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Biological methane oxidation in actively vented biofilters

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Abstract
Biological methane oxidation may be a suitable treatment option for gas emissions from old landfills and other sources with low gas production rates where energy recovery or flaring is not economically feasible.

In comparison to passively vented systems, the use of enclosed biofilters with forced gas flux offers better process control and verifiability of the actual efficiency. This paper provides design parameters and cost estimation for actively vented biofilters for methane oxidation.

Investigations were carried out at various methane concentrations and temperatures. Since severe material clogging due to the accumulation of exopolymeric substances (EPS) had occurred in earlier experiments, a special biofilter packing material composed of compost, wood fibres, and peat was developed and successfully tested. Because of its structural properties, this material was not subject to clogging and showed stable degradation rates over a period of one year.

From the experimental results, kinetic data were derived which allowed the calculation of the required size of a biofilter for given inlet concentrations, biofilter efficiencies, and operational temperatures. A biofilter volume of 800 m^3 at a temperature of 22°C and 370 m^3 at a temperature of 30°C is required to obtain 90% efficiency for the treatment of 50 m^3/h of landfill gas with a methane concentration of 20%, which is diluted by air to a concentration of 2.5% before entry into the biofilter.

A cost estimation revealed that biofilter operation at 30°C is cheaper than at 22°C with respect to both investment and operational costs due to the smaller biofilter volume and lower expenses for material exchange. The costs for the avoidance of greenhouse gas emissions were estimated to range from 43 to 68 € per Mg CO_2-equivalent.

1 Introduction
Landfill gas (LFG) emissions are a significant source of the potent greenhouse gas methane. Therefore, reduction of LFG emissions is an important measure of climate protection and may help to fulfil the goals set by the Kyoto Protocol. LFG

collection for the production of heat and electrical power has been established as state of the art in many countries. In other cases, flares are operated to burn the methane and organic trace compounds contained in the landfill gas. However, these processes can only be applied in cases where sufficient methane production rates are provided. By now, the mitigation of emissions with low methane concentrations and/or flow rates often is neglected.

Biological methane oxidation may be an option for these cases. For example, landfill covers can be designed to serve as methane oxidation layers (Humer and Lechner, 2001; Scheutz et al., 2004; Berger et al., 2005; Zeiss, 2006). This setup provides a large volume for methane oxidation. On the other hand, methane oxidation layers are strongly exposed to climatic influences and may show uneven flow distribution.

Another possibility is the use of biofilters with passive flow (Gebert and Gröngröft, 2006a; Powelson et al., 2006). Here, the LFG is forced through a fixed bed filled with a suitable packing material that contains methanotrophic bacteria. Unlike landfill covers, passively vented biofilters do not have to act as recultivation layer and therefore may be optimized for the single purpose of methane oxidation. Moreover, they may comply with legal requirements more easily because they allow for the installation of a conventional landfill cover. As in landfill covers for methane oxidation, the flow is controlled by the pressure difference between the landfill body and the ambient air. Flow rate and direction may therefore fluctuate considerably (Gebert and Gröngröft, 2006b). In passively vented systems, oxygen has to enter the substrate from the surface, i.e. in the opposite direction of the general LFG flow, which leads to the formation of a restricted methane oxidation zone.

In the investigations presented in this paper, actively vented biofilters for methane oxidation are used, where the LFG is mixed with ambient air and introduced into the biofilter by means of a blower. Thus, both methane and oxygen are present throughout the biofilter volume. Actively vented biofilters provide a constant flow rate, and temperature and moisture can be controlled more accurately. On the other hand, these biofilters require higher investment and operational costs than the biological systems with passive gas flux. The operation of actively vented biofilters may be especially suitable for applications where a gas extraction system is already in operation, but the gas production has decreased to levels where gas utilisation or flaring are no longer feasible.

2 Experimental setup

2.1 Experimental plant

The investigations discussed in this paper were carried out in a bench scale experimental plant consisting of four biofilters in two columns. Each filter contained 13 to 18 L of filter material. Methane was mixed with pressurised air, humidified in a counter-current column scrubber and introduced into the biofilter columns. The gas flow for each column was adjusted to 150 L/h. The biofilters were continuous-

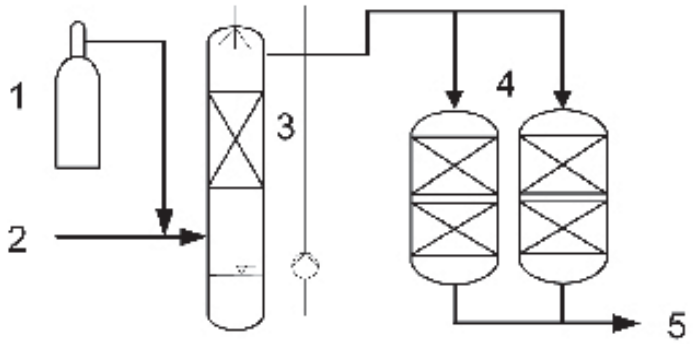


Fig. 1: Flow chart of the experimental plant for methane oxidation in biofilters. 1-methane; 2-pressurised air; 3-air humidifying scrubber; 4-biofilters; 5-exhaust.

Tab. 1: Operational data of the experimental biofilter plant used for microbial methane oxidation.

Biofilter volume (per filter)	[m^3]	0.013-0.018
Number of filter elements / parallel columns	-	4 / 2
Total biofilter volume (initial)	[m^3]	0.064
Surface area per filter	[m^2]	0.045
Total flow rate	[m^3/h]	0.3
Filter surface load	[$m^3/(m^2h)$]	3.3
Volumetric filter load	[$m^3/(m^3h)$]	5.6
Methane concentration (feed after dilution)	[Vol.-%]	0.3-3.2

ly operated for 340 days at different methane concentrations. A flow chart of the bench scale plant is given in Figure 1. Basic operational data of the experimental plant is given in Table 1.

2.2 Biofilter packing material

Previous investigations have focussed on the selection of an appropriate biofilter packing material for methane oxidation. It was found that material clogging due to accumulation of exopolymeric substances (EPS) was a major issue, and

maintaining a sufficient material porosity for mass transfer was crucial for the long-term performance of the biofilters. After testing different other biofilter materials, a mixture of compost, wood fibres, and peat was developed and successfully tested. Because of its structural properties, this material was not subject to clogging and showed stable degradation rates over a period of one year. Table 2 summarises some characteristics of this material (Streese and Stegmann, 2003).

Tab. 2: Characteristics of the optimised biofilter material consisting of compost, peat, and wood fibres. Values measured at the beginning of the experiment.

bulk density[a]	434 kg/m³	NH_4-N[b]	881 mg/kg
water content[a]	45.6 %	NO_3-N[b]	166 mg/kg
water holding capacity[a]	76.7 %	NO_2-N[b]	40.2 mg/kg
ignition loss [%][b]	52.1 %	TKN[b]	7370 mg/kg
TOC [%][b]	26.3 %	SO_4^{2-}[b]	363 mg/kg
pH	5.7	PO_4^{-}[b]	63.2 mg/kg
Conductivity	961 µS/cm		

[a] related to moist mass; [b] related to dry mass

3 Biofilter performance at different temperatures and concentrations

Considering first-order degradation kinetics, the required biofilter volume for a specific application can be calculated using Equation 1 (Streese et al., 2005):

$$V = \frac{1}{k} \cdot \dot{V} \cdot \ln \frac{c_{in}}{c_{out}} \quad \textbf{(1)}$$

Where: V, Volume of biofilter packing [m³]; k, first-order kinetic constant [h^{-1}]; $\dot{V}$, volumetric gas flow rate [$m^3\ h^{-1}$]; c_{in}, c_{out}: Methane concentrations at biofilter inlet and outlet, respectively [$g\ m^{-3}$].

The kinetic constant k has to be determined in biofiltration experiments under similar conditions as in the intended application.

Besides concentration, temperature has a strong influence on methane degradation rates. According to the Arrhenius equation, chemical reaction rates increase exponentially with temperature. For biological reactions, it has to be taken into account that at some point the enzymes are denatured. Furthermore, in biofilters and other technical applications, the biological processes are superposed by mass transfer processes. Here, two contrary effects occur: the diffusion velocity increases, whilst the solubility of the substrates methane and oxygen decreases. In total, an optimum temperature results. For methane oxidation, most authors report optimum temperatures around 35°C (e.g., Whalen et al., 1990; Bender & Conrad, 1995). Biofilters usually are operated below this optimum temperature.

In order to include in the design calculations the influence of temperature on methane oxidation, Equation (1) was extended by a temperature term describing the relative value of the kinetic constant k, and therewith the degradation rates, at

a given temperature as compared to a reference temperature (Streese, 2005). For practical reasons, 20°C was chosen as reference temperature. The temperature dependence of k was approximated by Equation (2).

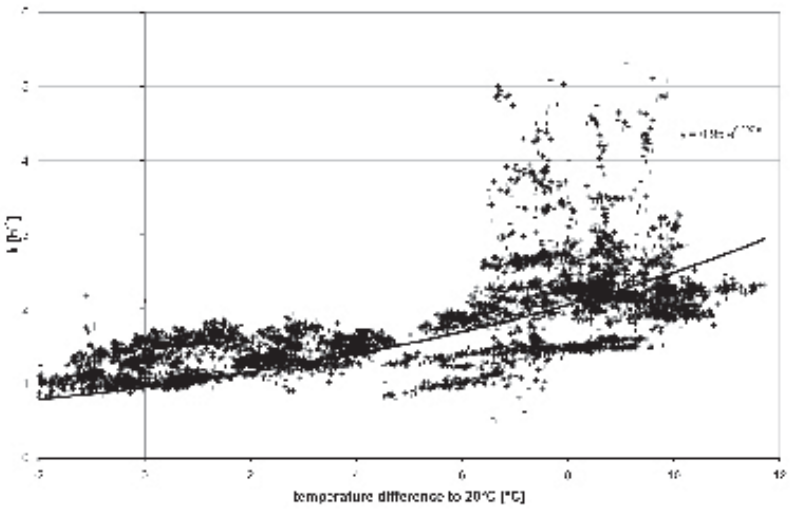
$$k(\vartheta_M) = k_{20°C} \cdot e^{k_t \cdot (\vartheta_M - 20°C)} \quad \textbf{(2)}$$

Where: ϑ_M, mean temperature in biofilter [°C]; $k(\vartheta_M)$, first-order kinetic constant at temperature ϑ_M [h^{-1}]; $k_{20°C}$, first order kinetic constant at 20°C [h^{-1}]; k_t, temperature coefficient [$°C^{-1}$].

Merging of Equations 1 and 2 allows for the calculation of the required biofilter volume for a given application and considering the operational temperature (Equation 3):

$$V = \frac{\dot{V}}{k_{20°C}} \cdot \ln \frac{c_0}{c_{out}} \cdot e^{-k_t \cdot (\vartheta_M - 20°C)} \quad \textbf{(3)}$$

The values for $k_{20°C}$ and k_t were derived from the data of the bench scale experiments described above, which had provided some 10,000 data sets, not counting data from the first 65 days of the experiment, which were regarded as adaptation phase. A "single-point kinetic constant" k was calculated from the respective deg-



radation rate and logarithmic mean concentration for each data set. The kinetic constants were then plotted against the difference between the mean biofilter temperature and the reference temperature (Figure 2). From this plot, $k_{20°C}$ and k_t

were derived by exponential regression. The values of these constants are 0.95 h^{-1} and 0.097°C^{-1}, respectively.

Fig. 2: Temperature dependence of methane degradation kinetics in biofilters. Data from laboratory scale investigations, biofilter volume 15 L, packing material: mixture of compost, peat, and wood fibres. See text for details.

The figure shows considerable data scattering. However, most of the larger deviations from the regression function occurred in the early phases of the experiment (Streese and Stegmann, 2005), resulting in a more reliable prediction of the long-term behaviour of the biofilter by the model. Considering the variance of the data, a safety factor should be included in the design calculations. For higher temperatures than 31°C, the data are not sufficient for reliable calculations.

4 Biofilter design calculation

In the following sections, a detailed estimation will be provided regarding the design and costs of a large scale biofilter utilising the above described packing material composed of compost, peat, and wood fibres. These calculations were done for a fictitious application. The chosen boundary conditions of this application are listed in Table 3.

Tab. 3: Boundary conditions for a fictitious application of a LFG biofilter

LFG collection rate	50 m^3/h
Methane concentration in LFG	20%
Methane concentration after raw gas dilution	2.5%
Flow rate of diluted LFG	400 m^3/h
Biofilter efficiency	90%

Using kinetic constants derived from Figure 2 and Equation 3, biofilters for methane oxidation can be designed for different operational temperatures. Figure 3 shows the biofilter efficiencies and corresponding residual methane concentrations in the cleaned gas which can be achieved with different biofilter volumes. The required volume to obtain 90% efficiency is 800 m^3 at a temperature of 22°C and 370 m^3 at a temperature of 30°C, respectively.

It was shown in a previous publication that the secondary emissions arising from the energy demand of a methane oxidation biofilter are much lower than the greenhouse gas emissions avoided by methane oxidation, even when the biofilter is heated to increase the operational temperature. By implementing a partial recirculation of the treated gas, heat losses and secondary emissions can be further decreased. For the above mentioned application, Streese and Stegmann (2005) calculated an annual avoidance of at least 1311 Mg CO_2-Equivalent of primary greenhouse gas emissions, whilst secondary emissions only ranged from 35 to 86 Mg CO_2-Equivalent per year depending on temperature and mode of operation.

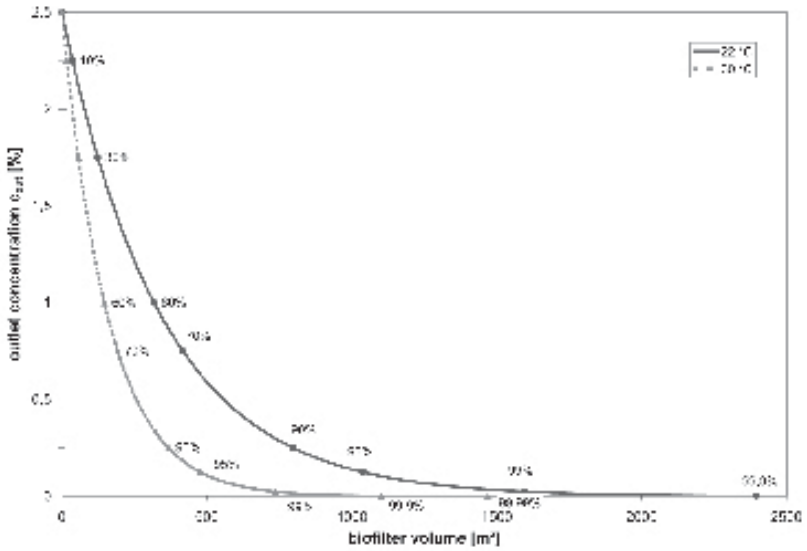


Fig. 3: Treatment efficiencies and corresponding residual methane concentrations achievable with different biofilter volumes. Values calculated for the treatment of 400 m³/h of diluted landfill gas with a methane concentration of 2.5% using an optimised biofilter packing material consisting of compost, peat and squeezed wood fibres.

5 Cost estimation

The investment and operating costs of a biofilter for methane oxidation may differ greatly for each case. Many factors are site-specific, such as the electricity costs, delivery costs for the components of the plant and the biofilter packing material, as well as the overall level of prices and salaries. Nevertheless, an estimation of the costs of the methane oxidation biofilter discussed above will be presented in the following. The calculations closely follow a sample calculation in Devinny et al. (1999). The costs of the equipment, machinery, and measurement systems were estimated from our own experiences. The costs for the biofilter building, which is designed as a prefabricated modular hall, are taken from an internet offer of a hall building company (Hansen, 2007). However, the authors have no intention to imply any preference for or state the suitability of the products of this company for the purpose discussed in this paper.

It is assumed that the landfill where the biofilter is to be installed is already equipped with a gas collection system and supply of electricity and water. This is the case at landfills where a landfill gas utilisation unit or a flare had been operated which was shut down due to low gas quality.

A safety factor of 1.2 for the biofilter volume is applied to allow for increased flow rates due to recirculation and the uncertainties resulting from the data quality (see above). The height of the filter bed is 1.6 m. To provide optimal gas flow distribution, coconut fibre mats of 10 cm thickness are installed at the inlet, outlet, and within the packing material at a distance of 50 to 60 cm to each other. The investment and operational costs are listed in Tables 4 and 5, respectively.
By installing a partial recirculation of the treated gas, considerable lower costs can be achieved because of the smaller volume required at higher temperatures, resulting in lower investment costs. Another important cost factor which is reduced at higher operational temperatures is the installation and exchange of the biofilter packing material. In comparison, the costs for additional electricity and water consumption are much lower. The additional costs for installations and control for this mode can be estimated to amount to about 4,000.00 €, which would amortise within the first year of operation.

Tab. 4: Investment costs of a biofilter for methane oxidation for different temperatures

operational temperature	(a) 22°C	(b) 30°C
A. Initial site preparation costs: levelling of 800 m² [(a)] / 500 m² [(b)] ground space; excavation of 0.5 m topsoil (mean) disposal of 400 m³ [(a)] / 250 m³ [(b)] excavated topsoil (assume non-hazardous soil) costs for machinery, labour, disposal fees: 3.90 € per m²	**3,120.00 €**	**1,950.00 €**
B. Biofilter hall (cf. Hansen (2007))		
basic costs	70,200.00 €	42,400.00 €
thermal insulation roof: Astrotherm-roof, 100 mm	10,800.00 €	6,000.00 €
thermal insulation walls: 80 mm, 310 m² [(a)]/225 m² [(b)]	16,500.00 €	11,900.00 €
zinc coating of the entire hall	2,000.00 €	1,000.00 €
additional corrosion protection inside, 1000 m² [(a)]/600 m² [(b)]	2,000.00 €	1,200.00 €
foundation concrete base, improved surface	27,800.00 €	15,900.00 €
gate (filling/exchange of material)	5,000.00 €	5,000.00 €
inspection door	1,000.00 €	1,000.00 €
openings for gas in- and outlet, 400 mm, 200 € each	400.00 €	400.00 €
Total	**135,700.00 €**	**84,800.00 €**
C. Equipment biofilter		
biofilter packing material: 35 €/m³	33,600.00 €	15,750.00 €
coconut fibre mats: 1.75 €/m²	4,200.00 €	1,890.00 €
support grid, recycled plastics, accessible	11,200.00 €	6,800.00 €
filling: machinery hire (wheel loader, conveyor...) + labour costs; 2.50 €/m³	2,400.00 €	1,125.00 €
Total	**51,400.00 €**	**25,565.00 €**

D. Devices and machinery		
counter current column scrubber, heatable	2,500.00 €	2,500.00 €
radial fan for ambient air	1,500.00 €	1,500.00 €
landfill gas pump (e.g., liquid ring pump), explosion proof	1,000.00 €	1,000.00 €
inlet air heating	800.00 €	800.00 €
Total	5**,800.00 €**	5**,800.00 €**
E. Measurement and automation		
gas analyser	4,500.00 €	4,500.00 €
explosion protection monitoring	1,200.00 €	1,200.00 €
tensiometers for material moisture control 6 x 400 €	2,400.00 €	2,400.00 €
flow meters	2,000.00 €	2,000.00 €
Computer / PLC	1,000.00 €	1,000.00 €
Total	**11,100.00 €**	**11,100.00 €**
F. Other: travel, spare parts, etc.	**5,000.00 €**	**5,000.00 €**
G. Piping: 10% of investment costs	**30,300.00 €**	**18,460.00 €**
H. Electrical: 4% of investment costs	**12,120.00 €**	**7,385.00 €**
I. Equipment installation (except packing material): 4% of investment costs	**12,120.00 €**	**7,385.00 €**
J. Engineering: 12% of investment costs	**36,370.00 €**	**22,155.00 €**
Total investment costs	**303,030.00 €**	**184,600.00 €**

[(a)]operational temperature: 22°C; [(b)]operational temperature: 30°C

Tab. 5: Operating costs of a biofilter for methane oxidation for different temperatures

operational temperature	**(a) 22°C**	**(b) 30°C**
A. Electricity: 0.15 €/kWh		
1. without recirculation:	**11,900.00 €/a**	**18,750.00 €/a**
2. with recirculation:	**7,655.00 €/a**	**11,300.00 €/a**
B. Water consumption: 1.46 €/m³		
1. without recirculation: 54.2 m³/a [(a)]/98.8 m³/a [(b)]	**80.00 €/a**	**144.00 €/a**
2. with recirculation: 29.8 m³/a [(a)]/54.3 m³/a [(b)]	**45.00 €/a**	**80.00 €/a**
C. Filter material exchange (see number C in investment costs)		
removal: machinery hire + labour + disposal: 4.50 €/m³	4,320.00 €	2,025.00 €
biofilter packing material: 35 €/m³	33,600.00 €	15,750.00 €
coconut fibre mats: 1.75 €/m²	4,200.00 €	1,890.00 €
filling: machinery hire + labour costs; 2.50 €/m³	2,400.00 €	1,125.00 €
Total	44,520.00 €	20,790.00 €
annual costs (replacement every 3 years)	**14,840.00 €/a**	**6,930.00 €/a**
D. Maintenance: 1 hour per working day at 30 €/h	**7,650.00 €/a**	**7,650.00 €/a**
E. Spare parts	**2,000.00 €/a**	**2,000.00 €/a**
F. Depreciation (10% of investment costs)	**30,300.00 €/a**	**18,460.00 €/a**

G. Interest (6% of investment costs)	**18,180.00 €/a**	**11,080.00 €/a**
Total operating costs:		
1. without recirculation:	**84,950.00 €/a**	**65,014.00 €/a**
2. with recirculation:	**80,670.00 €/a**	**57,500.00 €/a**

[a]operational temperature: 22°C; [b]operational temperature: 30°C

6 Conclusions

The applicability of actively vented biofilters for methane oxidation has already been shown in previous publications (Streese and Stegmann, 2003, 2004). In this paper, a detailed cost estimation is provided, allowing for a well-founded estimation of the cost/benefit ratio.

One important result is that operation of methane oxidation biofilters at 30°C is more cost-effective than at 22°C with respect to both investment and operating cost.

The specific costs for the reduction of greenhouse gas emissions range from 43 to 68 €/Mg CO_2-equivalent, with biofilter operation at 30°C and with partial gas recirculation being cheapest. For comparison, Melse and van der Werf (2005) have calculated specific costs of 26 to 31 US-$/Mg CO_2-equivalent for the treatment of methane emissions from a liquid manure storage in a biofilter. The differences result, at least partly, from differences of the design and operating conditions. These costs range approximately in the same order of magnitude as the costs for thermal treatment of lean gas emissions from landfills.

Acknowledgements

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References

Bender, M. and Conrad, R. (1995): Effect of CH_4 concentrations and soil conditions on the induction of CH_4 oxidation activity. Soil Biology & Biochemistry 27(12), pp. 1517-1527.

Berger, J.; Fornés, L. V.; Ott, C.; Jager, J.; Wawra, B. and Zanke, U. (2005): Methane oxidation in a landfill cover with capillary barrier. Waste Management 25(4), pp. 369-373.

Devinny, J. S.; Deshusses, M. A. and Webster, T. S. (1999): Biofiltration for air pollution control. CRC Press, Boca Raton.

Gebert, J. and Gröngröft, A. (2006a): Performance of a passively vented field-scale biofilter for the microbial oxidation of landfill methane. Waste Management 26(4), pp. 399-407.

Gebert, J. and Gröngröft, A. (2006b): Passive landfill gas emission - Influence of atmospheric pressure and implications for the operation of methane-oxidising biofilters. Waste Management 26(3), pp. 245-251.

Gebert, J.; Gröngröft, A. and Miehlich, G. (2003): Kinetics of microbial landfill methane oxidation in biofilters. Waste Management 23(7), pp. 609-619.

Hansen (2007): Hansen Hallenbau Modulhallen (09.01.2007). http://www.hansen-hallenbau.de/ modulhallen/modul0.htm

Humer, M. and Lechner, P. (2001): Microbial methane oxidation for the reduction of landfill

gas emissions. Journal of Solid Waste Technology and Management 27(3-4), pp. 146-151.

Melse, R. and van der Werf, A. W. (2005): Biofiltration for mitigation of methane emission from animal husbandry. Environmental Science & Technology 39(14), pp. 5460-5468.

Powelson, D. K.; Chanton, J.; Abichou, T. and Morales, J. (2006): Methane oxidation in water-spreading and compost biofilters. Waste Management & Research 24(6), pp. 528-536.

Scheutz, C.; Mosbaek, H. and Kjeldsen, P. (2004): Attenuation of methane and volatile organic compounds in landfill soil covers. Journal of Environmental Quality 33(1), pp. 61-71.

Streese, J. (2005): Abbau von Methan in aktiv durchströmten Biofiltern. Dissertation, Hamburger Berichte 25. Verlag Abfall aktuell, Stuttgart.

Streese, J. and Stegmann, R. (2003): Microbial oxidation of methane from old landfills in biofilters. Waste Management 23(7), pp. 573-580.

Streese, J. and Stegmann, R. (2004): Biologische Methanoxidation bei Altdeponien: Potenziale von Biofiltern mit aktiver Durchströmung. In: Deponietechnik 2004. Hamburger Berichte 22, pp. 301-315.

Streese, J. and Stegmann, R. (2005): Potentials and limitations of biofilters for methane oxidation. In: Proceedings Sardinia 2005, 10th International Waste Management and Landfill Symposium, Cagliari.

Streese, J.; Schlegelmilch, M.; Heining, K. and Stegmann, R. (2005): A macrokinetic model for dimensioning of biofilters for VOC and odour treatment. Waste Management 25(9), pp. 965-974.

Whalen, S. C.; Reeburgh, W. S. and Sandbeck, K. A. (1990): Rapid Methane Oxidation in a Landfill Cover Soil. Applied and Environmental Microbiology 56(11), pp. 3405-3411.

Zeiss, C. A. (2006): Accelerated methane oxidation cover system to reduce greenhouse gas emissions from MSW landfills in cold, semi-arid regions. Water, Air & Soil Pollution 176, pp. 285-306.