



Mitigation of methane and trace gas emissions through a large-scale active biofilter system at Glatved landfill, Denmark



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ABSTRACT

Biocover systems are a cost-effective technology utilised to mitigate methane (CH₄) and trace gas emissions from landfills. A full-scale biofilter system was constructed at Glatved landfill, Denmark, consisting of three biofilters with a total area of 3950 m². Landfill gas collected mainly from shredder waste cells was mixed with ambient air and fed actively into the biofilter, resulting in an average load of 60–75 g m⁻² d⁻¹ for CH₄ and 0.15–0.21 g m⁻² d⁻¹ for trace gases (e.g., aromatics, chlorofluorocarbons (CFCs), aliphatic hydrocarbons). The initial CH₄ surface screening showed uneven gas distribution into the system, and elevated surface concentrations were observed close to the gas inlet. Both positive and negative CH₄ fluxes, ranging from –0.36 to 4.25 g m⁻² d⁻¹, were measured across the surface of the biofilter. Total trace gas emissions were between –0.005 and 0.042 g m⁻² d⁻¹, and the emission flux of individual compounds were generally small (10⁻⁸ to 10⁻³ g m⁻² d⁻¹). Vertical gas concentration profiles showed that the oxidation of CH₄ and easily degradable trace compounds such as aromatics and aliphatic hydrocarbons happened in the aerobic zones, while CFCs were degraded in the anaerobic zone inside the compost layer. In addition, oxidation/degradation of CH₄ and trace gases also occurred in the gas distribution layer, which contributed significantly to the overall mitigation efficiency of the biofilter system. Overall, the biofilter system showed mitigation efficiencies of nearly 100% for both CH₄ and trace gases, and it might have the potential to work under higher loads.

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1. Introduction

Landfills containing organic waste produce landfill gas (LFG) consisting of 50–60% v/v methane (CH₄) and 40–50% v/v carbon dioxide (CO₂) as well as <1% v/v of trace components. CH₄ is an important greenhouse gas that contributes to climate change (Bogner et al., 2008). In addition, the trace compounds, albeit in small amounts, can pose a hazard to the environment and human health (Duan et al., 2021b), especially in the absence of an engineered gas collection and control system. Some species are often associated with odour problems and provoke opposition from surrounding residents (Fang et al., 2012; Lim et al., 2018; Palmiotto et al., 2014). Compounds such as benzene and vinyl chloride are known carcinogens (de Sá Borba et al., 2017; Durmusoglu et al., 2010; Yaghmaien et al., 2019), while chlorofluorocarbons (CFCs) can contribute to ozone depletion and global warming (Hodson et al., 2010; Liu et al., 2017; Wallington et al., 1994). Hence, the establishment of mitigation measures is necessary to reduce gas-emissions from landfills.

A common mitigation practice is to collect LFG for energy utilisation or flaring. However, this is not always feasible if LFG generation is low or gas quality is poor (e.g. at old sites or landfills receiving low-organic waste). As an alternative and cost-effective strategy, engineered biological mitigation systems (herein called 'biocover systems') are often preferred, in order to achieve enhanced microbial CH₄ oxidation and simultaneous trace gas mitigation (Huber-Humer et al., 2008). Such systems typically consist of a highly porous gas distribution layer overlain by an oxidation layer, which is constructed with organic-rich materials such as sewage sludge or green waste composts (Scheutz et al., 2009a). Depending on the design, they can be either passively loaded biocovers/biowindows that are often integrated within the landfill cover (Scheutz et al., 2014, 2011), or actively loaded open or closed bed biofilters (Fjelsted et al., 2020). In addition, air can be supplied into the system together with LFG to provide oxygen (O₂). CH₄ is oxidised to CO₂ by methanotrophs, while some trace gases such as sulphide, lower chlorinated compounds and aromatics can be co-metabolised and fully degraded to CO₂, due to the substrate specificity of methane monooxygenase enzyme (Scheutz et al., 2009a, 2004). In addition, if an anaerobic environment is formed, fully halogenated hydrocarbons can also be dechlorinated with

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the production of degradation products, which can further be degraded to CO₂ under aerobic conditions (Scheutz et al., 2010a, 2009b; Scheutz and Kjeldsen, 2003).

The Danish government launched a biocover subsidy scheme in 2015 in an effort to reduce greenhouse gas emissions (Danish EPA, 2015), and 178 million DKK was set aside to finance the establishment of approximately 100 biocovers at Danish landfills. To apply for the funding, landfill managers are first required to conduct a baseline study, which consists of two total CH₄ emission measurements and a surface screening (optional). If the baseline study shows total emissions of >2 kg CH₄ h⁻¹ (for sites with total emissions between 2 and 6 kg CH₄ h⁻¹, a surface screening is needed), the applicant may start to apply for grant for the implementation of a biocover project upon approval from the Danish Environmental Protection Agency. A biocover project generally consists of a detailed design based on the baseline study, establishment of a biocover system and follow-up CH₄ emission measurements, possible system improvement, four emission monitoring campaigns over two years and possible maintenance of the biocover system until the last subsidy-covered monitoring. By the end of 2020, 23 biocover systems had been established at different Danish landfills, including full-scale biocovers, biowindows and biofilters in different sizes.

The effects of lab- and full-scale biocover systems in mitigating CH₄ emissions have been well documented (e.g. Cassini et al., 2017; Roncato and Cabral, 2012; Scheutz et al., 2017, 2014, 2011). However, the mitigation efficiency of trace gases in biocover systems has not been sufficiently explored. Scheutz et al. (2009b) investigated the degradation of CH₄ and halocarbons in simulated landfill biocover systems containing compost materials, finding that all of the studied volatile organic compounds (VOCs) were degraded, thereby indicating the potential of compost materials to attenuate trace gas emissions from landfills. Barlaz et al. (2004) and Bogner et al. (2010) measured very small emissions from two biocover systems in the US, demonstrating that biocovers can reduce CH₄ and trace gas emissions from landfills, with or without gas collection systems. A more recent study conducted by Pecorini et al. (2020) evaluated the performance of two field-scale biofilter and biowindow systems (270 and 28 m²) in mitigating CH₄, trace gas and odour emissions from two Italian landfills. Their results showed that both systems performed well in removing trace gas and odour emissions (efficiencies >80%); nevertheless, the biofilter showed poor CH₄ oxidation efficiency (58%), indicating the need for further improvements. In Denmark, a few studies have been conducted focusing on the degradation of selected trace gases in landfill cover soils or inside the waste body (Scheutz et al., 2010a, 2010b, 2007, 2004), whereas no study was found on the trace gas mitigation in full-scale biocover systems. As the LFG produced from Danish landfills contains considerable amounts of trace gases (Duan et al., 2021c), and biocover systems have been established at many sites, it is necessary to investigate the attenuation of trace gases in such systems and quantitatively assess the mitigation efficiencies.

The objective of the present study was to quantify the efficiency of CH₄ oxidation and trace gas mitigation in a large-scale biofilter system actively loaded with LFG at a Danish landfill. Surface emission fluxes of CH₄ and trace gases were measured through static flux chambers, and attenuation processes were examined by installing gas probes. The efficiency of the biofilter system was subsequently determined based on measured surface emissions and input LFG loads.

2. Materials and methods

2.1. The landfill site

The biocover system was constructed at Glatved landfill, located in North-eastern Jutland, Denmark as part of the Danish biocover

subsidy scheme. Glatved landfill was established in 1981 and accepted mainly construction and demolition waste, refuse collections and non-recyclable bulky waste. Since 1997, Denmark banned the landfilling of organic and combustible waste, and thereafter Glatved received mostly non-combustible waste with low organic content and combustible wastes that are not allowed for incineration, such as contaminated soils, mineralised sewage sludge, construction waste, residues from street cleaning, asbestos, shredder waste, etc. A previous study has shown that significant quantities of landfill gas are produced at the landfill (Rosendal and Rolsted, 2015), especially from the shredder waste cells, which contained high concentrations of trace gases (Duan et al., 2021c). An LFG collection system was installed in the shredder waste cells with the initial purpose of utilising the gas for electricity production; however, this became impossible due to poor gas quality in the form of low CH₄ and high siloxane content. Hence, LFG collected from the shredder waste cell, together with excess gas from other parts of the landfill, was pumped into three biofilters constructed on cell I-C to remove CH₄ and trace gases (Fig. S1). A more detailed description on the disposal cells and LFG collection systems of Glatved landfill can be found in the [Supplementary information](#) and in a previous paper (Duan et al., 2021c).

2.2. Biofilter design

The biofilter system was constructed on cell I-C and consists of three biofilters (C1-3), each with a width of 14 m and a length ranging from 84 m (I-C1) to 100 m (I-C3) (Fig. S1), giving a total area of 3950 m². Prior to construction, part of the top cover soil on the waste cell was removed to flatten the surface, and soil walls were constructed around the three biofilters to minimize the horizontal migration of LFG, and to avoid the erosion and desiccation of the compost. The dimensions and cross-section of one of the biofilters are shown in Fig. 1. The gas distribution layer at the bottom was constructed with 0.3 m coarse gravel (32–64 mm grain size), and 13 slotted gas distribution pipes were embedded along the long side of the biofilter at 1 m intervals. On top of the gas distribution layer, a 0.8 m methane oxidation layer was placed, which consisted of mature compost originating from green waste. To feed the biofilters, LFG extracted from different waste cells was first directed to a gas manifold installed in a container, mixed with air and then equally distributed through a series of T-pipes and gas pipes into the three biofilters. It is expected that the mixing of O₂ with input LFG will enhance the efficiency of the biofilter system, since methanotrophs require oxygen to mitigate CH₄ as well as some trace compounds. The source of input LFG from different cells, as well as the LFG distribution system leading to the three biofilters, is illustrated in Table S1 and Fig. S2. Five field campaigns were conducted between March and June 2020. During the first campaign, screening of surface CH₄ concentrations was conducted for the three biofilters, and a test measurement of trace gas emission fluxes was conducted on biofilter I-C1. In the second to the fifth campaigns, CH₄ and trace gas emission fluxes, as well as vertical gas concentration profiles and temperature profiles, were monitored on biofilter I-C3. During the experimental period, the biofilters were gradually vegetated by different weeds, which at the end of the experimental period reached a height of up to 170 cm. Detailed descriptions of the measurements in each campaign are provided in the following sections.

2.3. Flow and composition of LFG entering the biofilters

The flowrate and composition of LFG entering the biofilter system were monitored inside the container through a series of manifolds (Fig. S2). In the first three campaigns, the combined flowrate of LFG into the three biofilters was kept constant at 78 m³ h⁻¹. For

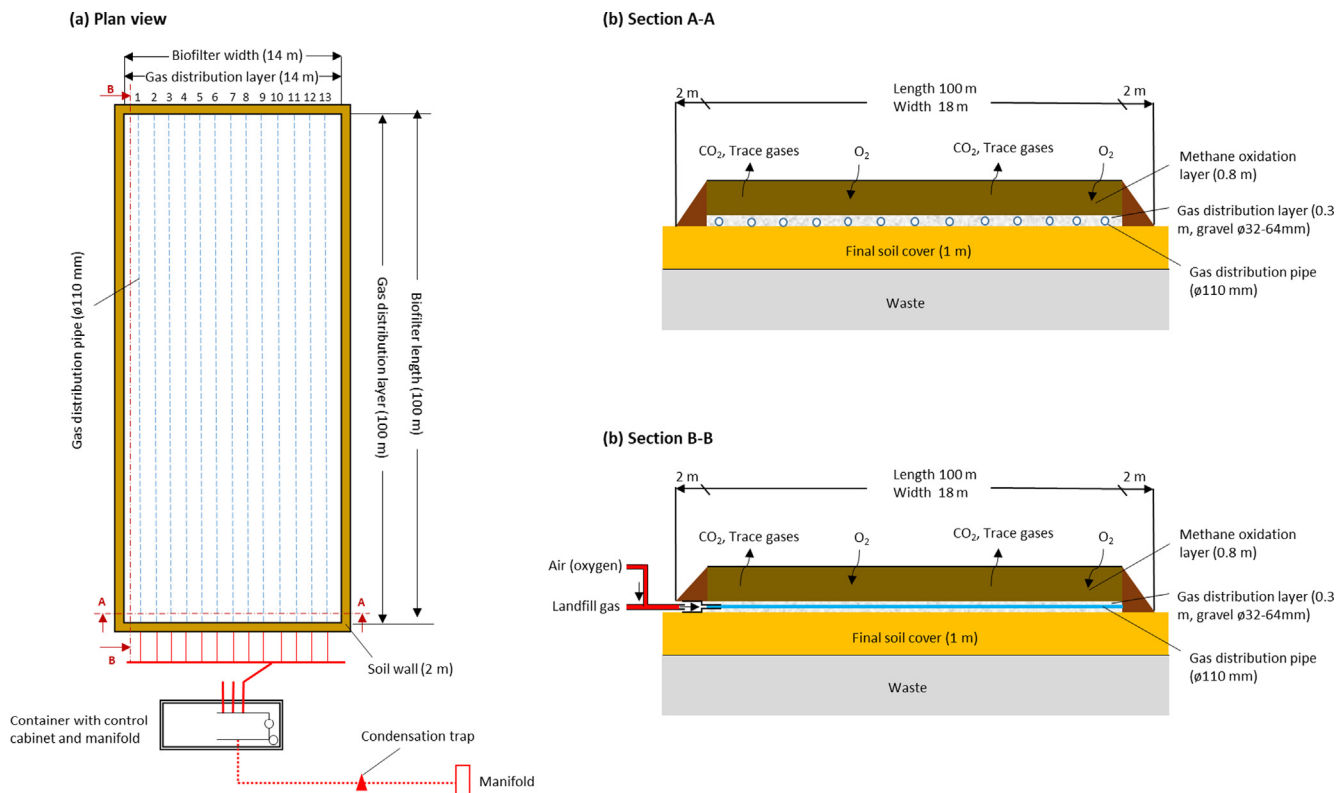


Fig. 1. (a) Plan view of biofilter I-C3, showing the gas distribution system with 13 slotted gas pipes connected to an un-slotted delivery pipe, (b) cross-section view of biofilter I-C3 and dimensions.

the fourth and fifth campaigns, the flowrate was increased to $95 \text{ m}^3 \text{ h}^{-1}$, which allows for monitoring biofilter efficiency under different loads.

LFG composition in the inlet of the biofilters (source gas) was monitored at the combined manifold before the gas was distributed between the biofilters (manifold no. 10 in Fig. S2). A Bio-gas 5000 portable gas analyser (Geotech, Warwickshire, UK) was used to measure the major gas components (CH_4 , CO_2 , O_2 , N_2) and H_2S on each measurement day throughout the four measurement campaigns. To determine the trace components in LFG, an ‘air toxics’ multi-sorbent tube (Markes, Lilantrisant, UK) was connected to the combined manifold, and 25 mL of the mixed LFG was extracted onto the sorbent tube via a low-flow SKC Pump (SKC Inc., USA) at a flowrate of 50 mL min^{-1} . The sample tube was then capped with Swagelok® end caps fitted with combined PTFE ferrules (Markes, Lilantrisant, UK) and stored in clean, airtight containers. After the field measurements, the sorbent tubes were transported to the laboratory and analysed within 3 days.

2.4. Surface screening and emission flux measurement of CH_4

To obtain a qualitative understanding about the spatial variability in surface emissions on the biofilter, surface screening was carried out during the first campaign by measuring point CH_4 concentrations at the surface of the three biofilters. A grid was created on each biofilter to divide the area regularly into 294–350 subareas of 4 m^2 each ($2 \text{ m} \times 2 \text{ m}$) (Fig. S3). Surface CH_4 concentrations were measured at the central spot of each subarea using a Laser One Portable Methane Leak Detector (Huberg, Italy). The probe was held 5 cm above ground for about 20 s, and CH_4 concentrations were recorded manually on a work sheet when the reading was stabilised. The weather conditions during the surface screening campaign were not optimal due to strong winds; although

the measurements were performed in periods with lower wind speed, high uncertainties are still expected in the surface screening results due to high spatial mixing.

Based on the surface screening results, it was decided to perform subsequent measurements at I-C3, as this biofilter had the largest dimensions and showed similar surface concentration distributions to the other two biofilters (Fig. S3). Quantitative measurements of CH_4 surface emission fluxes were conducted during the second to fifth campaigns, using a static flux chamber. The flux chamber was placed for every 4 m in the length, starting at 4 m and ending at 96 m. In the width, the chamber was placed at 4, 8 and 12 m, resulting in 72 measurement points equally distributed over the biofilter surface (sampling locations are shown in Fig. S4). The flux chamber used in this measurement was made of stainless steel and equipped with sampling ports as well as a manually operated fan to mix the gas inside the chamber during sampling. The chamber had a height of approximately 21 cm after installation and an inner diameter of 30 cm (total chamber volume $\sim 15 \text{ L}$). To measure the surface emission fluxes, a photoacoustic Multi Gas Monitor INNOVA 1512i (LumaSense Technologies, Denmark) was connected to the sampling port of the chamber, and five gas samples were taken over a period of five minutes to measure concentrations of CH_4 (one sample per minute). The concentration changes in each gas over time (dC/dt), and the chamber volume/chamber area ratio, were used to calculate the surface emission flux (Cassini et al., 2017; Mønster et al., 2019). The detection limit of the flux chamber measurements was $\pm 0.05 \text{ g m}^{-2} \text{ d}^{-1}$.

2.5. Installation of measurement stations

Based on the initial surface screening, five measurement stations were installed in selected grids representing high (E8 and F22), medium (C14) and low emission areas (D60 and B90)

(Fig. S3 and Fig. S4). Each measurement station contained one stationary flux chamber and five gas probes (Fig. S5). The flux chambers consisted of a metal drum (0.57 m ID) and a lid equipped with an electrical fan. The sides of the drum were pushed into the soil approximately 5–6 cm deep, and the lid was placed on the drum before each flux measurement. The dimensions of the flux chambers when installed in the field are reported in Table S3. Both CH₄ and trace gas emission fluxes were measured from the stationary flux chambers. To measure the CH₄ emission flux, the lid was placed on top of the drum and secured with hand clamps. The INNOVA was connected to the sampling port on top of the lid, and 10 gas samples were collected in approximately 10 min (one sample per min) for flux calculation. During sampling, the electrical fan was connected to a 12 V battery to ensure proper mixing of the air inside the chamber. Measurement of trace gas emission fluxes was conducted after CH₄ measurement. The lid was taken off the drum for 5–10 min before putting it back to allow equilibration of the headspace. A sorbent tube was then connected to the sampling port, and 200–300 mL gas was extracted from the chamber through an SKC pump operated at 50 mL min⁻¹, which represented time-zero concentration. Two more gas samples (100–300 mL each) were taken 30 and 60 min after re-installing the chamber, and the electrical fan was started about 5 min before each sampling to mix the gas. In all the campaigns, trace gas flux measurements were only conducted at four stations (E8, C14, D60 and B90) due to the limited number of sorbent tubes.

The gas probes were installed adjacent to the flux chambers to determine vertical gas concentration profiles inside the biofilter. At each measurement station, five stainless steel gas probes (5 mm ID, with slits over the lower 3 cm) were inserted to 10, 20, 30, 50 and 56–70 cm depths with about 40 cm lateral separations from each other and from the chamber (Fig. S5). The deepest probes were placed right on top of the gas distribution layer at different depths, due to varying compaction statuses of the compost layer at different locations. The main gas components (CH₄, CO₂, O₂, N₂) and H₂S at each depth were determined using the Biogas 5000. For trace gas measurement, 50–200 mL of gas samples were collected immediately onto sorbent tubes after main gas measurement at each probe. At each measurement station, the gas concentration profiles were measured at least 30 min before or after conducting the flux measurement. The probes were capped between different measurements. During the field campaigns, vertical temperature profiles were also measured at each station between 10 and 60 cm b.s., using a 12-channel Omega RDXL12SD temperature recorder (OMEGA Engineering, USA), and the meteorological conditions (temperature, barometric pressure, wind speed, etc.) were recorded based on data from an adjacent weather station. Temperature and barometric pressure ranges for each field campaign are presented in Table S4.

After collection of trace gas samples from flux chambers and gas probes, the sorbent tubes were capped with Swagelok® end caps fitted with combined PTFE ferrules and transported to the laboratory for analysis.

2.6. Trace gas sample analysis

The trace gas samples were analysed on a thermal desorption (TD) unit (TD 100Xr, Markes, UK) attached to a capillary gas chromatography (GC) and mass spectrometry detector (MS) (Models 6890/5973, Agilent Technologies, USA). The sorbent tubes were first heated to 320 °C for 12 min, during which time trace compounds were desorbed and refocused on a cold trap (U-T15ATA-2S, Markes, Lilantrisant, UK) maintained at 20 °C. The trap was then heated up quickly to 320 °C to inject the concentrated trace compounds into the GC column within 3 min (DB-624 UI, 60 m × 0.32 mm, 1.8 μm, Agilent). During thermal desorption, a

split ratio of 31:1 was applied when analysing high concentrated samples such as the source gas sample and the samples taken from deep gas probes, while no split was used for low-concentration samples (e.g., samples taken from flux chambers). The GC oven was operated at 35 °C for 5 min and then ramped up to 200 °C over 33 min. Trace compounds were identified by the MS in the selective ion monitoring (SIM) mode and quantified using the internal standard method. The analytical method was able to quantify 92 VOCs with detection limits between 0.01 and 2 ng for most compounds, and the precision and accuracy were generally within 20%. More information regarding the analytical method is available in Duan et al. (2021a).

2.7. Data analysis

Trace gas emission fluxes at each measurement station were determined by using the three gas samples taken at 0, 30 and 60 min after chamber installation. A linear regression was fitted to concentration versus time data to determine the concentration gradient as the slope of this relationship. In general, all three samples were included in the calculations; however, if R² < 0.65, only the first two samples (taken at 0 and 30 min) were used for flux calculation, since they were less affected by possible concentration buildup or vacuum formation and were thus more representative.

The overall CH₄ oxidation efficiency of the biofilter was determined by comparing the integrated CH₄ surface emissions from the biofilter to the average CH₄ load to the biofilter. Total CH₄ emissions from the biofilter surface were determined by multiplying the average of the 72 surface flux measurements with the total area of the biofilter. This was considered a reasonable step, due to the high number of measurement points and their equal distribution on the biofilter.

Trace gas emission fluxes measured at four measurement stations were scaled to the entire biofilter to calculate the overall biofilter mitigation efficiency (Huber-Humer et al., 2009; Yesiller et al., 2018). Based on the CH₄ surface screening results, the biofilter was divided into three categories: high emission areas (surface CH₄ concentration > 30 ppm), medium emission areas (surface CH₄ concentration between 10 and 30 ppm) and low emission areas (surface CH₄ concentration < 10 ppm) (Fig. S3). Scaling was conducted by using the relative areas of each emission category, which consisted of 6% high-emission, 41% medium-emission and 53% low-emission areas. The averaged emission flux measured at each monitoring station was thus multiplied by the corresponding total area of each emission category, and the overall trace gas emissions from all three biofilters were compared with the daily input into the biofilter to calculate trace gas mitigation efficiency. Note that the area of each emission category was determined based on one surface screening campaign and is therefore associated with uncertainty. In addition, the use of point measurements in the scaling as well as the sensitivity of the applied analytical method will add to the uncertainty in the estimated overall trace gas emission from the biofilter. Hence, the calculated trace gas mitigation efficiency is a rough estimate, and whole-site emission measurements are needed to estimate the biofilter efficiency more precisely.

3. Results and discussion

3.1. Major and trace gas composition of inlet LFG fed into the biofilters

The compositions of the inlet LFG fed into the biofilters measured in the exhaust manifold throughout the four campaigns are presented in Fig. 2. The major gas components turned out to be very stable, in spite of changes in barometric pressure between campaigns. On average, CH₄ and O₂ contents were 20% (v/v) and

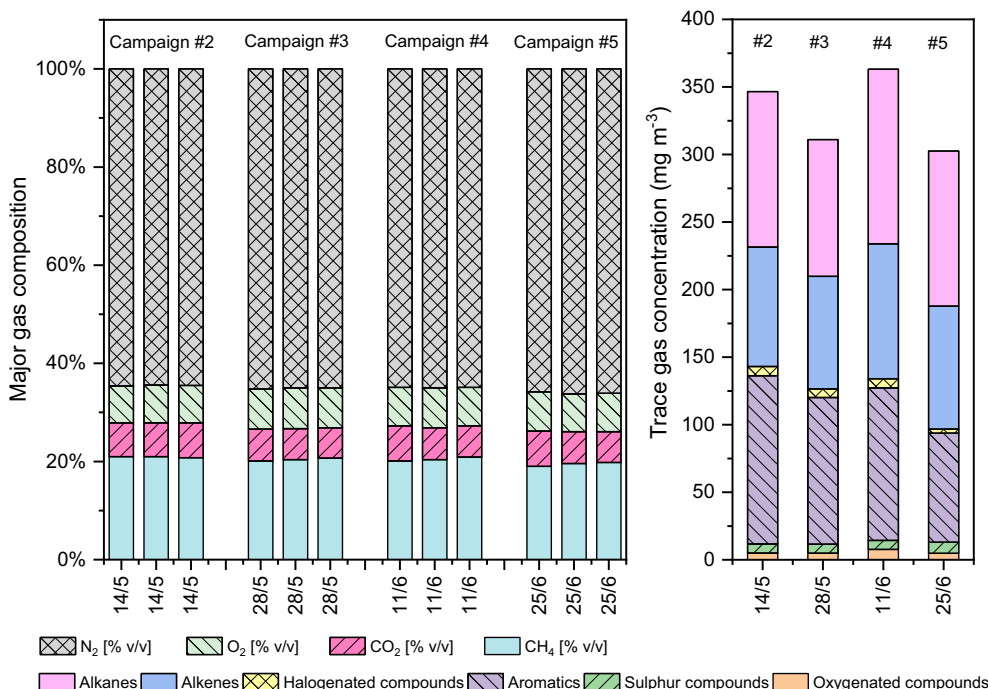


Fig. 2. Major and trace gas composition of inlet LFG fed into the biofilters.

7.9% (v/v), respectively, while a relatively low CO₂ content of 6.7% (v/v) was measured. Slightly larger fluctuations were observed in trace gas concentrations, which varied from 303 to 363 mg m⁻³ (concentration of individual compounds is presented in Table S5). Alkanes, alkenes and aromatics dominated total trace gas concentrations, whereas low concentrations of halogenated, sulphur and oxygenated compounds were measured. Toluene, ethylbenzene and xylenes (67–89 mg m⁻³) constituted >70% of the total aromatics (Table S5). Although the LFG was diluted before transporting it into the biofilters, the overall gas composition was similar to LFG measured at shredder waste cells in a previous study (Duan et al., 2021c), indicating that most of the LFG fed into the biofilters was produced from the shredder waste cells.

Based on the concentration of major and trace gases in the inlet LFG fed into the biofilters, the inlet LFG flowrate and the total area of the biofilter, the daily average load of the biofilter system was estimated at 61 g CH₄ m⁻² d⁻¹ and 0.16 g trace gas m⁻² d⁻¹ under a flowrate of 78 m³ h⁻¹. When the inlet LFG flowrate was increased to 95 m³ h⁻¹, average loads for the biofilter were 72 g CH₄ m⁻² d⁻¹ and 0.19 g trace gas m⁻² d⁻¹, respectively. The trace gas load was about one order of magnitude higher than that in a field-scale active biofilter system reported by Pecorini et al. (2020) (0.02 g m⁻² d⁻¹).

3.2. Surface emission fluxes of CH₄ and trace gases

The CH₄ emission fluxes obtained from the whole-surface measurement (72 points) varied between -0.36 and 4.25 g m⁻² d⁻¹, and most of them were below 0.5 g m⁻² d⁻¹ (Fig. 3), which are comparable with previous studies (Scheutz et al., 2017, 2011). The uptake of atmospheric CH₄ (negative fluxes) was observed in 19–46% of the measurements in different campaigns, slightly lower than those reported by Barlaz et al. (2004) and Bogner et al. (2010), who measured 52% and max. 60% negative fluxes at two biocover systems, although the magnitude of uptake rates were similar (10⁻²–10⁰ g m⁻² d⁻¹). The CH₄ surface emissions became slightly higher when the inlet LFG flowrate increased from 78 m³ h⁻¹ to

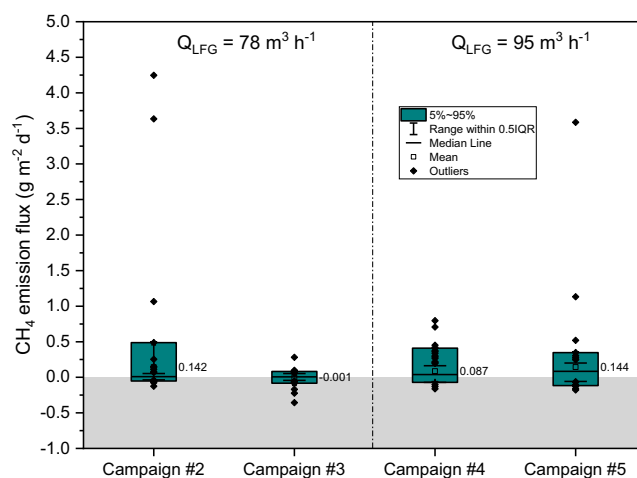


Fig. 3. CH₄ surface emission fluxes measured with static flux chambers across biofilter I-C3 during the four measurement campaigns (72 points equally distributed over the biofilter). The average CH₄ emission flux is shown for each individual campaign.

95 m³ h⁻¹, although no statistically significant difference was found between the emission fluxes measured under the two different flowrates. In addition, there was not a significant correlation ($p > 0.05$) between either ambient temperature or barometric pressure change and CH₄ flux (data not shown). Hence, the biofilter system seemed to be functioning well in removing CH₄ emissions, and the performance was not strongly affected by any change in external parameters.

The CH₄ and trace gas surface emission fluxes measured at the monitoring stations are summarised in Fig. 4, and all individual fluxes are presented in Table S6 and Table S7. Negative values indicate the uptake of atmospheric CH₄ and trace gases by the biofilter, as the concentration in chamber decreased over time. In general, the CH₄ fluxes were between -0.25 and 0.25 g m⁻² d⁻¹, except for a few cases at Station F22 (Table S6), and negative fluxes

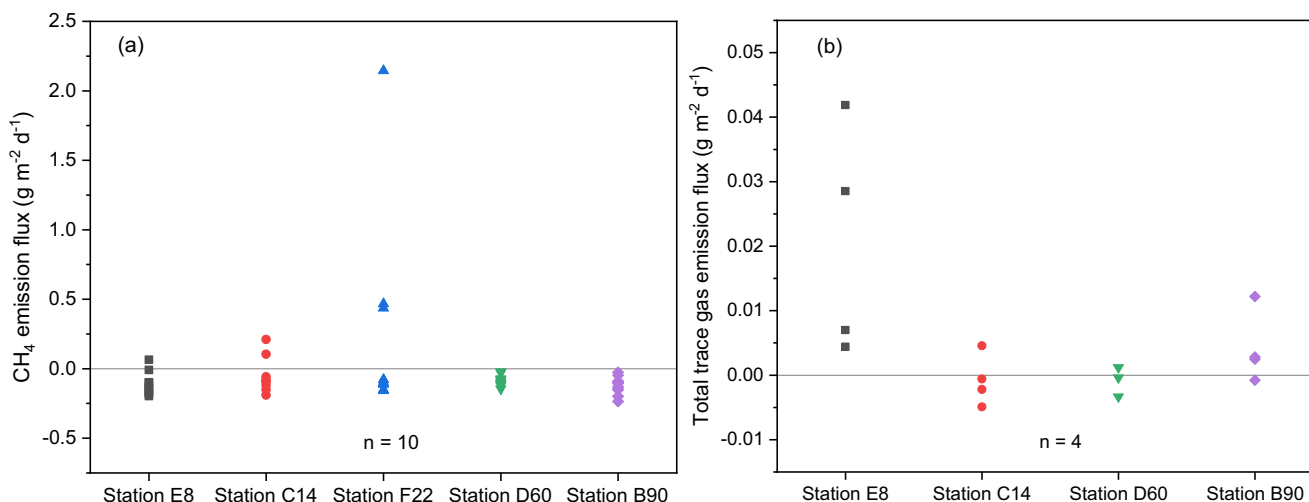


Fig. 4. CH₄ (a) and trace gases (b) emission fluxes measured at the five monitoring stations throughout four campaigns. The letter 'n' refers to the total number of measurements.

accounted for 88% of the total measurements. All the positive emission fluxes were measured at Stations E8, C14 and F22, which were close to the gas inlet to the biofilter, while Stations D60 and B90 only showed negative fluxes. The highest emission flux measured at Station F22 on one of the measurement days during the fourth campaign cannot be explained, but it was likely caused by an overload at that location during this specific measurement, indicating that the biofilter system was dynamic and the surface emissions varied over time. The overall distribution of CH₄ emission fluxes corresponded well with the surface screening results, suggesting uneven gas distribution into the compost layer.

Total trace gas emissions measured at the four stations were between -0.005 and 0.042 g m⁻² d⁻¹ (Table S7). The highest emission flux in each measurement campaign was always measured at Station E8, indicating the good representativeness of this spot as a high emission area. Nevertheless, low and mainly negative emission fluxes were observed at Station C14, even though it was installed in the medium emission area, while Station B90 showed occasionally elevated emissions. As previously mentioned, a biofilter is a dynamic system, and so LFG load at a specific spot can vary from time to time, thereby resulting in different surface emissions. In addition, the measurement stations were not placed in the exact same spots where surface screening was conducted, which was possibly another reason for the different surface emission patterns compared with surface screening.

The fourth and fifth campaigns showed slightly higher but statistically similar surface emissions of trace gases with the increased inlet LFG load (data not shown). In general, higher emissions were measured for alkanes and oxygenated compounds in all campaigns, while alkanes showed the lowest emissions (Table S7). This differed in terms of their percentage levels in the source gas, indicating that different trace compounds exhibited different oxidation/mitigation rates in the biofilter, and alkenes probably degrade faster than the other chemical groups. The emission fluxes of individual trace compounds were generally between 10^{-8} and 10^{-3} g m⁻² d⁻¹ (Table S7), with both positive and negative fluxes measured, which were comparable with previous studies conducted on biocovers or final soil covers in MSW landfills (Barlaz et al., 2004; Bogner et al., 2010; Scheutz et al., 2008, 2003). BTEXs showed higher emission fluxes (10^{-5} – 10^{-3} g m⁻² d⁻¹) compared to those reported by Scheutz et al. (2008) measured on the final soil cover of a French landfill, although they were still comparable to the surface emissions from two field biofilter systems in Italy

(Pecorini et al., 2020). CFC emission fluxes (in the order of 10^{-5} g m⁻² d⁻¹) were generally comparable with those measured by Barlaz et al. (2004), Bogner et al. (2010) and Pecorini et al. (2020), and negative fluxes accounted for 16% of the measurements.

Some oxygenated and halogenated compounds were not detected in the source gas but showed positive emission fluxes on the surface of the biofilter (e.g., ethanol, ethyl acetate, tetrachloride, trichloroethylene, tetrachloroethylene, etc.). Alcohols, ketones and aldehydes are typically produced during the oxidation of selected trace compounds such as alkanes and alkenes (Randazzo et al., 2020; Rojo, 2009; Tassi et al., 2009), leading to increased emissions of these compounds from the surface. In contrast, the 'increased' concentration of halogenated compounds was more likely the result of enhanced detection sensitivity, due to the larger sample volume as well as lower split ratios applied during the analysis of samples taken from flux chambers (as described in Section 2). As concentrations of the above-mentioned halogenated compounds were generally very low in the source gas and could not be quantified with the applied analytical method, it was not possible to determine the mitigation efficiencies for those compounds.

3.3. Vertical gas concentration profiles inside the biofilter

A representative example of gas concentration profiles from the measurement stations is shown in Fig. 5 (profiles for other campaigns are provided as Fig. S6–9). As evident in the figure, CH₄ content decreased steadily from the bottom of the compost layer towards the surface, and no CH₄ was measured in the upper 10 cm, except at Station E8 during the fourth campaign (Fig. S8). The CH₄/CO₂ ratio decreased from 55 to 70 cm b.s. and upwards at all stations, indicating CH₄ oxidation or the production of CO₂ through compost respiration (Fig. S10). Atmospheric O₂ penetrated the compost layer up to 30 cm below the surface, creating a broad aerobic zone that supported the rapid oxidation of CH₄ and trace gases. At the bottom of the profile, no or very low O₂ content (<2% v/v) was measured, even though the initial O₂ content in the inlet LFG was 7–8% v/v after mixing with air, thereby indicating that CH₄ oxidation was also occurring in the lower parts of the biofilter and in the gas distribution layer. In the fourth campaign, O₂ was present in the entire profile at Station B90, and CH₄ was below detection limits at all depths, suggesting that the gas load

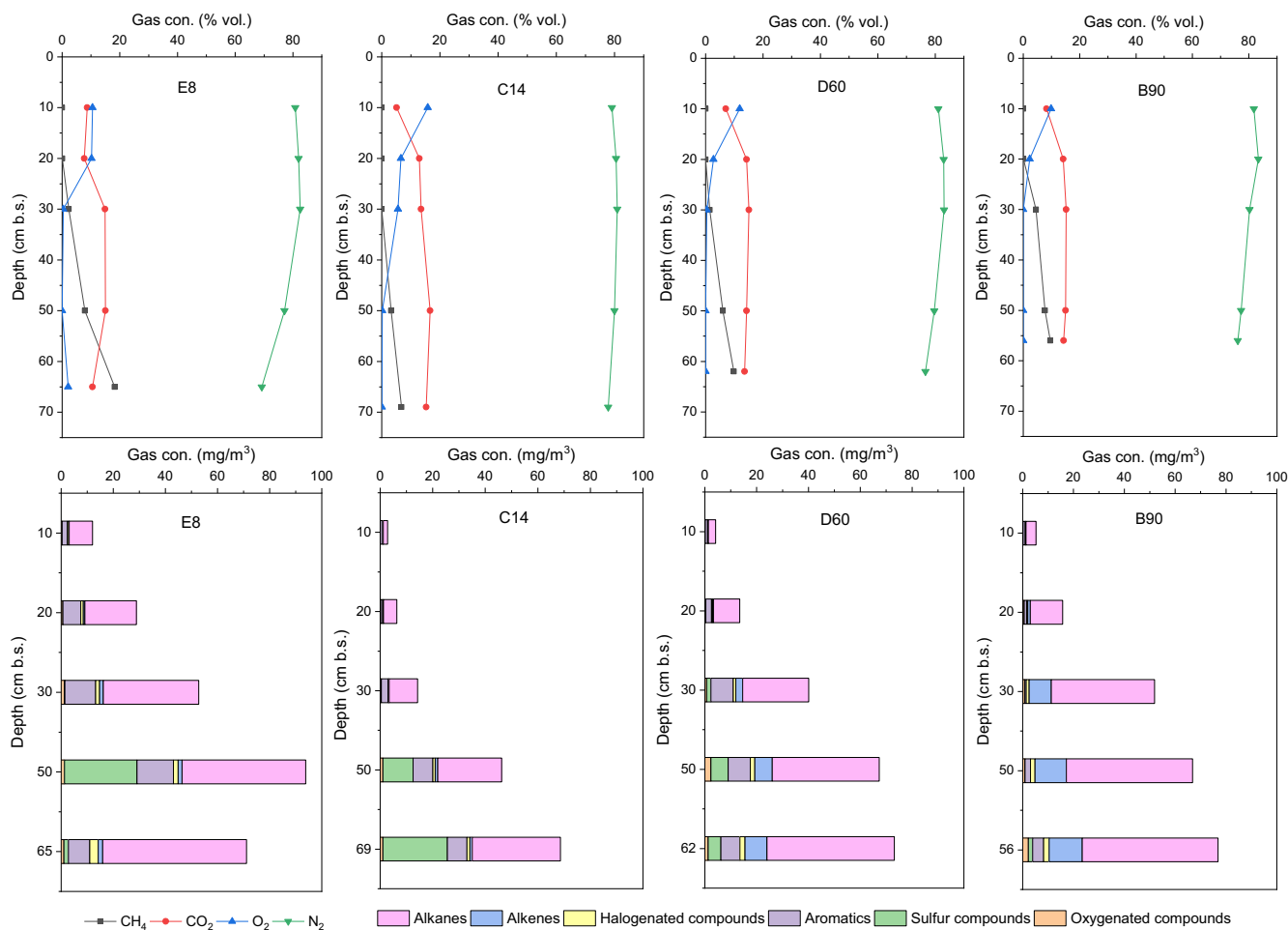


Fig. 5. Vertical gas concentration profiles at the four measurement stations from Campaign #2. Monitoring station E8 was placed closest to the biofilter gas inlet, while station B90 was the furthest away.

was very low at this spot. Combined with the high CH₄ content measured at Station E8 on the same day, which differed from other campaigns, the results further support the findings that the biofilter is a dynamic system and its gas distribution is uneven.

An anaerobic zone also developed inside the biofilter between 30 and 70 cm b.s., due to the absence of O₂. Large amounts of H₂S were produced at 50–60 cm b.s., probably due to the anaerobic degradation of aromatics and aliphatic hydrocarbons by sulphate reducing bacteria (Rojo, 2009; Spormann and Widdel, 2000; Widdel and Rabus, 2001), resulting in increased trace gas concentrations. The produced H₂S was then rapidly oxidised in the presence of O₂ or adsorbed onto compost material in the upper 30 cm. CFC reduction probably also occurred within this anaerobic zone, since it is well known that CFCs degrade under anaerobic conditions through reductive dechlorination (Scheutz et al., 2009b; Scheutz and Kjeldsen, 2003). The concentration profile of CFCs inside the biofilter showed a general decreasing trend from the bottom of the compost layer upwards (Fig. S11), and the reduction of CFCs in the lower 40 cm indicated anaerobic degradation, whereas the decreased concentration in the upper 30 cm probably resulted from dilution by atmospheric air. By comparing the surface emission fluxes of CFCs and their average loads into the biofilter, a reduction rate of 45–100% was found for CFC 12 and CFC 113, thus strongly supporting their anaerobic degradation inside the biofilter. However, it was not possible to distinguish the dechlorination by-products (e.g., chlorofluoromethane (HCFC-31)) in this study, due to limitations of the analytical method. A sharp decrease

in concentrations was observed for the other chemical groups in the upper 30 cm of the compost layer. At 10 cm b.s., total trace gas concentrations were mostly below 10 mg m⁻³, thus suggesting the strong oxidation of trace gases in the biofilter (dilution by atmospheric air was similar in 10–30 cm b.s., as suggested by a stable N₂ content).

It should be noted that CH₄ and trace gas concentrations measured at the deepest gas probes, which represented samples taken just above the gas distribution layer, were considerably lower than those in the inlet LFG. Elevated CO₂ concentrations (10.1–16.1% v/v) were observed at all stations at the bottom of the profiles, indicating CH₄ oxidation or the production of CO₂ through compost respiration (Scheutz et al., 2011). Fig. 6 shows how the CH₄/CO₂ ratio changes in line with increased distance to the biofilter gas inlet. As evident, the CH₄/CO₂ ratio was significantly lower at Station E8 compared to the source gas, and it decreased gradually as the LFG transported further into the biofilter. While the decreased CH₄/CO₂ ratio at the starting point (Station E8) was probably due to increased CO₂ concentration from compost respiration, the overall decreasing trend of CH₄/CO₂ ratio along the biofilter strongly suggests that CH₄ oxidation had already started in the gas distribution layer – as observed in other biocover systems (Scheutz et al., 2014, 2011). Most of the CH₄ in LFG might have been oxidised before reaching the end of the biofilter, which could explain the low CH₄ concentrations measured at Station B90. The low CH₄/CO₂ ratio at Station C14, on the other hand, was likely to be a result of low LFG load, due to uneven gas distribution.

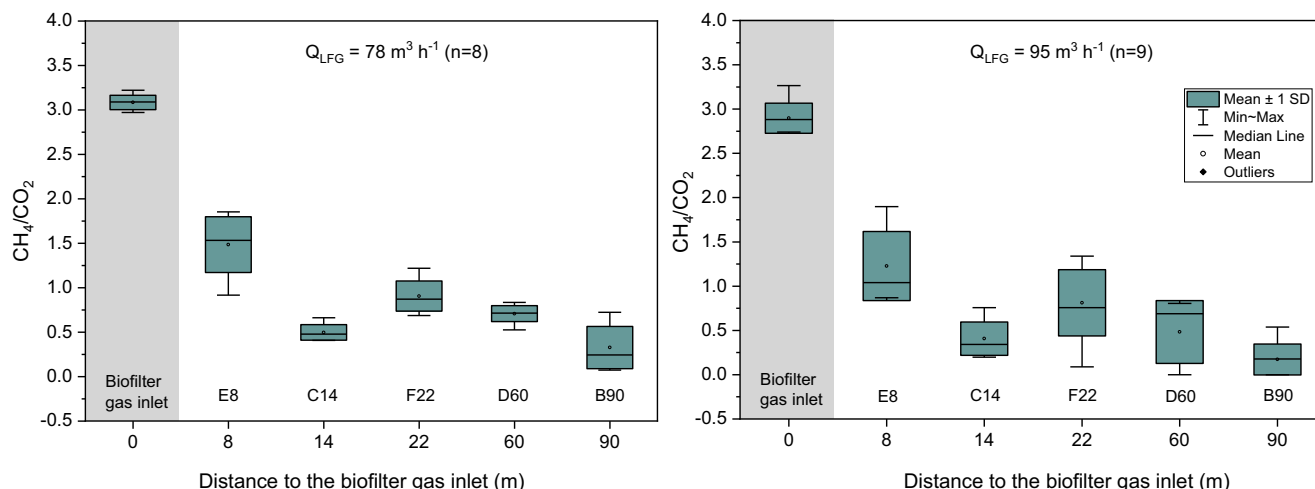


Fig. 6. CH_4/CO_2 ratio change in the gas distribution layer in line with increased distance to the biofilter gas inlet and different flowrates. The letter 'n' refers to the number of samples detected at a particular distance.

Total trace gas concentrations were generally 2–3 times lower than the inlet LFG, which might be attributed to dilution with atmospheric air or mitigation in the gas distribution layer. The CH_4 oxidation process reduces gas volume and can potentially create low pressure, which in turn may pull in atmospheric air (Kjeldsen, 1996; Scheutz and Kjeldsen, 2003), and the trace gas concentrations can be corrected for dilution by dividing with $[1 - (\text{N}_{2, \text{measured}} - \text{N}_{2, \text{inlet gas}})/\text{N}_{2, \text{air}} - 78\%]$ (Scheutz and Kjeldsen, 2003; Scheutz et al., 2008). Calculations for the dilution and up-concentration factors are provided in the [Supplementary Information](#) (Section S7), and the results are shown in Table S8. Trace gas concentrations after correction ($11\text{--}155 \text{ mg m}^{-3}$) were still far lower than the source gas, which clearly suggests that trace gas attenuation occurred in the gas distribution layer. As the gas distribution layer consisted of coarse gravel with a particle size of 32–64 mm, fine particles of the compost material might drop into the pore spaces and bring microorganisms into the gas distribution layer. The steady supply of LFG provided nutrients (CH_4 and trace gases) and O_2 that could support microbial growth, while water infiltrating from precipitation prevented material desiccation inside the system. Hence, the gas distribution layer acted as a small biofilter facilitating the removal of more than half of the trace compounds in LFG. It is also worth mentioning that the fourth and fifth campaigns showed elevated trace gas concentrations in the deepest gas probes as a result of increased LFG load, whereas such an increase was not seen for CH_4 . It seems that the gas distribution layer was more sensitive to high trace gas concentrations, and mitigation efficiencies tended to decrease under increased loads.

3.4. Vertical temperature profiles inside the biofilter

Temperature profiles in the biofilter system at the five measurement stations are shown in Fig. 7. Significantly elevated temperatures were measured at all stations in comparison to ambient temperatures, and overall temperatures decreased in line with increasing distance to the biofilter gas inlet. Temperatures at Stations E8, C14 and F22 increased in line with depth, with the highest temperatures reaching up to 60°C at 60 cm b.s. In contrast, Stations D60 and B90 had lower temperatures ($32\text{--}43^\circ\text{C}$), which peaked at a depth of between 30 and 40 cm. The different temperature profiles are likely to be a result of CH_4 oxidation, compost respiration in combination with heat transport from input LFG (Scheutz et al., 2014). CH_4 oxidation by methanotrophs is an exothermic process (Scheutz et al., 2009a), and the higher temperatures at the first three stations indicated higher CH_4 oxidation

activities, which was consistent with previous observations that most CH_4 oxidation happens in the first part of the biofilter. Similarly, the increased temperatures in Campaigns #4 and #5 were probably a result of enhanced CH_4 oxidation in the biofilter, due to increased LFG load into the system. Heat from input LFG might also have contributed to the elevated temperatures. The temperatures of input LFG were not measured, but they are expected to be much higher than normal soil temperatures (Coccia et al., 2013), which could explain the high temperatures at the bottom of Stations E8 and C14. The influence of LFG temperature was more pronounced at locations close to the biofilter gas inlet, as heat would be lost during transport of LFG in the gas distribution layer. At Stations D60 and B90, heat was probably only generated from CH_4 oxidation and compost respiration, and thus the highest temperatures were observed at 30–40 cm b.s., where most CH_4 oxidation occurred.

3.5. CH_4 and trace gas mitigation efficiency of the biofilter

The overall CH_4 oxidation efficiency of the biofilter was above 99%, determined by comparing the measured CH_4 inlet load to the biofilter with the integrated CH_4 surface emission from the biofilter, based on 72 point surface flux measurements (Table 1), indicating that the biofilter system was performing very well in mitigating CH_4 emissions from the landfill site. The single point CH_4 oxidation efficiencies at the 72 points as well as the five measurement stations were between 93% and 100%, which were generally higher than several other biocover systems reported in previous studies (Scheutz et al., 2017, 2014, 2011). It seems that the biofilter has the potential to oxidise a much higher CH_4 load, if needed.

Total surface emissions of trace gases from the biofilter system were estimated at between -1.1 and 27 g d^{-1} , which corresponded to an overall mitigation efficiency of 97–100% (Table 1) – higher than those reported by Pecorini et al. (2020) in two field-scale biofiltration systems. The system showed neglectable emissions of CFCs (0.07 g d^{-1} or 0.024 kg yr^{-1}), indicating the efficient reduction of those compounds inside the biofilter, although the load was also small. In addition, the biofilter showed an uptake of atmospheric BTEXs (-1.6 g d^{-1}) when operated at a lower LFG load, and even with increased load, the emission was still very small (0.1 g d^{-1}). Compared with the high input amount of BTEXs into the system ($152\text{--}205 \text{ g d}^{-1}$), it is clear that the biofilter is very efficient in reducing hazardous compound emissions from the landfill, as also documented in previous studies (Scheutz et al., 2004;

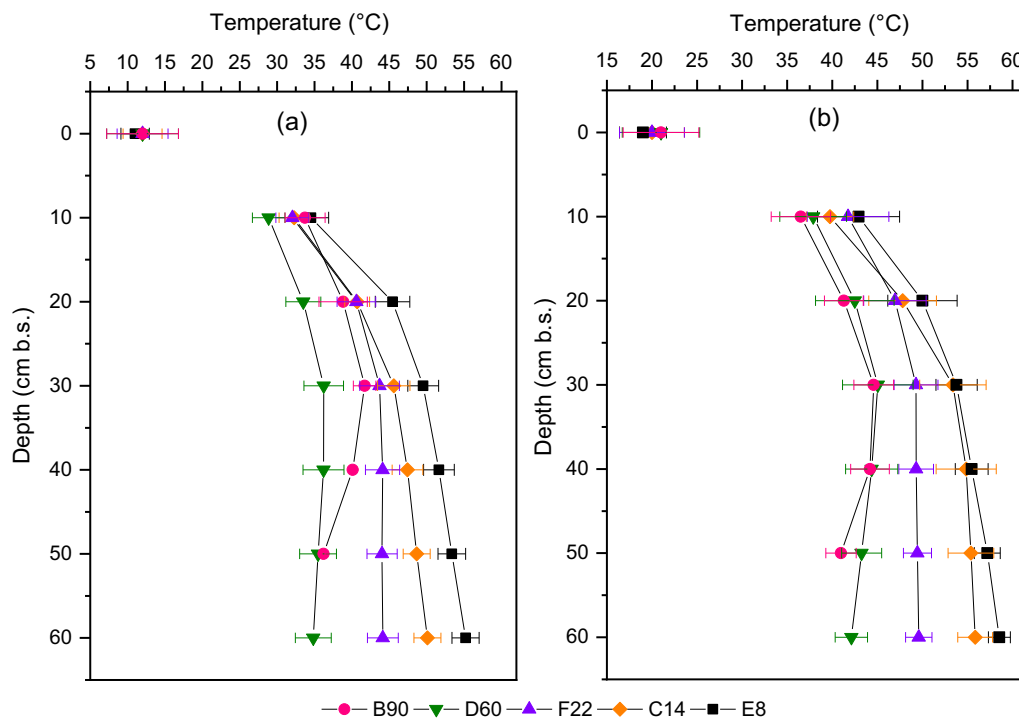


Fig. 7. Vertical temperature profile inside the compost layer (a) average of Campaign #2 and Campaign #3; (b) average of Campaign #4 and Campaign #5. Temperature at 0 cm indicates average ambient temperature during the measurement campaigns.

Table 1
Overview of biofilter CH₄/trace gas oxidation/mitigation efficiencies and CH₄/trace gas oxidation/mitigation rates obtained during four measurement campaigns.

Campaign	Inlet load		Total surface emission		Oxidation/mitigation rate		Oxidation/mitigation efficiency %	
	g CH ₄ d ⁻¹	g trace gas d ⁻¹	g CH ₄ d ⁻¹	g trace gas d ⁻¹	g CH ₄ d ⁻¹ (g m ⁻² d ⁻¹)	g trace gas d ⁻¹ (g m ⁻² d ⁻¹)	CH ₄	Trace gases
#2	244,806	649	560	2.7	244,246 (62)	646 (0.16)	99.8	100
#3	238,568	582	-5.2	-1.1	238,573 (60)	583 (0.15)	100	100
#4	291,514	828	343	27	291,171 (74)	801 (0.20)	99.9	96.8
#5	277,270	690	567	4.1	276,703 (70)	686 (0.17)	99.8	99.4

Scheutz and Kjeldsen, 2003), and it even has the ability to remove air pollutants in ambient air emitted from other sources (e.g. BTEXs emissions from landfill operation vehicles). However, when examining mitigation efficiency at individual measurement stations, low removal efficiencies were obtained for benzene and toluene at Station E8 during the second campaign (66% and 56%). In addition, low trace gas mitigation efficiency (79%) was found at the same station during Campaign #4, thus indicating a risk of high emissions from biofilter hotspots. Hence, additional measures may be needed to distribute the gas evenly into the biofilters, and to avoid the formation of emission hotspots in order to obtain an even higher mitigation efficiency.

4. Conclusions

This study investigated the efficiency of a large-scale biofilter system in mitigating methane (CH₄) and trace gas emissions at a Danish landfill. Landfill gas (LFG) produced mainly from shredder waste cells was mixed with air and fed into three biofilters with a total area of 3950 m², and the average loads were 60–75 g m⁻² d⁻¹ for CH₄ and 0.15–0.21 g m⁻² d⁻¹ for trace gases. Based on five field campaigns over four months, CH₄ emissions from the biofilter varied from -0.36 to 4.25 g m⁻² d⁻¹, with atmospheric uptake measured in 32% of the measurements. For trace gases, both posi-

tive and negative fluxes were measured, ranging between 10⁻⁸ and 10⁻³ g m⁻² d⁻¹, and neither CH₄ nor trace gas emissions increased significantly when the LFG load was increased. In general, higher emissions were measured close to the gas inlet, thereby indicating uneven gas distribution into the biofilter. Vertical gas profiles indicated that both aerobic and anaerobic zones formed inside the biofilter. CH₄ and easily degradable trace compounds such as alkenes and aromatics were oxidised in the aerobic zones, whereas chlorofluorocarbons (CFCs) were degraded anaerobically in the compost layer where O₂ was not available. In addition, the oxidation/mitigation of CH₄ and trace gases also occurred in the gas distribution layer, most likely due to the growth of microorganisms brought in by fine particles of compost. Overall, the biofilter showed a high CH₄ oxidation efficiency of about 100% and a trace gas mitigation efficiency of above 96%, indicating that the biofilter system was very effective in reducing emissions from the landfill and even had the potential to work under a higher load, if needed.

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 material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2021.03.023>.

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