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## Potential and Limitations of Passively Vented Biofilters for the Microbial Oxidation of Landfill Methane

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### 1 Introduction

In Europe, 30 % of the anthropogenic methane emissions are estimated to originate from landfills, constituting the second largest source (EEA 2006). The microbial oxidation of methane in biofilters or cover soils represents a viable and sustainable option for the treatment of landfill methane emissions that do not meet the calorific requirements for energetic utilization or flaring. Biofiltration as a means for the abatement of anthropogenic methane emissions may be applied at landfills in the initial phase of operation, old landfills or sites containing material of low gas generation rate.

Gas supply to biofilters may be active or passive. In active systems, gas is forcibly extracted from the landfill and can be supplied to the biofilter at favourable flow rates (e. g. Streese and Stegmann, 2003; Du Plessis et al., 2003, Haubrichs and Widmann, 2006). These systems are usually housed and allow for an operation at favourable conditions of water content and temperature. Passive systems receive landfill gas as a result of a positive pressure gradient between the landfill and the atmosphere and are operated as 'robust' open bed, sometimes with no additional heating or irrigation (e. g. Straka et al., 1999; Dever et al., 2005; Gebert and Gröngröft, 2006a). Capital and operational costs are therefore lower than for active systems. However, as operational conditions are not as manageable, passive biofilter operation is subject to particular constraints which need to be considered to warrant successful methane degradation.

Based on the results from a four-year research project that investigated the potential of a biofilter on a passively vented landfill in Northern Germany, both in field scale application and in supplementary laboratory experiments, this paper discusses fundamental issues connected to the particular case of passive biofiltration. To this end, previously published findings (Gebert et al., 2003; Gebert and Gröngröft 2005; Gebert and Gröngröft, 2006 a, b) are reviewed.

### 2 Materials and methods

#### 2.1 Biofilter setup

Figure 1 schematically illustrates the arrangement of gas wells, emission monitor-

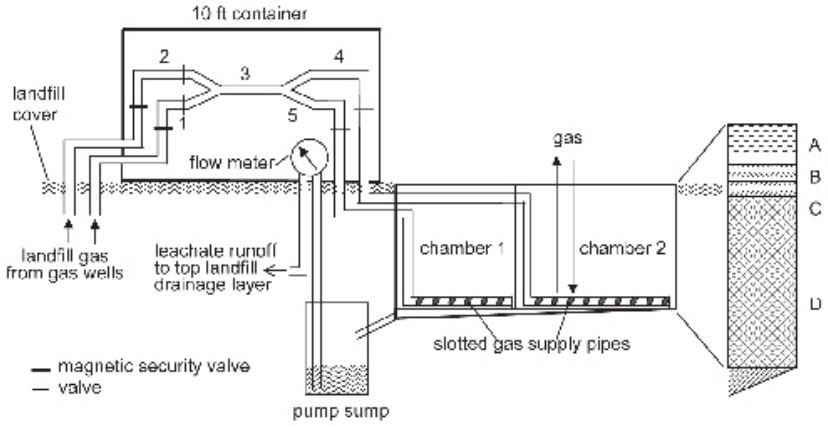



**Fig. 1:** Gas well and biofiltration unit setup at the harbour sludge disposal site Francop. 1-5, A-E: see text.

ing unit, and the biofilter, as operated from October 1999 to date.

Landfill gas from two gas wells venting different sections of the landfill (1, 2) was combined (3) and then split up again to be fed to two biofilter chambers (4, 5) at 6 $m^3$ and 9 $m^3$ size. The biofilter is an upflow system consisting of five layers (top to base) inside a 15 $m^3$ polyethylene container with an inclined base: A = topsoil (10 cm), covered with grass vegetation, B = sand (1.5 cm), C = gravel (1.5 cm), D = crushed porous clay (67 cm) and E = gravel for water drainage (10-30 cm). Landfill gas is distributed via horizontal and parallel slotted gas supply pipes embedded in the top centimetres of the drainage gravel layer (E). Oxygen is supplied by diffusion from the atmosphere via the biofilter surface. As the biofilter is integrated into the landfill cover without supplementary heating, biofilter temperature follows ambient temperature. The moisture content of the filter material solely relies on precipitation and moisture content of the landfill gas. More details on the filter material are given in Gebert et al. (2003). Information on sensor equipment and configuration is given in Gebert and Gröngröft (2006 b).

### 2.2 Experimental procedures

For all laboratory batch assays, freshly sampled biofilter material from the top 10 cm of the porous clay was used. Methane consumption rates were calculated from linear regression analysis of the change of headspace methane concentration over time (see Gebert at al., 2003). Information on the modelling of diffusive oxygen ingress are provided in Gebert and Gröngröft (2005), details on field measurements of biofilter performance are given in Gebert and Gröngröft (2006 a).

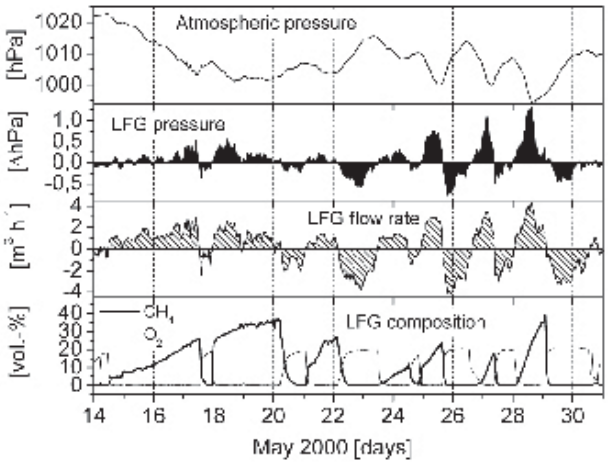



**Fig. 2:** Atmospheric pressure, landfill gas pressure, flow rate and composition in May 2000 as measured in the gas supply system (see Figure 1). LFG = landfill gas. Δ = difference.

## 3 Results and discussion

### 3.1 Features specific to passive landfill and biofilter ventilation

The curves in Figure 2 show examples of how the dynamics of atmospheric pressure, landfill gas pressure, gas flow rate and composition relate to each other, as measured in the biofilter gas supply piping.

Pressure differences in the gas supply pipe ("LFG pressure against the atmosphere") were small at -0.8 to 1.6 hPa during the entire period of operation. Figure 2 shows that they regularly alternated between over-pressure and negative pressure compared to atmospheric pressure. Over-pressure was related to falling atmospheric pressure and resulted in the emission of gas from the landfill via the biofilter into the atmosphere (positive values for "LFG flow rate"). Conversely, negative pressure in the gas supply system corresponded to phases of rising atmospheric pressure, drawing atmospheric air via the biofilter into the landfill (negative values for "LFG flow rate"). This was also reflected by the gas composition in the biofilter supply pipe (Figure 2, "LFG composition"): with decreasing atmospheric pressure, subsequent increasing landfill gas pressure and positive gas fluxes, the methane content increased steadily (maximum concentration in the years of 2000-2002: 56 vol.-%) while the oxygen content dropped to zero. When the flux direction changed, methane concentration in the supply pipe rapidly fell to zero whereas oxygen reached atmospheric concentration. Hardly any phases of constant and high positive fluxes and constant methane concentrations were observed. Gas emission usually collapsed after the methane content had reached

its maximum and pressures had levelled.

The data indicate that passive emission is driven by momentary over-pressure inside the landfill, which is not sustained over longer periods of time. Correspondingly, biofilters connected to a passively vented gas drainage system receive both highly variable methane loads and methane concentrations. For the biofilter under study, the methane load varied between 0 and 256 g $h^{-1}$ $m^{-3}$.

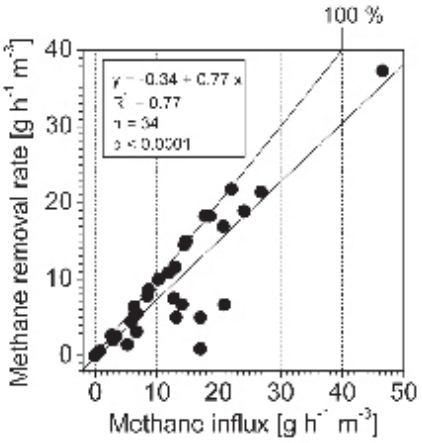



**Fig. 3:** Biofilter methane removal rates as determined from surface emissions (manual chambers). The 100 % line indicates complete oxidation of landfill methane entering the biofilter.

### 3.2 Potential of passively vented biofilters

Figure 3 shows methane influx and emission rates as determined by manual surface emission measurements (closed chamber):

Methane removal rates increased with an increasing influx of methane into the biofilter. For the observed cases, the limit of filter performance was therefore not reached and consequently its methane oxidation potential not fully tapped. The maximum methane removal rate

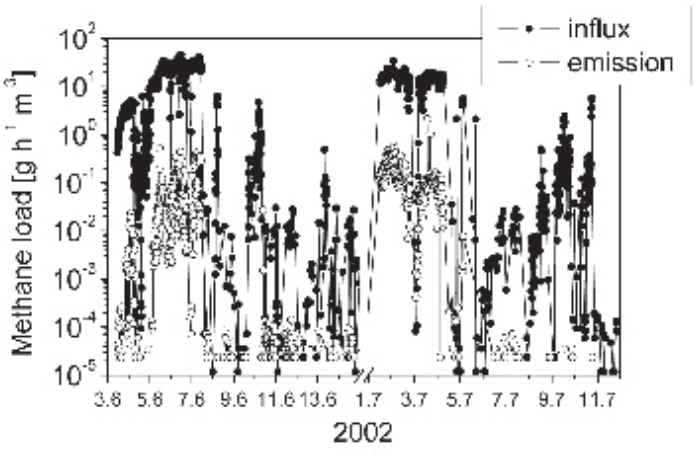



**Fig. 4:** Methane influx and emissions as measured with an automated closed chamber. Note the logarithmic y-scaling. Period covered by axis break = no data recorded.

observed for this data set was 37.3 g $h^{-1}$ $m^{-3}$.Methane removal rates could not be related to any potential factors of influence tested (e.g. temperature, gas flux). Rather, methane oxidation was primarily determined by the availability of methane itself. The temperature range for the dataset depicted in Figure 3 covered a wide span from 3 to 24 °C.

Figure 4 shows methane influx and emissions as determined by the automated chamber measurement installed in the summer of 2002. Methane emissions from the biofilter during the summer of 2002 were continuously more than two orders of magnitude lower than methane input into the biofilter. Methane oxidation rates were thus close to 100 %. Maximum input and therefore maximum absolute methane removal rates were around 80 g $CH_4$ $h^{-1}$ $m^{-3}$. Methane emissions in general only traced LFG emission dynamics in case of higher influxes and stronger influx variability.

The performance of the investigated biofilter is within the same order of magnitude as reported by other authors (Table 1). However, it must be noted that the removal rates observed here were achieved in the field and under in situ conditions, thus at lower temperatures and also not under conditions of controlled methane and oxygen supply.

**Tab. 1:** Methane removal rates by different actively vented biofilters.

| Author | Filter material | Temperature [°C] | Methane removal [g $h^{-1}$ $m^{-3}$] |
|---|---|---|---|
| Mennerich (1986) | compost | 30 | 38 |
| Sly et al. (1993) | glass tubes | ? | 21 |
| Figueroa (1996) | compost | 43 | 50 |
| Streese and Stegmann (2003) | compost | 30 | 65 |
| Wilshusen et al. (2004) | compost | room temp. | 4.1 g $h^{-1}$ $m^{-2}$ |

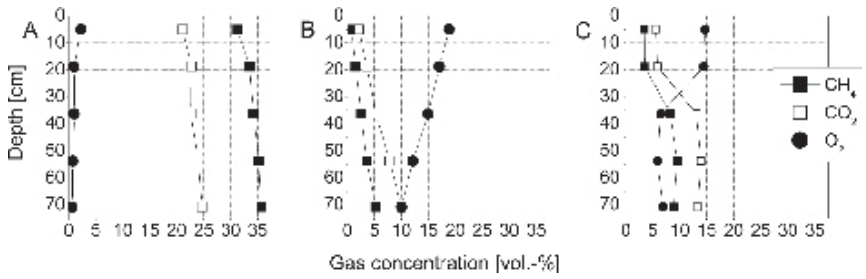



**Fig. 5:** Typical profiles of gas composition in the biofilter.

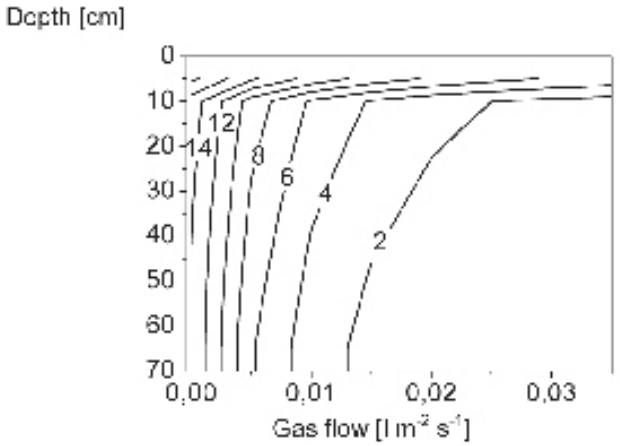



**Fig. 6:** $O_2$-concentration [vol.-%] with biofilter depth as dependent on landfill gas flow rate.

| | | | |
|---|---|---|---|
| Melse and Van der Werf (2005) | compost/perlite | 12 | 11-15 |

### 3.3 Limitations of passively vented biofilters

The emission dynamics described above caused variable vertical gas distribution within the biofilter (Figure 5), and subsequently induced variable conditions for methanotrophic activity, especially with respect to oxygen supply.
In situation A, hardly any methane oxidation is possible due low oxygen concentrations at high methane concentrations across the entire biofilter depth. In situation B, both methane and oxygen are available. Graph C depicts an intermediate situation with the development of a methane oxidation zone in the top 30 cm as often observed for landfill cover soils.

Using a simple box model, eight simulation runs were carried out to estimate the efficacy of diffusive oxygen input into the biofilter over a range of advective landfill gas flow between 0 and 0.035 l $m^{-2}$ $s^{-1}$. The latter equals 0.13 $m^3$ $m^{-2}$ $h^{-1}$, corresponding to the mean value of LFG charge to the biofilter in the field. A biofilter layering of 10 cm topsoil and 60 cm of porous clay material were assumed (for details see Gebert and Gröngröft, 2005). Figure 6 shows the isolines of oxygen concentration as dependent on the extent of advective LFG flow after a period of 24 hours. Note that the model assumes that the gas flow rate is continuous.

Oxygen concentrations >10 vol.-% were only reached at simulated LFG flow rates <0.0025 l $m^{-2}$ $s^{-1}$ (equals 0.009 $m^3$ $m^{-2}$ $h^{-1}$). On the investigated site these occur in only 14 % of all cases (avg. flow rate = 0.11 $m^3$ $m^{-2}$ $h^{-1}$, max. = 0.84 $m^3$ $m^{-2}$ $h^{-1}$), suggesting that principally oxygen-limited conditions prevail. Under conditions

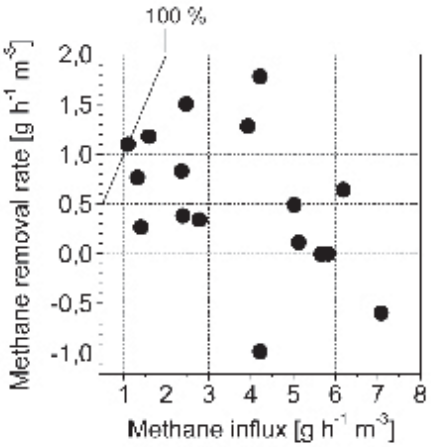



**Fig. 7:** Methane influx and methane removal rate as determined from mass methane backflow after LFG flux reversal. The 100 % line indicates complete oxidation of landfill methane entering the biofilter.

of continuous advective landfill gas supply, only very low gas flow rates would warrant effective diffusive oxygen ingress from the atmosphere into the biofilter.

However, the concentration profiles as measured in situ (compare Figure 5) and also the frequency distribution of oxygen concentrations in different depths, show that the actual input of oxygen into the biofilter is much higher than could be expected from the diffusive potential at the given flow rates. This is attributed to the high sensitivity of landfill gas emission (Figure 2) to atmospheric pressure variations, inducing an oscillation of the gas column in the biofilter and thereby an advective influx of atmospheric oxygen. An effectual oxygen supply to the deeper layers is also suggested by the very high density and activity of the methanotrophic population across the entire filter profile (Gebert et al., 2003) as well as in situ methane removal rates of up to 80 g $h^{-1}$ $m^{-3}$ (Figure 4).

Figure 7 shows methane influx and methane removal rates as determined for conditions of longer and steady gas emission. The data indicate that under these conditions biofilter performance is poor, despite the fact that before flux reversal methane influx into the biofilter was a lot less than for the cases shown in Figure 3. For the cases reported, methane concentrations fed to the biofilter on average were as high as 35 vol.-% (maximum: 50 vol.-%), it can thus be ruled out that the poor performance is a result of unfavourable kinetic conditions with respect to methane ($K_M$ = 15.1 µM, equals 1.2 vol.-% in the gaseous phase at field capacity water content, see Gebert et al., 2003).

The failing methane oxidation performance a is assumed to be due to insufficient oxygen supply to the biofilter methanotrophic organisms, occurring when the diffusive oxygen flux is lower than the counterdirective advective LFG flow

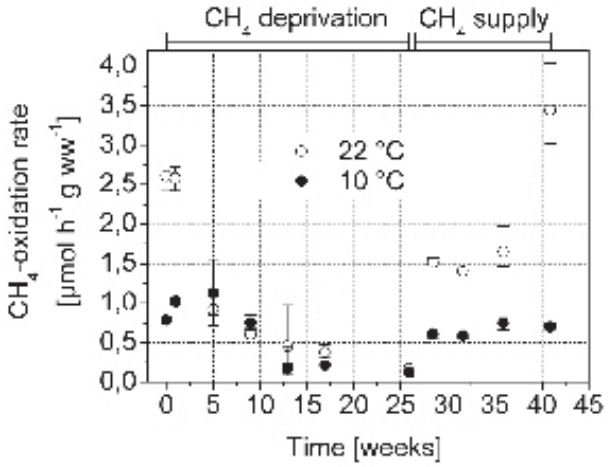



**Fig. 8:** Variation of methane oxidation rate with time of methane deprivation. Circles = average of three replicates, error bars = maximum and minimum.

or insufficient during longer phases of steady, continuous LFG emissions lacking the flow oscillations as observed for most other phases.

### 3.2 Influence of methane deprivation duration

The reversal of gas flux means that the biofilter methanotroph population is regularly deprived of methane, sometimes for longer periods. The data in Figure 8 show how, in a laboratory batch experiment, the methane oxidation rate reacted to the duration of methane deprivation at 10 °C and at 22 °C incubation temperature.

Deprivation of methane led to a significant decrease in methane oxidation rate within five weeks at 22 °C and within nine weeks at 10 °C incubation temperature. Interestingly, the rate at 22 °C dropped below the one at 10 °C after five weeks, even though initial rates were around 3.5 fold higher. This might be due to an overall higher metabolic activity leading to a faster consumption of storage compounds at 22 °C. The data show that fluctuation of methane supply within the frame of days or even a few weeks does not impair biofilter operation, especially at lower temperatures. Similar observations on the impact of substrate deprivation on methanotrophic activity have been reported elsewhere (Kightley et al., 1995; Reay et al., 2001).

## 4 Conclusions

On the investigated passively vented landfill, the direction of advective gas flow reverses more than once a day in reaction to atmospheric pressure variation. The

reaction of gas emission to atmospheric pressure changes even below 1 hPa is immediate and highly sensitive, resulting in a very high variability of gas flux into the biofilter connected to one of the gas wells. Correspondingly, the biofilter receives very variable loads of methane. Gas flow rate and emission phase duration not only govern methane supply to the methanotrophic bacteria but also determine the degree of counterdirective diffusive oxygen supply via the filter surface. Emission dynamics therefore strongly influence vertical gas distribution in the biofilter and thus conditions for successful operation. While the performance of the biofilter under study was found to be very high with rates up to 80 g $CH_4$ $h^{-1}$ $m^{-3}$, comparing well to actively vented systems, both the simulation of oxygen concentration profiles and on-site measurements revealed that in cases of constant gas feed diffusive oxygen ingress across the filter surface can be significantly reduced, thus impeding methanotrophic activity. Therefore, it must be concluded that the highly dynamic emission behaviour associated to passive landfill ventilation and the corresponding advective input of oxygen is highly relevant to the performance of the biofilter under study. Reversal of gas flow and subsequent deprivation of methane were found not to impact methanotrophic activity. For the investigated system, the frequency of changes in LFG flow rate and composition are sufficient to sustain very high densities and activities of methanotrophic bacteria as well as high methane removal rates for the biofilter as a whole.

The limitations connected to biofilter operation under conditions of diffusive oxygen ingress may be overcome by

- charging the gas onto a greater filter area, thus decreasing LFG charge onto the filter area until the diffusive flux equals or exceeds the advective LFG flux;
- mixing the raw LFG with atmospheric air prior to entering the biofilter. In this case, the problems of potential biofilter desiccation and the development of explosive mixtures have to be addressed.

Overall, passive biofiltration was shown to be an effective and low cost tool to mitigate low calorific landfill methane emissions. The design of robustly operated systems without heating or irrigation should observe that

- the pore size distribution of the filter material warrants a high gas permeability at simultaneous sufficient water retention;
- the filter material should be non-plastic, i.e. it should not shrink or aggregate to prevent preferential flow;
- the filter material itself is non-degradable to prevent compaction;
- the filter size has to be sufficient to allow a high performance at lower temperatures during winter time.

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