampling tube to the sampling port of the flux chamber and then taking five to ten gas samples over a period of 5–10 min. The samples were continuously analysed by the INNOVA, and CO_2, CH_4 and N_2O fluxes (in $g\ m^{-2}\ d^{-1}$) were calculated from the increasing concentrations over time inside the chamber (Scheutz et al., 2011). For more details on the flux measurements at the three biocover systems, refer to Duan et al. (2021, 2022) and Kissas et al. (2023).

2.5. CH_4 oxidation rates in individual locations, using a carbon mass balance approach

CH_4 oxidation rates at individual locations were determined using the carbon mass balance approach, which relies on biocover surface CH_4 and CO_2 fluxes and CH_4 and CO_2 concentrations measured at the interface between the gas distribution layer and the compost CH_4 oxidation layer (Scheutz et al., 2011, Duan et al. 2022). Gas compositions were measured by placing a stainless steel (5 mm ID) probe slotted in the lower 3 cm on the interface between the compost and the gas distribution layer. The probe was connected through a sampling tube to the Biogas 5000 instrument, and concentrations of CH_4 and CO_2 were then measured. These measurements, together with the measured CH_4

Table 1
Overview of biocover systems investigated at the three landfills.

Landfill	Biocover type	Supply of LFG	Gas distribution pipes	Name of investigated biofilter/biowindow	Total biocover area (m^2)	Investigated biofilter/biowindow area (m^2)	Number of flux chamber measure-ments	Number of measure-ment campaigns
Skellingsted	Biowindow/ Biofilter	Passive	Yes	B8	3225	460	12	5
Glatved	Biofilter	Active	Yes	IC3	3940	1400	5	10
Uggeløse	Biofilter	Active	Yes	CIII	1930	1930	17	2

and CO_2 surface fluxes (converted to mole m^{-2} d^{-1}) at the location, provided the input data for the carbon mass balance approach, which calculates the CH_4 oxidation rate in the unit mole CH_4 m^{-2} d^{-1} (confer Duan et al. (2022) for details of the method). The calculated CH_4 oxidation rates were re-calculated to the unit g CH_4 m^{-2} d^{-1} and compared to the emitted N_2O surface flux, using GWPs of 27 for CH_4 and 273 for N_2O, considering in each case a period of 100 years (Foster et al., 2021). Due to the biogenic origin of the CH_4 in this study (generated from organic biodegradable landfilled waste), we used the GWP for non-fossil CH_4.

2.6. Overall CH_4 oxidation rates for actively loaded biofilters

The biocover systems at Glatved and Uggeløse are actively loaded, i.e., generated LFG is pumped from extraction wells through a manifold facility and into the gas distribution layer forming the base of the biofilter. By comparing the filter's CH_4 load (in kg CH_4 d^{-1}) with the integrated surface CH_4 emission from the biofilter (in kg CH_4 d^{-1}), the overall amount of CH_4 oxidised in the biofilter was determined. Similarly, the amount of N_2O generated in the compost layer was determined by integrating surface N_2O fluxes from the biofilter surface. Overall CH_4 and N_2O emitted from the filter surface was calculated by integrating the surface flux measurements, using a weighted area for each flux measurement based on surface screenings. In addition, based on surface concentrations, the filter was divided into different emission zones, and fluxes measured in these zones were multiplied by the corresponding area of the zone.

3. Results and discussion

3.1. Comparison of CH_4 oxidation efficiency with N_2O emission in individual locations

Table 2 compares CH_4 oxidation rates and surface N_2O emission rates measured at individual locations during the campaigns at the three landfills by summarising the ranges and averages of CH_4 oxidation rates, using the carbon mass balance, together with the ranges and averages of measured N_2O surface fluxes. A broad range of CH_4 oxidation rates (0–100 g CH_4 m^{-2} d^{-1}) was observed, with low numbers mostly reflecting a low CH_4 load to the compost oxidation layer, albeit there were also some hotspots with high CH_4 loads resulting in high CH_4 oxidation rates.

High spatial variability of the surface N_2O emission was observed, with fluxes ranging from 0 to 0.37 g N_2O m^{-2} d^{-1}. Several potential correlations between the data were evaluated (N_2O emitted to CH_4 oxidised (mass based), N_2O emitted to CH_4 emitted (mass based), ratio of N_2O emitted to CH_4 oxidised (in CO_2-eq) correlated with CH_4 oxidised or emitted (in CO_2-eq)). No strong correlations were found; the one with the highest R^2 (0.39) was N_2O emitted to CH_4 oxidised (in CO_2-eq) (Fig. S5), revealing a weak tendency that N_2O emitted was higher when CH_4 oxidised was high. A recent laboratory study on landfill intermediate cover soils found that N_2O production was higher in incubations where CH_4 oxidation occurred as compared to incubations where no CH_4 was added (Lienhart et al., 2024).

In general, the observed surface N_2O fluxes from the biocovers were much lower (a factor of 10 to 100) compared to N_2O fluxes measured at the surface of garden waste windrows, which varied between 1.2 and 8.2 g N_2O m^{-2} d^{-1}, with the lowest surface fluxes observed at windrows with more stabilised compost material (after about 300 days of treatment) (Andersen et al., 2010a). A slightly lower range of N_2O surface fluxes (about 0.1–1.4 g N_2O m^{-2} d^{-1}) was measured from home composting units continuously loaded with food waste and garden waste with higher fluxes during spring and summer (Andersen et al., 2010b).

Fig. 1 compares average surface N_2O emissions (g CO_2-eq m^{-2} d^{-1}) to average CH_4 oxidised (g CO_2-eq m^{-2} d^{-1}) based on measurements in individual locations on the biocover for the three landfills. Detailed graphs for each location on the three biocover systems are provided in SM (Fig. S2-S4). The results clearly show that N_2O surface emissions are significantly lower compared to the amount of CH_4 oxidised in the same location. The estimated average ratio of N_2O emissions to CH_4 oxidised (in CO_2-eq) is 2.2 % for Glatved, 2.1 % for Skellingsted and 2.3 % for Uggeløse. Thus, for functional biocover systems using mature compost as the support material for CH_4 oxidation, N_2O produced in the compost material is insignificant and can be disregarded in overall greenhouse gas accounting when examining the mitigation performance of biocover systems.

3.2. Comparison of integrated CH_4 oxidation efficiency with N_2O emission

For two of the landfills, namely Glatved and Uggeløse, the overall CH_4 oxidation performance of the biofilter was compared to overall N_2O emitted from the biofilter surface. Table 3 shows the result of comparing CH_4 oxidised and N_2O emitted. At these two sites, LFG was actively pumped into the biocover system with average loading rates of 271.6 and 118.6 kg CH_4 d^{-1}, respectively. CH_4 emitted from the surface were 0.8 and 11.9 kg CH_4 d^{-1} for Glatved and Uggeløse, respectively, corresponding to an overall amount of CH_4 oxidised of 271 and 107 kg CH_4 d^{-1} (Table 3). The amount of N_2O generated in the compost layer was 0.17 and 0.12 kg N_2O d^{-1} for Glatved and Uggeløse, respectively (Table 3).

Overall, the results indicate that the average ratios of N_2O emitted from the biofilter surface to CH_4 oxidation in the biofilter range were 0.63 and 1.1 % for Glatved and Uggeløse, respectively (Table 3). The percentages in this regard are a bit lower than the averages obtained from comparing CH_4 oxidation rates with N_2O emissions in individual locations, because some of the single measurements in small hotspot areas, where N_2O emitted to CH_4 oxidation is higher, weigh less in the area integrated comparison.

The presented analysis assumes that surface fluxes originate solely from N_2O production in the compost layer and not also from N_2O originally contained in the LFG. This assumption, however, seems valid based on literature observations (as mentioned in the introduction) and general knowledge on anaerobic LFG formation. If LFG at the investigated landfills did contain trace-level concentrations of N_2O, the ratios of N_2O surface fluxes to CH_4 oxidation rates would be lower and the contribution of N_2O formation from compost of even less importance from a climate mitigation perspective. The results presented herein are thus conservative estimates of the significance of N_2O in compost-based CH_4 oxidation biocover systems in relation to the overall climate change mitigation balance.

Table 2
Observed ranges and averages of CH_4 oxidised (g CH_4 m^{-2} d^{-1}) and N_2O emitted (g N_2O m^{-2} d^{-1}) for all three biocover systems.

	Glatved	Uggeløse	Skellingsted
Range CH_4 oxidised (g CH_4 m^{-2} d^{-1})	0–100	0–93	0–92
Average CH_4 oxidised (g CH_4 m^{-2} d^{-1})	54.1	26.7	18.6
Range N_2O emitted (g N_2O m^{-2} d^{-1})	0–0.37	0–0.24	0–0.25
Average N_2O emitted (g N_2O m^{-2} d^{-1})	0.12	0.06	0.04

4. Conclusion

A detailed investigation at three full-scale landfill biocover systems using mature compost material as methane (CH_4) oxidation layers showed that N_2O emissions originating from processes occurring in the compost material constitute a tiny fraction (2.1–2.3 %) of the climate change mitigation gained by significant CH_4 oxidation occurring therein. Thus, there is little risk that the use of mature compost materials in biocover systems will negatively affect the greenhouse gas mitigation efficiency of these systems.

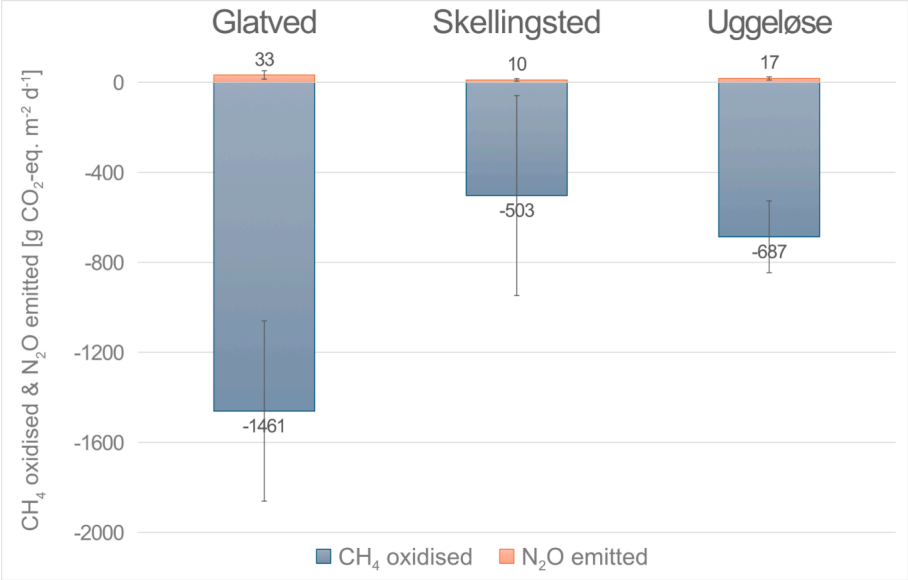

Fig. 1. Comparison of average CH_4 oxidised, calculated by the carbon mass balance, to the average measured surface N_2O emission given in CO_2-equivalents (g CO_2-eq. m^{-2} d^{-1}) for all three biocover systems. Values for N_2O (generated) and CH_4 (oxidised) are given above and below the columns.

Table 3
Comparison of total oxidised CH_4 in the actively loaded biofilters at Glatved and Uggeløse landfills to total emitted N_2O. Values are average values based several campaigns (Table 1).

	Glatved	Uggeløse
Input CH_4 (kg CH_4 d^{-1})	271.6	118.6
Integrated CH_4 emission (kg CH_4 d^{-1}) [a]	0.8	11.9
Oxidised CH_4 (kg CH_4 d^{-1})	270.8	106.7
Oxidised CH_4 (kg CO_2-eq d^{-1})	7311	2881
Emitted N_2O (kg N_2O-eq d^{-1})	0.17	0.12
Emitted N_2O (kg CO_2-eq d^{-1})	45.8	31.5
N_2O emitted in % of CH_4 oxidised [b]	0.63	1.1

[a] This number is based on an area-weighted average of all measured surface fluxes.
[b] Calculated in CO_2-eq based on global warming potentials for CH_4 and N_2O of 27 and 273, respectively, considering a 100-year period (Forster et al., 2021).

CRediT authorship contribution statement

Peter Kjeldsen: Writing – review & editing, Writing – original draft, Validation, Methodology, Data curation, Conceptualization. **Konstantinos Kissas:** Visualization, Investigation, Data curation. **Charlotte Scheutz:** Supervision, Project administration, Methodology.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Peter Kjeldsen reports financial support, administrative support, equipment, drugs, or supplies, and writing assistance were provided by Technical University of Denmark. Peter Kjeldsen reports a relationship with Technical University of Denmark that includes: employment. If there are other authors, they 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 material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2024.10.009.

Data availability

Data will be made available on request.

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