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Biotic systems to mitigate landfill methane emissions

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Landfill gases produced during biological degradation of buried organic wastes include methane, which when released to the atmosphere, can contribute to global climate change. Increasing use of gas collection systems has reduced the risk of escaping methane emissions entering the atmosphere, but gas capture is not 100% efficient, and further, there are still many instances when gas collection systems are not used. Biotic methane mitigation systems exploit the propensity of some naturally occurring bacteria to oxidize methane. By providing optimum conditions for microbial habitation and efficiently routing landfill gases to where they are cultivated, a number of bio-based systems, such as interim or long-term biocovers, passively or actively vented biofilters, biowindows and daily-used biotarps, have been developed that can alone, or with gas collection, mitigate landfill methane emissions. This paper reviews the science that guides bio-based designs; summarizes experiences with the diverse natural or engineered substrates used in such systems; describes some of the studies and field trials being used to evaluate them; and discusses how they can be used for better landfill operation, capping, and aftercare.

Keywords: landfill gas, methane oxidation, biocover, biofilter, bio-window, landfill aftercare, wmr 1317-2

Introduction

For many years, good landfill design focused on liner and routing systems to contain landfill leachate and prevent the methane produced from causing landfill fires or explosions. In recent years, it has become clear that there are other problems related to landfill methane production, namely, its likely contribution to global climate change. Methane emissions and migration from closed landfills are not uncommon, because many landfills were completed before requirements for rigorous capping procedures existed. Lateral migration of the gas out and around a liner or cover system can also occur, so that methane levels in buffer regions around a site can be higher than those on the cover (Christophersen & Kjeldsen 2001). Landfills are estimated to be responsible for 35–69 Tg CH₄ year⁻¹, and their emissions constitute 30 and 24% of the anthropogenic methane production in Europe and the

US, respectively (EEA 2006, IPCC 2007a, US EPA 2007). Further, for the past 25 years, global anthropogenic methane emissions have exceeded those from natural sources (IPCC 2007a). This realization has stimulated new academic research, updated policies, and of course, design and operational changes to reduce landfill methane emissions. Such changes have included efforts toward more efficient biogas capture, complete methane destruction in thermal (flares) or biotic systems, and good energy recovery from captured methane.

Whenever economically feasible, gas collection systems are recommended for landfill gas emissions control, and liner designs have been configured to prevent lateral biogas migration. Collection systems are not 100% efficient, however, and emissions may also escape preferentially from and around wells and along the routes of installed landfill equipment. A

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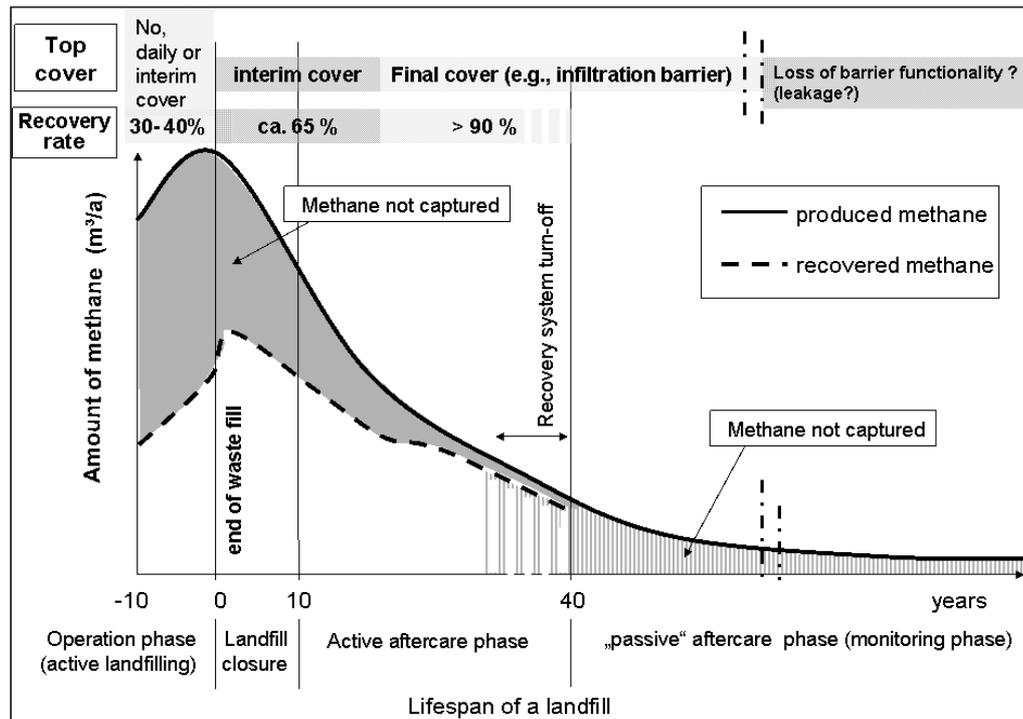


Fig. 1: Time-dependent methane production and recovery over a landfill lifetime. Methane emissions (methane not captured) are shown as a function of cover type and do not include methane oxidation removal: gas recovery rates according to Spokas *et al.* (2006) and (Huber-Humer 2007).

report from a German landfill study documented that the gas utilization and/or flaring systems associated with active gas collection began to falter or fail as gas production diminished, leaking considerable methane to the atmosphere (Krümpelbeck 1999). Historical practice suggests that collection systems may operate less than half the time that landfill gas (LFG) is produced, because they are only economically feasible when methane concentrations are high. As low-level gas production may continue for several decades up to 100 year, the net accumulation of methane during non-collection periods is not negligible with respect to global warming (Figure 1).

Once methane concentrations fall below 35–40% and total gas production rates are 30–50 m³ h⁻¹, treatment in combined heat and power plants (CHP plants) becomes technically and economically infeasible (Haubrichs & Widmann 2006). When methane concentrations reach 20–25% v/v and LFG flow-rates fall to 10–15 m³ gas h⁻¹, the most suitable treatment method becomes high temperature flares. Below these values, the treatment of poor landfill gas becomes more expensive and complex. Fluidized bed combustion or catalytic oxidation are two possible options, and if landfill gas is > 0.3% methane by volume, it can be oxidized in a non-catalytic reactor bed or subjected to an auto-thermal reaction under steady-state conditions (Stachowitz 2001).

One of the most promising and cost-effective options being employed for control of low-level methane emissions is the use of engineered bio-based systems. They were developed after numerous reports documented that landfill cover micro-organisms were removing methane from LFG as the gas migrated through aerobic regions above the buried waste (Mancinelli & McKay 1985, Whalen *et al.* 1990, Czepiel

et al. 1996, Liptay *et al.* 1998). In 2002, a group of engineers, scientists, and practitioners came together as CLEAR, the Consortium for Landfill Emissions Abatement Research, to organize their research efforts and collaborate to design bio-based methane removal systems for all stages of landfill life (Huber-Humer 2004a). The result was a toolbox of engineered systems, including biocovers, biofilters, biowindows and biotarps, all of which are based on microbial methane oxidation.

Bio-based systems have a wide range of applications. They can be used with gas collection to capture escaping emissions; or alone during new landfill start-up; at older landfills past their peak gas production period; at small sites where gas collection is not technically and economically feasible; at closed landfills to clean escaping methane emitted during post-closure forced in-situ aeration (a technique used to reduce waste volume and aftercare duration time); and in instances where the landfilled wastes have a low lifetime gas generation potential, so that gas collection is infeasible. An example of the latter are landfills containing mechanically and biologically pretreated (MBT) wastes. The purpose of this paper is to briefly review the science that guides these designs; describe some of the studies and field trials used to evaluate them; and discuss how they can be used for better landfill operation, capping, and aftercare.

Methane oxidation theory

The microbiology and ecology of microbes that can consume methane has been well-reviewed (Conrad 1996, Hanson & Hanson 1996), and only the most salient issues related to the design of landfill methane mitigation systems will be summarized here. Although there are some yeasts and nitrifying

bacteria that use methane (Wolf & Hanson 1980, Ward 1987), most microbial methane uptake in landfills is believed to be accomplished by methanotrophs, a group of obligate aerobes that can oxidize methane for energy (yielding carbon dioxide and water) and incorporate its carbon into biomass (Conrad 1996). Therefore, for simplicity, the organisms accomplishing methane uptake in landfills will be here referred to as methane oxidizers or methanotrophs, although it is acknowledged that some methane is assimilated and not oxidized, and not all uptake is mediated by methanotrophs.

Methanotrophs are diverse and ubiquitous in the environment (Whittenbury *et al.* 1970, Hanson & Hanson 1996). They are naturally occurring in many methane-influenced ecosystems and are particularly abundant at the interface of aerobic and anaerobic regions of wetlands (Harriss & Sebacher 1982, Boon & Lee 1997), rice paddies (Joulian *et al.* 1997, Dubey *et al.*, 2002), and peat bogs (Krumholz *et al.* 1995, Sundh *et al.* 1995). They also exist in most terrestrial soils, thriving on atmospheric levels of methane (Reay *et al.* 2001, Horz *et al.* 2002). Abundant numbers of methanotrophs have been found in landfill cover soil and biofilters (Jones & Nedwell 1993, Gebert *et al.* 2003, Nozhevnikova *et al.* 2003).

Methanotrophs are generally classified into type I and type II strains based on a variety of characteristics that differentiate them (Bowman *et al.* 1993, Hanson & Hanson 1996), and some of their differences that are relevant to design are: (1) some can co-metabolize non-methane organic substrates (Dalton & Sterling 1982, Linder *et al.* 2000, Scheutz *et al.* 2004); (2) the methane concentration that triggers the onset of oxidation varies among methanotrophs (Le Mer & Roger 2001, Bender & Conrad 1992, 1995); (3) their methane consumption rates vary (Czepiel *et al.* 1996, Bogner *et al.* 1997); (4) in some cases their oxygen requirements vary (Whittenbury *et al.* 1976); (5) their tolerance to temperature and moisture changes vary (Omelchenko *et al.* 1993); and (6) they have different propensities for producing exopolymeric substances (EPS) (Malashenko *et al.* 2004). A number of explanations have been suggested to explain why cells produce EPS, and for methanotrophs, it has been suggested that EPS production may be a metabolic response to excess carbon relative to other nutrients. Formaldehyde that might otherwise accumulate and poison the cell is shunted into sugar-based polymers that are then excreted (Linton *et al.* 1986, Wilshusen *et al.* 2004a).

Methanotrophs also consume some non-methane landfill gas emissions (Scheutz *et al.* 2004, Scheutz & Kjeldsen 2004), some of which are more than 1000-fold more greenhouse-active than methane (IPCC 2007a). Methanotrophic uptake of compounds other than methane was first demonstrated in aqueous systems, where methanotrophs removed several non-methane hazardous compounds from contaminated aquifers (Alvarez-Cohen & McCarty 1991, Broholm *et al.* 1993, Arcangeli *et al.* 1996, Chang & Alvarez-Cohen 1996). Their consumption of these non-methane compounds proved to be a form of co-metabolism, where biodegradation occurs, but there is no metabolic gain to the organism. It typically occurs

when an enzyme made by a bacterium is not highly specific for a particular substrate, and the enzyme binds and catalyzes reactions with alternate substrates. In methanotrophs, this enzyme is a soluble form of methane monooxygenase (sMMO).

There is some evidence that landfill methanotrophs also may be associated with N₂O formation (Mandernack *et al.* 2000), another greenhouse gas with a global warming potential of 289 over a 100-year period (IPCC 2007a). Nitrogen-rich environments (such as covers made of organic soil substrates or composts) and alternating aerobic and anaerobic zones are both factors that are believed to stimulate methanotrophic, N₂O production. The N₂O is generated when low-specificity sMMO catalyzes nitrification (Mandernack *et al.* 2000, Rinne *et al.* 2005). Other studies, however, report no increased N₂O formation with methane oxidation in organic landfill cover materials such as organic soils (Börjesson *et al.* 1998a) or a biowaste compost/gravel mixture (Watzinger *et al.* 2005), and it has been suggested that high soil moisture may have more impact, since it limits oxygen availability. Rinne *et al.* (2005) as well as Börjesson & Svensson (1997) concluded from the scale-up of their N₂O-measurements on different northern landfills that landfill N₂O emissions are likely of minor impact relative to the large land tracts of forests and agricultural fields producing this gas. This issue will need to be addressed through further research before a whole landfill greenhouse gas mass balance for all emerging greenhouse gases can be conducted.

There are broad differences among methanotrophs' response to different methane concentrations. One group, which are known as the upland soil cluster alpha USC α , are considered to be 'high affinity' methanotrophs because they can initiate uptake at low methane concentrations (0.8–280 nmol L⁻¹) and thereby consume atmospheric methane (1.7 ppm). Other 'low affinity' methanotrophs will not begin uptake until methane levels reach 0.8–66 mol L⁻¹. They tend to favour lower oxygen concentrations, and their kinetic parameters (V_{\max} and K_M) are usually high, resulting in high methane turnover rates (Bender & Conrad 1992, Henckel *et al.* 2000).

Environmental factors that are favourable for methane oxidation have been reviewed by Hanson & Hanson (1996). Microbial methane uptake rates are affected by moisture content (Castro *et al.* 1995, Boeckx & Van Cleemput 1996, Schnell & King 1996, Cai & Yan 1999, Reay *et al.* 2001), temperature (Whalen *et al.* 1990, Cai & Yan 1999, Gebert *et al.* 2003, Börjesson *et al.* 2004), and soil physical properties such as permeability and particle size (Bender & Conrad 1994, Kightley *et al.* 1995, Borjesson *et al.* 1998b). These physical and environmental factors can be addressed in an engineered system design, as will be evident for some of the systems described in subsequent sections. A final factor that can influence methanotroph performance in engineered systems is their propensity to excrete exopolymeric substances that can cover the cells and limit gas transfer (Hilger *et al.* 2000a, Humer & Lechner 2001b, Wilshusen *et al.* 2004a).

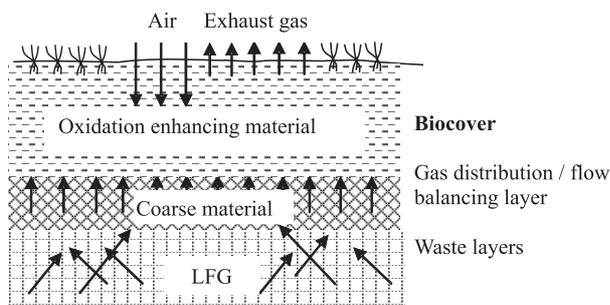


Fig. 2: Conceptual scheme of a biocover. LFG, landfill gas.

Early experiments seeking to optimize methane oxidation in landfill cover soils evaluated the use of amendments (Kightley *et al.* 1995, De Visscher & Van Cleemput 2003) and the effects of vegetation and pH (Hilger *et al.* 2000b) to see if external factors could be manipulated to increase microbial activity. Dramatic increases in oxidation rates were reported for media alternatives to soil, such as compost (Humer & Lechner 1999, 2001b, Wilshusen *et al.* 2004b) and inert manufactured media (Gebert & Groengroeft 2006b) due to more favourable physical and bio-chemical properties provided in the engineered media.

Biocovers

One of the first attempts to quantify methane oxidation in landfill cover soils was made by Whalen *et al.* (1990), who documented about $45 \text{ g CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ ($= 63 \text{ L CH}_4 \text{ m}^{-2} \text{ day}^{-1}$) at normal conditions: 1013 mbar; 0 °C) uptake rates in laboratory tests of soil cores taken from a closed municipal solid waste (MSW) landfill. Since then, various landfill sites and types of cover soils have been tested for their methane oxidation capacity in laboratory and field trials, the latter mostly using stable isotope techniques (Liptay *et al.* 1998, Chanton *et al.* 1999, Chanton & Liptay 2000, Borjesson *et al.* 2001). The findings generally converge to suggest that high oxidation capacity is associated with coarse, porous and well-structured substrates that are often rich in organic matter.

Humer & Lechner (1999) experimented with various composted materials as supports for methane oxidizers, and they developed some of the first prototype 'biocovers' for testing (Figure 2). Using mature compost proved to be important to avoid interference by heterotrophs competing for oxygen supplies (Humer & Lechner 1999). Mature compost can be operationally defined as compost with a 7-day oxygen demand below $8 \text{ mg O}_2 \text{ g}^{-1} \text{ dry matter (DM)}$ (Humer & Lechner 2001a). In laboratory soil column simulations, mature and porous compost enhanced methane uptake, showing markedly higher and rapidly increasing oxidation rates relative to conventional landfill cover soils. Another virtue of using mature compost is that organic mineralization and structural changes are minimized. Long-term observations showed that composts with initially mature and well-stabilized organic matter [initial total organic carbon (TOC) contents between 12–16% DM] mineralized relative slowly, with only a 10–

15% TOC reduction over more than 5 years (Huber-Humer 2004b).

Along with compost maturity, sufficient porosity was also identified as a key variable for optimum methane consumption (Humer & Lechner 1999). The porosity of a medium can be deduced from particle size distribution data and a measure of the air-filled pore volume. A substrate that combines the characteristics of fine texture and sufficient pore volume (e.g. sewage sludge mixed and composted with large wood chips) will foster good air diffusion from above and sufficient retention time of methane in the substrate. Suitably structured compost substrates investigated in the Austrian study (Huber-Humer 2004b) provided air-filled pore volumes of 30–45% v/v at moisture contents ranging between 40–50% w/w wet matter. Of course, the long-term structure of compost can change due to settling and biological activity. The degree of change depends mainly on the relative proportions of inert, easily biodegradable, and poorly biodegradable materials present. When MSW compost containing glass cullet, plastic parts and stones as the primary structural materials was compared over 3 years to a sewage sludge compost in which wood was the principal structural medium, the particle size distribution in the MSW compost had changed little whereas the sewage sludge compost clearly contained more finer particles than were present at the outset of the trials. The sewage sludge compost shifted from an initial distribution of 20% w/w < 2 mm and 50% w/w < 6.3 mm to 50% w/w < 2 mm and 80% w/w < 6.3 mm over the 3 years (Huber-Humer 2004b). Interestingly, the methane oxidation performance was not negatively impacted during the trial, which was attributed to the fact that the original sewage sludge compost was very coarse, and even after the particle sizes changed, it was able to provide higher porosity than the finely sieved MSW compost.

The results from these trials demonstrate that the nature and size of compost constituents and their propensity for degradation can all affect the ability of a medium to support and sustain good methane uptake. Differences in compost properties will, however, exist due to the natural heterogeneity of the input materials and various composting process variables. Thus, standardization of comprehensive quality criteria for their applicability for biocover construction is difficult and currently hardly practicable. Further research is needed on this issue.

Some of the first field trials to investigate compost covers were carried out on two different Austrian MSW landfills between spring 1999 and winter 2002. The purpose was to design a cover to enhance biotic methane removal as well as to minimize leachate generation under mid-European seasonal conditions. After testing various designs over several years, a simple but efficient two-part cover system proved most effective. It consisted of a layer of up to 1.2 m of mature, well-structured compost underlain by a 0.3–0.5 m coarse gravel layer to provide high gas permeability. While the function of the sub-layer was to homogenize gas fluxes, the porous upper layer served to support good methane oxidation activity by

providing, a balanced oxygen and methane supply, proper moisture content and viable temperature regimes.

Although a strong decline in methane oxidation was evident in conventional or shallow cover soils during the winter (Liptay *et al.* 1998, Chanton & Liptay 2000, Börjesson *et al.* 2001), there was no decrease in methane emission mitigation at the Austrian study site, which was probably due to the good insulation properties afforded by the biocover design. In optimally designed compost covers the year-round methane removal rate (relative to an adjacent open landfill reference cell) was 95–99%, depending on the kind of compost applied (Humer & Lechner 2001a, Huber-Humer 2004b). The high removal rate was mostly linked to the installation of a coarse gas distribution layer for balanced methane fluxes and the good insulation effect due to sufficient cover dimension and the use of proper substrates. In contrast, variants in the same trials that had shallower compost layers (about 30–40 cm) and no gas-balancing layer removed only 68–74% of the emitted methane.

Within the constraints of meeting certain permeability and stabilization requirements, biocover modifications can be made to adapt designs to meet local site-specific conditions and performance objectives. Thus, the dimensions and thickness of a biocover may vary depending on the nature of available materials, likely settlement behaviour (particularly when the covers are placed without any artificial compaction), climate conditions (precipitation, temperature, frost penetration depth), expected gas fluxes, the purpose of the cover (final or temporary), and the intended after-use of the site (vegetation, land use).

When a biocover was designed for a Florida (US) site, where sub-tropical conditions predominate, 50 cm of a 3-year-old yard and garden waste compost was laid on top of 10–15 cm of crushed recycled glass distribution layer and placed on an already existing interim cover made of about 65–75 cm of sandy clay and sandy loam (Bogner *et al.* 2005, Abichou *et al.*, 2006, Stern *et al.* 2006). The interim cover also acted as a control. Using isotope tracer studies, it was determined that the methane uptake in the biocover was almost double (64%) that of the control interim cover (30%). Methane emission rates from the biocover ($1.2 \text{ g CH}_4 \text{ m}^{-2} \text{ day}^{-1}$) were 10-fold lower than the $10.6 \text{ g CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ measured on the intermediate cover. The authors concluded that the thickness and higher moisture-holding capacity of the biocover increased the retention time of gases in the cover, and methane entered the biocover from below at a slower rate relative to the interim cover, so that a greater portion of the gross methane flux could be oxidized (Stern *et al.* 2006). Moreover, the biocover depth provided better protection against desiccation.

A field-scale biocover was also tested on an MSW landfill in Louisville, Kentucky that had an operating gas extraction system. Both a flat and a sloped biocover section were tested, where each was made of a 0.15 m clay layer overlain with a 0.15 m layer of tyre chips for gas distribution and a 1 m layer of yard waste compost. The control plots contained a 1 m

thick conventional clay cover (Barlaz *et al.* 2004). With the gas collection system off, methane emissions from the biocover cells did not increase, whereas those from the conventional soil cover rose significantly. When the collection system was operational, the soil cover generally performed well, although it occasionally released large quantities of methane thought to be mainly associated with desiccation cracks. No such cracks were observed in the biocover cells, leading the authors to conclude that compost-based biocovers not only reduce emissions through biotic mitigation, but also, through their propensity to resist erosion and cracking, can vent large gas flows.

Well-functioning biocovers can also act as a sink for atmospheric methane even when not paired with gas extraction systems. Several biocover studies report measuring negative gas fluxes in biocovers but not in conventional cover layer controls (Barlaz *et al.* 2004, Bogner *et al.* 2005, Stern *et al.* 2006). When a gas extraction system is functioning, it creates a negative pressure that draws air into the landfill through the cover, where atmospheric methane can be oxidized. Even in the absence of gas extraction, however, some atmospheric air can be drawn into a highly active biocover. This occurs due to the phenomenon whereby more moles of gas (methane and oxygen) are consumed in methane oxidation than are produced, because some of the water product exists as liquid rather than vapour.

Currently, Austria has at least five closed MSW landfills or sections of landfills covered with systems designed according to Humer & Lechner (2001a) (i.e. 0.5 m gravel gas distribution layer overlain by up to 1.2 m of mature, well-structured compost or waste substrates). These biocovers are serving either as the sole means to mitigate methane emissions on smaller, older sites or in combination with an operating gas extraction system as an additional measure to capture emissions that escape gas collection. Presently, these sites are fitted with a quasi state-of-the-art biocover design, the construction of which has been officially approved in Austria as an acceptable interim MSW landfill cover for a period of about 20 years. During this period, the biocover performance must be thoroughly monitored and documented. To date, the longest practical operating biocover is in Austria and has been monitored for 6 years. The data show that flat, undisturbed biocover areas have been consuming nearly 100% of the potentially emitted methane over the entire investigation period. Only in border areas and zones around physical installations such as drainage or gas wells are sporadically high methane emissions detected, particularly when the gas extraction system is turned off. Thus, site-specific trouble spots such as this will require particular attention during the life of a biocover.

The quality of a biocover can be checked with a flame-ionization detector (FID) mapping unit to detect surface methane concentrations that may indicate leaks, but for determining the overall effectiveness of such a system, the methane influx (reference flux or emission) into a biocover must be known, which is typically a more complex measure-

ment. These reference values can be defined by flux measurements on the site prior to biocover application, on adjacent control cells. In some instances, landfill gas production data for the specific site can be surveyed, calculated or modelled. Due to temporal and/or spatial variability, however, noticeable discrepancy can occur between reference influxes and day-to-day values.

Biofilters

Biofilters for methane mitigation are patterned on similar engineered systems for filtering air for odour or organic contaminants. They are configured as self-contained, fixed-bed reactors containing a packing material that can support and sustain a population of methane oxidizers. In contrast to biocovers, biofilters can be operated in combination with conventional caps, but the filters require either an active or passive gas collection system to feed the filter. Biofiltration is particularly appropriate when active landfill gas extraction and subsequent energy recovery or flaring is no longer or not yet viable. It is a suitable measure for all the situations described in the introduction to this paper. Several biofilter designs, media and gas flow regimes have been tested in laboratory and field experiments. The filters are operated as either open or fully contained beds. While active gas feed can be controlled, passive feed is driven solely by the pressure gradient between the landfill and the atmosphere (Straka *et al.* 1999, Dever *et al.* 2005, Gebert & Gröngroft 2006a). The gas can be directed through the filter either in up-flow or in down-flow mode (Figure 3).

Open bed biofilters can be integrated into the landfill cover system and vegetated. They are typically operated as 'robust' systems without supplementary heating or irrigation (Straka *et al.* 1999, Dever *et al.* 2005, Gebert & Gröngroft 2006b), with landfill gas supplied and distributed at the bottom and metabolized during upward flow. Oxygen is supplied either by diffusive ingress from the atmosphere or as a supplement to the landfill gas supply pipe.

In contrast to open beds, fully contained biofilters are enclosed, so that oxygen must be fed via the gas supply line (Streese & Stegmann 2003, Du Plessis *et al.* 2003) or by injection into different layers of the biofilter (Haubrichs & Widmann 2006). Fully contained designs are more highly engineered to control methane and oxygen fluxes and maintain optimum temperature and moisture conditions. Their capital and operating costs are, however, considerably higher than those for passively vented, robust open bed applications.

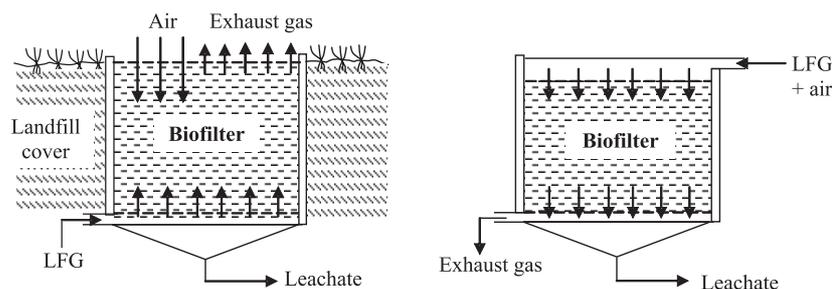


Fig. 3. Variants of biofilter design. Left: up-flow biofilter integrated into landfill cover; right: biofilter in down-flow mode. LFG, landfill gas.

As with biocovers, biofiltration media must offer a large gas permeable pore space, a large surface area, and good environmental conditions for the microbes. The latter would include factors such as sufficient water-holding capacity, appropriate pH, conductivity, and perhaps nutrient availability. High permeability is important to minimize pressure loss, particularly in passive systems. As too much settlement may reduce the permeability and promote the formation of anaerobic niches, the best medium will be inorganic or well-stabilized organic material that is resistant to microbial degradation. A good medium will also be homogeneous to avoid aggregation and segregation processes and to minimize preferential flow that can overload some filter sections.

To date, a variety of media have been laboratory-tested as candidates for methane biofiltration. The materials have included composts of various origins (Figueroa 1996, Straka *et al.* 1999, Streese & Stegmann 2003, Wilshusen *et al.* 2004b, Dever *et al.* 2005); wood chips, bark mulch or peat; inorganic materials such as glass beads (Sly *et al.* 1993), bottom ash (Maurice & Lagerkvist 2004) or porous clay pellets (Gebert *et al.* 2003); sands and soils (Park *et al.* 2002, Powelson *et al.* 2006); and mixtures of organic and inert materials (Du Plessis *et al.* 2003, Melse & Van der Werf 2005). In diverse laboratory column studies (including long-term investigations up to 375 days), methane oxidation rates of 20–60 g m⁻³ h⁻¹ were observed (Sly *et al.* 1993, Park *et al.* 2002, Streese & Stegmann, 2003, Wilshusen *et al.* 2004b, Haubrichs & Widmann 2006).

Good biofilter performance has also been reported in various field-scale applications. Streese (2005) found stable methane removal rates of 10–20 g CH₄ h⁻¹ m⁻³ (70 g m⁻³ h⁻¹ max) for an actively vented compost biofilter (four units of 1 m³ each) operated at 20 °C in down-flow mode. The filter was fed with landfill gas/air mixtures at CH₄ concentrations of around 2% v/v. Gebert and Gröngroft (2006b) investigated a passively vented open bed that was integrated into the landfill cover and operated under ambient temperature and humidity conditions. A medium of inorganic porous clay pellets topped by 10 cm of densely grassed topsoil achieved methane oxidation rates of up to 80 g m⁻³ h⁻¹.

When mixtures of compost and bark or wood chips were used in a passive up-flow open bed system operated under ambient conditions, methane removal rates of more than 90% were reported for loading rates of 1.1–2.5 m⁻³ h⁻¹ m⁻³ (Straka *et al.* 1999). A compost biofilter integrated into the cover of a MSW landfill in Western Canada added a passive

heat exchange system to transfer heat from inside the landfill to the filter bed. The filter operated at 14–18 °C and consumed 89% of the influent methane flux, or up to 40 g CH₄ m⁻² day⁻¹ (Zeiss 2006).

Together these studies have documented the good potential for methane biofiltration in landfills. They also reveal some of the key challenges to successful biofilter operation, especially for robust open beds. For example, high input LFG fluxes can limit biofilter performance if they impede diffusive oxygen ingress from the atmosphere (Gebert & Gröngroft 2006b). Therefore, the media diffusivity and anticipated LFG flux must be factored into biofilter sizing to avoid oxygen limitation. Low ambient temperatures will also slow methanotroph metabolism and methane uptake (e.g. Whalen *et al.* 1990, Gebert *et al.* 2003, Streese & Stegmann 2003, Börjesson *et al.* 2004, Scheutz *et al.* 2004), and the media of non-irrigated open beds need to have suitable water-holding capacity and be protected from desiccation. One clever design had differently textured sands installed in a fining upward gradient of particle sizes so that the fraction of gas-filled pore space increased downward (Powelson *et al.* 2006). As a result, water contents were higher near the top due to the higher matrix potential there, but lower near the base, which might otherwise be subject to water-logging.

A final problem to be avoided is the formation of exopolymeric substances (EPS), which can agglutinate the biofilter material and markedly limit mass transfer in the bed. EPS production has been reported in vented columns fed 36–130 g CH₄ m⁻³ h⁻¹ (Streese & Stegmann 2003, Wilshusen *et al.* 2004b, Haubrichs & Widmann 2006), but not in the passively vented open bed biofilters. As passive biofilters receive gas in relation to the prevailing pressure gradient between the landfill and the atmosphere, the LFG load tends to be variable or intermittent, and prolonged exposure does not occur. It may be possible to curtail EPS formation by controlling the rate of inlet fluxes to a landfill biofilter.

Biowindows

Whereas biocovers are designed to cover all or large sections of a landfill, biowindows are relatively small regions of cover on a landfill or open dump. They are useful when a full-expanse biocover is not warranted or economically feasible, and when no gas collection system is present that can connect to a biofilter. Biowindow media is often compost, and the windows are arranged in discrete sections integrated into the landfill cover (Figure 4). In contrast to biofilters, biowindows are usually not contained in a support structure. The windows receive biogas directly from the underlying waste. As with biocovers, gas migrating through the cover naturally routes itself through the windows, since the lower permeability there offers the path of least resistance. In Germany, many old dumps have been remediated using biowindows in combination with bentonite mineral liners. The Danish EU-Life project BIOCOVER (Kjeldsen *et al.* 2007) is currently investigating the efficiency of biowindows for the mitigation of methane emissions from an old Danish landfill. The objec-

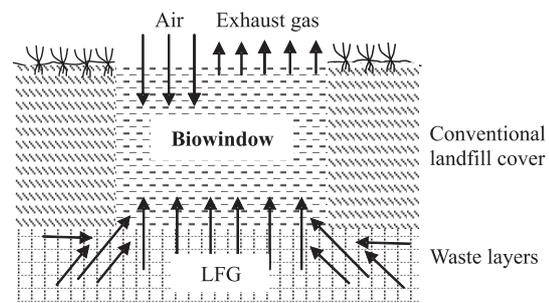


Fig. 4: Conceptual scheme of biowindows. LFG, landfill gas.

tives of the study are to use microbial methane oxidation in passively vented biowindows to reduce the greenhouse gas emissions; to demonstrate a method to document the emission reductions; to analyse the economic viability of the technology; and to develop guidelines on how to incorporate biowindows into the landfill gas management practices at European landfills. The emission measurement methods used during the baseline study and after placement of the biowindows include robust and established approaches such as the static flux chamber and soil gas profiles as well as advanced tracer-based plume analyses for quantifying whole-site emissions (Scheutz *et al.* 2007).

Methane mitigation before capping: biotarps

Biocovers, biowindows and biofilters are typically applied after landfill cells are completed and awaiting intermediate or final cover. However, methane is being produced during landfill cell construction, because soon after waste placement, anaerobic conditions prevail and promote the biochemical reactions that lead to methane genesis. It is well known anecdotally (Reinhart, personal communication) and documented by field reconnaissance data (Bogner, unpublished data), that methane release from open cells does occur (see also Figure 1).

Typically, only a small portion of a landfill is available for waste disposal at a given time. An open cell will be operational for some period of time as it is alternately layered with waste and then daily soil cover until it is filled to a predetermined height. During typical landfill operations, several cells may be filled to this elevation and then overlain with a 30–45 cm layer of intermediate soil cover for several months or years. Methane production and emission will occur during much of the time the waste is in place and awaiting final capping.

One line of research that aims to address methane releases during the active life of a landfill cell is focusing on the design of a 'biotarp' (Hilger *et al.* 2007). Conceptually, this would be a removable tarp impregnated with methanotrophs. While most landfills use a 15 cm layer of soil for daily cover, there are a variety of alternate daily covers (ADCs) that some landfills use because of favourable economics (SWANA 1996). Waste materials such as sewage sludge or paper and water slurries or commercial products such as foams and canvas

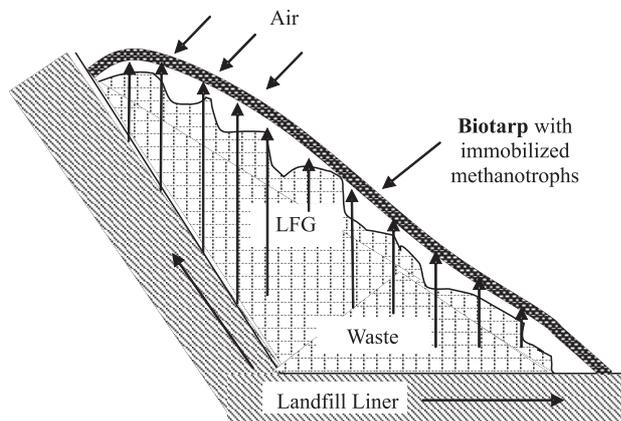


Fig. 5: Conceptual scheme of biotarps. LFG, landfill gas.

covers have all been employed as ADC on landfills (Haughey 2001).

The 'biotarp' is designed to cover the waste at the end of each working day. While in place, it would mitigate methane releases from the decaying wastes below, and then each morning it would be removed to allow for further waste placement (Figure 5). A variety of prototype tarp materials have been tested in batch for their ability to support a viable and active population of methane oxidizers under varying climate and methane availability conditions. As for the biocover and biofilter, moisture-holding capacity and porosity are important properties for the tarp matrix, but an equally potent variable is proving to be the surface area of the matrix available for colonization and the mass/volume density of the tarp material (Hilger *et al.* 2007). Unlike biocovers, biowindows and biofilters, the tarp must be manageably manipulated day-to-day. It must be moist enough to support good methane oxidation, but light enough to roll or fold. Size and weight constraints also limit the depth of the tarp, which affects the methane retention time that can be provided.

Laboratory trials have shown that methanotrophs adsorb readily to a variety of support materials and that once adhered, they are fairly resistant to removal. Even after 5 h of shaking at $4.31 \times g$ (where g is the gravitational 'force' that would cause the centrifuge tube mass to settle without centrifugation), approximately 30% of acclimated cells remained adsorbed to a geotextile support (Hilger *et al.* 2007). Methanotrophs were also readily immobilized in gel capsules and entrapped in calcium alginate beads, although these tended to desiccate quickly when not immersed in liquid. Adsorbed cells tested for their ability to withstand alternating 12-h intervals with or without methane showed that this variability in substrate supply will stress the cells and lower their uptake rates (Hilger *et al.* 2007). It may, however, be possible to select for methanotrophs that are most resistant to stress so that high rates of methane consumption will resume soon after a biotarp is emplaced.

One of the advantages of a biotarp over biocovers and some biofilters is that the support matrix is inert and not subject to biochemical degradation over time. Although some

wear and tear is expected and intermittent replacement will be needed, the tarp offers a stable base of support for microbial methane oxidizers and could be used for long-term intermediate cover alone or in tandem with biocovers and biofilters. Further, when it is no longer needed, no landfill volume will have been sacrificed to accommodate its use. In the near future, trials will be conducted to test the capacity of this methane capture method in the field. The methane flux at several locations on an operating landfill will be measured with and without prototype biotarps in place to assess their potential to mitigate early methane emissions from recently placed wastes.

Scope of applications of biological methane mitigation

Aftercare

At some point after landfill closure when gas production slows, gas collection or flaring is discontinued, but small amounts of biogas release continue. Biofilters can be connected to the existing gas drainage system to treat these low calorific emissions. Gas can be supplied actively or passively, with the latter requiring a collection system that offers a preferential pathway that can route a significant share of residual gas to the filter. Likewise, biocovers can be applied, especially in cases where there is no gas collection system. They can continue to act as a long-term evapo-transpiration and methane cleaning layer, particularly when vegetated with plants that will take up water and limit infiltration.

In addition to their use for capping or as a capping amendment, bio-based systems can be valuable elements in remediation schemes for closed landfills requiring aftercare intervention. For example, when in-situ aeration is used to accelerate and complete waste degradation in closed landfills, it generates mostly CO_2 , but some methane is produced in anaerobic pockets where the air does not fully penetrate (Prantl *et al.*, 2006). Biofilters can capture these low-level methane emissions, as has been demonstrated in field investigations (Scharff *et al.* 2003). Ritzkowski *et al.* (2006) proposed that regenerative thermal oxidation (RTO) be used until methane concentrations fall below 0.6%, and then a biofilter be used after that to capture residual methane emissions. Biocovers would be equally suitable for such applications.

Old landfills and dumps

Unlike old landfills, dump sites can be more problematic because they may have no top cover; they may still emit substantial quantities of methane; and there may be no legally responsible party to be held accountable. Some interesting low-cost biocover and biofiltration system trials have been conducted for such sites. At one Austrian site, the upper waste layer consisted of excavated soil, demolition waste, and decade-old excavated and relocated municipal solid waste. The well-decomposed material from 0.5–2 m deep in this layer was sieved and tested in 3-month bench scale studies to assess its suitability for use as a biocover. It proved to be about one-quarter as effective (approx. $15 \text{ L m}^{-2} \text{ h}^{-1} \text{ CH}_4$

Table 1: Methane oxidation efficiency of an old municipal solid waste material (WM) and mixtures of this material with 30 vol% (WM₃₀) and 50 vol% (WM₅₀) structural compounds tested in continuously charged laboratory columns

Methane flux	Oxidation efficiency in L CH ₄ m ⁻² h ⁻¹ and (%)		
	WM	WM ₃₀	WM ₅₀
4–4.5 L m ⁻² h ⁻¹	4.0–4.5 (100%)	4.0–4.5 (100%)	4.0–4.5 (100%)
9.5–10.5 L m ⁻² h ⁻¹	3.8–5.3 (40–50%)	5.2–6.8 (55–65%)	7.5–8.5 (80%)

uptake rate) as well-decomposed mature compost (Humer & Lechner 1999, 2001b; Huber-Humer 2004b), but when mixed with bulky wood components (shredded wood and yard waste), its methane oxidation capacity improved. The methane uptake rate of a 50 : 50 v/v mix was more than two-fold higher than that of the unamended waste (Table 1).

The increased porosity caused by the bulky compounds probably provided better gas distribution and exchange. Additional testing is underway to discern how much manipulation (sorting, sieving) and what kind of bulky-item amendments would be suitable to prepare degraded MSW for service as biocover media and to render it aesthetically pleasing for long term placement. Scheutz *et al.* (2005) also found high oxidation capacity in the upper 40 cm of a landfilled automotive shredder waste with low organic content and coarse structure. Old dumps with remaining methane production potential [e.g. TOC > 4% DM (Prantl 2007)] are also suitable candidates for in-situ aeration accompanied by a biofilter or biocover.

MBT landfills

MBT landfills are modern disposal sites filled with mechanically-biologically pre-treated waste. The MBT-strategy is being intensely pursued in some EU countries, particularly in Germany and Austria. These landfills have some unique features that make them a good fit for biotic methane oxidation systems. The organic content and usually the reactivity of the pretreated waste material in MBT fills are low, so that methane production will be marginal but not unappreciable with respect to climate impacts. In Germany and Austria, the national landfill directives allow passive degasification combined with biofiltration (in biofilters or biocovers) for MBT fill sites, based on the experiences from early and on-going laboratory and monitoring studies. Such studies showed that composted biosolids oxidized 144–196 L CH₄ m² day⁻¹, which well exceeded the anticipated MBT landfill methane emission rates of 24–48 L CH₄ m² d⁻¹ (Felske 2003), and trials with other compost types and even organic-rich soil are showing that there are a number of potential media for bio-based systems engineered for MBT landfills (Huber-Humer 2004b).

Moreover, mature MBT material itself can be used for biocover construction when the material contains bulky compounds to provide adequate porosity and when it is emplaced loosely to maintain that porosity. Einola *et al.* (2007) investigated MBT residuals that had been aerobically stabilized for 5–12 months. When tested in laboratory col-

umns and batch tests, it proved to be a good support medium for methane oxidation, even at low temperatures.

Since the fill in MBT landfills is compacted densely, cover design must include a minimum 3% slope to divert runoff and prevent infiltration that could contribute to very concentrated leachate. Such diversion is also essential to avoid geotechnical instability and slope failures of the MBT fill, which is vulnerable to slumping. As with biocovers for conventional landfills, a good gas distribution layer and sufficient thickness to insulate against large temperature changes are required for good performance. Felske (2003) recommends an 85 cm thickness for middle-European climates. When high annual precipitation is expected, a capillary barrier system (at slopes > 10% inclination) is recommended to reduce infiltration and serve as a gas distribution layer (Wawra & Holfelder 2003). Such designs must, however, avoid sharp interfaces between materials that could lead to water saturation and oxygen limitations in the biotic layers (Berger *et al.* 2005).

Landfills in low-income countries

Waste management practices are still quite rudimentary in many low-income countries. The high urban population growth and higher per capita waste generation rates predicted to accompany their increasing economic development means that there is, and will continue to be, an urgent need for low-cost waste disposal systems in the developing world. Such systems must provide sanitary conditions locally and also limit methane releases that contribute to global climate change. As biological methane oxidation systems can use natural materials, are relatively inexpensive, and can be easily constructed and maintained, they are promising options for countries with minimally developed waste management programmes. Biotic systems would be viable alone or as components of more sophisticated systems that may evolve through the clean development mechanism (CDM), which was proposed as part of the Kyoto Protocol.

The CDM is a means for developed countries to invest in sustainable projects through carbon credit purchases that would go toward reducing greenhouse gas emissions in the developing world (Lee *et al.* 2005). It has the potential to greatly accelerate installation of landfill gas recovery systems (Bogner 2006) that could be supplemented with biotic methane oxidation systems. In the absence of gas collection, a practicable and inexpensive alternative could be to use a simple mechanical-biological waste stabilization pre-treatment step (such as composting) accompanied by a biocover that could capture any remaining escaping methane produced.

Conversely, a portion of the composted product could be used for biocover construction.

The feasibility of using biocovers on landfills in India, where no gas collection or recovery systems exist, has been examined in the context of a typical urban landfill in Delhi (Mor *et al.* 2006b). Based on emission estimates for this landfill and on the steady-state oxidation activity of specific compost materials measured in laboratory batch tests (Mor *et al.* 2006a), it was deduced that a 28–55 cm compost layer could theoretically oxidize all methane emitted from the landfill. The authors recommend that additional cover thickness beyond that needed for complete oxidation be applied to prevent desiccation in lower cover regions and conserve moisture in dry periods.

Clearly, continued research will be needed to assess the performance and long-term functionality of biocovers in each geographical region where they are considered. Weather extremes such as drought and monsoon seasons will impact biotic and physical soil conditions available to support methanotroph growth. Previous study of landfill soil incubated at the high soil temperatures typical of tropical climates showed that such soil was very suitable for methane oxidation (Visvanathan *et al.* 1999). The authors suggested that methane uptake would probably be optimum immediately after a rainy season, and that high activity could be maintained through leachate recirculation during dry seasons. As compost offers both large pore spaces and higher water-holding capacity than soil, an alternate or supplementary option might be to use compost for cover construction or amendment.

High organic content materials such as composts not only absorb water but retain it over a longer time period than soil. When the upper layer (e.g. 10–20 cm) of the compost dries, there is a hydro-physical phenomenon that occurs whereby the desiccation makes the dry layer hydrophobic, which impedes the capillary rise of water and limits its evaporation from lower layers, particularly if the cover remains un-vegetated (Huber-Humer 2004b).

Conclusions and outlook

Bio-based landfill methane mitigation systems are well-suited and cost-effective for a variety of applications where prolonged low-level methane emissions occur. Biotic systems can be readily configured to meet site-specific topographic, climatic, and logistical conditions and needs. They exploit the natural propensity of methanotrophs to consume methane by optimizing conditions for their habitation and providing a means to route LFG to them. Biocovers offer the advantage of full-landfill coverage, so that the flux burden is dispersed over a large surface area, and the risk of untreated emissions is minimized. Further, they provide good water-holding capacity and porosity for vegetation, which limits rainwater infiltration and promotes evapo-transpiration. Biowindows may be sufficient when emissions are quite low, when only a few 'hot spots' are releasing biogas, or when the supply of support media is limited. When gas collection lines

exist, biofilters may be appropriate because of their small footprint and high uptake capacity. It is likely that other bio-based methane oxidation systems, such as the biotarp, will continue to appear to meet targeted geographical, technical, or regulatory demands. A summary table comparing different bio-based systems is provided in Table 2.

A good body of knowledge has amassed characterizing the capacity of these low-cost options to reduce landfill methane emissions and thereby mitigate landfill climate change impacts. Many of the key factors for good performance have been identified. Nevertheless, there is still much work to be done to translate these findings into technical design and performance assessment guidelines that will ensure good methane removal but allow for continued innovation and cost reductions. Some of the issues yet to be resolved include defining the acceptable level of variability among compost or other media batches, specifying acceptable compost feedstocks, assessing the effects of different kinds of vegetation on cover performance, estimating the life expectancy of a cover, assessing the mitigation potential of different bio-systems under different climate conditions, and documenting the ability of bio-based systems to reduce non-methane organic compounds.

A number of full-scale research projects in Germany, Denmark, Canada and Australia are already underway to address some of these questions. In Germany, the MiMethox project (Microbial Methane Oxidation in Landfill Covers) will develop and test cover designs to sustainably reduce methane fluxes from landfills generating low calorific gas, including sites where fill is mechanically and biologically pretreated waste (Gebert *et al.* 2007). In Canada and Australia, biofilter test cells of different layering and material have been constructed on landfills to evaluate the methane abatement potential under the particular environmental conditions posed by nordic and arid climates (Dever *et al.* 2005, Philopoulos *et al.* 2006, Cabral *et al.* 2007). An interdisciplinary project on remediation of old sites and dumps (NUTZRAUM) started in Austria in spring 2007 is focusing on the design of landfill covers (including biocovers) on the top of sites subjected to in-situ aeration to optimize both methane emission reductions and water infiltration. Finally, field trials will commence in the US to test a variety of prototype biotarps against conventional intermediate soil covers for their relative methane uptake capacities.

It is likely that new policy initiatives will be undertaken as the potential contributions of bio-based methane oxidation systems at landfills are better understood. It is anticipated that these initiatives will in turn drive more interest in, research about and adoption of such systems in the near future. One policy change is already anticipated in Germany, where it is expected that, for the first time, legislation will set the biogas emission levels allowed in order for sites to be released from aftercare requirements. The draft legislation stipulates that the maximum allowable methane flux from a recultivation layer be $0.5 \text{ L CH}_4 \text{ m}^{-2} \text{ h}^{-1}$ and the total allowable residual methane production level be $25 \text{ m}^3 \text{ CH}_4 \text{ h}^{-1}$.

Table 2: Summary of some specific features of diverse bio-based oxidation systems: benefits and drawbacks of systems are related and compared.

	Biofilters		Biowindows	Biocovers	Biotarps
	actively vented	passively vented			
Field of application	With gas extraction as interim or final measure, during landfill operation, aftercare or on old landfills	Without gas extraction as interim or final measure, during landfill operation, aftercare or on old landfills	Interim or final measure within (existing) cover, mostly on old landfills	Interim or final cover, in addition or alternative to gas extraction; during landfill operation, after-care or on old landfills, remediation of old landfills	As daily cover during operational phase
Materials used (examples)	In-organic or organic engineered materials (e.g., compost, bark mulch, manufactured clay pellets, peat & sand mixtures)		In-organic or organic engineered materials (like biofilters)	Coarse soils, composts, soil/compost mixes, engineered waste materials	Roll- or foldable inert matrix (e.g., geo-synthetic mats) impregnated with methanotrophs
Benefits	<ul style="list-style-type: none"> effectiveness can be readily monitored operational conditions can be manipulated and controlled. 	<ul style="list-style-type: none"> operational conditions (except gas supply) can be manipulated and well controlled no gas extraction system needed 	<ul style="list-style-type: none"> installs quickly and easily no gas extraction system needed a good option for point source leaks 	<ul style="list-style-type: none"> suitable for long duration but low flux rate high surface area yields low local gas loads and resists EPS formation high gas retention and oxidation efficiency supports vegetation and evapotransport 	<ul style="list-style-type: none"> mitigates pre-capping emissions provides daily cover conserves landfill capacity
Drawbacks	<ul style="list-style-type: none"> unsuitable for fugitive gas; requires gas collection sys. risk of methane overload (EPS) energy required for heating, irrigation, aeration requires an operating gas extraction system 	<ul style="list-style-type: none"> unsuitable for fugitive gas; requires gas collection sys. risk of methane overload (EPS) effectiveness more complex to evaluate requires connection to an existing gas extraction system 	<ul style="list-style-type: none"> limited coverage area for fugitive emission capture risk of methane overload and EPS formation effectiveness difficult to evaluate 	<ul style="list-style-type: none"> can be limited by substrate quantity demand monitoring is laborious effectiveness assessment is complex limited control of operational conditions 	<ul style="list-style-type: none"> more expensive than conventional ADC no field data available; research is addressing: <ul style="list-style-type: none"> desiccation retention time effectiveness

These levels could reliably be treated with well-designed bio-based methane oxidation systems (Stegmann *et al.* 2007). Perhaps in anticipation of such drivers, the most recent IPCC

Working Group III assessment report lists biocovers and biofilters as the key mitigation technologies and practices projected to be commercialized before 2030 (IPCC 2007b).

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