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MASTER THESIS

Optimizing Microbial Methane Oxidation Systems for Various Landfill Configurations: A Study on Biofilters, Biowindows, and Biocovers.

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To all the future generations
of humanity...

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Abstract

Landfills are significant contributors to global greenhouse gas emissions, primarily methane (CH_4), a potent greenhouse gas. This thesis explores the efficacy of Microbial Methane Oxidation Systems (MMOS) in mitigating these emissions through the detailed study of biofilters, biowindows, and biocovers. Additionally, the thesis investigates the policy and regulatory framework governing sustainable waste management and landfill emissions in the European context. The research aims to elucidate the structural and operational nuances of MMOS, focusing on their design features, functionality, and the complex interplay between physical, chemical, and biological factors that influence their methane oxidation efficiency. The thesis work addresses critical questions regarding the impact of system characteristics on methane oxidation efficiency, the key factors influencing MMOS selection, and the challenges associated with their design, monitoring, and maintenance. By advancing knowledge on MMOS, this research contributes to informed decision-making and innovative solutions for the mitigation of greenhouse gas emissions from landfills, offering a pivotal step towards sustainable waste management practices.

1 Introduction

Anaerobic decomposition of organic waste within landfill sites leads to the generation of landfill gas, primarily comprising methane (CH_4; around 60%) and carbon dioxide (CO_2; around 40%). Methane is recognized for its significant global warming potential (GWP), being 28 times more potent than CO_2 over a 100-year period and 84 times within a 20-year span (Aghdam et al., 2019). Landfills stand as the largest direct source of greenhouse gas emissions within solid waste management, contributing an estimated 630 million tonnes of CO_2 equivalent globally (Fedrizzi et al., 2018) according to the fifth evaluation by the IPCC.

The anticipated increase in global landfill emissions is concerning, particularly given the projected growth in waste production, especially in developing countries, where substantial amounts of biodegradable organic waste are disposed of in landfills (Héroux et al., 2010). Effective reduction of emissions from landfills is crucial due to the high short-term GWP of methane, the concentrated nature of landfills as a point source, and the magnitude of greenhouse gas emissions they produce.

Despite regulations in the European Union (EU) strictly governing waste management (such as the Waste Framework Directive and the Landfill Directive), old landfills, even those already closed, still have the potential to release significant amounts of methane. In the EU, various options exist for reducing CH_4 emissions from landfills, ranging from measures prior to landfilling (such as separate collection of biogenic waste for composting or anaerobic digestion) to procedures during or after landfilling. These include landfill gas extraction, in-situ techniques to reduce the amount of deposited biodegradable waste (Kjeldsen & Scheutz, 2018).

While gas collection systems are recommended for controlling and recovering landfill gas emissions, they are not always 100% efficient, and emissions may escape from wells and along routes of installed landfill equipment. Additionally, energy recovery from landfill gas becomes technically and economically infeasible when methane concentrations fall below 35–40% v/v and total gas production rates are 30–50 $m^3\ h^{-1}$. In such cases, high-temperature flares become the most suitable treatment method. When methane concentrations decrease further to 20–25% v/v and landfill gas flow rates fall to 10–15 $m^3\ h^{-1}$, controlling methane emissions becomes more complex. Therefore, there is an imperative need to explore and optimize alternative methane mitigation strategies, and Microbial Methane Oxidation Systems (MMOS), emerge as the most promising technology (Huber-Humer et al., 2008).

Microbial Methane Oxidation Systems (MMOS) represent a sophisticated, engineered approach designed to mitigate the release of landfill gases by channeling them through a multi-layered bed of biofilter media that supports the growth of methanotrophic bacteria - microorganisms that consume methane as their primary energy and carbon source - to convert methane (CH_4) into carbon dioxide (CO_2), water (H_2O), and biomass. MMOS can be implemented in various configurations, tailored to specific landfill conditions and objectives. The three main types of MMOS utilized in landfills include: Biofilters, Biowindows and Biocovers.

This thesis investigates the structural and operational nuances of Microbial Methane Oxidation Systems (MMOS) in landfill covers, focusing on biofilters, biowindows, and biocovers. Through a detailed exploration of their design features, functionality, and the complex interplay between physical, chemical, and biological factors, this research aims to address several critical inquiries that underpin the effectiveness and suitability of these systems for various landfill scenarios. The overarching research questions guiding this study are:

- How do structural characteristics and operational mechanisms of biofilters, biowindows, and biocovers impact their methane oxidation efficiency?
- What are the key factors that influence the selection of MMOS, and how can these factors guide the optimization of MMOS for specific landfill scenarios?
- How do the structural and operational differences between biofilters, biowindows, and biocovers influence their suitability for specific types of landfills or waste compositions?
- What are the challenges associated with the design, monitoring and maintenance of MMOS and what are the strategies to overcome them?

By addressing these questions, the thesis aims to advance the knowledge base on MMOS, facilitating informed decision-making and innovative solutions in the mitigation of greenhouse gas emissions from landfills.

2 Sustainable Waste Management: Policy and Regulatory Framework

2.1 Current Waste Production and Policy

Waste management stands as a critical challenge worldwide, faced with the dual pressures of escalating volumes of waste and its environmental consequence. These challenges are however more pressing, peculiar, and prominent in developing countries, where institutional frameworks are often incomplete, and waste is poorly managed. The urgency of sustainable waste management as a global environmental agenda cannot be overstated, marking it as a pivotal concern for the 21st century. Sustainable development mandates a reduction in pollution emissions and the establishment of sustainable waste management practices. Aiming for environmentally sound waste management, particularly of hazardous waste, is a key target of the United Nations' Sustainable Development Goals.

In Europe, during 2020, waste production from all economic sectors and households amounted to a staggering 2,153 million tonnes, equivalent to 4,813 kg per capita, with the construction sector alone accounting for 37.5% of the total number. Meanwhile, municipal waste generation per capita in the EU stood at 505 kg, as depicted in Figure 1, leading to a total of 225.7 million tonnes of municipal waste—a 1% increase from 2019, and marking a 14% rise since 1995, equivalent to an additional 27.7 million tonnes.

However, the projections by the World Bank (2018) indicate that, driven by population growth and urbanization, global annual waste generation is expected to jump to 3.4 billion tonnes over the next 30 years. While constituting only 16% of the global population, high-income nations collectively produce over a third (34%) of the world's waste. The region of East Asia and the Pacific is accountable for approximately one quarter (23%) of total waste production. Projections indicate that by the year 2050, waste generation in Sub-Saharan Africa will increase by over threefold from present rates, whereas South Asia is expected to see a more than twofold rise in its waste output.

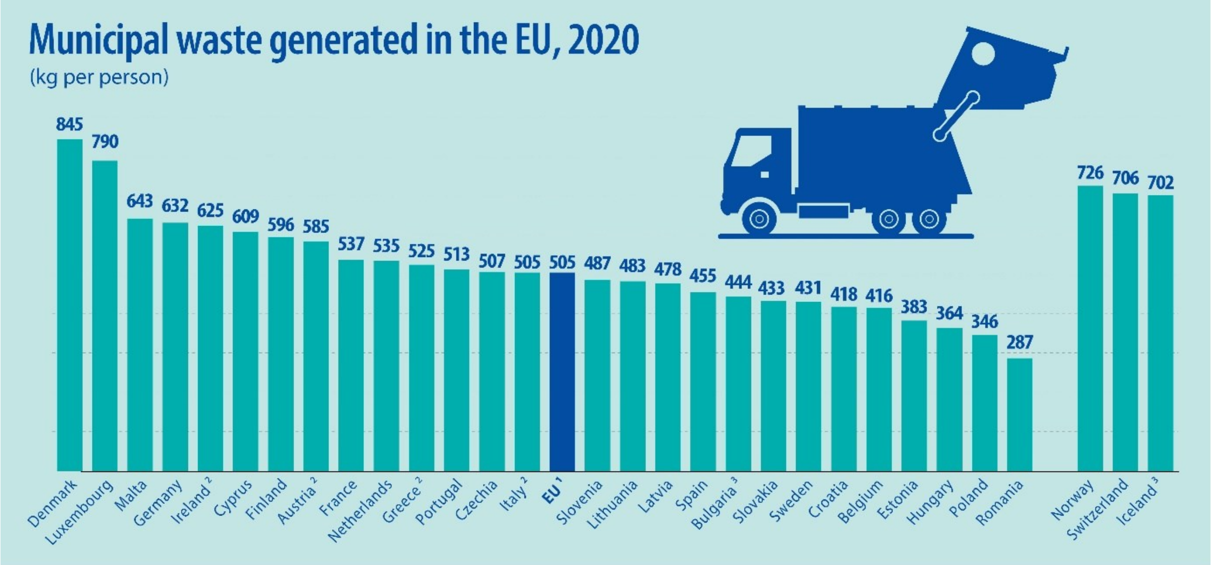
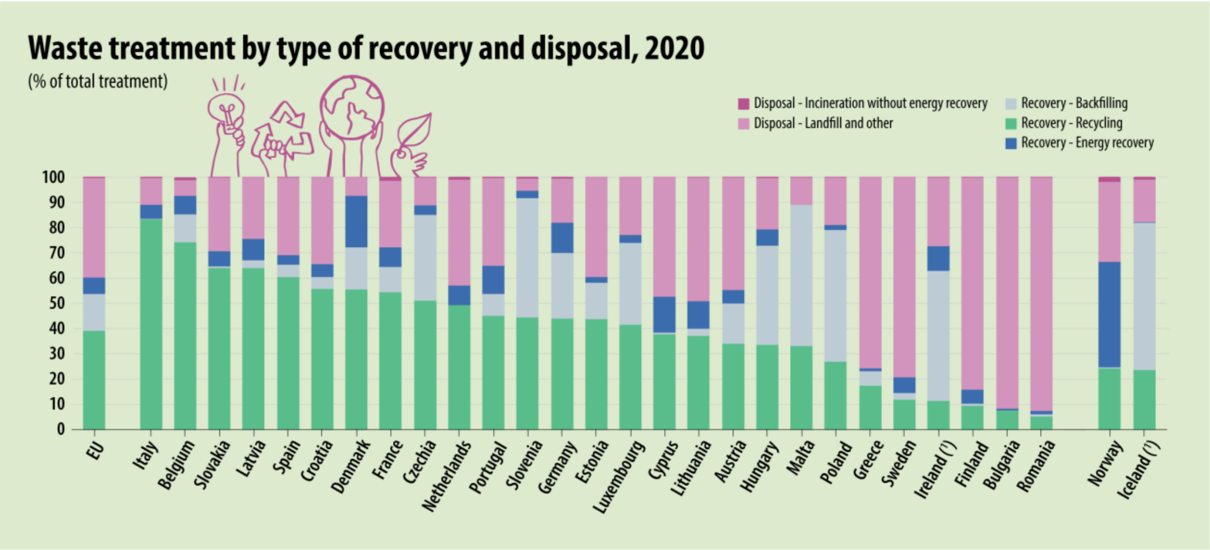


Figure 1. Municipal Waste Generated in the EU, 2020 (Eurostat, 2023)

European waste policy is based on the Circular Economy concept, emphasizing product value retention, resource conservation, and minimization of waste. In the countries of European Union (EU) in the year 2020, a significant portion of the overall waste generated, which amounted to 59.1%, was treated through various recovery processes. These operations included recycling, accounting for 39.9% of the total treated waste, backfilling at 12.7%, which involves utilizing waste in excavated areas for purposes like slope reclamation, safety measures, or engineering applications in landscaping, and energy recovery at 6.5% as documented by Eurostat, 2023).The remaining 40.9% of the waste was subjected to different methods, with 32.2% being landfilled, 0.5% incinerated without energy recovery, and 8.2% disposed of in other ways. The differences in the distribution of recovery and disposal proportions among EU member countries are outlined in Figure 2, based on data provided by Eurostat in 2023.



(1) 2018 data

ec.europa.eu/eurostat

Figure 2. Waste Treatment by Type of Recovery and Disposal, 2020 (Eurostat, 2023).

The Circular Economy concept has significantly bolstered separate waste collection, viewed by many as a panacea for disposal issues. However, this perspective often sidesteps the waste management hierarchy's nuanced demands—Prevention, Reuse, Recycling, Energy recovery, and Landfilling—leading to a simplistic and sometimes moralistic approach to waste management technologies. Notably, incineration and landfilling have been stigmatized, overshadowing their potential utility in waste management. This prevailing narrative neglects several critical points: not all waste is suitable for recycling; materials cannot be recycled indefinitely; recycling processes themselves generate waste; and hazardous substances can accumulate in recycled materials, posing health and environmental risks. The negative perception of landfills and incineration, fueled by EU communication strategies, has led to public resistance against the establishment of necessary facilities (Cossu et al., 2020).

To align with the principles of environmental sustainability, landfills should incorporate strategic measures aimed at managing waste stability and immobilizing contaminants effectively. This involves:

- the adoption of specific pre-treatments to reduce the volume and potential emissions of contaminants
- the optimization of biogas and leachate extraction processes to control mobile contaminants
- the implementation of in situ treatments to enhance waste stabilization, and the deployment of physical barriers like bottom liners, drainage systems, and emissions collection mechanisms.

These practices, as outlined by Cossu et al., 2022, are essential for minimizing environmental impact, ensuring the sustainable operation of landfills, and mitigating the risk of pollution from waste disposal activities.

2.2 European and Italian Regulations on Waste and Landfill Management

The management of waste and landfilling activities is strictly regulated through specific legislations across countries to mitigate pollution across various environmental compartments. Central to this regulatory landscape is the Directive 2008/98/EC of the European Parliament and of the Council, also known as the Waste Framework Directive, alongside the Directive 1999/31/EC concerning the landfill of waste. The Waste Framework Directive provides the legal groundwork for the European Union's waste management, emphasizing the need for waste to serve a useful purpose either through recovery operations or by fulfilling a specific function within the broader economy.

The Directive 1999/31/EC on the landfill of waste seeks to mitigate the adverse effects of landfill operations on various environmental facets, including surface and groundwater, soil, air, and human health. This directive categorizes landfills into three distinct groups based on the type of waste they accommodate: hazardous, non-hazardous, and inert waste. It introduces stringent measures such as the progressive reduction of biodegradable waste allocated to landfills by 2024, mandating that only pre-treated waste may be disposed of in these facilities. Moreover, certain materials, including used tires and waste deemed liquid, flammable, explosive, corrosive, or originating from medical practices, are expressly prohibited from landfill disposal.

Subsequent directives and decisions, including Decision 2003/33/EC and Directive (EU) 2018/850, have further refined these regulations. The 2018 amendment introduces a significant reduction in landfilling of recyclable or recoverable waste by 2030 and sets an ambitious target to limit municipal waste landfilling to no more than 10% by 2035. Additionally, it mandates landfill operators to harness landfill gas for energy production or safely flare it, contributing to the mitigation of methane emissions, a potent greenhouse gas.

Parallel to European directives, Italy's approach to waste management and landfill regulation is encapsulated in the D. Lgs. n. 152/2006 – Testo Unico Ambientale. This comprehensive legislative framework encompasses waste management principles, authorization procedures, and emission standards, including those for volatile organic compounds (VOCs), dust, and combustion plants. Specific regulations for landfills in Italy are further detailed in the D. Lgs n. 121/2020, aligning with the European Directive (EU) 2018/850. This Italian legislation mirrors the European directive's emphasis on waste treatment before landfilling, without setting specific limit values for air emissions from landfills. Such limits are determined on a case-by-case basis, particularly for landfills receiving more than 10 Mg of waste per day or with a total capacity exceeding 25,000 Mg, with the exception of inert waste landfills, as stipulated in Directive 2008/1/EC on integrated pollution prevention and control.

By setting progressive targets for waste reduction, treatment prior to landfilling, and emission control, these regulations embody a comprehensive strategy aimed at minimizing the environmental footprint of waste disposal operations. Further enhancements to these regulations, especially concerning landfill gas management, are anticipated to yield additional reductions in methane emissions, thereby contributing to global efforts to mitigate climate change impacts.

3 Fundamental Considerations for Design of Microbial Methane Oxidation Systems

This chapter delves into the fundamental considerations crucial for the effective design of MMOS. It encompasses a comprehensive examination of the aerobic methane oxidation process, elucidates the role of methanotrophic bacteria, explores the dynamics of water retention and gas transport within these systems, and outlines the laboratory methods employed to assess methane oxidation potential. Through an exploration of these core components, the chapter aims to provide a solid foundation for understanding the intricacies of MMOS and the vital factors that influence their efficiency and performance in landfill cover systems.

3.1 Aerobic Methane Oxidation in Landfill Covers

3.1.1 Fundamentals of the Methane Oxidation Process

Methane is produced within landfills through the anaerobic decomposition of the organic fraction of waste. The rate of methane production depends on several factors, including the volume of landfilled waste, the composition and degradability of the organic waste fractions, the age of the waste, and environmental conditions such as temperature, moisture content, nutrient availability, and the presence of inhibiting compounds (Scheutz et al., 2009a).

A portion of the methane produced within the landfill is recovered through gas extraction systems and subsequently utilized for energy generation or flared. The efficiency of these systems is typically reported to be around 50-60% (Duan et al., 2022) The remaining unrecovered methane can either be emitted into the atmosphere or intercepted by other mitigation methods.

Methane emissions from landfills can occur through various mechanisms, including:

- Diffusion: The major process, driven by the concentration gradient between the soil and ambient air, where the methane concentration is diluted.
- Advection via Darcy flow: Caused by pressure gradients due to changing barometric pressure or the pressure generated by landfill gas generation.
- Wind-induced advection: Caused by pressure gradients induced by wind (Scheutz et al., 2009).

Mechanism of Aerobic Methane Oxidation

Aerobic methane oxidation is a biological process mediated by methanotrophic bacteria, which utilize methane as their sole source of carbon and energy. These bacteria belong to the phyla Proteobacteria and Verrucomicrobia and are classified into two main groups: Type I and Type II, based on their metabolic pathways for formaldehyde assimilation.

The aerobic methane oxidation reaction can be represented by the following equation:

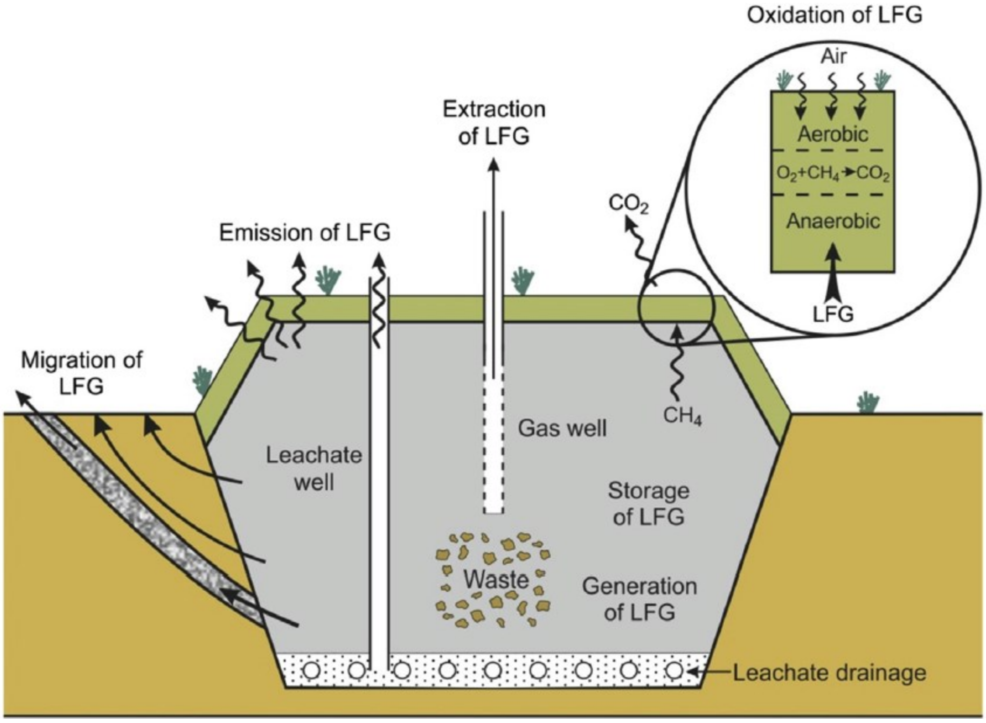


Figure 3 Conceptual model of the generation, transport, and oxidation of landfill gas (Kjeldsen & Scheutz, 2018)

$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$ (+ biomass + energy) (Kriipsalu et al., 2023)

This exergonic reaction, with a standard Gibbs free energy change ($\Delta G°$) of -780 kJ mol^{-1} CH_4, is thermodynamically favorable and provides energy for the growth and maintenance of methanotrophic bacteria.

In landfill environments, aerobic methane oxidation occurs within the landfill cover soil, where methane diffuses from the decomposing waste, and oxygen is available from the ambient air. The vertical distribution of methane and oxygen concentrations creates an idealized gas concentration profile, with the highest oxidation potential in the upper part of the landfill cover soil, where both gases are present in sufficient quantities.

As illustrated in Figure 4 the methanotrophic active zone, where aerobic methane oxidation occurs, typically extends from the soil surface to a depth of 30–40 cm, with maximal oxidation activity occurring between 15–20 cm depth. At depths greater than 60 cm, aerobic methane oxidation is limited by low oxygen concentrations (Scheutz et al., 2009a). The depth of the active zone is dynamic and varies depending on the flow rate of landfill gas, with higher flow rates pushing the zone closer to the surface.

Kinetics of Aerobic Methane Oxidation

The kinetics of aerobic methane oxidation by methanotrophic cultures and soils is well described by Michaelis-Menten kinetics, represented by the following equation:

$$r = V_{max}[CH_4] / (Km + [CH_4])$$

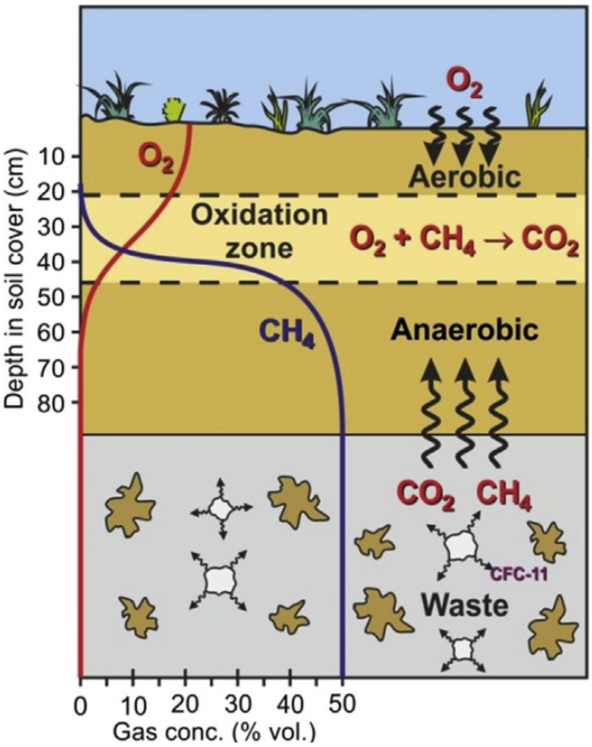


Figure 4. Idealized gas concentration profile in landfill cover (Kjeldsen & Scheutz, 2018)

Where r is the methane oxidation rate, V_{max} is the maximum methane oxidation rate, Km is the Michaelis-Menten constant (representing the methane concentration at which the oxidation rate is half of Vmax), and [CH4] is the methane concentration (Scheutz et al., 2009a).

Methane Mass Balance Equation

The methane mass balance in a landfill can be described by the following relationship (Scheutz et al., 2009a):

$$CH_4\ production\ [mass\ t^{-1}] = CH_4\ recovered\ [mass\ t^{-1}] + CH_4\ emitted\ [mass\ t^{-1}]$$

$$+\ Lateral\ CH_4\ migration\ [mass\ t^{-1}] + CH_4\ oxidized\ [mass\ t^{-1}]$$

$$+\ \Delta\ CH_4\ storage\ [mass\ t^{-1}]$$

This equation represents the conservation of mass for methane within the landfill system, where the total methane production is balanced by the sum of methane recovered, emitted, laterally migrated, oxidized, and the change in methane storage within the landfill.

3.1.2 Methanotrophic Bacteria and the Aerobic Methane Oxidation Pathway

Methanotrophic bacteria are a unique group of microorganisms that play a crucial role in the global methane cycle. These bacteria can utilize methane as their sole source of carbon and energy, making them essential for mitigating methane emissions in various environments, including landfills.

Classification and Diversity of Methanotrophic Bacteria

Methanotrophic bacteria are a subset of a physiological group known as methylotrophs, which are aerobic bacteria that utilize one-carbon compounds more reduced than formic acid as sources of carbon and energy. These bacteria assimilate formaldehyde as a major source of cellular carbon.

Aerobic methanotrophs belong to three main phylogenetic groups:

- Gammaproteobacteria (Type I): This group includes the families Methylococcaceae and Methylothermaceae.
- Alphaproteobacteria (Type II): This group comprises the families Methylocystaceae and Beijerinckiaceae.
- Verrucomicrobia: The family Methylacidiphilaceae belongs to this phylum (Guerrero-Cruz et al., 2021).

Physiological Characteristics

Type I and Type II methanotrophs exhibit distinct physiological characteristics and environmental preferences:

- Type I methanotrophs utilize the ribulose monophosphate (RuMP) pathway for formaldehyde assimilation, while Type II methanotrophs employ the serine pathway.
- Most Type I bacteria are unable to fix atmospheric nitrogen and are typically associated with low methane concentrations and high oxygen and nutrient levels.
- In contrast, Type II methanotrophs are capable of nitrogen fixation and can thrive under conditions of high methane concentrations and low oxygen and nutrient levels (Pedersen, 2010).

In landfill cover soils, where high methane concentrations are prevalent in the soil gas phase, Type II methanotrophs are typically the dominant group (Röwer, 2014).

Aerobic Methane Oxidation Pathway

The aerobic methane oxidation pathway, illustrated in Figure is a multi-step process initiated by methane monooxygenases (MMOs), which require both oxygen and reducing equivalents for their activity (Conrad, 1996). MMOs are classical monooxygenases that utilize two reducing equivalents to cleave the O-O bonds of dioxygen. One oxygen atom is reduced to water (H2O), while the other is incorporated into methane to form methanol (CH3OH).

Methanol is subsequently oxidized to formaldehyde (HCHO) by a periplasmic methanol dehydrogenase (MDH) in methanotrophs. The formaldehyde produced is then assimilated by methanotrophic bacteria to form intermediates of the central metabolic pathways, which are subsequently utilized for the biosynthesis of cellular material.

The pathways for formaldehyde assimilation differ between Type I and Type II methanotrophs:

- Type II methanotrophs employ the serine pathway, in which formaldehyde and carbon dioxide are utilized to form a three-carbon intermediate.

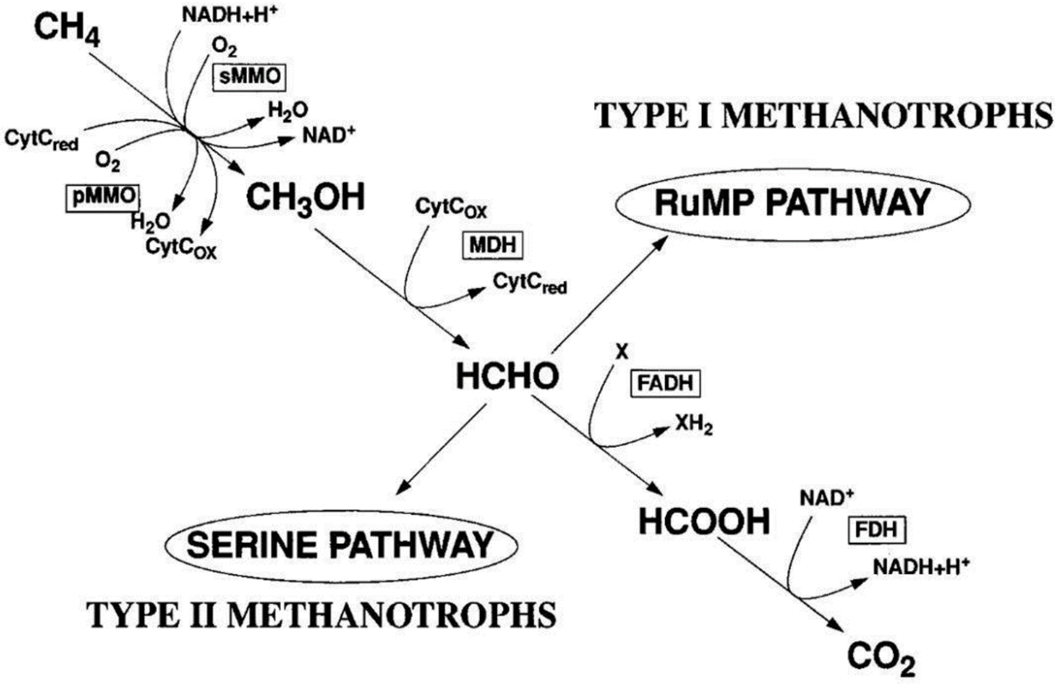


Figure 5. Pathways for the aerobic oxidation of methane and assimilation of formaldehyde. Abbreviations: CytC, cytochromec; FADH, formaldehyde dehydrogenase; FDH, formate dehydrogenase (Hanson and Hanson, 1996).

- Type I methanotrophs utilize the ribulose monophosphate (RuMP) cycle, where formaldehyde is incorporated into a three-carbon intermediate. In this pathway, all cellular carbon is assimilated at the oxidation level of formaldehyde (Hnson and Hanson, 1996).

The aerobic methane oxidation pathway culminates with the oxidation of formaldehyde to carbon dioxide, catalyzed by a NAD-dependent formate dehydrogenase (FDH) in methanotrophs (Hanson and Hanson, 1996).

3.1.3 Influencing Factors

This chapter explores the critical influencing factors of methane oxidation, highlighting their interdependencies and the implications for the design of MMOS.

Temperature: Temperature profoundly influences the activity of methanotrophic bacteria, with most aerobic methanotrophs being mesophiles, showing optimal methane oxidation at around 25°C in peat soils. However, methane oxidation occurs even at temperatures as low as 0 to 10°C, indicating a wide temperature adaptability among methanotrophs (R. S. Hanson and Hanson, 1996). In colder conditions or during winter with temperatures below 5–10°C, methane oxidation may reduce significantly or halt, emphasizing the importance of temperature as a controlling factor (Scheutz et al., 2009b).

pH: Soil pH is a critical determinant for the growth and activity of methanotrophic bacteria. Aerobic methanotrophs exhibit optimal methane oxidation in soils with pH values ranging from

5.5 to 8.5. The adaptability of methanotrophs to varying pH levels suggests a relatively broad operational range for MMOS in different landfill cover materials (Scheutz et al., 2009b).

Inorganic Nitrogen Content: The availability of inorganic nitrogen, in forms such as ammonium (NH4+) and nitrate (NO3-), can either stimulate or inhibit methane oxidation, depending on the concentration and form of nitrogen, methane concentrations, pH, and the type of methanotrophs present. High concentrations of NH4+ can inhibit aerobic methane oxidation by acting as a competitive inhibitor towards methane monooxygenase (MMO) enzymes (Pedersen, 2010). Conversely, the N-AOM process benefits from increased nitrogen input, enhancing the activity of nitrate-dependent methanotrophs (Yang et al., 2023).

Availability of Electron Acceptors and Oxygen Exposure: The presence of suitable electron acceptors is essential for methane oxidation. Aerobic methanotrophs can function efficiently at low oxygen concentrations, with methane oxidation starting at oxygen levels above 1.7–2.6% (Scheutz et al., 2009). The adaptation of anaerobic methanotrophs to environments with intermittent oxygen exposure further underscores the role of electron acceptors in methane oxidation.

Inhibitory Substances: Certain substances, such as difluoromethane, dichloromethane, methyl fluoride, and others, can inhibit methane oxidation. The extent of inhibition is influenced by the concentration of these substances, methane levels in landfill gas, and the composition of the methanotrophic community (Scheutz et al., 2009).

The efficiency of methane oxidation in landfills is governed by a complex interplay of environmental, physical, and biological factors. Understanding these influences is vital for optimizing landfill designs to enhance methane oxidation, thereby reducing greenhouse gas emissions. Tailoring landfill management practices to account for temperature, pH, inorganic nitrogen content, availability of electron acceptors, and the presence of inhibitory substances can significantly improve the performance of MMOS in mitigating methane emissions.

3.2 Water Retention and Transport

The efficiency of microbial methane oxidation systems (MMOS) in reducing CH_4 emissions is intricately linked to the movement of gas and water within their layered structure. The pore size distribution, influenced by soil texture and compaction, plays a pivotal role in determining the soil's water retention capacity and its ability to facilitate both diffusive and advective gas transport (van Verseveld & Gebert, 2020a). Additionally, the soil's thermal properties affect these transport mechanisms. Central to MMOS design are several critical aspects: the water retention curve (WRC), which outlines the relationship between soil moisture and suction; the hydraulic conductivity function (k-fct), detailing water permeability; and the air/gas permeability function (kG-fct), which describes how gas permeability varies with volumetric air porosity. This section briefly reviews the corresponding fundamental processes and mechanisms that form the basis for proper design.

3.2.1 Water Retention Curve (WRC):

The Water Retention Curve (Figure) represents the constitutive relationship between moisture in the soil and the negative water pressure (soil suction) caused by capillarity (Barbour, 2011). Soil suction, quantified by variables such as gravimetric water content (w), volumetric water content (θw), or the degree of saturation (Sr), measures the tendency of soil to retain water against external forces, such as gravity. This curve is crucial for understanding how water is held in the soil matrix, the conditions under which water is retained or released, and how these processes impact the physical and hydraulic properties of soil.

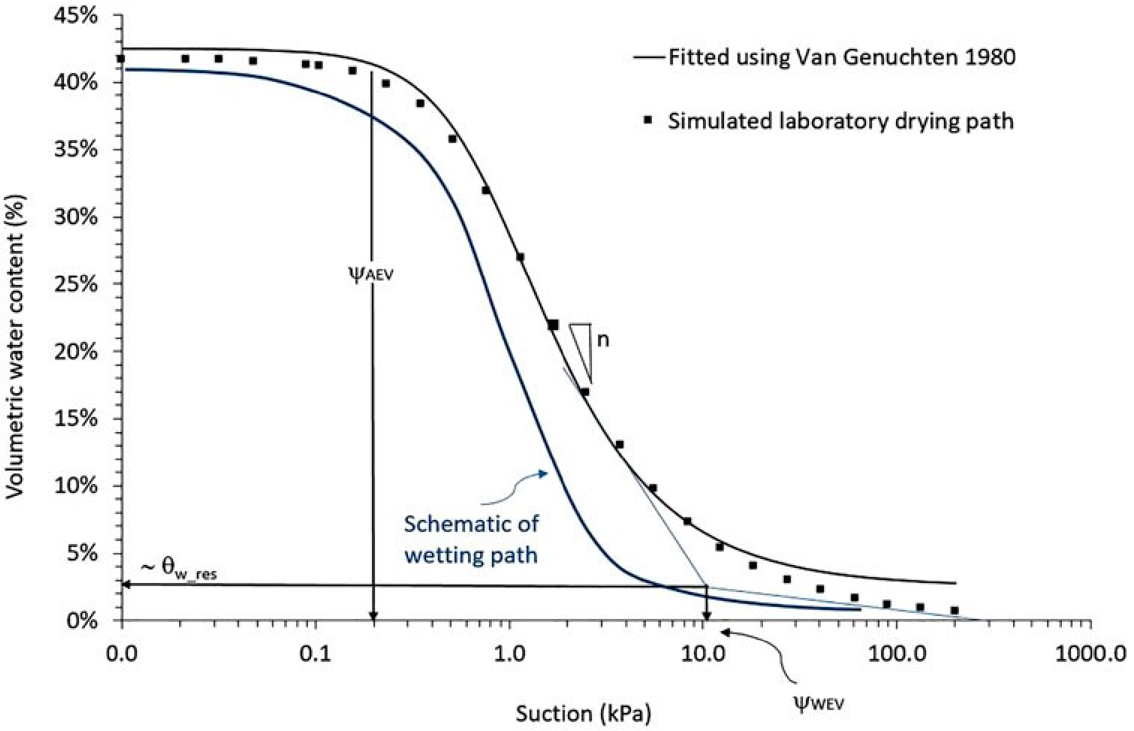


Figure 6. Water retention curves of simulated laboratory experiments on two materials forming a capillary barrier: sand (the moisture retaining layer; or MRL) and gravel (the capillary break layer; or CBL) (Gebert et al., 2022).

The WRC is characterized by several key features:

- Suction at Air Entry Value (ψ_{AEV}) marks the threshold at which air starts to penetrate the largest pores in the soil, displacing water and thereby decreasing the soil's moisture content as suction increases.
- Desaturation Curve Slope (n) represents the rate at which soil moisture decreases with increasing suction beyond the ψAEV.
- Residual Water Content (θ_{w_res}) and the Water Entry Value (ψ_{WEV}) indicate the minimal water content that soil can retain and the suction level at which this state is achieved, respectively.

Soil exhibits hysteresis; thus, its moisture retention capability varies depending on whether the soil is undergoing wetting or drying. This behavior underscores the importance of adopting a conservative approach in environmental design, using the drying curve to model the worst-case scenario for water content at a given level of suction.

3.2.2 Hydraulic Permeability Function (k-fct)

Hydraulic permeability, fundamentally, encapsulates the capacity of a soil matrix to facilitate water transit under a differential pressure regime. The hydraulic permeability function (k-fct, Figure 12) describes how this ability to allow water flow (permeability) changes with soil suction. Soil suction is the measure of how much negative pressure is required to draw water into the soil or pull it through the soil pores.

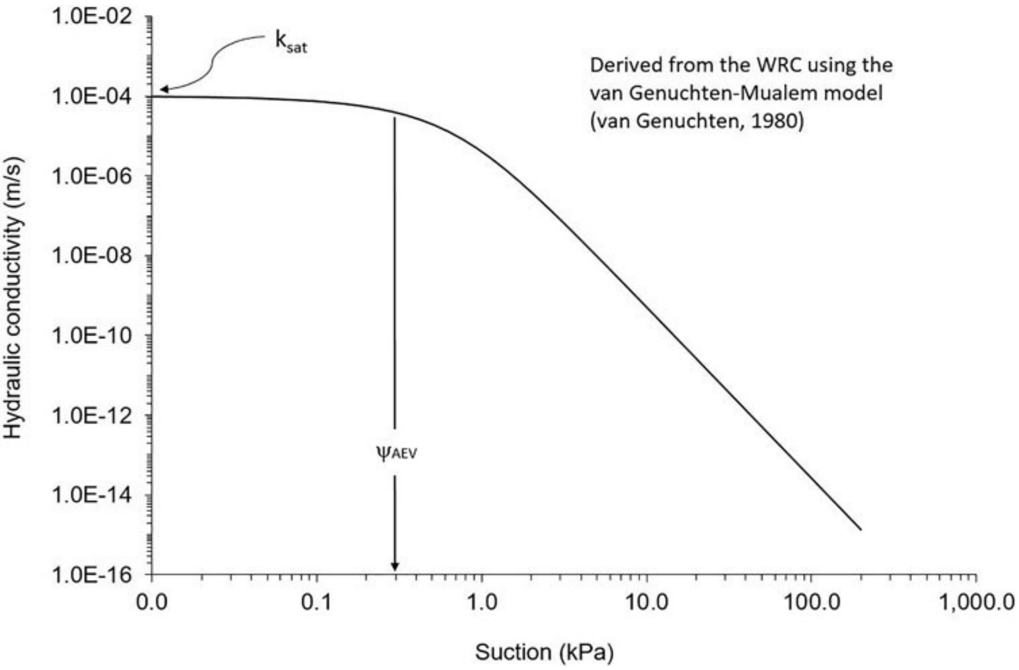


Figure 7 Hydraulic permeability function (k-fct) of the same soil. (Gebert et al., 2022)

Fundamental Attributes of k-fct: At zero suction, when the soil is fully water-saturated and devoid of air, the saturated permeability (ksat): represents the peak hydraulic permeability. This state underscores the maximum water flow potential through the soil matrix. Desaturation Curve Slope delineates the rate at which soil permeability declines as it transitions from a saturated to a less saturated state. The slope is indicative of how soil properties, such as pore size distribution and connectivity, influence water flow resistance as moisture decreases. Air Entry Value (ψ_{AEV}) signifies the suction threshold beyond which air penetrates the soil pores, displacing water and thus markedly reducing hydraulic permeability. This value, consistent with that obtained from the Water Retention Curve (WRC), forms a critical link between soil moisture retention and permeability characteristics.

Relevance to MMOS Design: The k-fct's role extends into the practical arena of MMOS design, where it informs the simulation and optimization of unsaturated water flow. Such flow dynamics directly impact gas transport within the soil, affecting the efficiency of methane oxidation processes in landfill covers.

By leveraging the k-fct, we can simulate water movement through the landfill cover's soil layers, gaining insights into moisture dynamics that influence gas diffusion and biological oxidation conditions. Through detailed modeling, it is possible to pinpoint the interface between the Methane Oxidation Layer (MOL) and the filter layer where air occlusion occurs. Optimizing this aspect is crucial for maintaining conducive conditions for methane oxidation. In the design of Methane Mitigation Systems (MMOS), understanding the WRC and k-function is paramount for optimizing gas transport. This involves:

Optimizing the Base Area for Methane Oxidation: By leveraging the concept of air-filled porosity at the point of occlusion of pores with water (θ_{a_occ}), we can determine the effective area required for receiving the gas load and calculate the length of unrestricted gas migration (LUGM) along a slope. This optimization ensures the spatial homogenization of methane load across the Methane Oxidation Layer (MOL) and the Gas Distribution Layer (GDL), enhancing the efficiency of methane oxidation.

Optimizing for Atmospheric Oxygen Ingress: The effective diffusivity of a soil is a measure of its ability to transport gases, such as atmospheric oxygen, which is essential for the oxidation of methane. By understanding the WRC and applying the k-fct, we can optimize the soil structure to maximize the ingress of atmospheric oxygen into the methane oxidation layer. This involves adjusting the soil's water content to levels that favor gas diffusion, typically by maintaining a water content at or below field capacity, which corresponds to the soil's maximum ability to hold water against gravity while still allowing for adequate gas transport.

The application of WRC and k-fct in MMOS design involves a detailed analysis of soil properties, including moisture content, suction pressure, and hydraulic permeability, to create conditions conducive to efficient gas transport and methane oxidation. This includes:

- **Experimental Determination and Modeling:** Ideally, the WRC and k-fct of the materials used in the MOL and filter layers are determined experimentally. When direct determination is not feasible, pedotransfer functions and models like the Mualem-van Genuchten model are applied to estimate these properties.
- **Numerical Modeling:** Key input data from the WRC and k-fct are used in numerical models to simulate unsaturated flow of water and gas transport in soils. These models help in the iterative process of testing different material combinations and configurations to identify the most effective designs for methane oxidation and gas transport.
- **Optimization of Porosity for Gas Transport:** By understanding the distribution of pore sizes (as given by the WRC) and the soil's hydraulic behavior (as described by the k-fct), engineers can tailor the soil structure to optimize air capacity and field capacity. This ensures that a sufficient volume of pores is available for gas transport, particularly under conditions drier than field capacity.

3.2.3 The Impact of Material Layering

The contrasting of materials with varying textures introduces a significant variation in unsaturated hydraulic conductivity, leading to moisture accumulation at the interface of these materials, specifically where finer materials overlay coarser ones. This phenomenon, recognized as the capillary barrier effect, can hinder the transport of gases across the interface. The degree of moisture accumulation is influenced by the characteristics of the materials involved, climatic conditions, and the physical attributes of the cover system, including the slope's angle and length. Capillary barriers are strategically designed to mitigate infiltration into waste bodies by channeling water laterally within the finer capillary layer, positioned above the coarser capillary block layer (Williams et al., 2011).

In the context of Methane Mitigation Overlay Systems (MMOS), the inherent contrast in the hydraulic properties of layered materials naturally facilitates the formation of capillary barriers. The migration of water downslope can lead to moisture levels increasing to a point where soil pores above the MOL-GDL (Methane Oxidation Layer-Gas Diffusion Layer) or MOL-filter interface become waterlogged, creating a barrier to gas flow. This condition underscores a pivotal challenge in MMOS design, wherein the management of water and gas transport processes presents a conflict.

Water-filled pores reduce the permeability of the soil to gas, resulting in a disparity in gas flow directionality—diminished downslope and enhanced upslope. This differential leads to gas ascending within the GDL, bypassing areas where pores remain unobstructed. Zones of lower moisture, predominantly near the summit of the sloped MMOS, become conduits for gas migration, a phenomenon observed by (Berger et al., 2005) in their study of a laboratory-scale capillary barrier system. Similarly, field studies Geck et al., 2016 have shown that preferential upward gas migration results in higher methane concentrations upslope and increased surface emissions. Through experimental and numerical analyses, these studies have illustrated the dynamic between diffusive and advective gas flows within landfill covers, highlighting the variability in gas diffusivity and conductivity across different slope positions.

3.3 Gas Transport Dynamics in MMOS

3.3.1 Advective and Diffusive Gas Transport

The flow of methane (CH_4) and oxygen (O_2) through the Methane Oxidation Layer (MOL) is dictated by the soil or material's gas transport capabilities, playing a pivotal role in the performance of methane oxidation systems. The design of these systems must, therefore, prioritize understanding and optimizing gas transport properties and their influencing factors. Methane oxidation systems feature both diffusive and advective gas transport mechanisms: diffusive transport, propelled by concentration gradients, moves CH_4 from its source to the atmosphere and draws atmospheric oxygen into the system. Advective transport, driven by pressure differences, moves landfill gas across the system, with wind or barometric pressure changes also contributing to this flow. Moreover, the methane oxidation reaction itself creates a vacuum by using up more gas (3 moles of CH_4 and O_2) than it produces (1 mole of CO_2), enhancing the advective movement of both landfill gas and atmospheric oxygen.

Gas transport rates depend on the interconnected volume and geometry of the soil's water-free pore space, known as air-filled porosity or volumetric air content. This porosity, calculated as total porosity minus the volumetric water content, indicates the proportion of porosity available for gas movement. The soil's particle size distribution and degree of compaction determine total porosity and its geometric attributes, which in turn define soil permeability, a constant property irrespective of moisture or the medium. Diffusive gas transport takes into account soil moisture and gas viscosity, leading to effective gas diffusivity, while advective transport considers moisture and kinematic gas viscosity, resulting in a measure of gas conductivity. Gas conductivity reflects the medium's resistance to gas movement under a pressure gradient, a common scenario in methane oxidation systems where landfill gas moves through the Gas Distribution Layer (GDL) due to pressure differences.

Design and sizing of methane oxidation systems should also account for factors that could reduce air-filled porosity, affecting diffusivity and gas conductivity. These factors include moisture changes due to weather conditions, soil settlement from self-weight consolidation or construction compaction, and long-term soil structure changes due to physical and biological processes. Monitoring these processes during operation is crucial for maintaining system performance.

The gas conductivity or permeability function links air-filled porosity to gas conductivity, a relationship determined through laboratory procedures. This function demonstrates how soil moisture or compaction affects gas conductivity, essential for system design. Identifying the required effective porosity and appropriate compaction levels is key to achieving optimal gas transport.

3.3.2 Soil Moisture's Impact on Gas Permeability

Figure illustrates two distinct responses to changes in soil moisture that can affect gas permeability, relevant during periods of increased rainfall or reduced evaporation. For fine sand, a significant decrease in gas conductivity (k_{Gas}) occurs when water fills the interconnected pores crucial for advective gas flow, disrupting the continuity of air-filled spaces. This condition, where air content drops sharply, is identified as θ_{a_occ}. Conversely, in a compost-sand mixture, the graph shows two reductions in k_{Gas} upon watering, indicating a bimodal pore size distribution where larger pores, less affected by capillary action, quickly release water, allowing gas conductivity to partially recover.

Ahoughalandari et al., 2018 recommend a conservative approach for materials displaying the latter behavior by marking the first conductivity drop as the conservative occlusion point, $\theta_{a\_Cons\_occ}$. Their findings suggest that soil compaction levels (100%, 94%, and 89%) minimally influence $\theta a_{\_occ}$ or $\theta a_{\_Cons\_occ}$ but slightly affect the k_{Gas} function.

In designing Methane Oxidation Systems (MMOS), identifying the air content at which occlusion occurs, θ_{a_occ} (or $\theta_{a\_Cons\_occ}$), is crucial. Ahoughalandari et al., 2018 detail methods to determine θ_{a_occ} using the gas permeability function of MOL materials, including a quicker approach via the Standard Proctor curve in the absence of a ka-function. For sandy soils, θ_{a_occ} roughly matches the soil's capacity for air at a specific matric potential (−6 kPa).

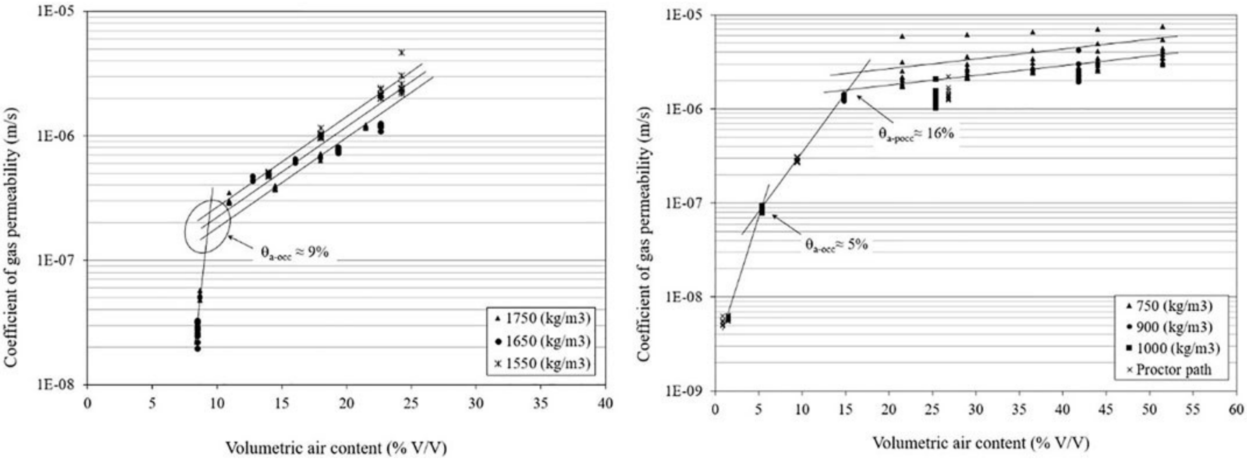


Figure 8. Gas permeability function with volumetric air content (or air-filled porosity ε_a) adjusted by soil moisture: single drop (fine sand, left) and dual-drop (compost-sand mixture, right). Fluid = air. (Gebert et al., 2022)

The gas permeability function, influenced by soil moisture, depends on the soil's texture and compaction level, which dictate pore size distribution. Adding water to soil typically saturates finer pores before larger ones due to capillary suction order. Larger pores, essential for gas flow, ensure less susceptibility to moisture variations in coarser substrates compared to finer sands.

3.3.3 Soil Compaction and Gas Conductivity

Soil compaction primarily affects the large-diameter pores, crucial for advective gas flow. With increased compaction, smaller pore diameters are also impacted, diminishing the soil's gas conductivity (k_{Gas}). This relationship underscores the critical role of pore size, following Hagen-Poiseuille's law, where the radius significantly influences advective flow.

Studies, including those by Richard et al., 2001 and van Verseveld & Gebert, 2020b, have documented a log-linear decrease in k_{Gas} with increased compaction effort. Notably, this trend has been observed across seven different soils used in Methane Oxidation Layers (MOL), as depicted in Figure , indicating variability in conductivity changes due to the differing proportions of large-diameter pores. The degree of MOL compaction is pivotal for the spatial distribution of landfill gas within the Gas Distribution Layer (GDL) beneath the MOL. This necessitates incorporating soil compaction metrics into MMOS design to ensure efficient gas distribution.

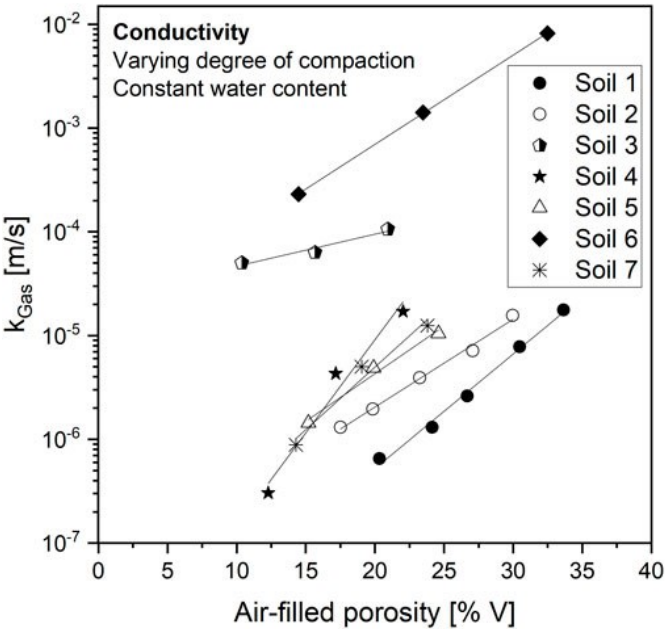


Figure 9. Gas permeability function (fluid = air) with air-filled porosity (εa) adjusted by degree of soil compaction in different sandy candidate MOL soils. (Gebert et al., 2022)

3.3.4 Diffusive Gas Transport

Contrary to gas conductivity, diffusive gas transport is influenced by the total share of water-free pores, rather than pore size distribution. This aspect highlights the insignificance of Knudsen diffusion in coarsely textured soils typical in MMOS, where gas molecule-pore wall collisions are minimal. Studies have shown that gas diffusivity decreases almost linearly with reduced air-filled porosity (θa), particularly affected by soil moisture. This trend is evident in the analysis of five sandy soils (Figure , left), where diffusivity diminishes at a near-linear rate until a critical porosity level is reached.

Seasonal changes in moisture content significantly impact diffusive gas transport, affecting the system's oxygen ingress and methane oxidation capacity. The comparative analysis (Figure 0, right) illustrates that increased moisture and compaction similarly reduce air-filled porosity and, consequently, gas diffusivity. Some researchers have identified a non-linear decrease in diffusivity with lower air-filled porosity levels. This phenomenon, attributed to increased tortuosity and reduced pore connectivity due to moisture or compaction, suggests a heightened effect in finer-textured soils (van Verseveld & Gebert, 2020b). The findings indicate a more pronounced impact of compaction on advective gas transport than on diffusive transport, as evidenced by the differential slopes of specific diffusivity (D_{eff}/θ_a) and specific conductivity (k_{Gas}/θ_a) in Figure (right). This underscores the importance of considering pore size distribution in MMOS design.

Design Implications for Methane Oxidation Systems: Design considerations for methane oxidation layers must account for these dynamics, utilizing relationships between soil particle size, compaction, and moisture retention to predict effective diffusion coefficients. This approach, detailed in supplementary materials, is essential for customizing MMOS properties to meet specific methane oxidation targets, with adjustments necessary for compost-based materials.

Figure 10. Effective gas diffusivity (D_{eff}) in relation to air-filled porosity (ε_a), adjusted by soil moisture (left) and by degree of compaction (right) in different sandy candidate MOL soils. Fluid = artificial landfill gas mixture (Gebert et al., 2022).

3.4 Laboratory Tests for Methane Oxidation Potential Assessment in MMOS

Assessing the methane oxidation potential within landfill cover systems is crucial for understanding the effectiveness of MMOS and their impact on greenhouse gas emissions. Various laboratory techniques have been developed to investigate methane oxidation, including batch incubation experiments, continuous gas flow systems (packed soil column tests), and stable carbon isotope ratio analysis. Each method offers unique advantages and challenges in simulating methane oxidation processes under controlled conditions. This chapter explores these laboratory techniques, highlighting their methodologies, applications, and considerations for interpreting results.

3.4.1 Batch Incubation Experiments

The batch incubation technique is a widely used method for assessing the methane oxidation potential of the methane oxidation layer in landfill cover systems. This technique involves setting up closed batch reactors, where initial gas concentrations can be adjusted, and the temperature is maintained constant throughout the experiment. The actual gas concentrations are measured regularly over the course of the incubation period (Sindern et al., 2013).

Batch incubation experiments are technically simple, have lower costs, and are less laborious to conduct compared to other methods, such as column experiments. Due to their small dimensions, batch tests are particularly useful when a large number of samples need to be analyzed or when the goal is to investigate the impact of different environmental parameters on methane oxidation. The batch conditions can be easily manipulated, allowing for controlled variations in factors like temperature, moisture content, and initial gas concentrations (Scheutz et al., 2009a)

However, it is important to note that batch incubation tests do not accurately simulate the dynamic gas transport processes that occur in landfill cover systems. Additionally, these tests may not adequately represent the consequences of long-term gas exposure, which can influence the methane oxidation potential over time (Scheutz et al., 2009).

Sample Collection: The first step in conducting a batch incubation experiment is to collect representative samples from the methane oxidation layer of the landfill cover system. This layer is typically located at the interface between the cover material (e.g., soil) and the landfill waste. Care must be taken to ensure that the samples are representative of the entire methane oxidation layer by collecting multiple samples from various locations within the landfill cover.

Experimental Setup: Once the samples are collected, they are transferred to batch reactors, which are typically airtight containers or bottles. The batch reactors are sealed to create a closed system, and the initial gas concentrations within the headspace (the air space above the soil sample) are adjusted to the desired levels (Sindern et al., 2013). Common gases used in these experiments include methane, oxygen, and sometimes carbon dioxide, as these gases play crucial roles in the methane oxidation process.

The batch reactors are then incubated at a controlled temperature, typically within the range suitable for methanotrophic bacteria activity. Throughout the incubation period, gas samples are regularly extracted from the headspace of the batch reactors and analyzed to monitor the changes in gas concentrations over time (Sindern et al., 2013).

Fourier-Transform Infrared (FTIR) Spectroscopy Analytical Technique: Various analytical techniques can be employed to measure the gas concentrations in the extracted samples, however, the Fourier-transform infrared (FTIR) spectroscopy is the most commonly used method for quantifying methane, oxygen, and carbon dioxide concentrations. (Scheutz et al., 2009a).

A sample of the headspace gas is typically extracted through a septum or sampling port and introduced into the FTIR spectrometer's gas cell. FTIR spectroscopy is accomplished through the utilization of an interferometer, which enables the scanning of all the frequencies inherent in the IR radiation that emanates from the source. This scanning process is made possible by means of a movable mirror that, through its motion, introduces an optical path difference. This difference then results in either constructive or destructive interference with the beam reflection from a fixed mirror. Consequently, the representation of intensity in the time domain is obtained. Subsequently, the intensity representation in the frequency domain, or the infrared spectrum, is acquired through the application of the Fourier transform. The application of FTIR is employed in the investigation of soil chemical processes. Specifically, within the mid-infrared (mid-IR) range, vibrations arise from numerous environmentally significant molecules, such as organic acids, soil organic matter, and mineral phases (Peak, 2005).

Soil samples can be analyzed via FTIR spectroscopy through various methods. The most commonly utilized methods are transmission, diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS), and attenuated total reflectance (ATR). In particular, ATR-FTIR spectra offer insights into the functional groups that are in close proximity (approximately 1 μm) to the surface of an internal reflection element (IRE). One of the key advantages of ATR-FTIR is its ability to accurately collect spectra of samples in the presence of water. Overall, this technology provides a considerable degree of experimental and analytic flexibility.

3.4.2 Continuous Gas Flow Systems – Packed Soil Column Tests

Column tests are more suitable for investigating methane oxidation potential in the presence of heterogeneous materials, such as compost, and for higher mass input scenarios. These tests are designed to simulate the dynamic gas transport processes that occur in landfill soil covers and the effects of long-term gas exposure, which are not adequately represented in batch incubation experiments (Scheutz et al., 2009).

Experimental Setup: In a typical column test setup, a soil column is packed with the cover material or compost sample under investigation. Aerobic conditions are maintained by supplying air at the top of the column, while controlled methane influx is introduced from the bottom (Huber-Humer, 2004). The experimental conditions, such as temperature, are kept constant throughout the duration of the experiment. A typical column setup is reported in Figure 1.

Column tests offer the advantage of providing results that are more representative of real landfill cover systems compared to batch tests. However, it can be more challenging to unravel the diverse processes and conditions within these complex column microcosm tests (Huber-Humer, 2004).

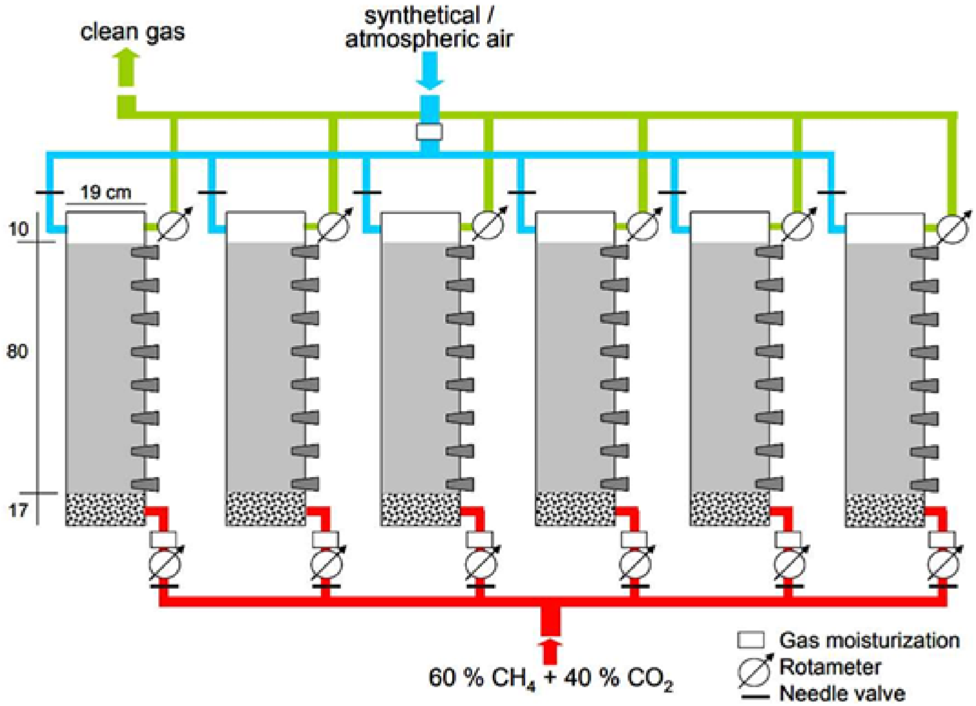


Figure 11. Schematic column setup (Gebert et al., 2008)

Typical Operating Conditions: Column experiments are typically operated with methane inlet concentrations of 50% or 100% v/v (volume per volume) and methane loads ranging from 200 to 300 g CH_4 $m^{-2}day^{-1}$ (Scheutz et al., 2009). This range of methane loads is considered to be in the middle to high range of reported landfill methane fluxes. It is important to note that active landfills can have gas fluxes up to 1300 g CH_4 $m^{-2}day^{-1}$, while older landfills or sites with gas collection systems typically exhibit methane fluxes of approximately 85 g CH_4 $m^{-2}day^{-1}$ (Scheutz et al., 2009).

Analytical Techniques: Similar to batch incubation experiments, various analytical techniques can be employed to measure the gas concentrations in the column effluent streams. Gas chromatography and Fourier-transform infrared (FTIR) spectroscopy are commonly used methods for quantifying methane, oxygen, and carbon dioxide concentrations. The FTIR spectrometer can provide real-time or near-real-time monitoring of methane concentrations in the effluent gas stream, allowing for continuous monitoring of methane production in the soil column.

3.4.3 Measurement of Methane Oxidation Using Stable Carbon Isotope Ratios

The stable carbon isotope ratio technique is based on the principle of isotopic fractionation, where lighter isotopes (12C) react more readily than heavier isotopes (13C) during chemical and biological processes. In the case of methane oxidation, methanotrophic bacteria preferentially consume the lighter 12C-methane, leading to an enrichment of the remaining methane in the heavier 13C isotope. This isotopic fractionation can be quantified by measuring the δ13C value, which represents the relative difference in the 13C/12C ratio between the sample and a reference standard, expressed in parts per thousand (‰).

To measure methane oxidation in landfills using the stable carbon isotope ratio technique, soil gas samples are collected from different depths within the landfill cover soil using probes or sampling ports (Börjesson et al., 2001). Additionally, atmospheric methane samples are collected from the landfill surface or nearby locations to establish the initial δ13C value of the methane source.

The collected soil gas and atmospheric samples are processed to extract and purify the methane component. Common techniques employed for methane extraction and purification include cryogenic trapping and gas chromatography. The purified methane samples are then introduced into an isotope ratio mass spectrometer (IRMS) for isotopic analysis. The IRMS measures the relative abundance of the 13C and 12C isotopes in the purified methane samples. The resulting δ13C values represent the deviation of the 13C/12C ratio in the sample from a reference standard, typically expressed in parts per thousand (‰).

As methane diffuses upward through the landfill cover soil, it undergoes microbial oxidation, leading to an enrichment of the remaining methane in the heavier 13C isotope. Consequently, the δ13C values of methane increase with increasing depth within the cover soil, reflecting the extent of methane oxidation. The extent of methane oxidation can be quantified by comparing the δ13C values at different depths within the cover soil and applying isotope fractionation models. These models account for factors such as the initial δ13C value of the landfill methane source, the isotopic fractionation associated with methane oxidation, and the transport mechanisms involved (Chanton et al., 2000).

When interpreting the stable carbon isotope ratio data for quantifying methane oxidation in landfills, several factors need to be considered:

- The isotopic composition of the initial landfill methane (δ13C value at the source) must be known or estimated accurately.
- Other processes, such as methane transport mechanisms, oxidation kinetics, and cover soil properties, may influence the isotope fractionation patterns and need to be accounted for in the interpretation (Chanton et al., 2000).
- The stable carbon isotope ratio technique is often combined with other methods, such as flux chamber measurements and microbial community analysis, to gain a comprehensive understanding of the methane oxidation processes in landfill environments (Cébron et al., 2007).

4 Design and Efficiency of Microbial Methane Oxidation Systems (MMOS) in Landfills

In the realm of landfills, gas collection systems are advised for the control and recuperation of landfill gas emissions whenever it is economically viable. However, the efficiency of these collection systems is not 100% efficient, leading to potential escape of emissions particularly preferentially from the wells and along the pathways of landfill equipment installation. Additionally, the feasibility of energy recovery diminishes when methane concentrations in landfill gas drop below 35–40% v/v and total gas production rates range from 30–50 m^3 h^{-1}; under these circumstances, the utilization of high-temperature flares becomes the technically and economically preferred approach (Huber-Humer et al., 2008). As methane concentrations decline further to 20–25% v/v and landfill gas flow rates decrease to 10–15 m^3 h^{-1}, the most appropriate treatment method shifts towards high-temperature flares (Huber-Humer et al., 2008). Below these thresholds, the management of substandard landfill gas becomes more costly and intricate, and the Microbial Methane Oxidation Systems (MMOS) emerge as the most promising technologies for mitigating methane emissions from landfills (Huber-Humer et al., 2008). Research on microbial methane oxidation in landfills dates back to the 1980s, with the formulation of a proper MMOS design being endorsed in 2002 by the Consortium for Landfill Emissions Abatement Research (CLEAR). Subsequent to this, there has been a proliferation of studies on this subject, leading to an increased adoption of these systems in various landfill sites.

This chapter delves into the various configurations of MMOS, including biofilters, biowindows, biocovers, and the innovative breathing biocovers. Through detailed examination of their structure, functionality, and the materials involved, this chapter aims to provide a comprehensive overview of the operational mechanisms these systems employ to mitigate methane emissions effectively. The subsequent sections offer a critical comparison of methane oxidation efficiencies across different MMOS installations, presenting a clear picture of their performance in existing landfill settings. This analysis is not only instrumental in identifying the strengths and limitations of each system but also helps to understand how structural characteristics and operational mechanisms MMO systems impact their methane oxidation efficiency.

4.1 Typical Configurations of Microbial Methane Oxidation Systems

Microbial Methane Oxidation Systems (MMOS) are designed to mitigate methane emissions through a multi-layered approach (Figure), typically comprising three to four distinct layers. Each layer serves a unique purpose, contributing to the efficient oxidation of methane within these systems. The composition and functionalities of these layers are detailed below.

4.1.1 Foundation Layer (MMOS Bed)

For below-grade systems such as biowindows and some biofilters, constructed within an existing landfill's top cover, the foundation often consists of the current cover soil or a soil layer directly applied on the waste. In contrast, for above-grade systems like biofilters, this layer forms the structure's base, designed to be gas-impermeable. Where landfills lack a pre-existing cover, a compacted layer of inert waste may serve as the foundation, structured to support the MMOS (Gebert et al., 2022).

4.1.2 Gas Distribution Layer (GDL)

The Gas Distribution Layer (GDL) serves dual critical functions within methane oxidation systems:

- Uniform Distribution of Methane (CH_4) Loading: It ensures that methane or landfill gas is dispersed evenly across the base of the Methane Oxidation Layer (MOL), preventing system overload and the formation of hotspots (Kjeldsen & Scheutz, 2018).
- Drainage of Seepage Water: It facilitates the removal of water that has percolated through the layers, maintaining optimal conditions for gas flow.

Key Properties Required for GDL Materials:

- Materials should be stable over the long term and inert to reactions that could alter their pore structure.
- To prevent the precipitation of carbonates that reduce pore volume, materials with low to no organic and inorganic carbon content are preferred.
- In areas of fluctuating redox conditions, materials should not contain iron that can dissolve, mobilize, and precipitate as oxides/hydroxides, potentially clogging the pore spaces.
- The materials should promote horizontal gas flow, enhancing uniform CH_4 distribution and minimizing the impact of capillary barriers at material interfaces (Gebert et al., 2022).

Suitable Materials for the GDL:

- Carbonate-Poor Gravels, Crushed Glass, and Coarse Quartz Sands: These are ideal due to their stability, inertness, and appropriate particle size.
- Recycled Coarse Materials: If free from carbonates, materials from construction, demolition, and renovation industries, or even tire shreds, are viable options, provided there's no risk of pollution or undesirable gas emissions.
- Geosynthetics: Geo-nets and nonwoven geotextiles may be used, with caution regarding their compression under the MOL load which could reduce their conductivity.

Design and Construction Considerations: For maximum conductivity, materials should have particle diameters akin to coarse sands or larger, ensuring free drainage and minimal water retention. While a few centimeters of material might be sufficient, practical construction considerations usually dictate a thickness of 0.1–0.3 meters to accommodate the use of standard construction equipment (Gebert et al., 2022).

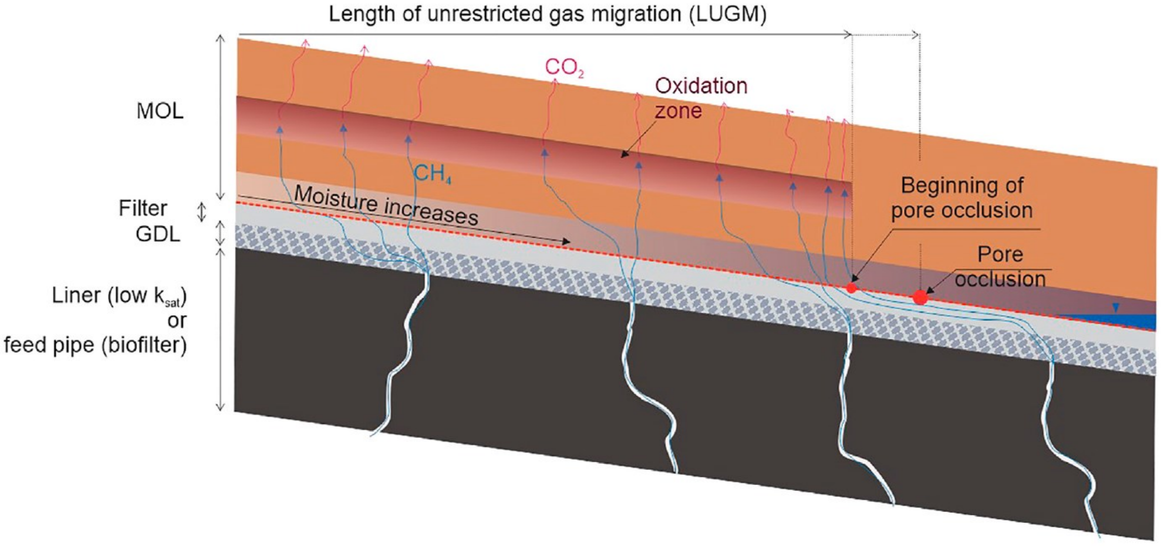


Figure 12. Schematic representation of the Typical MMOS Layering and the Length of Unrestricted Gas Migration (LUGM) (Gebert et al., 2022).

4.1.3 Filter Layer (FL)

The Filter Layer (FL), positioned between the coarser Gas Distribution Layer (GDL) and the finer Methane Oxidation Layer (MOL), serves a crucial function in methane oxidation systems. Its primary objective is to prevent mechanical clogging by intercepting particle migration from the MOL to the GDL (Cassini et al., 2017). This interception is pivotal in ensuring uninterrupted gas flow and preventing the redirection of this flow, which could otherwise result in operational inefficiencies or hotspots within the system.

Essential Properties of FL Materials: The FL must effectively stop particles from the MOL from infiltrating the GDL, upholding continuous gas flow and system integrity. By reducing the hydraulic difference between the GDL and MOL, the FL plays a significant role in minimizing the capillary barrier effect, which could otherwise impede gas flow and lead to preferential pathways. Similar to the GDL, the FL operates in an environment with fluctuating redox conditions, necessitating the use of materials that are free from dissolvable iron to prevent clogging from iron oxide/hydroxide precipitation (Gebert et al., 2022).

Suitable Materials for the FL:

- Granular Materials: The use of granular materials that conform to established conservative filter criteria recommended (Messerklinger S, n.d.). These materials are chosen to prevent the erosion of the MOL material while still allowing for adequate water and gas permeability/diffusivity.
- Geosynthetic Materials: In cases where geosynthetics are employed as part of the FL, stringent filter criteria must be adopted (e.g. Giroud, 2010). However, the use of geosynthetics carries an increased risk of interface effects, such as clogging from fine particles, iron oxides/hydroxides, or biofilm growth.

Design Considerations: The thickness of the FL is influenced by practical considerations related to constructability within the methane oxidation system. Ensuring the materials used for the FL meet Terzaghi's filter criteria is essential for effective particle migration prevention (Cassini et al., 2017). The decision to include an FL depends on the contrast in unsaturated hydraulic conductivity between the GDL and MOL, underscoring the necessity of a tailored approach based on specific system conditions.

4.1.4 Methane Oxidation Layer (MOL)

The Methane Oxidation Layer (MOL) plays a critical role in methane management systems by facilitating the oxidation of CH_4 into less harmful compounds. This layer must be shielded from environmental extremes such as excessive heat or cold, and variations in moisture levels, to preserve its CH_4 oxidizing efficiency. The MOL is typically structured into two sublayers (Gebert et al., 2003):

- **Vegetated Top Part**: Composed mainly of humic topsoil, serving as the primary rooting zone for vegetation.
- **Subsoil Layer**: Acts as a support layer beneath the vegetated top part.

These sublayers work together to create a conducive environment for methane oxidation, influenced by atmospheric air penetration, landfill gas composition, and environmental conditions. Some of the key requirements that the MOL must provide are:

- Support both advective and diffusive gas transport to regulate CH_4 and O_2 levels effectively.
- Retain sufficient moisture to support the microbial and plant life critical for CH_4 oxidation.
- Provide a physical environment conducive to the growth of methanotrophic bacteria and their cohabitants.
- Maintain temperature within a range that supports methanotrophic activity.
- Offer a geochemical environment suitable for methanotrophic processes.
- Supply essential nutrients for the sustained activity of microorganisms and plants.

Balancing gas diffusivity and water retention is a central design challenge, necessitating materials that can accommodate both needs without compromising the system's functionality.

Requirements for MOL Material:

- Materials should contain organic matter to support nutrient availability and soil structure but minimize biodegradable content to preserve gas permeability.
- It's crucial to manage the capillary barriers within the MMOS to prevent water clogging and ensure efficient gas transport.
- The MOL's gas conductivity (kGas) must be lower than that of the GDL to ensure uniform CH_4 distribution.

- High air capacity is essential for O2 ingress and CH_4 oxidation, influenced by the soil's pore structure and water retention capabilities.
- MOL materials should resist excessive compaction to maintain air-filled porosity and gas diffusivity.

Physical and Hydraulic Properties:

- Materials for the MOL should promote gas exchange with the atmosphere and within MMOS layers, necessitating a focus on soil physical and hydraulic properties.
- Compaction of MOL material should be minimized to prevent loss of gas permeability and physical instability.
- The pore-size distribution must be considered in design to ensure effective water retention and gas transport.

Chemical Properties:

- pH Value: Should range between 5.5 and 8.5 to support methanotrophic activity.
- Organic Matter Content: Between 2 and 8% for topsoil (1-4% TOC) and less than 1% for subsoil.
- Electric Conductivity: Should be lower than 4mS/cm to prevent methanotrophic activity decline.
- Ammonium Levels: High NH_4+ concentrations can inhibit CH_4 oxidation and should be avoided (Gebert et al., 2022).

Suitable Materials for MOL: Suitable materials include natural humus in mineral topsoils, sand-dominated textures, and artificial substrates like porous clay or perlite, considering their physical stability, gas and water conductivity, and nutrient supply capabilities (Gebert et al., 2022). Materials prone to cracking or those with a significant clay content are not ideal due to potential gas migration issues.

Use of MOL Materials Rich in Organic Matter

Organic materials, notably compost, are extensively utilized in the Methane Oxidation Layer (MOL) either solely or as an amendment to mineral covers (Gebert et al., 2022). Their application spans various functions, including serving as filter beds (mixed or not with soils or other structuring materials) for biofilters or as components of biowindows. The primary advantages of using compost in MOL include:

- Offering abundant nutrients essential for microbial colonization and activity.
- Providing high surface area conducive to microbial growth.
- Exhibiting high water retention capacity beneficial for microbial and plant life within the MOL.
- Despite high moisture retention, compost's coarse pore structure ensures adequate gas diffusivity and water and gas conductivity.

- Compost's low thermal conductivity aids in maintaining optimal temperatures for CH_4 oxidation, especially in cooler climates by leveraging the exothermic nature of methane oxidation (Huber-Humer, 2004).
- Combining compost with structuring materials like large wood chips or mineral soils can mitigate issues related to reduced diffusivity or waterlogging.
- In various research papers high CH_4 oxidation rates up to 24 g m^{-2} h^{-1} have been observed for composts (Huber-Humer, 2004; Schuetz et al., 2003) and up to 33 g m^{-2} h^{-1} for compost mixed with sand (Roncato & Cabral, 2011).
- Compost materials are usually easily available at a low cost.

Challenges with Organic Materials:

- Immature compost may lead to undesirable temperature increases within the MOL, potentially hampering methane oxidation in warmer conditions.
- The microbial degradation of organic materials can result in settlement, reducing gas permeability, conductivity, and diffusivity over time.
- The aerobic degradation of organic matter competes with methane oxidation for oxygen, necessitating the use of stable, not easily degradable organic substances.
- High organic content that is not sufficiently humified may foster anaerobic conditions, leading to methane generation within the MOL or landfill cover.

Use of Biochar as an Amendment: Research indicates biochar's potential to enhance CH_4 oxidation through its high surface area, improved soil porosity, and water retention capabilities. Biochar amendments have shown to boost methane oxidation rates, attributed to enhanced methanotroph populations due to better water sorption compared to control soils (Tolaymat et al., 2010).

Pairing GDL and MOL Materials for CH_4 Load Distribution

The objective of strategically pairing Gas Distribution Layer (GDL) and Methane Oxidation Layer (MOL) materials is to ensure a uniform distribution of methane (CH_4) load across the MOL. This uniformity is crucial for preventing system overloading and the formation of hotspots, which can compromise the efficiency of methane oxidation systems. It's essential to design the system in a way that maximizes the length of unrestricted gas migration (LUGM) (Figure) along the sloped interface between the MOL/GDL or Filter Layer (FL). This approach aims to expand the area through which biogas can enter the MOL, enhancing the spatial homogenization of CH_4 load (Gebert et al., 2022). Achieving minimal variance in pressure loss between the longest and shortest paths of landfill gas before it reaches the MOL base is critical. This condition is met when the gas conductivity (k_{Gas}) of the GDL surpasses that of the MOL, necessitating careful assessment of the difference in k_{Gas} values during the design phase. The design goal focuses on equalizing pressure loss across the GDL to prevent preferential pathways of gas flow, ensuring a balanced CH_4 load distribution.

4.2 Types of Microbial Methane Oxidation Systems

Microbial Methane Oxidation Systems (MMOS) represent a sophisticated, engineered approach designed to mitigate the release of landfill gases by channeling them through a multi-layered bed of biofilter media that supports the growth of methanotrophic bacteria - microorganisms that consume methane as their primary energy and carbon source - to convert methane (CH_4) into carbon dioxide (CO_2), water (H_2O), and biomass. MMOS can be implemented in various configurations, tailored to specific landfill conditions and objectives. This chapter delineates the predominant MMOS: methane oxidation biofilters, biowindows, and biocovers, offering a comprehensive analysis of their operational conditions, application and efficiencies.

4.2.1 Biofilters

Biofilters are self-contained fixed-bed reactors where methane produced in landfills undergoes oxidation by methanotrophic bacteria residing in the filter material. These systems can be utilized alongside traditional surface sealing methods and necessitate an active or passive gas supply system for directing landfill gas to the filter. The configuration of two open systems is depicted in Figure 13, with one being below-grade and the other above-grade. Biofilters can function as an open reactor (Gebert et al., 2003) or as closed reactors (Streese & Stegmann, 2003). In open systems, atmospheric oxygen permeates through diffusion from the surroundings. This system is especially appropriate for passive operation, where the gas flow through the filter occurs due to the differences in pressure between the waste material and the surrounding atmosphere. Factors like temperature, humidity, and pressure are regulated by the local climate conditions, consequently displaying seasonal fluctuations; nonetheless, the impacts of extreme conditions are mitigated by the presence of vegetation on the top, which additionally serves as a protective system against erosion.

Conversely, in closed reactors, oxygen needs to be introduced either prior to gas loading or through aeration of the filter bed, unless the gas mixture already contains sufficient oxygen. This scenario typically arises when treating landfill gas from a site undergoing in situ aeration. In closed bed biofilter systems, variables such as temperature, humidity, and pressure can be regulated (Gebert et al., 2022). Ultimately, the employment of closed bed biofilters might face limitations due to the total gas load; in fact, under elevated gas loading conditions, the overall size of a biofilter must be substantial, potentially becoming economically infeasible (Kjeldsen & Scheutz, 2018).

Applications: The application of biofilters is particularly beneficial in landfill scenarios where existing infrastructure, such as gas collection systems or similar energy conversion units, are aged and associated with high operational and maintenance costs. In such contexts, the inefficiency and financial impracticality of replacing these energy conversion systems necessitate the exploration of alternative solutions. In this case, biofilters emerge as a viable and cost-effective option.

Furthermore, the closed bed biofilter systems are designed with the capability to regulate internal environmental conditions, including temperature, humidity, and pressure. This is

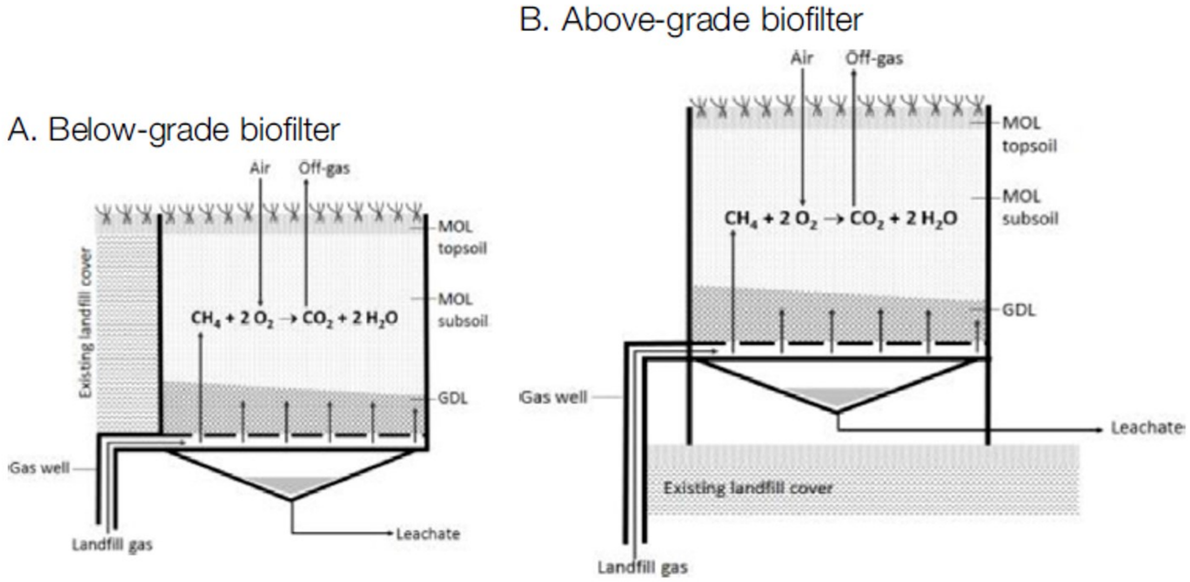


Figure 13. Biofilters conceptual scheme (Gebert et al., 2022)

particularly advantageous for landfills located in regions with substantially low temperatures during winter seasons. The capacity for environmental control within the biofilter ensures the comfortable conditions for methanotrophic bacteria to consume CH_4 which optimizes methane oxidation efficiency.

4.2.2 Biowindows

Biowindows are open compartments incorporated into the landfill top cover, characterized by higher permeability than the surrounding material (Figure 14). They can be pre-planned elements of the landfill design or subsequently installed to remediate localized emission hotspots which emerge, for example, because of cracks caused by settlement or drying, or by differences in degree of compaction of the cover soil, as well as cracks in the interface area between waste and overlaying soil. Biowindows function similarly to open biofilters, with atmospheric oxygen diffusion as the primary source but do not require connection to a gas extraction system.

Applications: Biowindows are especially suitable for old landfill sites that do not have gas extraction system and are already capped with soil. In cases where a geosynthetic liner is incorporated into the final cover, existing gas wells can serve to channel the landfill gas through the liner and into the window. Biowindows have the potential to substitute gas wells in

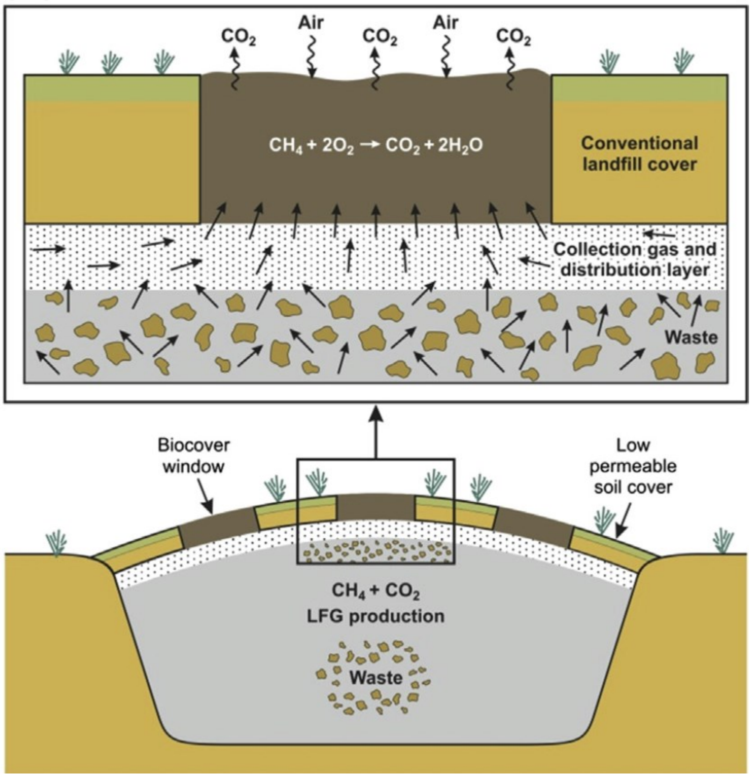


Figure 14. Biowindows conceptual scheme (Kjeldsen & Scheutz, 2018)

situations where the operational gas extraction system in sealed landfills needs to be turned off (Gebert et al., 2022).

4.2.3 Biocovers

Biocover, a specifically engineered soil cover, serves a variety of purposes, including recultivation, maintaining water balance, and possessing suitable properties for the intended after-use. Typically, it extends over the entire landfill area or a significant portion of it (Röwer, 2014). Landfill gas is typically passively loaded into biocovers, either directly from the underlying waste or through a previously placed cover (see Figure 15.). In cases where the biocover is implemented in an established landfill with an impermeable liner or a compacted soil layer capping the waste, landfill gas might be directed into the biocover via gas supply pipes (Gebert et al., 2022).

Biocovers offer an extensive surface area for remediation, leading to decreased spatial CH4 load compared to biofilters and biowindows. However, constructing an entire landfill cover requires large quantities of suitable materials, which can result in significant costs or even limitations in availability. Nevertheless, a significant obstacle associated with biocovers lies in the need to ensure a uniform spatial distribution of the CH4 load. This scenario is likely to occur if the cover is directly placed on top of the waste mass. Moreover, in terms of moisture distribution, significant challenges arise in achieving uniform loading over great lengths due to potential accumulation of moisture at the interface between the GDL and the MOL, especially in areas with a downward slope. One approach to tackle this issue is the incorporation of

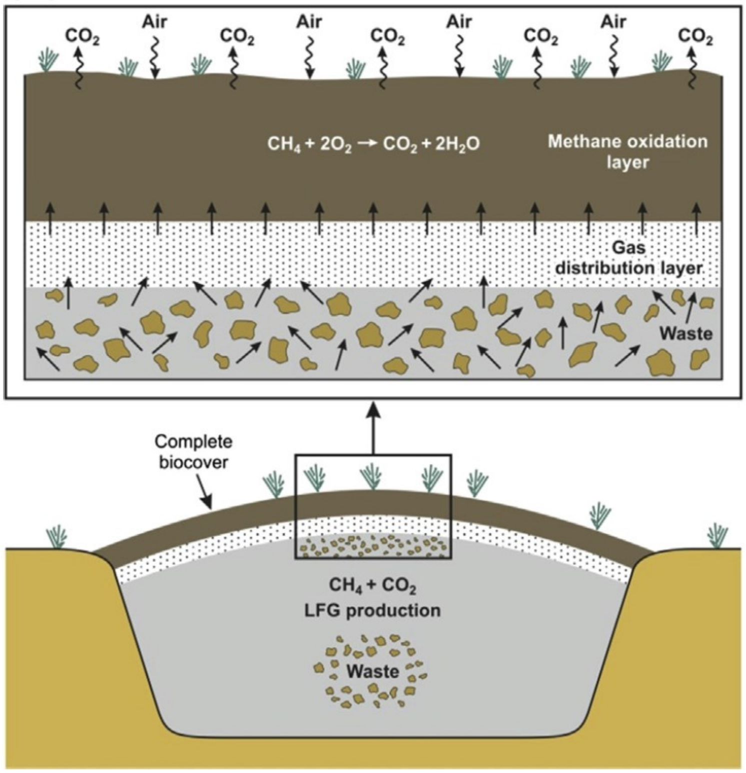


Figure 15. Biocovers conceptual scheme (Kjeldsen & Scheutz, 2018)

drainage points at specified intervals along the interface to remove accumulated moisture. Another strategy could involve introducing a sloped interface between these layers. Alternatively, the selection of substrate and construction techniques should prioritize higher methane loadings, thereby enhancing the oxidation capacity in the upslope region.

Applications: Biocover systems present an optimal solution for the mitigation of residual methane emissions from old landfills after the determination of gas extraction infrastructure. Their implementation can also be integrated into the design phase of new landfill operations handling waste streams with inherently low methane generation potential, such as those with the waste mass subjected to mechanical and biological pre-treatment processes or designated for the remediation of hazardous materials. Furthermore, biocovers can serve as a complementary measure at landfills with deployed gas extraction systems.

4.2.4 Breathing Biocovers

To address the challenges associated with O2 diffusion in landfill covers and open biofilters, researchers prefer using multi-layered beds, composts, soil, or combinations thereof. These porous materials enhance O2 distribution; however, they fail to resolve the core issue of inadequate O2 concentration. This limitation stems from the restricted capacity for atmospheric O2 to diffuse downward, typically resulting in an oxygenated zone of no more than 0,6 m from the top cover (Scheutz et al., 2009a). While forced aeration can bypass this limitation by utilizing compressor pumps to supply continuous air, this approach necessitates additional energy inputs.

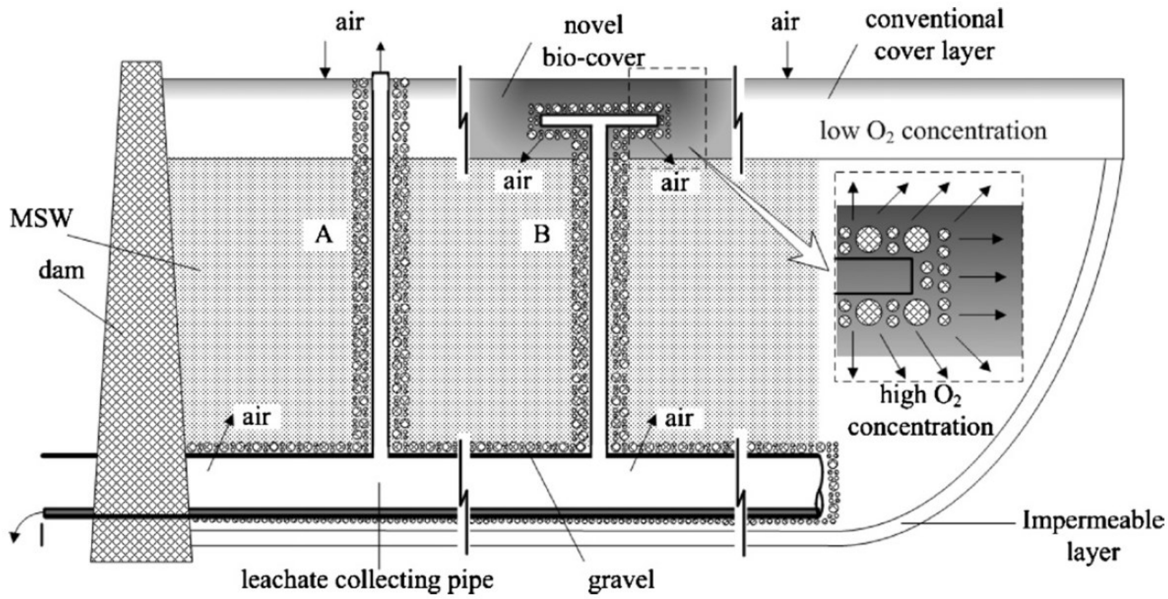


Figure 16. Breathing Biocover in Semi-Aerobic Landfill. Conceptual scheme (Lu et al., 2011)

Addressing the need for a more energy-efficient solution, the paper by Lu et al. (2011) introduces a novel "breathing biocover system" designed to facilitate continuous O2 supply through passive diffusion in semi-aerobic landfills. Central to this system is a passive air venting system, grounded in aerothermodynamic principles derived from semi-aerobic landfill practices (Figure 16). The passive air venting system serves as a supplementary oxygen source to the biocover, enhancing O2 availability beyond what atmospheric diffusion can achieve alone.

The research demonstrates significant improvements in O2 concentration across the entire 1-meter profile of a microcosm equipped with the modified passive air venting system. Specifically, under simulated landfill gas flow rates of 771 g m-3 d-1 and 1028 g m-3 d-1, the O2 levels within the modified passive air venting system exhibited a gradual increase, stabilizing at atmospheric concentrations after just 10 days. Furthermore, a 100% CH4 oxidation rate at simulated landfill gas flow rates up to 1285 g m-3 d-1was observed (Lu et al., 2011). The introduction of the breathing biocover system offers a promising solution to the pervasive problem of insufficient O2 in cover layers of semi-aerobic landfills.

4.3 Comparative Analysis of Methane Oxidation Efficiencies of MMOS in Existing Landfills

In line with recommendations from Heyer et al., 2013, engineered MMOS are advised for diminishing residual methane emissions when the methane load on the cover layer exceeds 0.5 L CH_4 m^{-2} h^{-1} (0.36 g CH_4 m^{-2} h^{-1} or 8.64 g CH4 m^{-2} day^{-1} under standard temperature and pressure conditions). It is assumed that lower CH_4 loads can be oxidized by any cover soil or layer, even those that are not specifically optimized for microbial methane oxidation. However, relatively high CH4 loads exceeding the oxidation capacity of the medium may result in excessive advective methane bottom flux that hinders diffusive influx of oxygen from the surface (Gebert et al., 2011). To identify the methane fluxes that can be optimally oxidized by MMOS, a comparative analysis encompassing both laboratory and field-scale evaluations of methane oxidation efficiencies in landfill covers was undertaken.

Roncato and Cabral (2011) assessed the methane oxidation efficiency of substrate materials (a mixture of sand and compost) under both field conditions and in the laboratory using column tests. The field tests achieved a maximum oxidation rate of 576 g CH4 m^{-2} day^{-1}, significantly higher than the laboratory results of 115 g CH4 m^{-2} day^{-1}, with an oxidation efficiency of 96%. The discrepancy between field and laboratory outcomes remains unexplained, though it was hypothesized that vegetation on the surface of the passive methane oxidation biocovers (PMOBs) could significantly enhance methane oxidation efficiencies in field settings.

Cabral et al. (2009) reported on an experimental PMOB constructed on the final cover of the St-Nicéphore landfill, utilizing a substrate mix of sand and compost 0.80 m thick. The maximum methane load applied was 27 g CH4 m^{-2} h^{-1} or 648 g CH4 m^{-2} day^{-1}. The study found that nearly all the methane input was oxidized, with absolute removal rates linearly correlated with methane loading, indicating that the PMOB's maximum oxidation capacity was not reached during the study period.

The results suggest that the maximum oxidation capacity of biocovers can surpass 648 g CH4 m^{-2} day^{-1}. However, it is critical to consider that the study did not encompass the examination of factors such as media compaction, the potential for clogging, and the long-term degradation of the biocover materials and its effect on methane oxidation capacity.

Additionally, Table 1 provides a comprehensive compilation of 4 reported in scientific literature MMOS, with details on the landfill setting, scale, system type, monitoring methods, and mitigation efficiency. The reported gas loading rates exhibit significant variability, ranging from 32 g CH_4 m^{-2} day^{-1} to greater than 620 g CH_4 m^{-2} day^{-1}. However, a clear correlation between gas loading rate and mitigation efficiency is lacking. Hence, it is evident that there is much more to be discovered regarding the key environmental factors that govern the effectiveness of bio-mitigation systems.

Table 1. Compilation of established MMOS in existing landfills reported in literature (Adopted from Kjeldsen & Scheutz, 2018)

Landfill, Country	**Scale[a]**	**System Type**	**Typical Gas Composition[c] (%CH_4 /%O_2)**	**Active Material**	**Gas Distribution Layer[e]**	**Reported Gas Load [f] GL g/m^{-2}d^{-1}**	**Total Efficiency [f] ME(%)**	**Reference**
Aikkala, Finland	FS	Full surface biocover (FSB) with gas wells	31-72/ 1-5	Compost, 39,000 m2, 50 cm depth	50 cm	32-216	25-46%	Einola et al., 2008
Landfill x, United Kingdom	FS	Biofilter active, open bed (BF-AO)	20-40/ n.r.	Compost and expanded clay pellets, 4x150 m2, 130 cm depth	30 cm	530	55-99%	Parker, 2013
Wieringermeer, Netherlands	PS	Biofilter active, open bed (BF-AO)	Sewage sludge gas	Soil, 510 m2, 100 cm depth	20 cm	43	30-96%	M. Geck, 2013
Two French Landfills, France	PS	Biofilter active, closed bed (BF-AC)	2-2.5/ 18-20	17 m3 compost	30 cm	620	15-17, max60%,	Ducrocq, 2013

[a] FS: Full Scale, PS: Pilot scale.

[c] Composition of CH_4 and O2 in LFG.

[e] D: thickness of gas distribution layer (cm).

[f] GL: reported average gas load to biocover system; ME: Reported methane removal efficiency.

[g] Methods supporting performance evaluation: WLEM: whole landfill emission measurement using tracer dilution approach or other method; FC: flux chamber measurements; PGP: pore gas profiles; MSS: methane surface screen; SCIC: stable carbon isotope composition; MF: mass flow (gas flow and CH_4 content).

[h] Efficiency approach: CMB: carbon mass balance; PBE: profile based efficiency; TMMB: total methane mass balance; TEMBA: total CH4 emission measurement before and after. n.r. = not reported

5 Key Considerations in the Selection of Microbial Methane Oxidation Systems

The selection of the most appropriate MMOS is a nuanced process, intricately linked to the landfill's operational, environmental, and physical characteristics. This chapter delves into the essential considerations for choosing and designing MMOS, focusing on aspects such as waste composition, climate conditions, landfill configuration, and the challenges and opportunities associated with MMOS implementation. The decision tree, informed by comprehensive research, guides stakeholders through a series of pivotal queries, each leading to informed decisions tailored to specific landfill scenarios. The endeavor here is not just to outline the procedural steps but to contextualize them within the broader environmental and operational parameters, thus paving the way for effective, site-specific methane mitigation strategies.

5.1 Waste Composition

Organic Content and Biodegradability: Landfills containing a high amount of biodegradable organic waste (e.g., food waste, garden waste) produce significant quantities of methane (CH_4) during the anaerobic decomposition process. The presence of such materials requires an MMOS capable of handling high methane fluxes, necessitating designs that enhance microbial methane oxidation capacity and ensure efficient gas distribution and water management within the system.

Moisture Content: Optimal moisture conditions facilitate the maximum biological activity of methanotrophs. If the waste body is too dry, it can limit the bacteria's ability to oxidize methane, leading to lower MMOS efficiency. Conversely, overly saturated conditions may create anaerobic pockets, reducing oxygen availability necessary for methanotrophs and potentially leading to increased methane production rather than oxidation (Dizon et al., 2023). For instance, biocovers, which rely on soil or compost layers for methane oxidation, may be more suitable for sites with moderate moisture, leveraging the natural water retention capabilities of the cover material. In contrast, engineered systems like biofilters might be preferred in drier conditions, where moisture can be more precisely controlled and supplemented if necessary.

The main design considerations for moisture regulation include the integration of irrigation systems in biofilters to maintain optimal moisture or the design of drainage systems within biocovers to prevent waterlogging (Gebert et al., 2022). Effective leachate management becomes a critical operational aspect, as leachate recirculation can be used to adjust moisture content beneficially. In drier climates or periods, leachate or external water sources might be applied to increase moisture, while in wetter conditions, excess water may need to be drained or evaporated to prevent oversaturation.

Continuous monitoring of moisture levels within the MMOS and the waste body is crucial to maintain the optimal conditions for methane oxidation. Systems may need to be adjusted based on seasonal variations, changes in landfill operations, or observed fluctuations in moisture content.

Physical Properties of Waste: The compaction and texture of the landfill waste affect the porosity and permeability of the landfill, impacting gas migration paths and the distribution of methane and oxygen. Uneven compaction or the presence of large voids can lead to uneven gas distribution, creating "hotspots" of methane emissions (Gebert et al., 2011).

Age of the Waste: The age of the waste affects both the quantity and quality of methane produced. Older landfills, especially those that have undergone partial stabilization, might produce less methane but over a prolonged period. The choice of MMOS, in this case, could lean towards passive systems like biocovers, which are suitable for lower methane fluxes.

Current Gas Production and Methane Load: The volume of gas produced directly informs the capacity requirements for the MMOS. Systems must be designed to handle peak methane production levels to ensure efficient methane oxidation and minimize emissions. Landfills with high methane loads are more likely to implement MMOS that are integrated with energy recovery systems, such as gas-to-energy plants, since the higher the methane content, the more energy can be generated from the gas (EL-FADEL et al., 1996).

High gas production rates might favor more active MMOS designs, such as biofilters, which can be engineered to manage larger volumes of methane. Conversely, lower production rates might be adequately addressed with passive systems like biocovers. The cost of implementing and operating MMOS is closely tied to the scale of gas production. Larger, more complex systems required for high gas production volumes involve higher capital and operational costs. Accurate assessment of current gas production enables cost-effective system design, balancing the initial investment against the expected performance and lifespan of the system. Landfill gas production rates change over time as waste decomposition progresses (Tolaymat et al., 2010). Systems designed with an understanding of current and projected gas production can adapt more easily to these changes, maintaining high oxidation efficiency throughout the landfill's lifecycle.

5.2 Climate Conditions

This table provides an overview of how each system adapts to varying climatic conditions, highlighting their strengths and weaknesses in hot and cold climates, as well as during rainy and dry seasons:

Climate Condition	Biofilters	Biowindows	Biocovers
Hot Climates	- Perform well with enhanced microbial activity and methane oxidation rates. - Crucial to maintain optimal moisture due to rapid evaporation.	- Similar performance to biofilters. - Potentially lower maintenance but requires moisture management.	- Very effective with a thick, moisture-retaining layer. - Offers better resilience to drying out.
Cold Climates	- Efficiency decreases as low temperatures slow down microbial metabolism. - May require insulation or passive solar heating.	- Challenges due to reduced microbial activity. - Integration into landfill cover may provide some insulation.	- Might perform better than biofilters and biowindows. - Potential for deeper, more insulated layers that retain heat.
Rainy Seasons	- Risk of waterlogging. - Need for adequate drainage.		- Susceptible to waterlogging but offers better drainage and resilience. - Design to prevent runoff and erosion is crucial.
Dry Seasons	- Challenge to maintain moisture for microbial activity. - May require frequent watering.		- More resilient due to greater volume and organic matter. - Still requires monitoring and potential moisture addition.

5.3 Landfill Configuration

Landfill Topography and Inclination: The physical layout, including the size, slope, shape, and topographical features of a landfill, dictates the type of MMOS that can be effectively implemented. Passive systems like biocovers might be more suitable for landfills with moderate slopes, where natural microbial activity can be leveraged without extensive engineering modifications. In contrast, engineered systems like biofilters may be necessary for landfills with steep inclinations or complex geometries, where precise control over gas migration and moisture is needed (Gebert et al., 2022). MMOS designs may need to be adaptable or modular to accommodate the unique geometry and inclination of the landfill. This could include segmented systems that target specific areas of the landfill or flexible installation techniques that can accommodate varying slopes.

Existing Infrastructure Availability: The selection process involves balancing the technical feasibility, environmental compliance, and cost-effectiveness of integrating MMOS with the existing infrastructure to optimize methane oxidation and reduce greenhouse gas emissions from landfill sites.

- **Gas Extraction Pipes**: Landfills with efficient gas extraction and collection systems may opt for MMOS that can be integrated with these systems to treat the collected methane. For example, the collected gas can be actively or passively routed to an external biofilter for treatment. This setup is particularly suitable for in-situ aerated landfills that operate a gas collection system, as it allows for the management of high gas fluxes and low CH_4 concentrations (Gebert et al., 2022).
- **Leachate Collection Systems:** The design and operation of MMOS must also consider the impact on and integration with existing leachate collection systems. The presence of a leachate collection system underneath the landfill cover can influence the moisture content and hydraulic properties of the cover material, which in turn affects the gas transport and microbial activity within MMOS. Proper integration ensures that the methane oxidation process does not negatively impact the leachate collection system's efficiency or the overall environmental safety of the landfill (Gebert et al., 2022).
- **Existing Landfill Cover/Sealing Layer:** For landfills with an impermeable cover layer or liner, modifications might be needed to allow gas to reach the MMOS effectively. This could involve creating penetrations in the liner for gas routing or opting for biowindows or biocovers.

Landfill Closure and Aftercare Plans: The intended after-use of the landfill site plays a crucial role in selecting MMOS. For instance, if a site is destined for recreational use, a biocover that supports vegetation and integrates aesthetically with the surroundings might be preferred. Conversely, if the site will host structures or requires a sealed surface, systems that can be covered or adapted to these needs, such as certain types of biofilters, may be more suitable. The MMOS chosen must align with the aftercare plans to ensure that the landfill's final land use is not compromised by the system's presence or maintenance requirements.

5.4 Exemplary Decision Tree on Use of MMOS

In a research paper done by (Gebert et al., 2022) a decision tree that facilitates the determination of the most suitable MMOS for a specific site has been presented (Figure 17). This decision tree encompasses various crucial factors that are relevant to the decision-making process, including recoverable CH_4 flow, availability of gas extraction wells, and landfill geometry. The user is guided through the decision tree by responding to technical questions that can be answered with a simple yes or no. The consideration of potential after-use is an important component of the final decision

The starting point, which is also the most significant aspect, involves determining whether there is still gas formation and/or gas emission potential that justifies the implementation of MMOS to mitigate CH_4 emissions at the landfill site. In accordance with recommendations by Heyer et al., 2013 regarding criteria for releasing landfills from aftercare, engineered MMOS are suggested for reducing residual emissions when the approximate CH_4 load to the cover layer is still greater than 0.5 L CH_4 m^{-2} h^{-1} (0.36 g CH_4 m^{-2} h^{-1} under standard temperature and pressure conditions). It is assumed that lower CH_4 loads can be oxidized by any cover soil or layer, even those that are not specifically optimized for microbial methane oxidation. The specific magnitude of the residual flux that guarantee transitioning from active gas extraction and technical treatment (such as flare or energetic use) to the treatment of CH_4 in MMOS is dependent on the characteristics of the site, including the size of the landfill and the total CH_4 flux, as well as the efficiency of the existing active gas extraction and technical treatment, and the intended after-use of the site.

Other significant aspects that have been discussed in this chapter are also addressed in the decision tree, such as the inclusion of gas wells and gas extraction systems, the consideration of existing cover layers and bottom liners for leachate collection, the analysis of landfill geometry, and the implementation of measures to protect groundwater. The decision-making process also takes into consideration various considerations and questions related to subsequent and/or supplementary technical adaptations of the site.

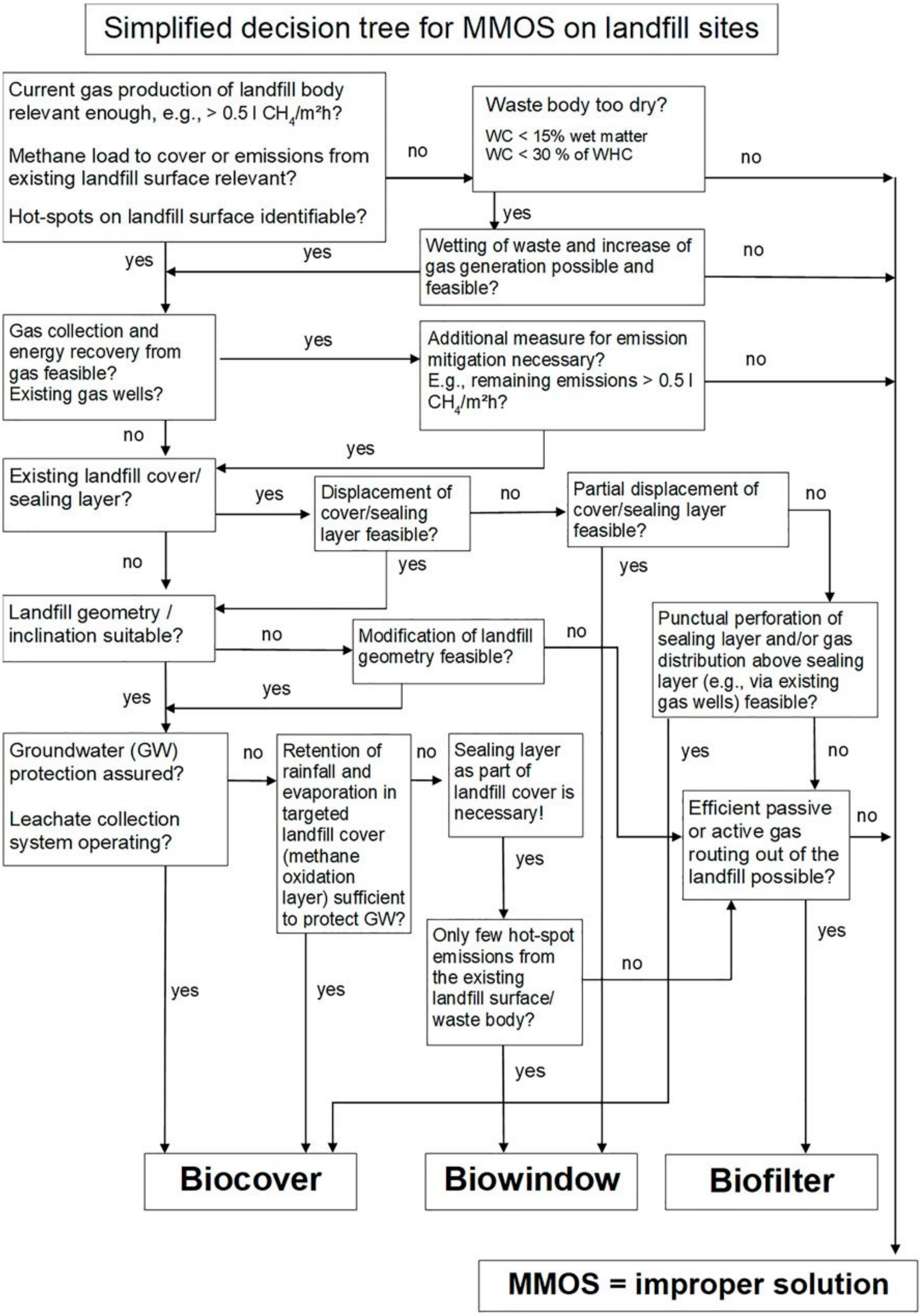


Figure 17. Exemplary decision tree on use of MMOS.

5.5 Examples of MMOS Applications for Various Landfills Configurations

Old Waste Dumps Without Final Top Cover and Without Gas Collection System

Worldwide, uncovered and largely unregulated waste dumps continue to pose a significant challenge. These sites frequently employ covers composed of whatever materials were accessible to the operators at the time of establishment. Given that the majority of emissions typically stem from localized hotspots, a pragmatic approach would involve addressing these specific areas through the installation of methane oxidation windows. It is feasible that, in numerous instances, a well-designed passive MMOS (such as a biocover or biowindow) using locally available materials could be utilized as an alternative or supplement to the current covering. Such simplified maintenance systems present a far superior option to taking no action, especially in regions where legal mandates and financially viable modern technical solutions are lacking.

Sanitary Landfills With a Final Top Cover And Gas Collection System

For sites belonging to this classification, the application of a MMOS serves as a supplementary measure during the active gas extraction and technical treatment of gas, especially when the top liner has not been installed or as a final solution after technical gas treatment is being terminated. Existing gas wells can be utilized to actively or passively supply biofilters or biowindows, which can also be integrated into the landfill cover in retrospect.

In situations where biocovers is intended, and there is already an existing top liner, careful consideration must be given to the sufficient spatial distribution of the load above the existing liner by utilizing a gas piping system within or beneath the gas distribution layer. If a top liner is already present, it must be perforated to direct landfill gas towards a gas distribution layer situated below a methane oxidation layer. For landfills treating mechanically-biologically pretreated waste, which typically have low gas production, passively vented biowindows (integrated into surface liners) or biocovers are considered as the most suitable decisions.

Closed Sanitary Landfills With Temporary Covers

For older sanitary landfills approaching landfill closure but still containing significant amounts of biodegradable organic waste, the use of biocovers can be a practical approach in certain regions. These covers should be designed to allow water infiltration into the waste mass to facilitate the degradation processes of the waste materials. However, these temporary covers must also effectively mitigate CH4 emissions.

Old Landfills During and/or After In-Situ Aeration

Old municipal solid waste landfills, which still contain biodegradable waste materials and are operated under in-situ aeration to accelerate the processes of mineralization and stabilization, have the option of being covered with a biocover to reduce the remaining methane emissions (Ritzkowski and Stegmann, 2012; Laux, 2015). Landfills with in-situ aeration systems that also have a gas collection system can choose to direct the collected air-gas mixture to an external biofilter, either actively or passively. The effectiveness of these filters is challenged by the high gas fluxes and low concentrations of CH4. Any gas emissions left post the in-situ aeration process can be mitigated through the use of a properly engineered biocovers or biowindows.

Landfills for Contaminated Soils and Mechanically and Biologically Pretreated Wastes

Landfills falling within this classification typically exhibit a low gas generation potential, making them conducive for the passive gas collection through and treatment using biofilters. Strategically positioned biowindows incorporated into the current top cover could also present a feasible alternative. In the same vein, biocovers emerge as a viable choice provided that the landfill gas can be directed to a gas distribution layer via penetrations in the current liner, if required.

5.6 Challenges and Opportunities Associated with the Implementation of MMOS in Landfills

Material Selection and Layering

Selecting appropriate materials plays crucial role in the performance of Methane Oxidation Systems (MMOS). These materials must support methanotrophic bacteria, provide physical stability, and meet gas and water transport requirements. However, enhancing the biological methane oxidation process is challenging due to its dependence on various factors, including specific microbial communities, environmental conditions, and substrate availability. The efficiency of methane oxidation can fluctuate significantly with changes in temperature, moisture content, and nutrient availability, necessitating careful consideration in MMOS design. Moreover, the layering of materials must prevent compaction and clogging, maintain porosity for gas exchange, and support moisture retention without saturating the system.

Achieving uniform spatial distribution of CH4 across the MMOS is a complex design consideration. Spatial inhomogeneity of landfill gas may arise from factors such as uneven distribution at the MMOS base, creation of secondary macropores within the system, or disparities in compaction and moisture content of the materials. These preferential pathways often lead to CH4 loads exceeding the oxidation capacity of the medium, resulting in excessive advective methane bottom flux that hinders diffusive influx of oxygen from the surface (Gebert et al., 2011). This phenomenon creates "hotspots" - areas with high CH4 emissions or uncontrolled fluxes on the landfill surface. Ensuring the uniformity of CH4 distribution significantly enhances emission control, with CH4 oxidation rates reaching close to 100% of the input load (Gebert et al., 2022).

This highlights the opportunity for innovative MMOS design which could involve the integration of engineered systems to resolve the issue associated with reduced oxygen uptake into methane oxidation layer. The systems might be designed as air blowers with connected wind turbines locally placed over the surface of the top cover soil for passive oxygen ingress. This strategy could provide methanotrophic bacteria with favorable ambient and enhance the overall methane oxidation capacity of MMOS. However, it is essential to consider the potential escaping of methane through these systems.

Another significant challenges in designing MMOS is finding a balance between maintaining sufficient moisture for microbial activity while ensuring adequate gas diffusivity for methane and oxygen transport within the system. In MMOS designed following the typical layering sequence outlined in the chapter Typical Layouts of Methane Oxidation Systems, the difference in hydraulic properties among the components results in the development of capillary barriers. As water moves downhill, heightened moisture levels near the interface of MOL-GDL (or MOL-FL) may cause the pores within the soil to be blocked by water, thus impeding gas flow. Consequently, a primary obstacle in the design of MMOS lies in managing the potentially conflicting mechanisms of water and gas transport processes (Gebert et al., 2022). This opens up an opportunity for advanced material engineering, aiming to develop or enhance materials that optimize methane oxidation under varying conditions. Advanced composite materials, engineered biochar, or specifically designed porous media could provide enhanced physical and biochemical properties. Such materials could be tailored to maintain an ideal balance of moisture and gas diffusivity, crucial for maximizing methane oxidation rates (Tolaymat et al., 2010).

Emission Monitoring and Evaluation of Methane Oxidation Efficiency of MMOS

Landfill emissions exhibit significant heterogeneity, with spatial and temporal variations arising from the dynamic nature of waste decomposition, top cover soil properties, and environmental conditions. This variability poses a challenge in accurately quantifying and characterizing emissions using traditional point-based monitoring techniques such as static surface flux chambers. Despite their widespread use, surface chambers possess a restricted surface area (typically <1 m2), resulting in coverage of only a very small portion (<1%) of the total landfill area, even after multiple measurements. Moreover, a significant portion of CH4 emissions arises from localized hotspots, which are unlikely to be captured by a grid of chamber measurements, leading to an underestimated total gas production (Kjeldsen & Scheutz, 2018).

Further complicating the monitoring process, a substantial portion of landfill emissions can occur through subsurface migration and diffusive pathways (Njoku et al., 2023) These subsurface emissions, traveling laterally through the waste mass or underlying soil layers, are challenging to detect and quantify using surface-based monitoring methods, potentially leading to underestimation of total emissions. Moreover, various meteorological and environmental factors, such as precipitation, temperature, atmospheric pressure, and wind speed and direction, influence landfill emissions (Njoku et al., 2023). These factors affect the generation, transport, and dispersion of emissions, introducing additional complexities in monitoring and interpreting the data accurately.

This underscores the opportunity to integrate advanced monitoring and data analytics for MMOS performance optimization, employing next-generation remote sensing techniques, such as satellite-based hyperspectral imaging and airborne light detection and ranging (LiDAR). These techniques enable large-scale, high-resolution mapping of emission hotspots (Papale et al., 2023). Combining remote sensing data with ground-based measurements and atmospheric modeling can provide a comprehensive understanding of landfill emissions and their environmental impacts. Furthermore, the integration of distributed sensor networks and Internet of Things (IoT) technologies has enabled real-time, high-resolution monitoring of landfill emissions (Moltchanov et al., 2015) These systems leverage low-cost, low-power sensors and wireless communication protocols to collect and transmit data, allowing for continuous monitoring and early detection of emission anomalies. Advanced data processing techniques, such as machine learning algorithms, can be applied to these data streams for predictive analytics and optimized monitoring strategies (Behera et al., 2015).

Accurate and comprehensive emission monitoring techniques are essential for quantifying the methane flux before and after it passes through the MMOS. This allows for the determination of the oxidation efficiency, which is a key performance indicator for assessing the effectiveness of these systems in mitigating landfill methane emissions (Huber-Humer et al., 2008). Furthermore, emission monitoring data can provide insights into the influence of various environmental and operational factors on the oxidation efficiency, informing the optimization and design of MMOS for enhanced methane oxidation. Thus, advancements in emission monitoring techniques can contribute to a better understanding and improvement of MMOS for landfill gas mitigation.

Long-term Performance and Maintenance of MMOS

Over time, the physical and chemical properties of biofilter media and cover materials undergo significant transformations due to various factors, including weathering, microbial degradation, and chemical reactions. To ensure the optimal performance of these systems, continuous monitoring coupled with regular maintenance is required. This proactive approach allows for the identification and resolution of issues like media compaction, clogging, and material degradation, which, if left unaddressed, could compromise the efficiency of methane oxidation.

Based on the insights gained from continuous monitoring, it may become evident that operational adjustments are necessary to maintain or enhance the MMO system's performance. Such adjustments can range from altering the moisture content of the biofilter media, making nutrient amendments to possible replacement old biocover material that lost their ability to oxidize methane due to material degradation. However, the maintenance of these systems is not without its challenges. It demands a significant investment of resources, including time, labor, and materials. For landfill operators, the task then becomes one of carefully balancing these operational costs against the environmental benefits of efficient methane oxidation. This balance is crucial, as it underpins the sustainability and effectiveness of landfill operations.

Thus, the narrative of managing MMO systems in landfills is one of continuous adaptation and careful resource management. Landfill operators are tasked with not only maintaining the technical efficiency of methane oxidation processes but also with navigating the complex interplay between operational costs and environmental benefits.

The conversation around challenges and opportunities associated with MMOS design highlights the evolving landscape of MMOS implementation, signaling a move towards more innovative, efficient, and adaptive solutions. The integration of these considerations holds the promise of not only enhancing methane oxidation efforts but also contributing significantly to the global initiative of reducing landfill-related greenhouse gas emissions.

6 Conclusion

In landfills where the utilization of energy recovery system or high-temperature flares becomes technically and economically infeasible, but the approximate CH_4 load to the cover layer is still greater than 0.5 L CH_4 m^{-2} h^{-1} (0.36 g CH_4 m^{-2} h^{-1} under standard temperature and pressure conditions) in accordance with recommendations by Heyer et al. (2013) Microbial Methane Oxidation Systems (MMOS) are suggested for reducing residual emissions. It is assumed that lower CH_4 loads can be oxidized by any cover soil or layer, even those that are not specifically optimized for microbial methane oxidation.

Designing Microbial Methane Oxidation Systems (MMOS) is a multifaceted challenge that hinges on the careful selection of materials which must meet the complex requirements for the support of methanotrophic bacteria, ensuring physical stability, and facilitating appropriate gas and water transport. This delicate balance is complicated by the biological methane oxidation process's sensitivity to a variety of factors, including environmental conditions, substrate availability, and fluctuations in temperature, moisture content, and nutrient availability. Additionally, achieving an even distribution of methane throughout the MMOS to avoid "hotspots" of methane emissions, which could compromise the overall efficiency, requires meticulous planning and design.

The strategic choice and optimization of MMOS, supported by a deep understanding of waste composition, climatic impacts and landfill configuration, are becoming key factors determining efficiency in this area. While the focus of this thesis includes only technical fundamentals and proposes a principled framework for system selection and deployment, it does not consider the evaluation of the financial aspects or cost-efficiency of MMOS implementation.

Biofilters are most suitable for landfills with high methane flux, in particular closed bed biofilters offer a regulated environment beneficial for methane oxidation in landfills experiencing cold winters, optimizing the conditions for methanotrophic bacteria. Biowindows serve well in older landfills lacking active gas collection systems, providing a low-maintenance solution for addressing localized emission problems without needing a gas extraction connection. Biocovers, adaptable to various landfill types, offer broad coverage for methane mitigation but may involve higher costs due to the significant amount of materials required for their extensive surface area.

From the data reported in the scientific literature it was observed that MMOS can successfully oxidize methane gas loads as high as 648 g CH4 m^{-2} day^{-1} (Cabral et al., 2009). The study found that nearly all the methane input was oxidized, with absolute removal rates linearly correlated with methane loading, indicating that the maximum oxidation capacity of the passive methane oxidation biocover was not reached during the study period.

As we move forward, it is imperative that the waste management community continues to leverage advanced technologies, embrace adaptive strategies, and foster cross-disciplinary collaborations. Doing so will not only enhance our ability to mitigate methane emissions effectively but also contribute to the broader goal of achieving sustainable waste management and environmental protection.

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