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REVIEW PAPER

Elimination of methane generated from landfills by biofiltration: a review

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Abstract The production of biogas in landfills, its composition and the problems resulting from its generation are all reviewed. Biofiltration is a promising option for the control of emissions to atmosphere of the methane contained in biogas issued from the smaller and/or older landfills. A detailed review of the methane biofiltration literature is presented. The microorganisms, mainly the methanotrophs, involved in the methane biodegradation process, and their needs in terms of oxygen and carbon dioxide utilization, are described. Moreover, the influence of nutrients such as copper, nitrogen and phosphorus, and the process operating conditions such as temperature, pH and moisture content of the biofilter bed, are also presented. Finally, the performance of various filter beds, in terms of their elimination capacities, is presented for laboratory scale biofilters and landfill covers.

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1 Introduction

Biogas results from the anaerobic degradation of organic wastes. Every year, thousands of tons of the greenhouse gas (GHG), methane (CH_4), are produced in landfills, some of which escapes directly to the atmosphere. Even if GHG emissions associated with landfills represent only a small percentage (3.4% for Canada) of the national total of GHG emissions from all sectors, it is important to note that landfills generally constitute the most important sources of anthropogenic CH_4. For example, in Canada and the United States, around 25% and 34% respectively, of the total methane emissions are directly related to landfill installations (Environnement Canada 2006; EPA 2006). About 10,000 landfills presently exist in Canada and the average waste production per inhabitant in year 2000 was 1,020 kg, of which some 73.2% was discarded to landfills. The wastes have generated in year 2001 GHG emissions, mainly in the form of CH_4, at a level around 25×10^6 metric tons, when expressed as the carbon dioxide (CO_2) equivalent (Environne-

ment Canada 2006). The recent ratification by Canada of the Kyoto protocol forces this country, along with several others, to find new alternatives for the control of CH_4 emissions. Indeed, Canada has committed itself to reduce its GHG emissions by 6%, compared to the 1990 level, during the period from 2008 to 2012, by targeting some particular gaseous compounds, such as CH_4, for major attention (Kyoto protocol 1998).

Methane, as a GHG, is some 21–25 times more detrimental to the environment than CO_2 and its lifespan in the atmosphere is ~12 years (Hütsch et al. 1994; Goossens 1996; Hettiaratchi and Stein 2001; Kumar et al. 2004). Various technologies such as combustion can be used to control the CH_4 emissions issued from landfills but, for the older and/or smaller landfills, traditional technologies are not very applicable and thus the biofiltration approach could be a promising solution. This process is one of the oldest of biotechnologies used in the treatment of polluted air. In the beginning, the process was employed only for the elimination of odors (March 1994). Thereafter, biofiltration, applied to contaminated air, proved to be also reliable for the elimination of volatile organic compounds (VOCs) and volatile inorganic compounds (VICs) (Jorio et al. 2003; Delhoménie and Heitz 2005).

The idea of using biofiltration for CH_4 elimination derives from the fact that some bacterial species are able to degrade CH_4 while generating oxidation by-products such as water (H_2O), CO_2, salts and biomass, all products much less harmful for the environment than the initial substrate. On an annual basis, at least 10–25% of the total CH_4 emitted from landfills is oxidized by microorganisms (Nozhevnikova et al. 1993; Mancinelli 1995; Chanton and Liptay 2000; Christophersen et al. 2000; EPA 2005; Stralis-Pavese et al. 2006). Moreover, biofiltration creates environmental problems (such as CO_2 production) to a lesser extent, in comparison with regular chemical oxidation processes. Also, biofiltration often offers the advantage of being performed at normal atmospheric pressure and temperature, thus resulting in lower ranges operational costs than traditional technologies (Ottengraf 1986).

2 Sanitary landfills

A sanitary landfill is an installation arranged to receive wastes and to retain the products of their decomposition so that they cease to constitute a threat for human or animal health (Popov 2005; Zamorano et al. 2006). Several types of landfills presently exist, some, known as closed-landfills, prevent the migration of liquid phase species from these sites towards the exterior environment. They are often used for the long-term storage of dangerous wastes. However, the majority of landfills are only partially closed, thereby allowing the collection and treatment of the leachate, or kept open, leading to the gradual migration and dispersal of the leachate within the immediate ecosystem (Warmer Bulletin 2000; Nikiema et al. 2004a; Zamorano et al. 2006). Sanitary landfills can receive and process, over the period of their active life, more than a million metric tons of wastes (Desideri et al. 2003; Zamorano et al. 2006; Spokas et al. 2006). For small cities and towns of less than 35,000 inhabitants, a municipal landfill of 20–30 m in depth is able to receive up to 200,000 m^3 of waste during its lifetime and is classified as a small landfill (Börjesson et al. 2001; Park et al. 2004). The choice of a suitable site must be the subject of quite detailed attention. Factors commonly taken into account are; the long term availability of the site, which will be devoted to this exclusive use over a period of at least 30 years; its geological stability and characteristics. The site must also be of suitable size, and be located as far as possible from both residential and commercial areas, though remaining of easy access and servicing (Gielecki 1997).

Wastes, after their arrival on the site, are dehumidified if necessary, and moderately compacted, generally using bulldozers, to reduce their density to values bordering on 0.7–0.9 m^3 per metric ton before storage (Warmer Bulletin 2000; Zamorano et al. 2006). At the end of each day's operations, the densified wastes are covered with an inert layer: e.g. compacted mineral material, such as clay soil, of about 0.15 m height, in order to control the harmful effects of waste's decomposition (such as odors) and losses, and to reduce the risk of fires. When an operational section of the site is completely filled, a final cover, com-

posed of 0.6–1.0 m of clay and 0.2–0.6 m of soil, is applied to isolate it. The goal of this operation is thus to limit and even prevent the infiltration of H_2O into the thus deposited wastes (Zamorano et al. 2006).

3 Biogas

3.1 Biogas composition

Once stored in landfills, wastes degrade biologically, thereby generating biogas (Popov 2005). This biogas contains mainly CH_4, a colorless and odorless GHG, explosive when its concentration lies in the range 5–15% v/v in air (Perry et al. 1997; Tagaris et al. 2003), and CO_2, able to cause respiratory problems when its concentration is greater than 0.5% for a prolonged exposure (Toutant 1994; Reginster 1999; Nikiema et al. 2004a). The CH_4 concentrations in biogas, as mentioned in the literature, generally vary from 30 to 70% v/v while the CO_2 concentration varies between 20 and 50% v/v (Humer and Lechner 1999b; Kallistova et al. 2005; Murphy and McCarthy 2005; Tsai 2006; Zamorano et al. 2006).

In the biogas, some sulfur compounds are present in small proportions (typically less than 0.2% v/v), such as hydrogen sulfide (H_2S), mercaptans and thiols. These are responsible for the unpleasant odors that often emerge from poorly maintained landfills and can cause to humans and animals nausea, illness and in extreme cases death (Ma et al. 1996; Reginster 1999; Warmer Bulletin 2000). The biogas also generally contains some chlorinated compounds (less than 40 ppmv), among which are vinyl chloride, dichloromethane and tetrachloroethylene, all carcinogenic for humans and animals (Brosseau and Heitz 1994; Reginster 1999; Warmer Bulletin 2000; Scheutz et al. 2000; Zamorano et al. 2006).

Biogas can also contain trace amounts of various VOCs (less than 70 ppmv), such as benzene, a carcinogenic compound, toluene and the xylenes. Hydrogen (H_2), a by-product of the waste decomposition, can also be found in biogas at small concentrations, < 0.2% v/v, along with nitrogen (< 5% v/v) and sometimes oxygen (O_2) (< 1% v/v) (Reginster 1999; Warmer Bulletin 2000; ZWA 2006). Moreover, biogas is generally water saturated (Warmer Bulletin 2000; Spokas et al. 2006). Even when all of these compounds are found in biogas of various origins, their concentrations can be very variable and depend on the type of the stored waste and the age of the landfill. Table 1 presents typical concentrations for several compounds generally found in biogas.

Table 1 Typical composition ranges for biogas produced in a landfill (Reginster 1999; Humer and Lechner 1999b; ZWA 2006; Tsai 2006)

Important compounds	
Percentage[a] (%)	
Methane	30–70
Carbon dioxide	20–50
Nitrogen	1–5
Oxygen	0.1–1.0
Ammonia	0.1–1.0
Sulfur compounds	0–0.2
Hydrogen	0–0.2
Carbon monoxide	0–0.2
Other trace compounds	0.01–0.6

[a] These concentrations are expressed on a dry weight basis

3.2 Biogas production

One metric ton of municipal waste can generate between 135 and 375 m^3 of biogas (Humer and Lechner 1999b; Warmer Bulletin 2000; Aye and Widjaya 2006; Murphy and McCarthy 2005; Zamorano et al. 2006). Many parameters influence the quantity and the rate of biogas production over time (Goossens 1996; Ozkaya et al. 2006). First, the age of the site is a determining factor in the production of biogas, due to commencement of waste decomposition, which can begin approximately 3 months after the waste storage installation and is subsequently spread over some 20–50 years (Bajic and Zeiss 2001; Zamorano et al. 2006). During the early years of a sanitary landfill's life (when it is being established and filled), the rate of generated biogas released increases rapidly, from 0 to 11 m^3 metric ton^{-1} $year^{-1}$ (Reginster 1999; Kumar et al. 2004) and thereafter, a slow and continuous decline in the gas emission follows. After some 30–50 years, rates of biogas production become very low and almost cease (Reginster 1999).

The rate of biogas production also depends on the waste bed internal temperature and, to a lesser extent, on the external climatic conditions, such as the ambient temperature (Kumar et al. 2004). The optimal temperature for the production of biogas is 35–37°C (Kettunen and Rintala 1997). The lowering of the temperature to 24°C in a controlled environment, such as within a digester, causes a reduction in the rate of biogas production of ~50% (Crill 1991; Nguyen et al. 2006). On the other hand, according to Chanton and Liptay (2000), variations in the production of biogas from an older landfill, as caused by seasonal temperature changes, are weak because the composting reactions of the organic wastes, located inside the deeper installed beds, ensures a near constant year round temperature of ~50°C (Straka et al. 1999; Hudgins and Green 2000).

Another important parameter is the waste's moisture content that should ideally remain between 50 and 60% wt/wt. This factor can be controlled during the wastes' initial compaction, i.e. just before their placement in the long-term storage. The wetter the wastes, the greater their rate of degradation. However, a waste bed that is excessively wet (i.e. more than 65% wt/wt moisture content) may cause settlement in the site material and produces substantial amounts of leachate needing to be handled. On the other hand, when wastes are not wet enough (less than 30% wt/wt moisture content), they degrade more slowly because the microbial activity is inhibited. Therefore, it results in an increase of the lifespan of the wastes. However, the mechanical stability of the landfill is good, reducing the risk of safety hazards generation (Reinhart and Al-Yousfi 1996; Warmer Bulletin 2000; Hudgins and Green 2000).

The type of waste stored in the landfill can also influence both the composition and the quantities of the generated biogas produced. Organic wastes produce a biogas principally containing CH_4 and CO_2, in contrast to synthetic wastes that can be practically inert, like glass, or introduce into the biogas specific substances such as H_2S, in the case of certain plastics degradation (Brosseau and Heitz 1994). Finally, the physical characteristics of the landfill, e.g. the bed depth, and its chemical characteristics, such as the pH, also play important roles in determining the production rate of the biogas. For maximum biogas production, the bed must be of sufficient depth to ensure that its interior regions provide for an anaerobic environment in which the relevant microorganisms can thrive, and the pH must also generally be close to neutral, i.e. between 6.8 and 7.2 (Yongzhi and Hu 2002; Kettunen and Rintala 1997).

3.3 Methane in the biogas

Methane, the atomically simplest and most stable hydrocarbon, is one of the important components in biogas. Its synthesis in organic waste beds is performed in three steps. Initially, polymers of the organic matter are hydrolyzed by the heterotrophic bacteria to form monomers. These molecules are then subject to fermentation which leads to the production of the organic and soluble products, composed mainly of acetates, formates and alcohols. By-products arising during this process step are CO_2 and H_2 (Le Mer and Roger 2001). These by-products are then converted to acetate in the presence of acetogenic bacteria, with simultaneous acidification, according to the following reaction:

$$2CO_2 + 4H_2 \rightarrow CH_3COO^- + H^+ + 2H_2O$$

All of these steps are strictly anaerobic. Acetate and other organic acids are then decomposed to CH_4 and CO_2 by the methanogenic microorganisms, all belonging to the domain Archaea (Hudgins and Green 2000; Le Mer and Roger 2001; Ozkaya et al. 2006). These microorganisms are strictly anaerobic (i.e. the tolerated dissolved oxygen concentrations do not excede the low micromolar range) and they are widely found in various environments such as anaerobic digestors, anoxic sediments, flooded soils and landfills. The acidification and methane generation steps are synchronized and mutualistic associations of microorganisms belonging to different genera are often observed at this late stage of methanogenesis, creating reciprocally favorable conditions, each moving the reaction equilibrium of the other in the most favorable direction (Whitman et al. 1999; Le Mer and Roger 2001).

3.4 Biogas valorization

Some landfills have active biogas collection systems (made as gas wells) but even in these cases, the quantities of recovered gases are usually only between 40% and 60% of the actually produced gas quantities (Humer and Lechner 1999a, b; Bajic and Zeiss 2001; Christophersen and Kjeldsen 2001; Popov 2005; Zamorano et al. 2006; Spokas et al. 2006). Newer more efficient techniques, including the use of synthetic cover materials, now allow for up to 90% gas collection effectiveness to be reached (Spokas et al. 2006). The biogas thus collected can subsequently be used in a variety of processes.

Combustion: This option is applicable only if the generated CH_4 concentration in the biogas and the overall biogas quantities are important, i.e. more than 30% (which occurs during the first 25 years of the landfill) and 50 m^3 h^{-1}, respectively (Reginster 1999; Bajic and Zeiss 2001; Streese et al. 2001; Haubrichs and Widmann 2006). The calorific value of biogas is typically around 20,000 kJ m^{-3}, i.e. about half that of the calorific value of natural gas and thus, the hot gases generated from biogas combustion can be best used as an energy source for the production of electricity and/or to generate hot water or steam (Goossens 1996; Desideri et al. 2003; Tsai 2006; Zamorano et al. 2006; Spokas et al. 2006). This valorization process allows at least, the partial meeting of the energy demand for the wastes processing site and for other clients located in its neighborhood. The investment cost required to install and operate such technology, considering a global collection and energy recovery efficiency of 50%, in a landfill, already equipped with biogas collection systems, is 3.1$ US/ton CO_2 equivalent of CH_4 eliminated (Ayalon et al. 2001). Estimates made by the Environmental Protection Agency (EPA) in 1996 indicated that the recovered energy from biogas, issued from the landfills across the whole USA, could be used to meet the needs of some 2.3 million homes (Goossens 1996). However, this solution is not universally economic at present because of the low cost of natural gas. Moreover, the addition of biogas to the natural gas network may deteriorate the quality and lifetime of the latter (Brosseau and Heitz 1994; Ewall 1999).

Other alternatives: A catalytic flow reversal reactor technology concept was developed by Natural Resources Canada (USDE 2005). The main goal of this process is the elimination of CH_4 when its concentration in air lies between the values of 0.1–1% v/v. The methane is oxidized in a packed bed reactor, the exit product gases having a temperature ranging from 600 to 800°C. Heat can then be recovered from it, either to produce electricity or to satisfy various local heating needs. Another alternative for the CH_4 content in biogas valorization consists of transforming this compound into methanol. This latter product can then be sold to chemical processors (Ewall 1999; Popov 2005).

3.5 Biogas elimination

Flaring: Sometimes, collected biogas is simply burned in flares. This CH_4 elimination method is done with minimal facilities and without energy recuperation, the objective being to avoid the risk of explosion caused by the presence of CH_4 in the air. However, this disposal method can be environmentally harmful, when dangerous compounds, such as dioxins, are generated during the combustion and are released to the atmosphere (Gielecki 1997; Jaffrin et al. 2003). Flaring of landfill biogas requires about 1.2 $ US/ton eq CO_2 of CH_4 eliminated (Ayalon et al. 2001). This treatment process can be used only when the amounts of biogas to be treated exceed 10–15 m^3 h^{-1}, while the biogas CH_4 concentration remains greater than 20% v/v (Haubrichs and Widmann 2006).

Biological oxidation: Many landfill installations are, even today, still deprived of collection systems for the biogas produced. And even where such systems are in place, it is still difficult, and usually uneconomic, to utilize traditional valorization techniques for the older or smaller landfills (Bajic and Zeiss 2001). In these cases, other processes may need to be used to eliminate the dangers created by the CH_4 presence in the atmosphere-released biogas. A possible solution is the use of biofiltration, a biological oxidation process. This idea comes from the fact that some

bacteria are able to degrade air pollution compounds, such as CH_4. This process already provides for the elimination of some 10–100% of the CH_4 escaping from the upper layers of landfills, depending on local climatic conditions (Nozhevnikova et al. 1993; Kightley et al. 1995; Czepiel et al. 1996; Chanton et al. 1999; Christophersen et al. 2000; Bajic and Zeiss 2001; EPA 2005; Stralis-Pavese et al. 2006).

4 Methane biofiltration

4.1 Configuration

A biofilter is a three-phase bioreactor: the filter bed constitutes the solid phase, the biofilm, the liquid phase and the gaseous pollutants, the gas phase. Contact between the microorganisms and the polluting CH_4 takes place in the biofilm, immobilized on the filter bed. The majority of biofilters, as used in lab-scale experiments, are closed systems. The air supply is ensured by a forced ventilation system. Gases circulation in the biofilter can be effected from either top to bottom or conversely. Closed biofilters are compact systems that can be assembled from several stages. Different performance parameters like Inlet load (IL), Elimination capacity (EC) and conversion (X) used in biofiltration are defined in Table 2. In a closed biofilter, maintaining the operational parameters unchanged is also a relatively easy practice, resulting in good performance, with CH_4 X values as high as 90% (Dammann et al. 1999; Streese et al. 2001; Gebert et al. 2001; Du Plessis et al. 2003; Nikiema et al. 2005). The biofilter can also be an open system generally organized within the landfill covers. Usually, in this case, the flow of the polluted gas in the bed proceeds upwards, while the O_2 diffuses from the ambient air into the bed (passive ventilation). The main disadvantage of this process lies in the difficulty of controlling the operational parameters, such as the temperature and moisture levels. Moreover, transfer of O_2 to the bed's lowest layers is a very important limiting factor for the overall performance (Kjeldsen et al. 1997; Gebert et al. 2001). For example, removal efficiencies of up to 60% can be obtained, when the empty bed residence times (EBRT) is at least an hour, with an open biofilter, installed on a landfill site (Du Plessis et al. 2003; Gebert and Groengroeft 2006a, b).

Laboratory-scale experiments, using a forced ventilation at the top of the biofilter in order to simulate the natural behavior of landfill covers, have been reported by several authors (Hilger et al. 2000a, b; Hettiaratchi and Stein 2001; Stein and Hettiaratchi 2001). The best EC obtained with this operational mode was achieved in the range of 325 and 400 g m^{-2} d^{-1} (Hettiaratchi and Stein 2001). The IL of CH_4 is another important parameter. Various ILs have been tested at the laboratory scale and are reported in the literature, as presented in Table 3, ranging from 200 to 1700 g m^{-2} d^{-1}. For an IL close to 300 g m^{-2} d^{-1}, a conversion of 50% was obtained, as against 100% when the IL was only of 186 g m^{-2} d^{-1} (Hettiaratchi et al. 2000). An experiment reported by Humer and Lechner (1999b) on a sandy soil bed, showed the same tendency. However, according to Humer and Lechner (1999b), a flow rate of too low value could lead to poor performance if the filter bed porosity is not high enough.

Table 2 Performance parameters used in biofiltration

IL: Surfacic inlet load (g m^{-2} d^{-1})	$IL = \frac{C_{(CH_4)in} \times Q}{S}$
IL: Volumetric inlet load (g m^{-3} d^{-1})	$IL = \frac{C_{(CH_4)in} \times Q}{V}$
X: Conversion (%)	$X = \frac{C_{(CH_4)in} - C_{(CH_4)out}}{C_{(CH_4)in}} \times 100$
EC: Elimination capacity (g m^{-2} d^{-1} or g m^{-3} d^{-1})	$EC = IL \times \frac{X}{100}$

Where $C_{(CH_4)}$: Methane concentration in g m^{-3}; Q: Volumetric flow rate of gases in m^3 d^{-1}; S: Biofilter bed cross-section in m^2; V: Biofilter bed volume in m^3

Table 3 Experiments on the topic of methane biofiltration

Filter bed	Operating conditions	Inlet load	Elimination capacit or conversion	Authors
Compost and soil	Aerated at the top	IL = 202 g m^{-2} d^{-1}	EC = 80–90 g m^{-2} d^{-1}	Bajic and Zeiss (2001)
Clay and landfill cover soil	Mixture 45% v/v CH_4, 45% v/v CO_2		EC = 40–50 g m^{-2} d^{-1}	
Soil and sand	Optimal water content for all experiments		EC = 15–20 g m^{-2} d^{-1}	
Soil			EC = 5–7 g m^{-2} d^{-1}	
Multi-layers: Compost + sand (top) and sand (0.9 m)	Aerated at the top Mixture 50% v/v CH_4, 50% v/v CO_2	IL = 288 g m^{-2} d^{-1}	EC = 164–283 g m^{-2} d^{-1}	Berger et al. (2005)
Agricultural soil	Aerated at the top Mixture 50% v/v CH_4, 50% v/v CO_2	IL = 214 g m^{-2} d^{-1}	EC = 171 g m^{-2} d^{-1}	De Visscher et al. (1999)
Landfill cover soil		IL = 368 g m^{-2} d^{-1}	EC = 240 g m^{-2} d^{-1}	
Compost of pine bark	Aerated at the bottom	IL < 420 g m^{-2} d^{-1} EBRT > 0.85 h	$X \geq 70\%$	Du Plessis et al. (2003)
Multi-layers(from top to bottom): humic topsoil (0.1 m) + sand (0.02 m) + gravel (0.02 m) + clay (0.67 m) + gravel (0.1–0.3 m).	Pilot-scale open biofilter	IL = 0–6,000 g m^{-3} d^{-1}	EC ≤ 1,900 g m^{-3} d^{-1} X = 62% (annual basis)	Gebert and Groengroeft (2006a)
Compost	Aerated at the bottom (3 air input level)	IL = 590 g m^{-2} d^{-1}	EC = 530–590 g m^{-2} d^{-1}	Haubrichs and Widmann (2006)
Recycling paper pellets	Mixture 30% v/v CH_4, 70% v/v CO_2	IL = 105 g m^{-2} d^{-1}	EC = 47 g m^{-2} d^{-1}	
Compost + recycling paper pellets		IL = 105–485 g m^{-2} d^{-1}	EC = 47 g m^{-2} d^{-1}	
Compost and wood chips		IL = 485 g m^{-2} d^{-1}	EC = 475 g m^{-2} d^{-1}	
Compost of leaves	Aerated at the top	IL ~ 500 g m^{-2} d^{-1}	EC = 325–400 g m^{-2} d^{-1}	Hettiaratchi and Stein (2001); Wilshusen et al. (2004)
Compost of municipal waste	Pure CH_4, 99% v/v		EC = 200–250 g m^{-2} d^{-1}	
Compost of garden residues			EC = 200–250 g m^{-2} d^{-1}	
Compost of wood ships			EC < 50 g m^{-2} d^{-1}	

Table 3 continued

Filter bed	Operating conditions	Inlet load	Elimination capacit or conversion	Authors
Peat	Aerated at the top Optimal water content	IL = 160–320 g m^{-2} d^{-1}	EC < 186 g m^{-2} d^{-1}	Hettiaratchi et al. (2000)
Soil 1 (Sand 70%, clay 15%, silica 15% wt/wt)	Aerated at the top Mixture 60% v/v CH_4, 40% v/v CO_2	IL = 95 g m^{-2} d^{-1}	EC = 62 g m^{-2} d^{-1}	Hettiaratchi et al. (2000)
Soil 2 (Sand 70%, clay 25%, silica 5% wt/wt)		IL = 345 g m^{-2} d^{-1} IL = 95 g m^{-2} d^{-1} IL = 435 g m^{-2} d^{-1}	EC = 121 g m^{-2} d^{-1} EC = 49 g m^{-2} d^{-1} EC = 87 g m^{-2} d^{-1}	
Landfill cover soil	Aerated at the top Water content: 15% wt/wt Mixture: 50% v/v CH_4, 50% v/v CO_2	IL = 281 g m^{-2} d^{-1}	EC = 125–140 g m^{-2} d^{-1} (Peak) EC = 42–55 g m^{-2} d^{-1} (Steady operation)	Hilger et al. (2000a, b)
Fresh compost	Aerated at the top Temperature: 18°C	IL = 135–170 g m^{-2} d^{-1}	X = 60% from day 25 to day 50	Humer and Lechner (1999b)
		Q = 4 –5 ml min^{-1}		
Mature compost			X = 100% after 55 days	
Soil			X = 100% after 15 days X = 30–40% from day 10 to day 50	
Compost of municipal waste	Aerated at the top Temperature: 18–20°C	IL ~ 235 g m^{-2} d^{-1}	EC = ~188 g m^{-2} d^{-1}	Humer and Lechner (2001)
Compost of clarification sludge		Q = 4 –7 ml min^{-1}	EC = ~175–190 g m^{-2} d^{-1}	
Soil			EC = ~82 g m^{-2} d^{-1}	
Landfill cover soil			EC = ~95 g m^{-2} d^{-1}	
Garden soil			EC = ~175 g m^{-2} d^{-1}	
Mature compost	In situ	2,400 L biogas m^{-2} d^{-1}	X = 100%	Hupe et al. (1998)
Inorganic material	Aerated at the bottom 7,000–7,500 ppmv CH_4	IL ~ 1,700 g m^{-2} d^{-1}	EC = ~ 700 g m^{-2} d^{-1}	Nikiema et al. (2004b)
Compost			EC = ~ 300 g m^{-2} d^{-1}	
Soil	Aerated at the top Pure CH_4, Optimal conditions	IL = 525 g CH_4 m^{-2} d^{-1}	EC = 435 g m^{-2} d^{-1}	Park et al. (2002)
Glass tubes	Aerated at the bottom Percolation 0.25–1.0% v/v CH_4	IL = 200 g CH_4 m^{-2} d^{-1} IL = 750 g CH_4 m^{-2} d^{-1}	X = 96–98% X = 27–28%	Sly et al. (1993)

Table 3 continued

Filter bed	Operating conditions	Inlet load	Elimination capacit or conversion	Authors
Peat	Aerated at the top	IL = 320 g m^{-2} d^{-1}	EC = 96–160 g m^{-2} d^{-1}	Stein and Hettiaratchi (2001)
Landfill cover soil	Pure CH_4 99% v/v	IL = 320 g m^{-2} d^{-1}	EC = 64–130 g m^{-2} d^{-1}	
Agricultural soil	Water content: 10% wt/wt	IL = 310 g m^{-2} d^{-1}	EC = 93–155 g m^{-2} d^{-1}	
Compost + landfill cover material	In situ open biofilter	IL = 18,500–42,800 g m^{-3} d^{-1}	X ≥ 90%	Straka et al. (1999)
Compost	Bench-scale open biofilter	IL = 288–3120 g m^{-3} d^{-1}	EC = 1,500 g m^{-3} d^{-1}[a]	Streese and Stegmann (2003)
Compost + peat + wood fibers			EC = 960 g m^{-3} d^{-1}	
Multi-layers			EC = 720 g m^{-3} d^{-1}	
Compost	Large-scale open biofilter	IL = 288–3120 g m^{-3} d^{-1}	EC = 960 g m^{-3} d^{-1}[a]	Streese and Stegmann (2003)
Compost + peat + wood fibers			EC = 480 g m^{-3} d^{-1}	
Soil 1	Aerated at the top Mixture 60% v/v CH_4, 40% v/v CO_2	–	EC = 40–100 g m^{-2} d^{-1}	Visvanathan et al. (1999)
Soil 2			EC = 75–100 g m^{-2} d^{-1}	

[a] This EC was the maximal value obtained. After 5 months of operation, the CH_4 oxidation rate in the biofilter decreased.

In the case of methane biooxidation, EBRTs are typically between a few minutes to several hours, due to methane's low-biodegradability (Dammann et al. 1999; Hettiaratchi and Stein 2001; Du Plessis et al. 2003; Nikiema et al. 2004b, 2005). In contrast, for VOCs and VICs biofiltration, EBRTs are in general, between 30 and 120 s (Jorio and Heitz 1999). The required operating volumes can reach as much as 100 times those used for treating the same amount of odors (Streese and Stegmann 2003). Indeed, the size of the biofilter should be at a scale of at least 1 m^3 of filter bed for achieving flow rates of CH_4 in the range of 0.01–2.5 $m^3\ h^{-1}$ (Straka et al. 1999; Stresse and Stegmann 2003; Haubrichs and Widmann 2006). The height of the open biofilters with passive ventilation, used for CH_4 elimination, must also be lower than 1 m (Kjeldsen et al. 1997; Boeckx and Van Cleemput 2000; Stein and Hettiaratchi 2001; Stein et al. 2001; Park et al. 2002; Tagaris et al. 2003). Open systems are usually less expensive, at least 15%, than closed systems. In 2001, for the non-easily degradable, volatile organic pollutants, the costs for the installation of open biofilters were between 0.25 and 0.4$ for each $m^3\ d^{-1}$ of polluted gas to be treated (we assume this cost will probably be similar to that for CH_4). In addition, the industry consensus on capital and operating costs must be considered, and recently, these costs were 0.5–1.8$ and 0.07–0.1$ per $m^3\ d^{-1}$ of polluted gas, respectively (Janni et al. 2001).

4.2 Microorganisms

4.2.1 Methanotrophs

The specific bacteria responsible for the decomposition of CH_4 are known as methanotrophs and constitute a sub-group of the methylotrophs, i.e. bacteria specialized in the degradation of those compounds having only one carbon atom. Earlier, methanotrophs were identified only according to their morphology, their intracytoplasmic membranes structure and some of their physiological characteristics. Since then, DNA analysis has aided the identification of the genera of methanotrophs (Hanson and Hanson 1996; Lidstrom 2001).

There are three basic steps in the decomposition of CH_4. The first reaction step consists of the oxidation of CH_4 to methanol, utilizing the enzyme MMO (Hanson and Hanson 1996; Auman and Lidstrom 2002). The methanol thus obtained is transformed into formaldehyde. The latter compound can be subsequently used in a dissimilatory pathway (i.e. being oxidized to CO_2, with formate as an intermediate) or via several types of assimilatory pathways, leading to the synthesis of cell components, necessary for the growth of methanotrophs (Hanson and Hanson 1996).

The genera of methanotrophs are grouped into three main types. The genera *Methylomonas*, *Methylomicrobium*, *Methylobacter*, *Methylocaldum*, *Methylophaga*, *Methylosarcina*, *Methylothermus*, *Methylohalobius* and *Methylosphaera* belong to type I. They assimilate formaldehyde by the ribulose monophosphate pathway and their cellular membranes are mainly made up of fatty acids with 16, or sometimes 14 atoms of carbon (Hanson and Hanson 1996; Tsubota et al. 2005, Kalyuzhnaya et al. 2005; Heyer et al. 2005; Stralis-Pavese et al. 2006). *Methylocystis*, *Methylocella*, *Methylocapsa* and *Methylosinus* constitute the type II and they use the serine pathway for their formaldehyde assimilation. Their cellular membranes contain fatty acids of 18 carbons, arranged around the cell periphery (Hanson and Hanson 1996; Börjesson et al. 1998; Dedysh et al. 2000; Dedysh et al. 2002; Nikiema et al. 2005). *Methylococcus*, known as type X, combines the properties of types I and II i.e. fatty acids with 16 carbons and the assimilation of formaldehyde through both the ribulose monophosphate cycle and the serine pathway. The recently completed genomic sequence of *Methylococcus capsulatus* confirmed the presence of genes directing both pathways (Hanson and Hanson 1996; Wise et al. 1999; Kelly et al. 2005). Aerobic methanotrophic bacteria are essentially unable to grow on substrates containing C–C bonds as the only carbon source and thus can be considered as obligate C_1 metabolizers. The genus *Methylocella* seems however to be an exception to this rule, being able to use compounds such as acetate, pyruvate, succinate, malate, and ethanol (Dedysh et al. 2005; Horz et al. 2005).

Methylococcus (type X), *Methylothermus* and *Methylocaldum* (type I) are moderately thermophilic and their optimal growth temperatures vary from 42°C, for the majority, to 62°C. *Methylomonas*, *Methylobacter* and *Methylosphaera*, all of type I, are psychrophilic, developing over a range of temperatures, from 5 to 15°C (Trotsenko and Khmelenina 2002). *Methylobacter*, type I bacteria, have an optimum growth temperature of around 6°C, while *Methylosphaera* develop better, between 10 and 13°C, in sea water (Berestovskaya et al. 2002). Mention is made that several methanotrophic communities have the capability of adapting to various temperatures, as long as these lie between 0 and 55°C. However, at temperatures lower than 0°C, the multiplication of the bacteria stops (Humer and Lechner 1999b). *Methylocystis* and *Methylosinus*, bacteria composing type II, are acidophilic. They exhibit a maximum growth rate in acidic media, in the pH range from 5 to 5.5. *Methylomicrobium* (type I bacteria) are distributed between the group of halophilic, being at ease in saline media having sodium chloride concentrations ranging from 0.5 to 5.6% wt/wt, and that of the alcaliphilic, for which the optimal pH ranges between 7.5 and 10 (Trotsenko and Khmelenina 2002).

Methane monooxygenase enzyme A specific enzyme known as methane monooxygenase or MMO characterizes the methanotrophs. The MMO is the key enzyme allowing methanotrophs to perform the decomposition of CH_4 (Hanson and Hanson 1996). This enzyme exists in two forms: particulate MMO (pMMO) and soluble MMO (sMMO). The pMMO enzyme can be both found in and synthesized by all methanotrophs, except *Methylocella*, but the sMMO is almost always present in bacteria of type II and X. However, some *Methylomonas* strains (type I), possessing the sMMO enzyme, have already been found (Auman and Lidstrom 2002).

It is known that methanotrophs containing pMMO (mainly type I) grow more rapidly and are more specific to CH_4 than those having the sMMO (type II and X) (Henckel et al. 2000; Reay and Nedwell 2004). These differences are noticed when the CH_4 concentration is lower than 1,000 ppmv of CH_4 (Segers 1998). Thus, type I bacteria with pMMO develop quickly when the experimental conditions permit and become dominant in environments when such rapid growth is allowed (Henckel et al. 2000). However, they are sensitive to variations in nutrients availability, mainly the nitrogen and copper, and in the CH_4 concentrations. On the other hand, populations of type II and X bacteria, having the sMMO, are quasi-steady and very stable in various environments, such as the landfill covers (Henckel et al. 2000; Crossman et al. 2004). In addition, sMMO also has affinities for a variety of compounds, such as methanol, several chlorinated compounds and hydrocarbons, among which are the alkanes, olefinic hydrocarbons and aromatic compounds (Hanson and Hanson 1996; Dunfield et al. 1999; Vorholt 2002; Hilger and Humer 2003; Erwin et al. 2005; Hesselsoe et al. 2005; Lindner et al. 2005).

Oxygen and carbon dioxide needs of methanotrophs All of the methanotrophs species can be found in small quantities in any environments exposed simultaneously to significant amounts of CH_4 and O_2 (Börjesson et al. 1998; Dammann et al. 1999). For example, *Methylomonas* and *Methylobacter* (type I), *Methylocystis* and *Methylosinus* (type II) as well as *Methylococcus* (type X) have already been isolated from the cover soils of several landfills (Börjesson et al. 1998). However, the distribution of methanotrophs within a filtering material is not a random process since each type of bacteria develops preferentially in that portion offering the most advantageous conditions for its growth (Henckel et al. 2000; Gebert et al. 2003). An O_2 concentration of 21% v/v, associated with a CH_4 concentration less than 1,000 ppmv better supports the growth of type I bacteria. On the other hand, when the CH_4 concentration is superior to 1% v/v and the concentration of O_2 is low (about 1% v/v), type II bacteria develop better (Hanson and Hanson 1996; Henckel et al. 2000; Crossman et al. 2004). However, there are exceptions to this scheme and some type I bacteria have their growth stimulated only in

the presence of an appreciable concentration of CH_4 (> 1% v/v), and correspondingly, a low amount of O_2 (< 1% v/v) (Henckel et al. 2000; Erwin et al. 2005). Bender and Conrad (1994), Czepiel et al. (1996) and Stein and Hettiaratchi (2001) have shown that, by increasing the O_2 concentration from 3 to 20% v/v in the gas mixture, the CH_4 conversion varies only slightly (less than 10%). However, a decrease of O_2 concentrations from 3 to 1% causes the fall off of CH_4 oxidation of more than 50%. However, during the experiments of Stein and Hettiaratchi (2001), the maximal CH_4 elimination was obtained at O_2 concentration between 0.75 and 1.6%.

The presence of CO_2 in a biofilter at the same time as the CH_4 modifies the behavior of the microorganisms present. According to Acha et al. (2002), the activity of the methanotrophs, using the serine pathway for the assimilation of formaldehyde obtained during the decomposition process of CH_4, requires some CO_2 input (partial pressure of CO_2 around 11.6 kPa) (Acha et al. 2002).

4.2.2 Non-methanotrophic bacteria

Nitrifying bacteria, responsible for the decomposition of ammonia (NH_3), can also degrade CH_4, but their performance rate is less than 5% that of the pure methanotrophic populations (Hanson and Hanson 1996; Bodelier and Frenzel 1999). Also, some bacteria involved in the decomposition of methanol are capable of degrading CH_4, but only if the CH_4 concentrations remain below 10% v/v. The optimal growth temperature for these bacteria is around 35°C (Hughes et al. 2002). There are also certain anaerobic bacteria that are able to degrade CH_4. Such bacteria are active when immersed in aqueous media. These bacteria work in tandem with those involved in reducing sulfates, the reaction requiring additional sources of carbon such as acetate or lactate (Hanson and Hanson 1996; Kotelnikova 2002; Valentine 2002). The minimal sulfate concentration in the system must be approximately 1 mmol l^{-1} (Segers 1998). The hypothesis of coupling between sulfate reduction and anaerobic methane oxidation is also supported by studies on a landfill-leachate plume (Grossman et al. 2002) and in ground water (Van Stempvoort et al. 2005). However, experiments to isolate these anaerobic bacteria remain unsuccessful to date (Conrad 1996; Segers 1998; Kotelnikova 2002). Recently, a microbial consortium has been isolated, found to be performing methane oxidation, coupled to nitrate reduction, in the absence of oxygen. The consortium includes two microorganisms: a bacterium and an archaeon, belonging to as yet unknown species (Raghoebarsing et al. 2006).

4.3 Inoculation and incubation

When contact is created between methanotrophs and CH_4 in a biofilter, an induction step, during which X is weak (0–10% of the steady state conversion), always precedes the optimal system functioning. This lag phase is due to the activation and growth of the methanotrophic bacteria (Bender and Conrad 1995; Henckel et al. 2000) and its duration is determined by the operating conditions (CH_4 concentration, temperature and moisture of the filter bed). During the experiments carried out by Henckel et al. (2000) in microcosms maintained under a CH_4 continuous flow environment, some 6 and 19 days were required to reach steady X, respectively for high (10,000 ppmv) and low (1,000 ppmv) CH_4 concentrations. In order to aid the establishment of the specific and competitive methanotrophic population in the filter bed, inoculation of the bed by selected methanotrophic bacteria is usually performed, even if the success of this practice is not guaranteed.

At the laboratory scale, another common practice involves incubation, consisting of a prolonged exposure (several days or weeks) of the filter bed to significant CH_4 concentrations, ranging between 1,000 and 200,000 ppmv. The higher the CH_4 concentration, the more the growth of the methanotrophs is promoted. The consequence then is a rapid increase in the oxidation rate (Bender and Conrad 1995; Hanson and Hanson 1996; Henckel et al. 2000; Le Mer and Roger 2001; Crossman et al. 2004; Mor et al. 2006). For example, the oxidation rate for a CH_4 at initial concentration of 100,000 ppmv is around 0.8 g

CH_4 kg $soil^{-1}$ d^{-1} which is 10 times higher than the value observed for a CH_4 initial concentration of 10,000 ppmv (Bender and Conrad 1995). Since all bacteria do not develop within the same range of CH_4 concentrations, the choice of the incubation parameters must be made judiciously. At the end of the induction phase, a peak value in the conversion up to 3 times that obtained for a steady operation (e.g. $X = 64\%$) can be noted (Hettiaratchi and Stein 2001; Abichou et al. 2006a).

4.4 Parameters

4.4.1 Filter bed

The filter bed is the solid phase on which the biofilm containing the microorganisms is to be formed. It must present sufficient space for the development of microorganisms and it should also have a texture providing a great moisture-holding capacity, in addition to appropriate bacteriological and mechanical properties. It must also be inexpensive (Humer and Lechner 1999a, b; Bajic and Zeiss 2001; Nikiema et al. 2004b). Various experiments, conducted at the laboratory scale, have been performed to test various filter bed structures, using natural materials such as soils and composts or synthetic materials. The results obtained are presented in Table 3 and will be expressed in terms of the IL, EC and X. Composts of various origins (solid wastes, vegetable wastes, clarification sludges...) were tested during the CH_4 biofiltration. Compost, made from mature yard wastes yielded the best results with EC up to 590 g m^{-2} d^{-1} and at values for X of between 90 and 100%, during more than 100 days of continuous filter operation (Haubrichs and Widmann 2006). Compost, made from dead leaves, also yielded good results (Hettiaratchi and Stein 2001; Wilshusen et al. 2004). In addition, the time required to reach 100%, conversion is less for the mature compost than that for freshly generated compost, being some 15 and 55 days respectively. This result makes the mature compost a preferred framework for the biofiltration of CH_4 (Humer and Lechner 1999b).

The soils most often employed are those of landfills covers (Hettiaratchi et al. 2000; Hilger et al. 2000a), but agricultural soils, soils derived from mountains, forests and rice plantations, peat bogs and swamps, have also been tested in CH_4 biofiltration (Dobbie and Smith 1996; Hütsch 1998b; Del Grosso et al. 2000; Hettiaratchi et al. 2000; Cai and Mosier 2000; Nozhevnikova et al. 2001; Stein and Hettiaratchi 2001; Novikov and Stepanov 2002; Kravchenko 2002). All of these soils contain different proportions of sand, clay, silica and organic matter. The most effective soils for CH_4 elimination are those taken directly from the upper layers of landfill covers. An EC of 435 g m^{-2} d^{-1}, corresponding to an X value of greater than 80%, has been reported in the literature (Park et al. 2002). The addition to a soil of organic residues, such as vegetable residues (beet leaves, wheat straw), clarification sludge or composts, can improve its CH_4 elimination. The EC values, reported from these modifications (100–200 g m^{-2} d^{-1}), correspond to some 40–100% of CH_4 conversion, and remain below the EC obtained during similar experiments with compost-based beds (Börjesson et al. 1998; De Visscher et al. 1999; Humer and Lechner 1999b; Park et al. 2002). The mean size of the soil particles must preferably lie between 0.5 and 2 mm (Bender and Conrad 1995; Kightley et al. 1995; Börjesson et al. 1998; Hettiaratchi et al. 2000; Min et al. 2002). Indeed, when particle sizes are less than 0.02 mm, the bed tends to become packed, preventing the effective diffusion of pollutants in the gas phase and then negatively affecting the conversion (Bender and Conrad 1995; Le Mer and Roger 2001; Min et al. 2002).

With either synthetic or inert filter materials, a few interesting results were obtained during the CH_4 biofiltration. An experiment, involving biofiltration by percolation with glass particles, has been reported (Sly et al. 1993). For a residence time of 20 min and an IL of around 200 g m^{-2} d^{-1}, more than 95% of CH_4 conversion was achieved. But the best EC reported in the literature is 700 g m^{-2} d^{-1}, obtained by Nikiema et al. (2004b) during their experiments with an inorganic-packed bed biofilter of 0.018 m^3, the gas flow rate being 6 m^3 d^{-1} and the CH_4 concentration maintained at between 7,000 and 7,500 ppmv.

4.4.2 Nutrients

Nutrients such as copper, nitrogen and phosphorus are strong determining factors for the success of CH_4 biofiltration, since they are necessary for the growth of the microorganisms (Trotsenko and Khmelenina 2002). These nutrients, unless already present in the filter bed in a bioavailable form, must be added to the solution used to humidify the filter bed (Nikiema et al. 2005).

Copper It has been shown that, while copper inhibits the sMMO enzyme at concentrations superior to 1μ mol l^{-1}, it supports the synthesis of the pMMO for concentrations between 1 and 5μ mol l^{-1} (Hanson and Hanson 1996). Thus, by adjusting the bed copper concentration, it is possible, in various cases, to develop a medium rich in bacteria of types I or II (Wise et al. 1999; Erwin et al. 2005). It has also been noted that, in adding around 0.02 g of copper, in the form of $CuCl_2$, per kg of paddy soil, CH_4 oxidation is slightly stimulated (an increase of around 5%) (Mohanty et al. 2000).

Nitrogen compounds Nitrogen element is an important nutrient for the methanotrophic bacteria. This element is usually provided to microorganisms in an inorganic form: e.g. nitrate (NO_3^-), ammonium (NH_4^+) or nitrite (NO_2^-) ions. Various tests have been performed and described in the literature to determine the influence of each of these compounds. Usually, they were undertaken with soils from various environmental sites, such as landfills, rice paddies, containing indigenous populations of microorganisms. The influence of NH_4^+ and NO_3^- seems to be variable (Hütsch 1998a, b; Bodelier and Laanbroek 2004; Reay and Nedwell 2004). The sources of NH_4^+ most frequently tested are ammonium chloride, ammonium sulfate and urea. For NO_3^-, sodium nitrate and potassium nitrate are the most studied. On some occasions, ammonium nitrate was used as a nitrogen source (Kightley et al. 1995; Hettiaratchi et al. 2000). Hettiaratchi et al. (2000) reported an example of improvement of CH_4 elimination by some 100%, following the addition of nitrogen (25 mg N per kg soil) in the form of NH_4^+ or NO_3^-. But, according to Chiemchaisri et al. (2001a), 30 mg N per kg soil or more, added in the form of NH_4^+ or NO_3^- inhibit the CH_4 elimination. In the case of NH_4^+, many authors also report the risk of competition between CH_4 and NH_4^+ when the latter was provided as a nitrogen source (Mancinelli 1995; Boeckx and Van Cleemput 1996; Humer and Lechner 1999b; Sitaula et al. 2000; Novikov and Stepanov 2002). Indeed, methanotrophs can convert NH_4^+ to NO_2^-. During the experiments conducted by Novikov and Stepanov (2002), 12–28% of the methanotrophic population was dedicated to a nitrification step instead of the CH_4 oxidation. In soils however, the decrease of CH_4 elimination rate was observed only after the nitrogen concentration reached 10–200 mg $N–NH_4^+$ kg $soil^{-1}$ (Bronson and Mosier 1994; Cai and Mosier 2000; Hettiaratchi et al. 2000; Novikov and Stepanov 2002; Park et al. 2004). But, the importance of this inhibition depends on the type of soil (Novikov and Stepanov 2002; Wang and Ineson 2003; Reay and Nedwell 2004) and can be further accentuated if other operating conditions, such as moisture content, are not satisfactory (Cai and Mosier 2000). Generally, an increase of the $N–NH_4^+$ concentration results in a higher percentage of inhibition at constant CH_4 concentration. Conversely, an increase of CH_4 concentration results in a lower percentage of inhibition at constant $N–NH_4^+$ content (De Visscher et al. 1999; Cai and Mosier 2000; Kravchenko 2002). Therefore, the inhibitory effect of NH_4^+ could be minimized if higher CH_4 concentrations were continuously provided to the filter media.

For the case of equal nitrogen supply, NH_4^+ will be less inhibiting than NO_3^- (Kravchenko 2002; Wang and Ineson 2003). But, according to Mancinelli (1995), NO_3^- instead of NH_4^+ is the preferred source of fixed nitrogen for the methanotrophs. Le Mer and Roger (2001) stated that the presence of NO_3^- can improve CH_4 elimination. Potassium nitrate has been used for the culture of methanotrophs since 1970 as a component of the ''nitrogen minimal salt'' (NMS) nutrient solution, which includes 0.14 g of $N–NO_3^-$ per liter (Whittenbury et al. 1970). During exper-

iments with an inorganic filter material, conducted by Nikiema et al. (2005), the authors noted that increasing nitrogen content supplied as sodium nitrate, from 0.14 to 0.75 g N l^{-1} in the nutrient solution, led to 5 times increase in the EC, from 130 to 700 g m^{-2} d^{-1}. However, a further increase of nitrogen content (> 0.75 g N l^{-1}) resulted in a decrease of the CH_4 oxidation conversion. During other experiments in soils, variations in the nitrogen supply between 25 and 100 mg N–NO_3^- kg $soil^{-1}$ did not show any noticeable influence on the biological elimination of CH_4 (Boeckx and Van Cleemput 1996; Park et al. 2002). A NO_3^- inhibition in soils was reported for high concentrations of around 2,500 mg N kg $soil^{-1}$ (Kumaraswamy et al. 2001).

Nitrite is well known as an inhibiting compound for methane elimination by methanotrophs (King and Schnell 1994; Mancinelli 1995; Boeckx and Van Cleemput 1996; Hanson and Hanson 1996). This compound can be generated when incomplete nitrification processes occurs in the filter media (Dunfield and Knowles 1995; Kravchenko 2002).

Sometimes, the inhibitory effect associated with the nitrogen content is otherwise caused by the salt effect. Indeed, the addition of salts containing inorganic nitrogen can change the overall ionic content of the soil (Hanson and Hanson 1996; King and Schnell 1998; Kravchenko 2002). Also, the influence of the nitrogen content is noticeable, especially in the case of low CH_4 concentrations, less than 100 ppmv (King and Schnell 1994).

Finally, it is important to mention that some methanotrophs are capable of N_2 fixation and express nitrogenase (Murrell and Dalton 1983; Kim and Graham 2001; Dedysh et al. 2002; Bodelier and Laanbroek 2004). Until recently, only Type II methanotrophs and Type X *Methylococcus* were thought to be capable of nitrogen fixation (Oakley and Murrell 1988; Dedysh et al. 2000). More recent work has revealed nitrogenase activity by the acetylene reduction route, and the presence of *nifH* genes generated by polymerase chain reaction amplification in a variety of methanotrophic species, belonging to both Types I and II (Auman and Lidstrom 2001; Boulygina et al. 2002). These data suggest that methanotrophs can play a significant role in nitrogen fixation in several natural environments, such as freshwater lakes (Zani et al. 2000). However the importance of N_2 fixation during biofiltration of methane remains to be assessed.

Phosphorus Generally speaking, phosphorus is of universal importance in promoting the growth of bacteria. However, despite its evident importance, it appears (from a close examination of the relevant literature) that only Kightley et al. (1995) have tried to obtain basic understanding of this element's contribution to the CH_4 biofiltration process. In their published studies, they have shown that the addition of a quantity of clarification sludge nutrient to an ordinary soil-based filter bed (final nutrient concentrations present in the soil: 0.1 g P per kg and 0.1 g N per kg) increased the rate of conversion of CH_4 by ~26%. On the other hand however, the addition of some 0.1 g of P–K_2HPO_4 nutrient per kg of the same soil did not result in any noticeable effect on promoting the CH_4 elimination property of the soil (Kightley et al. 1995; Hettiaratchi et al. 2000; Le Mer and Roger 2001). Thus, the role and activity of phosphorus, in the above described circumstances, remains unclear and further investigations will therefore be required to elucidate the mechanisms leading eventually to either the promotion of the bacterial growth or to its inhibition.

Other elements Potassium sulfate or manganese oxide increases the oxidation of CH_4 (Kumaraswamy et al. 2001). Addition of lime provides a soil-based bed with a neutral pH and thus appears to be interesting for CH_4 biofiltration (Hilger et al. 2000b). Excessive concentrations of sodium chloride and potassium chloride are both CH_4 elimination inhibitors (Cai and Yan 1999; Kravchenko 2002; Gebert et al. 2003), probably due to their osmotic effects.

4.4.3 Operating conditions

Temperature Methane oxidation is exothermic and, theoretically releases about 880 kJ per mole

CH_4. In case of bio-oxidation, the larger portion of this energy is used for the anabolic reactions during CH_4 biodegradation. The other portion is transferred to both the filtering material and to the mixture of gases that traverses it. The reaction heat released creates a temperature gradient in the biofilter, between its lower and upper surfaces (Humer and Lechner 1999b; Nikiema et al. 2004b; Nikiema et al. 2005). The significance of this thermal gradient depends on the input gas flow rate, the conversion, the type of filtering material and various other influential parameters. For example, a temperature change of around 4°C is noted for an inlet gases flow rate of 3.6 m^3 d^{-1}, when the volumetric EC in a compost-based biofilter is 840 g CH_4 m^{-3} d^{-1} (Streese et al. 2001). With an inorganic material, Nikiema et al. (2004b, 2005) did not observe any temperature gradient in the biofilter.

Tests on the influence of temperature during CH_4 biofiltration were conducted with common filter materials, such as soils and composts. In general, the optimal bed temperature is usually found to lie between 29 and 30°C for composts (Dammann et al. 1999; Streese et al. 2001; Mor et al. 2006) and between 25 and 36°C for soils (Whalen et al. 1990; Bender and Conrad 1995; Hanson and Hanson 1996; Boeckx and Van Cleemput 1996; Visvanathan et al. 1999; Cai and Yan 1999; Christophersen et al. 2000; Min et al. 2002; Mingxing and Jing 2002; Park et al. 2004). Apart from these intervals, the decrease in the conversion was important. For example, it fell by around 50% when the temperature was reduced from 30 to 20°C or from 29 to 24°C (Dammann et al. 1999; Streese et al. 2001). Between –5 and 10°C as the ambient temperature, the biological elimination of CH_4 in an opened biofilter system (landfill cover soil) is considerably decreased, i.e. more than 80% compared to the value at 15°C (Christophersen et al. 2000; Le Mer and Roger 2001). Therefore, the influence of temperature on the biological process constitutes the major limit for open biofilters, mainly during the winter season, when temperature falls to values lower than the limit that can be tolerated by the microorganisms consuming the CH_4 (Humer and Lechner 1999b).

On the other hand, if higher temperatures (>35°C) stimulate the activity of some methanotrophs, it should be noted that in such cases, the biofilter beds dry more quickly; this in turn leading to a decrease in the conversion rate (Visvanathan et al. 1999).

pH of the filter bed From a practical viewpoint, the pH of the filter bed is a parameter of lesser importance because the biodegradation of CH_4 does not generate intermediate or final products capable of influencing significantly the pH. The optimal pH values for the oxidation of CH_4 are in fact the same as those promoting the growth in the majority of methanotrophs bacteria. These are, in general, neutrophiles but they can, according to Hanson and Hanson (1996), tolerate pH values between 5.5 and 8.5. However, abrupt variations in the pH are adverse to methane elimination. A permanent inhibition was noted when the pH of the soil was changed by around 2 units, from 6.8 to 4.7 or from 6.8 to 9.0. This inhibition was partial for a unit variation, from 6.8 to 5.9 or 6.8 to 7.7, over the same operating conditions. This observation brought these present authors to propose a more restricted range of operating pH values, being that from 5.9 to 7.7 (Arif et al. 1996). In soil-based filter beds, the optimum pH ranges between the values of 6.7 and 8.1 (Bender and Conrad 1995) while for peat, the range lies between 5 and 6.5 (Le Mer and Roger 2001).

Filter bed moisture The filter bed moisture content is another key factor that determines the performance of the biofilter (Börjesson et al. 1998). When the moisture is too high, it acts as a rate-limiting factor by preventing the flow and transfer of CH_4 and O_2 (Humer and Lechner 1999b; Cai and Yan 1999; McLain 2000; Mingxing and Jing 2002; McLain et al. 2002; Park et al. 2002). The optimal filter bed water content depends on both the gas flow rate and the type of filter bed (soil, compost or other material employed) (Christophersen et al. 2000). Table 4 presents some typical water contents suggested in the literature. Optimal moisture content of soil materials (from the upper layers of landfills) ideally lies between 13 and 15.5% wt/wt, on a dry

Table 4 Optimal water content for some filter beds for methane elimination

Filter bed	Water content:% wt/wt	Authors
Compost	25–50	Humer and Lechner (1999b)
Landfill cover soil	13–30	Boeckx and Van Cleemput (1996), Park et al. (2002), Stein and Hettiaratchi (2001), Visvanathan et al. (1999), Giani et al. (2002)
Meadow soil	30–50	Mingxing and Jing (2002)
Woodland soil	18–33	Mingxing and Jing (2002)
Various soils	11–35	Bender and Conrad (1995), Christophersen et al. (2000)

basis (Whalen and Reeburgh 1996; Boeckx and Van Cleemput 1996; Chiemchaisri et al. 2001b; Stein and Hettiaratchi 2001; Jäckel et al. 2001; Park et al. 2002, 2004). However, Giani et al. (2002) reported a case for which the optimal moisture content of the landfill cover soil, used for biofiltration, was 25–30% wt/wt on a dry basis (at moisture values lower than 15%, the EC of CH_4 was reduced by 50% or more, compared to the maximal value). For composts or biological residues, optimal bed moisture lies between 25% and 50% wt/wt (Humer and Lechner 1999b). Methane conversion levels in soils that are less wet than the optimum level are lower than those attained in greater moisture content soils (Boeckx and Van Cleemput 1996; Cai and Yan 1999; Stein and Hettiaratchi 2001). Indeed, for a moisture content of around 745 g kg paddy soil^{-1}, i.e. approximately 265% of the optimal moisture (280 g kg paddy soil^{-1}), the conversion was only 24% of the maximum conversion. When the moisture content of the same material was changed to around 150 g kg soil^{-1}, the conversion fell to only 1% of that of the soil at its optimal moisture conversion (Cai and Yan 1999).

5 Landfill covers

Open biofilters are an attractive alternative for the older or smaller landfills, when gas collection systems cannot be installed for biogas valorization or elimination (Du Plessis et al. 2003; Berger et al. 2005). To our knowledge, there are no industrial applications related to the CH_4 biofiltration process in North America at the present time. However, at least the subjects of 3 patents registered worldwide, are more or less related to landfill biogas treatment, using in situ filters (Bergmann et al. 1998; Lee et al. 2002; Contec and Landkeis 2004). Landfill covers that permit a natural biological elimination of CH_4 could be considered as natural open biofilters. These covers are usually made of soils, sand or clay and represent the daily and final cover of the wastes in the landfill. Methane elimination in such covers is caused by the presence of methanotrophic populations. The behavior of a landfill cover is similar to that of an open biofilter equipped with passive aeration, except that the IL is usually low. Indeed, the mean IL of CH_4 in landfills covers generally lie between 50 and 340 g CH_4 m^{-2} d^{-1} (Jones and Nedwell 1993; Bogner et al. 1997; Humer and Lechner 2001; Perera et al. 2002; De Visscher and Van Cleemput 2003; Park et al. 2004; Gebert and Groengroeft 2006a; Abichou et al. 2006a, b). Börjesson and Svensson (1997) have noted that diurnal CH_4 emissions are up to 100% higher than the daily values, depending on the ambient temperature and air pressure. Also, CH_4 fluxes are themselves very variable and are usually not evenly distributed (Börjesson et al. 1998; Segers 1998; Gebert et al. 2001; Gebert and Groengroeft 2006a). Important rates of irrigation of the biofilter bed by rain may cause a decrease in the EC, up to 40%, by preventing the flow of biogas (Berger et al. 2005; Horz et al. 2005). On the other hand, even with a well constructed collection system, leaks always exist in such landfill covers, leading to the development of very important levels of emission in certain zones, up to 9,000 g CH_4 m^{-2} d^{-1} (Maurice et al. 1999; Chanton and Liptay 2000; Bajic and Zeiss 2001; Spokas et al. 2006). The covering of the whole landfill with a 0.1–0.6 m layer of mulch or compost helps to avoid uncontrolled CH_4 emissions from older landfills, when IL < 90 g m^{-2} d^{-1} (Chanton and Liptay 2000; Mor et al. 2006).

The performance of the landfill cover in the treatment of CH_4 is influenced by two main parameters: the temperature and the available

oxygen concentration. During winter, the CH_4 conversion within landfill covers is reduced to around 3–10% (Chanton et al. 1999; Chanton and Liptay 2000; Giani et al. 2002; Spokas et al. 2006). However, at an ambient temperature of 2°C, Christophersen et al. (2000) have noted that it was still possible to biodegrade all of the CH_4 produced in older landfills if IL is inferior to 70 g m^{-2} d^{-1}. Indeed, the microbial activity, combined with the isolation effect of the bed, contributes to keeping the inner bed layer at temperatures 5–8°C higher than the ambient temperature (Berger et al. 2005). In summer, the CH_4 conversion can reach 50% or more (Börjesson et al. 1998; Chanton et al. 1999; Chanton and Liptay 2000; Perera et al. 2002; Spokas et al. 2006). On the other hand, the diffusion of atmospheric O_2 is limited and generally, an oxygenated zone of only 0.6–0.8 m is observed (Nozhevnikova et al. 1993; Börjesson and Svenson 1997; Klusman and Dick 2000; Christophersen and Kjeldsen 2001; Chiemchaisri et al. 2001b; Perera et al. 2002; Tagaris et al. 2003; Crossman et al. 2004; Kallistova et al. 2005).

The landfill cover height must be at least 0.7 m for achieving best results (Giani et al. 2002). In order to reduce the influence of temperature, and the problems related to O_2 diffusion, on the landfill covers and also for open biofilters, many authors have favored the use of multi-layer beds (Bajic and Zeiss 2001; Streese and Stegmann 2003; Berger et al. 2005). For example, at the lowest bed level (0.25–0.9 m above entry point), a material, with the mean porosity such as soils or sand, is provided. This layer is employed for the retention of the filter bed humidity, in order to avoid quick bed drying events. The most important part of the overall CH_4 elimination process (typically 60%) will take place in the second layer, made of compost, for example (Bajic and Zeiss 2001; Berger et al. 2005). On a landfill site, the use of composts of ~0.3–0.6 m deep, instead of soils as an oxidation layer, can double the overall CH_4 elimination because of the availability of nutrients for the bacteria, while the higher porosity level leads in turn to a more satisfactory diffusion of the O_2 uptake (Hilger and Humer 2003). A third layer may be used at the top of the biofilter as a heat retention blanket, which will provide a particularly important practical feature to the biofilter when the atmospheric temperature falls during the winter season (Straka et al. 1999; Kallistova et al. 2005).

6 Conclusion

An important source of GHG emissions is that related to methane contained in biogas and released from sanitary landfills. In the present paper, a brief review of the composition, the production and valorization of the biogas is described. When this valorization is not possible, an alternative treatment lies in the biofiltration remediation of CH_4 emissions, particularly from older and smaller landfills. The main part of this paper focuses on this biotechnology.

The biofilter can be either an open or a closed system, equipped with either an active or a passive oxygen feed system. The use of open systems, whilst being more financially interesting, can also permit methane conversions of 60% and even more in specific cases, even if control of the process operational laboratory scale parameters is barely feasible. But, to our knowledge, there is no application in North America for this landfill technology. However, landfill covers play the role of a natural biofilter and eliminate up to 320 g CH_4 m^{-2} m d^{-1} On the other hand, closed systems are often more compact and provide for the better management and control of the operational parameters. The bed volume required for CH_4 control in a biofilter is at least 1 m^3 bed for a CH_4 gas flow in the range of 0.01–2.5 m^3 h^{-1}.

The majority of authors appear to agree on the point that matured compost constitutes a satisfactory filter material for supporting the biofiltration of CH_4. Indeed, both the presence of nutrients in the compost, in addition to its physical properties supports the growth of methanotrophs. The filter bed optimal temperature appears to lie between 29 and 30°C, and its optimal moisture level is found to lie between 25% and 50% wt/wt, on a wet basis. However, interesting results could also be obtained when using an inorganic-based bed biofilter.

Methane biofiltration is, at the same time, both a simple and a complex process. Indeed, even if the overall phenomenon of the reaction seems to be well known, many aspects are still misunderstood and contradictory theories are proposed, especially in relation to reaction optimization like nutrients, for long-term operations.

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