

# **Quantification of landfill gas emissions in biocovers – an experimental simulation in lysimeters**

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Marlies Hrad  
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# KURZFASSUNG

Autor: Marlies Hrad

Thema der Diplomarbeit: Quantifizierung von Deponiegas in Methanoxidationssystemen („biocovers“) – eine experimentelle Simulation in Lysimetern

Im Rahmen des Forschungsprojektes „Nutzraum“ („**IN**novative **In-SitU** **Me**thoden **Zur** **Sanie**rung von **Alt**ablagerungen **Und** **konta**minierten Standorten“) wurden Groß-Lysimeterversuche am Standort Seibersdorf (AIT Austrian Institute of Technology) konzipiert und errichtet. Ziel dieses Projektes ist es, einen optimalen technischen Aufbau einer Deponieabdeckschicht, die gleichzeitig als Methanoxidations- und Wasserhaushaltsschicht dient, für das Verfahren der In-Situ Aerobisierung zu finden. Insgesamt wurden drei Methanoxidationssysteme („biocovers“) mit unterschiedlichen Substraten zur Forcierung der methanoxidierenden Prozesse sowie eine Kontrollschicht parallel in Lysimetern eingebaut.

Inhaltlich beschäftigt sich die vorliegende Diplomarbeit mit der relativen Emissionsminderung der passiven Deponieabdeckschichten vor der In-Situ Aerobisierung. Die Oxidationsleistung der Abdeckschichten wurde anhand von Gaskonzentrations- und Temperaturprofilen sowie mithilfe von Haubenmessungen und oberflächennahen FID-Vermessungen bestimmt. Darüber hinaus wurde an ausgewählten Messtagen die Oxidationsleistung basierend auf der Methode der Isotopenfraktionierung quantifiziert. Die zum Einsatz gekommene Messhaube wurde eigens für dieses Projekt konstruiert und beruht auf dem Prinzip der geschlossenen, dynamischen Kammermethode. Die Schwierigkeit bestand darin, die bestehende Belüftungslanze (D = 6 cm) in das Design der Messeinrichtung zu integrieren um die gesamte Lysimeteroberfläche (4 m<sup>2</sup>) abzudecken und dabei die Messungen nicht zu beeinträchtigen. Die Eignung des Messsystems wurde für bekannte Methan- und Kohlendioxidströme zuvor im Labor validiert.

Aus den Ergebnissen geht hervor, dass nur bei mineralischen Abdeckschichten (Lysimeter C und D) Methanemissionen nachgewiesen werden konnten, wohingegen bei Abdeckschichten aus reifem Klärschlammkompost bzw. einer Sand-Kompost-Mischung (Lysimeter A und B) keine Methanaustritte detektiert wurden. Die Abdeckschichten wurden direkt mit Deponiegas aus den darunterliegenden Abfallkörpern versorgt, wodurch eine Kontrolle der Methanfracht nicht möglich war. Des weiteren ist zu beachten, dass die Methanbildung in den Abfallschichten sehr unterschiedlich verlief und es zu keiner einheitlichen Gasbildung in den Lysimetern A, B, C und D kam. Die Isotopenfraktionierung sowie die Gaskonzentrations- und Temperaturprofile von Lysimeter A und B weisen darauf hin, dass ein Großteil der Methanfracht bereits in der Gasverteilungsschicht und im Abfall selbst reduziert wurde. Die Abdeckschicht aus Klärschlammkompost in Lysimeter A zeigte aber grundsätzlich vielversprechende Ergebnisse im Bezug auf die Umgebungsbedingungen für methanotrophe Bakterien. Im Vergleich zu den anderen Abdeckschichten, konnte im Klärschlammkompost während des gesamten Untersuchungszeitraumes ein optimales Feuchte- und Temperaturmilieu für die Methanoxidation gehalten werden.

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# ABSTRACT

Author: Marlies Hrad

Topic of the thesis: Quantification of landfill gas emissions in biocovers – an experimental simulation in lysimeters

Within the framework of the joint project on remediation of old sites and dumps (“Nutzraum”), a lysimeter experiment was set up at the Austrian Institute of Technology in Seibersdorf, Austria, in February 2008. The project aims to derive design criteria for optimised composition of landfill covers regarding water balance and methane emission mitigation, in combination with in-situ aeration. Therefore, three experimental CH<sub>4</sub> oxidation biocovers using different substrates to enhance methane uptake as well as one control cover have been installed in parallel in lysimeters.

The diploma thesis presents data concerning the relative emission reduction effect of the passive biocovers prior to in-situ aeration. The methane oxidation efficiency of the biocovers was determined by gas composition and temperature profiles measured within the lysimeters, and by surface flux measurements as well as surface methane concentration screenings. In addition, methane oxidation was quantified at selected dates using stable isotope methods. The surface flux measurements were conducted using a closed dynamic chamber, which was constructed specially for this project. The challenge was to include the aeration pipe ( $D = 6\text{ cm}$ ) of the lysimeter into the design of the accumulation chamber in order to cover the whole lysimeter surface ( $4\text{ m}^2$ ) without disturbing the measurement procedure. Laboratory tests were conducted to verify the performance and accuracy of the measurement system with different known methane and carbon dioxide fluxes.

Among the four biocovers studied, CH<sub>4</sub> emission fluxes could only be detected from the biocovers with mineral soil covers (lysimeters C and D), whereas no CH<sub>4</sub> emissions were measured on the biocovers with mature sewage sludge compost and sand/compost mixture, respectively (lysimeters A and B). The biocovers were fed with biogas directly from the waste mass; therefore, it was not possible to control the upward biogas flux. It should be noted that methane production developed differently in the solid waste layers and methane load of lysimeters A and B did not indicate the same range as in lysimeters C and D. Stable isotope analyses as well as gas composition and temperature profiles of lysimeters A and B revealed that methane was reduced in the gas distribution system and in the waste itself. However, the mature sewage sludge compost (SSC) placed in lysimeter A showed, in principle, very promising results regarding the optimal ambient conditions for methanotrophic bacteria. In contrast to the other biocovers, the SSC-cover was capable of retaining the moisture content and the temperature profiles at an optimum level during the investigation period.

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# 1 INTRODUCTION

## 1.1 Motivation

Methane ( $\text{CH}_4$ ) is an important greenhouse gas (GHG), being 25-fold more radiatively active than carbon dioxide ( $\text{CO}_2$ ) over a 100-year period (IPCC, 2007a). Landfills are regarded as a major source of anthropogenic methane emissions, globally releasing between 30 and 70 Tg  $\text{CH}_4$  per year (Reay et al., 2007) or 10 – 20 % of the total global annual anthropogenic  $\text{CH}_4$  emissions (IPCC, 2007b). In developed countries landfill  $\text{CH}_4$  emissions have been stabilised during the last 20 years as a result of increased landfill  $\text{CH}_4$  recovery, decreased landfilling and waste generation, increased composting of source-separated bio-waste as well as the tendency towards either thermal or mechanical-biological pretreatment of municipal solid waste (MSW) (Bogner et al., 2007). For example, due to the implementation of the EU Landfill Directive (CEC 1999) landfilling of organic waste in Europe has progressively been reduced. On the other hand, methane generation from landfill sites is likely to rise in future as a result of rapid increase in population and urbanisation in developing countries (Bogner et al., 2007) (see Figure 1.1).

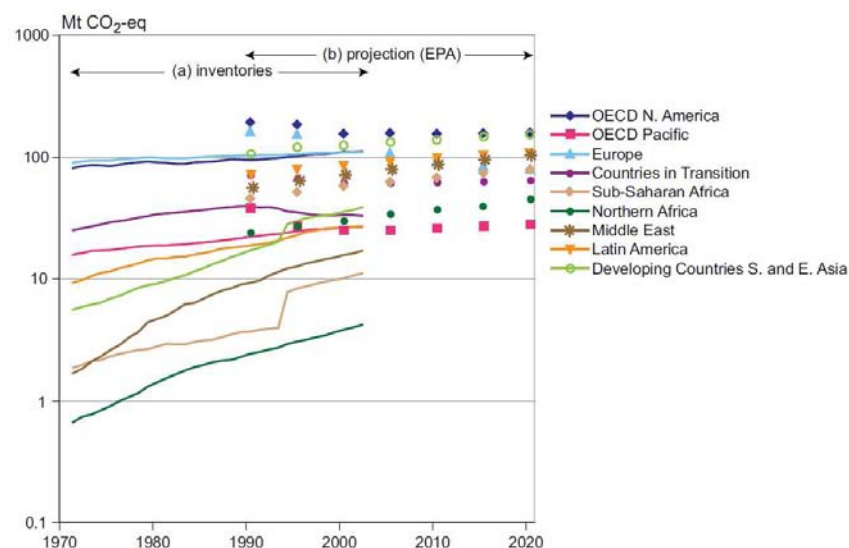


Figure 1.1: Comparison of regional emissions estimates for five-year intervals from 1990-2020 to annual historical estimates from 1971 – 2002 (from Bogner et al., 2007)

Depending on the landfilling technology applied, the waste and site characteristics as well as the environmental conditions, landfill gas will continue to be generated for periods between 15 and 30 years after final depositions of the waste (Williams, 2005). However, low-level gas production may continue up to 100 years after waste emplacement (Huber-Humer et al., 2009; Williams, 2005). Figure 1.2 shows the time-dependent methane production and recovery over a landfill lifetime. Engineered landfills with landfill gas (LFG) utilisation and control systems lead to considerably reduced  $\text{CH}_4$  emissions and recovery of energy. According to Haubrichs and Widmann (2006) and Mosher et al. (1999) gas recovery systems can reduce about 80 – 90 % of  $\text{CH}_4$  emission to the atmosphere depending on the boundary conditions of each landfill and its gas collection and control system. In contrast, Humer and Lechner (1999a) reported that in practice only 40 to 60 % of the landfill gas can be

controlled over the entire landfill lifetime by a gas recovery system as emission may also escape preferentially from and around wells and along the routes of installed landfill equipment.

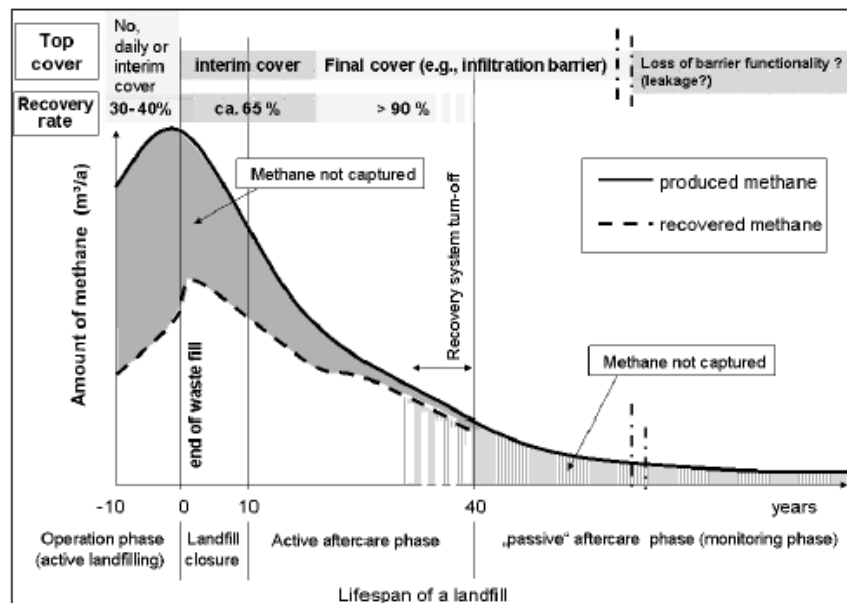


Figure 1.2: Time-dependent methane production and recovery over a landfill lifetime. Methane emissions (methane not captured) are shown as a function of cover type and do not include methane oxidation removal (from Huber-Humer et al., 2008b)

In the long term recovery facilities are not economically and technically suitable as only high methane concentrations ( $> 45\%$ ) can be used for heat and power production (Haubrichs and Widmann, 2006). Once  $\text{CH}_4$  concentrations reach  $20 - 25\%$  and total gas production rates are  $10 - 15 \text{ m}^3/\text{h}$ , the gas is flared with risk of producing toxic combustion products, or is just escaping to the atmosphere (Scheutz et al., 2009; Huber-Humer et al., 2008b). In addition, installation of a gas to energy system at old and small landfills is technically difficult and in most cases not feasible as LFG emissions are too low for energetic use (Damman et al., 1999).

Microbial methane oxidation, as a natural biogeochemical process, seems to be a preferable option to reduce, in particular, low-level  $\text{CH}_4$  emissions with a simple, passive landfill cover system. A considerable amount of methane can be biologically oxidised in the landfill cover to carbon dioxide, which has a lower global warming potential than methane. In recent years, specialised landfill “biocover” designs engineered to optimise  $\text{CH}_4$  oxidation have demonstrated huge potential to mitigate residual landfill  $\text{CH}_4$  emissions (Stern et al., 2007; Barlaz et al., 2004; Huber-Humer, 2004). Engineered biocovers usually consist of a coarse gas distribution layer to homogenise and distribute landfill gas to the overlying substrate layer. Such landfill cover systems can be potentially applied in the absence of a gas collecting system or as a complementary strategy for the control of  $\text{CH}_4$  emissions escaping gas collection, as well as to mitigate residual emission from mechanically and biologically pretreated (MBT) waste or old and small landfills with low  $\text{CH}_4$  production potential. Moreover, biocovers can also be combined with forced in-situ aeration as a postclosure remediation method for closed, old landfills. Importantly, biocovers offers a low-cost alternative for methane reduction especially required in developing countries.



The Austrian joint project on remediation of old sites and dumps (“Nutzraum”<sup>1</sup>) aims to derive design criteria for optimised composition of landfill covers in combination with in-situ aeration regarding water balance and emission mitigation. Within this framework, three experimental CH<sub>4</sub> oxidation biocovers using different substrates to support growth of methanotrophic bacteria as well as one control cover have been installed in parallel in lysimeters. The lysimeters, each measuring 2 m x 2 m x 3 m, consisted of 1 m thick substrate layer underlain by a 0.2 m gas distribution layer and 1.5 m layer of fresh municipal solid waste. In contrast, the “control” lysimeter included only a 0.5 m substrate layer (see section 3.3.1).

This diploma thesis covers only a part of the above mentioned project and presents the evaluation of the passive biocover performances under natural conditions by monitoring of the relative emission reduction effect. Therefore, the remaining methane and carbon dioxide emissions are determined and compared to the control cover.

## **1.2 Tasks and Objectives**

The present diploma thesis aims to assess the efficiency of different passive biocovers in reducing methane emission before the underlying waste is actively aerated for rapid waste stabilisation. Therefore, the residual landfill gas emission will be quantified. Further subgoals are:

- the development of a measuring system to quantify CH<sub>4</sub> and CO<sub>2</sub> surface emission from the different landfill covers in the lysimeters
- monitoring the development of the LFG generation and remaining CH<sub>4</sub> and CO<sub>2</sub> surface emission as well as CH<sub>4</sub> oxidation in the cover

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<sup>1</sup> see more information at [www.wau.boku.ac.at](http://www.wau.boku.ac.at)

## 2 BACKGROUND INFORMATION

### 2.1 Landfill gas generation

Landfill gases (LFG) are produced during the microbial anaerobic decomposition of waste containing degradable organic components. The biological decomposition of one tonne of deposited municipal solid waste produces roughly 160 to 250 m<sup>3</sup> of landfill gas (Abichou et al., 2004; Humer and Lechner, 1999). These gases are comprised typically of approximately 55 vol. % CH<sub>4</sub>, 44 vol. % CO<sub>2</sub> and numerous trace gases including hydrogen sulphide (H<sub>2</sub>S) and volatile organic compounds (VOC) resulting from microbiological as well as physico-chemical processes. Hence, one tonne of deposited municipal solid waste results in approximately 88 to 138 m<sup>3</sup> of CH<sub>4</sub>. However, the generation of landfill gas depends heavily on the landfilled waste volume, the solid waste properties, waste age, landfill operation and water content of the landfill waste (Steinlechner et al., 1994; Scheutz et al., 2009).

#### 2.1.1 Waste degradation processes

Waste degradation in landfills is a multi-stage process and can be divided into five main stages (Williams, 2005). Every stage is accomplished by a certain group of bacteria and has different requirements regarding pH and temperature. Figure 2.1 shows the different degradation steps and corresponding by-products during the decomposition of biodegradable waste. Until the waste has reached the final stage, the different stages may be proceeding simultaneously because of the heterogeneous nature of waste (Williams, 2005).

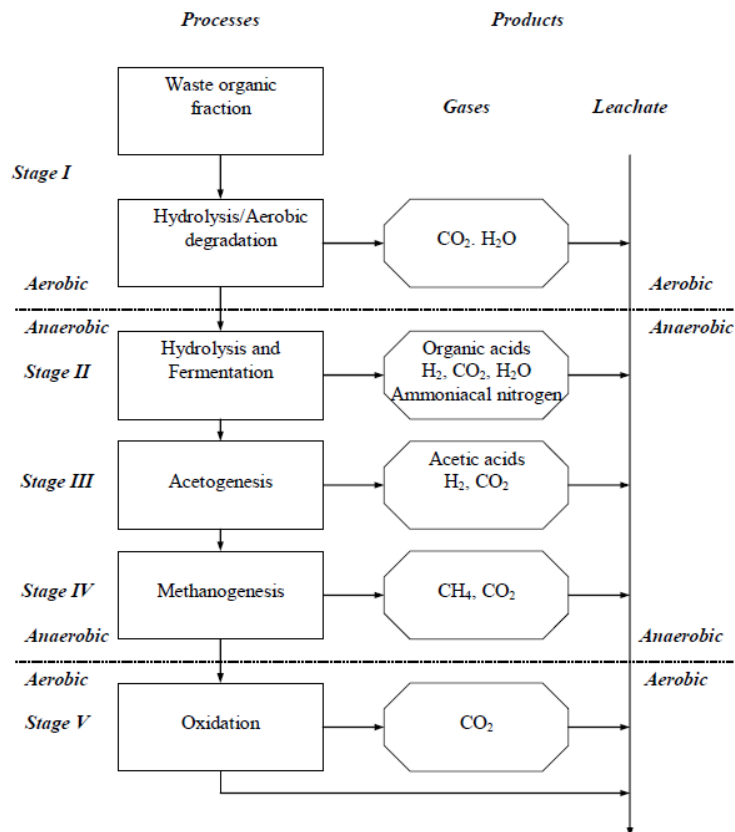


Figure 2.1: Major stages of waste degradation in landfill (Williams, 2005)

Aerobic degradation in the landfill occurs during the emplacement of waste and for a period thereafter. In this stage, aerobic bacteria, which only live in the presence of oxygen, are responsible for decomposition of the waste organic fraction. Thereby readily degradable organic matter is decomposed to CO<sub>2</sub>, water and heat (Steinlechner et al., 1994). CO<sub>2</sub> may be released as a gas or is transformed to carbonic acid (H<sub>2</sub>CO<sub>3</sub>) when it dissolves in water. However, the aerobic stage lasts only for a short time (days or weeks) (Williams, 2005), as the ability of oxygen to diffuse into the waste mass is lost within the waste body. The predominant part of the landfilled waste will soon become anaerobic.

During the second stage, carbohydrates (cellulose, starch and sugar), lipids and proteins are hydrolysed to glucose, amino acid, fatty acids etc. by means of extra-cellular enzymes, which are produced by facultative and obligatory anaerobic bacteria (fermenters). The hydrolysis process is a very important process in the landfill environment, as the solid organic compounds must be solubilised before the microorganisms can convert them. The resulting dissolved organic fragments are then fermented to CO<sub>2</sub>, hydrogen (H<sub>2</sub>), ammonia and organic acids like butyric acid, propionic acid, formic acid, valeric acid, etcetera. As ammonia largely derives from the deamination of proteins, the leachate contains high concentrations of ammoniacal nitrogen.

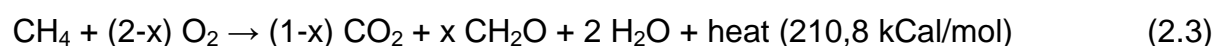
In the acidifying stage, organic acids formed in the second stage are transformed into acetic acid (CH<sub>3</sub>COOH), CO<sub>2</sub> and H<sub>2</sub> by acetogen microorganisms. Some bacteria are also able to directly decompose carbohydrates to acetic acid in the presence of CO<sub>2</sub> and H<sub>2</sub>. During the anaerobic stages, hydrogen sulphide may also be produced by the reduction of sulphate compounds by sulphate-reducing microorganisms. Due to the presence of organic acids, a pH level of 4 or even less is generated (Williams, 2005).

During the fourth stage, which is also the longest time stage, CO<sub>2</sub>, H<sub>2</sub> and acetic acid are consumed by methanogenic microorganisms in the following way:



About 70 % of the methane produced originates from splitting up of acetic acid although the responsible bacteria are the most sensitive ones in the whole process (Le Mer and Roger, 2001; Jeris and McCarty, 1965). During the methanogenic stage LFG can be generated over a temperature range of 30 – 65 °C as two classes of microorganisms (mesophilic and thermophilic) are active (Williams, 2005). Ideal for methanogenic microorganisms is a pH range from 6.8 to 7.5. However, microbial activity seems to continue down to pH 5 and up to pH 9 (Williams, 2005).

In the final stage, new aerobic microorganisms slowly replace the anaerobic forms as the acids are used up in the production of the LFG (Williams, 2005). Methanotrophic microorganisms utilise CH<sub>4</sub> as their only source of carbon and energy. While some of the CH<sub>4</sub> is consumed for energy yield gained from its oxidation to CO<sub>2</sub>, another fraction is incorporated into biomass. The exothermic process is catalysed by enzymes (see section 2.3) and can be simplified to the following stoichiometric equation:



where x is the fraction of carbon that is assimilated into biomass (CH<sub>2</sub>O). Methane oxidation causes a net decrease in the number of gas molecules as two molecules of

oxygen are consumed per oxidised methane molecule. The resulting volume reduction enhances the ability of oxygen to diffuse deeper into the waste mass (Huber-Humer et al., 2008a). Under aerobic conditions also hydrogen sulphate may be formed in waste with high concentration of sulphate.

### 2.1.2 Landfill gas composition over time

The gas composition changes with each of the five phases of degradation between different landfills (see Figure 2.2). The first aerobic phase continues until available oxygen is depleted. Depending on how much oxygen is present during the emplacement of waste in the landfill, the first stage lasts only for a short period of time. In the hydrolysis and fermentation stage carbon dioxide is the main constituent of landfill gas, which may rise to levels up to 80 %. The presence of  $\text{CO}_2$  and  $\text{H}_2$  reduces the content of nitrogen ( $\text{N}_2$ ).  $\text{H}_2$  and  $\text{CO}_2$  levels start to decrease throughout the third stage, as methanogenic bacteria start to grow producing  $\text{CH}_4$ . The anaerobic methanogenic phase is characterised by a fast increase in the production of  $\text{CH}_4$  followed by an extended period where gas production is relatively stable. This stage is the main LFG production phase, with typical LFG composition of approximately 55 %  $\text{CH}_4$  and 44 %  $\text{CO}_2$  and low concentrations of  $\text{H}_2$ . The maximum of LFG production is reached when the majority of degradable refuse will decompose (Thomas and Ferguson, 1999). The methanogenic stage of a landfill site is typically established within a half to one year and can last a few years or decades, depending upon environmental conditions as well as site and waste characteristics and landfilling technology used (Steinlechner et al., 1994; Williams, 2005; Scheutz et al., 2009). The last stage marks the end of the degradation reactions and a return to aerobic conditions.  $\text{N}_2$  is now present in significant concentrations in the gas.

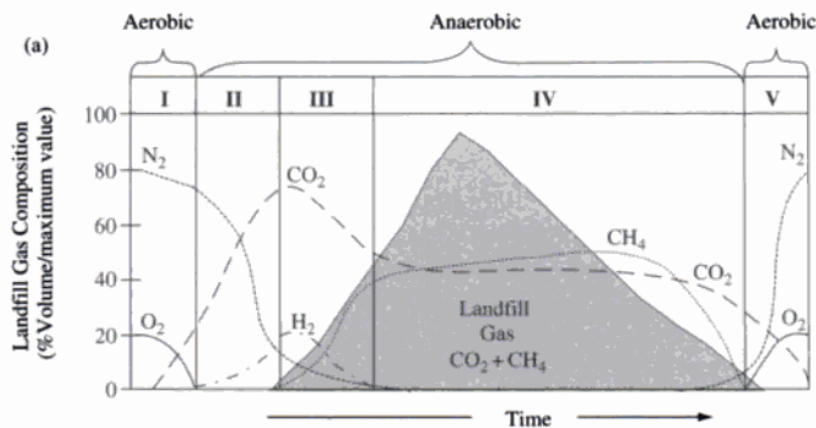


Figure 2.2: Landfill gas composition over time (Williams, 2005)

## 2.2 Landfill gas emission

Emissions from landfills vary roughly by seven orders of magnitude, from less than 0.0004 to more than 4000 g  $\text{CH}_4/\text{m}^2$  per day (Bogner et al., 1997a). This natural variability range of net emissions results from production (methanogenesis), consumption (methanotrophic oxidation) (see section 2.3), temporary storage (e.g. in pores, dissolved in water) and gaseous transport processes in a landfill (Bogner et al., 1997a). Several factors affect gaseous transport processes and control LFG emissions (Kjeldsen, 1996), as follows:

1. Meteorological conditions (e.g. barometric pressure, precipitation, temperature, wind);
2. Soil conditions (e.g. cracks and fissures, permeability, diffusivity, porosity, water content, organic matter content); and
3. Waste and landfill condition (e.g. gas production rate)

The major transport mechanisms for gas emissions from landfills include diffusion and advection. While diffusion is driven by variations in gas concentrations in the soil, advection is caused by pressure gradients induced by changes in atmospheric pressure (Gebert and Gröngroft, 2006; Christophersen and Kjeldsen, 2001; Christophersen et al., 2001; Latham and Young, 1993), landfill gas production (Kjeldsen, 1996) and wind turbulence (Poulsen, 2005). A decline in atmospheric air pressure will draw out gas from the landfill body, while an increase in barometric pressure will lead to reduced gas release. Figure 2.3 depicts the release of CH<sub>4</sub> through the top cover of a landfill following alteration in barometric pressure (Pirkle et al., 1993, cited in Kjeldsen, 1996). Kjeldsen and Fischer (1995) investigated the changes in gas composition in a well of a Danish landfill under a decrease in barometric pressure from 1022 mbar to 1010 mbar over a 33 h period. The authors observed that CH<sub>4</sub> concentration increased at 80 cm depth from below 1 % to near 40 % while CO<sub>2</sub> concentration showed only an increase of approximately 20 %.

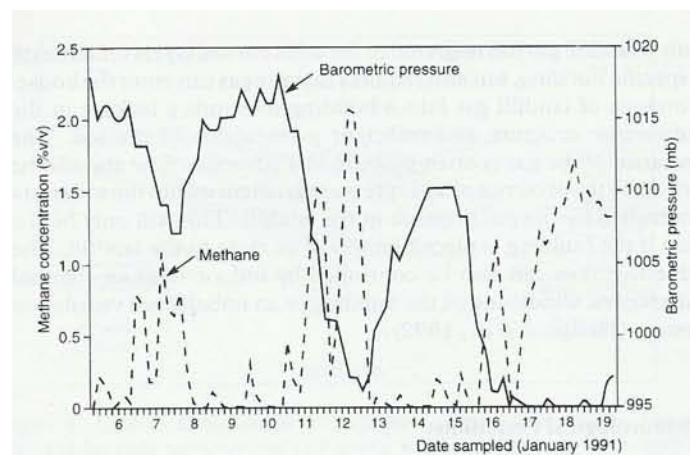


Figure 2.3: Methane concentration measured under a groundsheet placed on a landfill top cover and barometric pressure vs time (Pirkle et al., 1993, cited in Kjeldsen, 1996)

Czepiel et al. (2003) found a robust negative relationship between landfill CH<sub>4</sub> emissions and barometric pressure, as can be seen in Figure 2.4.

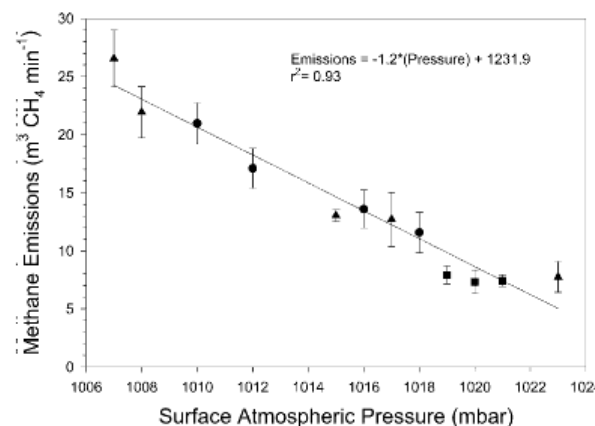


Figure 2.4: CH<sub>4</sub> emissions as a function of atmospheric pressure (Czepiel et al., 2003)

In some cases surface LFG emission even becomes negative during rising barometric pressure, which causes an influx of atmospheric air through the biofilter (Gebert and Gröngroft, 2006) and the top layer of landfill (Nastev et al., 2001), respectively.

Investigations by Poulsen (2005) showed that wind induced emissions can substantially contribute to total gas emissions, especially in moist soils and at wind speeds exceeding 5 m/s (see Figure 2.5).

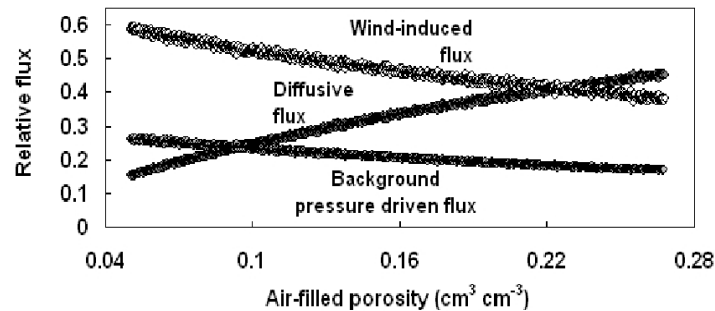


Figure 2.5: Relative fluxes of CO<sub>2</sub> caused by background pressure gradients, molecular diffusion and wind induced gas transport at a Danish landfill (Poulsen, 2005)

These results confirmed that the amount of gas emission from landfills is also dependent on both moisture content and physical components of the soil cover such as grain size, soil texture and porosity which affect the mobility of gas within the soil cover (Teclé et al., 2009; Poulsen, 2005; Börjesson and Svensson, 1997). At low air-filled porosities advective gas transport is the most important transport mechanism while diffusive flux dominates under higher air-filled porosity at drier conditions (Poulsen, 2005). The amount of precipitation, therefore, influences gaseous transport processes both in the long and the short term since high moisture content reduces the permeability and diffusivity of soil. In the short term, large amounts of precipitation can lead to water saturation of the top soil resulting in increased lateral gas migration. Heavy rainfalls often follow a substantial decrease in barometric pressure. This unfavourable combination has been reported as the main factor at a landfill gas explosion in a house close to a Danish landfill (Kjeldsen and Fischer, 1995).

Air temperature has direct effects on soil temperature, which has a close relationship to biological CH<sub>4</sub> oxidation in landfill covers (see section 2.4.5). Soil temperature also controls soil physical characteristics such as swelling and contracting (Teclé et al., 2009). These are important factors on regulating moisture content and LFG emissions throughout the soil cover.

Cracks and fissures, resulting from waste settlement or desiccation of the landfill cover during dry periods, can serve as pathways for methane. Previous studies have found that main emissions occurred at a few areas or hot spots associated with heterogeneities in the cover soil (Nozhevnikova et al., 1993, cited in Börjesson and Svensson, 1997). Bergamaschi et al. (1998) observed that 70 % of CH<sub>4</sub> emissions through landfill cover soils in Germany and The Netherlands were attributed to cracks.

In general, the gaseous transport process is governed by several processes and factors in an often complex network of connections. The transport mechanism is controlled by both diffusive and advective forces. It is difficult to evaluate which of the two mechanisms dominates in a specific case (Kjeldsen, 1996). In some cases pressures in landfills could be substantially higher than barometric pressure,

especially under saturated cover soils or under low permeability covers (e.g. clay or geomembrane composite) (Scheutz et al., 2009). Pressures as high as 2 kPa with respect to atmosphere have been recorded (Hartless, 1995). There have been also instances in a coarse sandy top cover where only very small pressure differences are needed for advective flux to dominate the diffusional flux (Kjeldsen, 1996; Christophersen and Kjeldsen, 2001). Several authors claim that the assumption of a preponderance of an advective flux is supported by observations of a robust inverse relationship between CH<sub>4</sub> emissions and barometric pressure at several landfills. (Stern et al., 2007, De Visscher et al., 2004; Czepiel et al., 2003). In particular, this is true for landfill sites without a gas collection system.

Kjeldsen (1996) concluded that the most important factors controlling CH<sub>4</sub> emission from landfills, besides the gas production intensity, are barometric pressure changes and microbial methane oxidation.

## 2.3 Microbial methane oxidation – general principles

Microbial CH<sub>4</sub> consumption by methanotrophic bacteria is an important biogeochemical sink for the removal of methane from the biosphere thereby reducing the amount of methane released to the atmosphere (Topp and Hanson, 1991). Methanotrophic bacteria, therefore, play an important role in the global methane-cycle (Hanson and Hanson, 1996) as they control the balance between CH<sub>4</sub> sources and sinks in both natural and anthropogenic settings such as landfills (Hanson and Hanson, 1996). Microbial CH<sub>4</sub> oxidation generally occurs at an interface of aerobic and anaerobic zones, where combined conditions of temperature, moisture content, CH<sub>4</sub> and O<sub>2</sub> concentrations are suitable (Zeiss, 2006).

The methane oxidising microorganisms, or methanotrophs, are a subset of a highly diverse group of bacteria known as methylotrophs which are able to metabolise a variety of different one-carbon compounds including methane, methanol, methylamines and halomethanes (Hanson and Hanson, 1996). Most of these bacteria are obligate methanotrophs and strict aerobes (Hanson and Hanson, 1996). However, a genus of facultatively methanotrophic bacteria (*Methylocella*) has been recently identified which also can use two-carbon compounds (Theisen and Murrell, 2005). Beside methanotrophs there are also some ammonia oxidising nitrifying bacteria (nitrifiers) and yeasts that are able to oxidise CH<sub>4</sub> (Bedard and Knowles, 1989; Wolf and Hanson, 1979).

In general, there are two known pathways used by methanotrophs for the conversion of methane to energy and biomass: the ribulose monophosphate path (RuMP) and the serine path (Hanson and Hanson, 1996). The following Figure 2.6 depicts the complete pathway for the oxidation of CH<sub>4</sub> to CO<sub>2</sub> and the assimilation of formaldehyde (CH<sub>2</sub>O) of both pathways. The first step of methane oxidation by methanotrophic bacteria is the conversion of dissolved methane to methanol (CH<sub>3</sub>OH) which is catalysed by a key enzyme known as methane monooxygenase (MMO). The MMO exists in two forms, a soluble, cytoplasmic form (sMMO) and a membrane-bound or particulate form (pMMO). While all known methanotrophs (except *Methylocella* sp.) are capable of forming pMMO, only certain methanotrophs have the ability to produce sMMO linked to low copper concentration (Hanson and Hanson, 1996). Formaldehyde produced from the oxidation of methane and methanol is then used either for biosynthesis or for catabolism (Lengeler et al., 1999). During biosynthesis, formaldehyde enters the serine or the RuMP pathway. In the

serine pathway 2 mol of formaldehyde and 1 mol of  $\text{CO}_2$  are utilised for the synthesis of multicarbon compounds whereas in the RuMP cycle 3 mol of formaldehyde are assimilated. In the catabolic reactions, formaldehyde is further oxidised via formate into  $\text{CO}_2$ , which constitutes the last step of the  $\text{CH}_4$  metabolism.

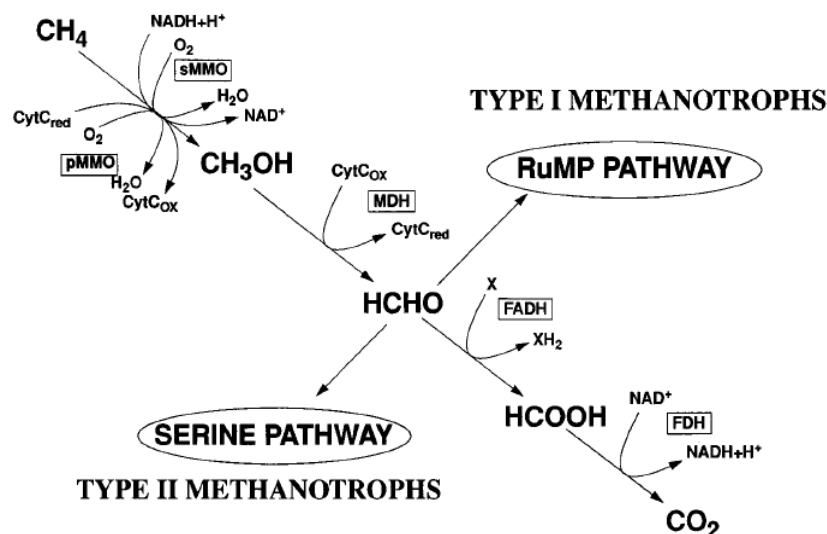


Figure 2.6: Pathways for the oxidation of methane and assimilation of formaldehyde. (Abbreviations: CytC, cytochrome c; FADH, formaldehyde dehydrogenase; FDH, formate dehydrogenase), (Hanson and Hanson, 1996)

Taxonomically, methanotrophs can be classified into two main distinct groups: type I and type II, belonging to the alpha and gamma subdivision of the Proteobacteria (Hanson and Hanson, 1996). Methanotrophs of the Proteobacteria differ in their carbon assimilation pathway, morphology, biochemistry, affinity for  $\text{CH}_4$ , membrane type and composition (Hanson and Hanson, 1996; Börjesson et al., 2004; Vishwakarma et al., 2009). Type I methanotrophs are represented by ten genera including *Methylobacter*, *Methylobacter*, *Methylobacterium*, *Methylosarcina*, *Methylosphaera*, *Methylosoma*, *Methylohalobium*, *Methylococcus*, *Methylocaldum* and *Methylothermus* (Vishwakarma et al., 2009). Type II includes the genera *Methylosinus*, *Methylocystis*, *Methylocella* and *Methylocapsa*. While type I methanotrophs assimilate carbon via the RuMP cycle, type II methanotrophs use the serine pathway.

According to methane oxidation kinetics and affinities, two methanotrophic populations have been observed (Dunfield et al., 1999). The first group, commonly referred to as the upland soil cluster (USC) alpha, are considered as 'high-affinity' bacteria because of their ability to oxidise  $\text{CH}_4$  at concentrations close to that of the atmosphere ( $< 12$  ppm) (Le Mer and Roger, 2001; Huber-Humer et al., 2008b). The second group, known as 'low-affinity' methanotrophs, is only active at  $\text{CH}_4$  concentrations higher than 40 ppm (Le Mer and Roger, 2001).

Hanson and Hanson (1996) suggest a classification of the methanotrophs according to the  $\text{CH}_4$  and  $\text{O}_2$  concentrations. Type I methanotrophs, which are mainly found in the upper parts of the soil, prefer to grow at low  $\text{CH}_4$  levels and high  $\text{O}_2$  concentrations (Hanson and Hanson, 1996). By contrast, the growth of some type II methanotrophs is favoured in deeper zones when  $\text{CH}_4$  concentrations are high together with low levels of nitrogen and oxygen (Hanson and Hanson, 1996). These methanotrophs are more typical of those encountered in landfill cover soils (Kightley



et al., 1995). However, type II methanotrophs have also been detected in an organic soil oxidising methane at atmospheric level (Dunfield et al., 1999)

Due to the multiplicity of methane sources, such as wetlands, microbial CH<sub>4</sub> uptake is ubiquitous (Hanson and Hanson, 1996). Biological CH<sub>4</sub> oxidation has been reported to occur in variety of ecosystems, as in temperate (Bradford et al., 2001), tropical (Butterbach-Bahl et al., 2004), desert (Angel and Conrad, 2009; Striegl et al., 1992) and arctic soils (Liebner et al., 2009; Whalen and Reeburgh, 1996) as well as in aquatic environments (Le Mer and Roger, 2001). However, large methanotrophic populations and the highest rates of CH<sub>4</sub> oxidation have been observed in landfill cover soils (Scheutz et al., 2003; De Visscher et al., 1999; Kightley et al., 1995; Whalen et al., 1990). One of the first journal publications to report methane oxidation activity in landfill cover soils was made by Whalen et al. (1990) using laboratory batch tests. The authors found the highest rates of CH<sub>4</sub> consumption observed in any soils before that time (45 g/m<sup>2</sup>d). Recent studies have observed the capacities for CH<sub>4</sub> oxidation of landfill cover soil in column experiments, with rates as high as >200 g CH<sub>4</sub>/m<sup>2</sup>d (Scheutz et al., 2003; De Visscher et al., 1999). Even higher steady state CH<sub>4</sub> removal rates (> 400 g CH<sub>4</sub>/m<sup>2</sup>d) have been found in simulated landfill covers rich in organic matter such as mature compost materials (Haubrichs and Widmann, 2006; Humer and Lechner, 2001b). Numerous studies, therefore, have supported the concept of microbial CH<sub>4</sub> removal by oxidation as an effective tool for greenhouse gas reduction in landfill emission (Stern et al., 2007; Zeiss, 2006; Hilger and Humer, 2003; Humer and Lechner, 1999a). Reported values for whole landfill CH<sub>4</sub> oxidation rates had a wide range which can be attributed to physical heterogeneities in the cover, different CH<sub>4</sub> levels, and seasonal climate change (Hilger and Humer, 2003; Börjesson et al., 2001; Chanton and Liptay, 2000). Rates of CH<sub>4</sub> removal in landfill covers can range from negligible to 100 % (Börjesson et al., 2001, 2007; Stern et al., 2007; Abichou et al., 2006; Huber-Humer, 2004). Under certain conditions, landfill covers even take up rather than emit methane, resulting in the landfill cover functioning as a sink for atmospheric methane (Barlaz et al., 2004; Bogner et al., 1995 and 1997a, b; Börjesson and Svensson, 1997; Boeckx et al., 1996). In general, steady state CH<sub>4</sub> consumption rates for landfill cover soils are between 100 to 150 g CH<sub>4</sub>/m<sup>2</sup>d, indicating 30 to 60 % CH<sub>4</sub> removal, whereas maximum rates of 200 to 250 g CH<sub>4</sub>/m<sup>2</sup>d can contribute to 80 – 100 % CH<sub>4</sub> oxidation (Scheutz et al., 2009).

Methanotrophs can also co-metabolise a variety of non-methane compounds including some halogenated hydrocarbons and aromatics (Bogner et al., 2010; Scheutz and Kjeldsen, 2004) due to the broad substrate specificity of the MMO enzyme (Bogner et al., 2010).

There have also been instances of methanotrophic activity contributing to production of nitrous oxide (N<sub>2</sub>O) (Mandernack et al., 2000), itself a greenhouse gas with a global warming potential of 289 over a 100-year period (IPCC, 2007). Additions of nitrogenous fertiliser or nitrogen-rich environments (i.e. covers made of organic soil substrates or compost) and alternating aerobic and anaerobic zones have resulted in increased N<sub>2</sub>O emission rates (Lee et al., 2009; Zhang et al., 2009; Huber-Humer, 2008b; Mandernack et al., 2000; Boeckx and Van Cleemput, 1996). In contrast, no or little N<sub>2</sub>O emissions were measured in organic landfill cover materials consisting of a mixture of biowaste compost and gravel (Watzinger et al., 2005) and mechanically–biologically treated municipal solid waste (Einola et al., 2008). However, it is still unclear what the relative contributions of methanotrophs are to N<sub>2</sub>O formation in landfill covers or how to minimise them. Besides nitrification by methanotrophs, also

nitrification and denitrification by ammonia-oxidising bacteria or archaea play a part in the  $\text{N}_2\text{O}$  production processes (Lee et al., 2009).

## **2.4 Controlling factors on methane oxidation**

Several laboratory and field studies have shown a wide range of  $\text{CH}_4$  removal rates (Stern et al., 2007; Zeiss, 2006; Huber-Humer, 2004; Chanton and Liptay, 2000; Whalen et al., 1990). This wide variation can be attributed to numerous factors including, among others, soil temperature and moisture, methane and oxygen supply, nutrient level, organic matter content and soil physical properties such as permeability and particle size which control microbial  $\text{CH}_4$  uptake (Boeckx and Van Cleemput, 1996; Boeckx et al., 1996; Kightley et al., 1995; Castro et al., 1994; Whalen et al., 1990). Although methanotrophic microorganisms are fairly adaptable and resilient to changing conditions, landfill covers have to ensure optimum ambient conditions in order to achieve sufficient  $\text{CH}_4$  oxidation rates. Thus, management of these physical and environmental factors in engineered biocover systems can contribute to enhanced methanotrophic performance, resulting in reduced GHG emissions. In this chapter the controlling factors on  $\text{CH}_4$  oxidation are presented in more detail.

### **2.4.1 Moisture content**

The moisture content in substrates appears to be a crucial factor controlling  $\text{CH}_4$  oxidation because it affects both the movement of gases through the cover soil and microbial activity. Soil moisture influences both gas permeability and diffusivity of soil as the available pore space for gaseous transport and diffusion are affected at different moisture contents (Tecle et al., 2009). Higher moisture levels may decrease the  $\text{CH}_4$  oxidation capacity of the landfill cover due to limiting of gaseous transport processes of  $\text{O}_2$  and  $\text{CH}_4$ . As a consequence, gas-phase diffusion is shifted to aqueous-phase diffusion, which is  $10^4$  fold less rapid, resulting in limited substrate delivery to the microbes (Boeckx et al., 1996). Nesbit and Breitenbeck (1992) showed that the average rate of  $\text{CH}_4$  oxidation was reduced by 56 % at water saturation. Sitaula et al. (1995) reported a significant decrease in  $\text{CH}_4$  uptake rate by 35 % to 50 % when increasing the soil moisture content from 32 % to 42 % in silty sand forest soil. In general,  $\text{O}_2$  can penetrate much deeper into unsaturated substrate layers compared to saturated ones. Hence, unsaturated zones serve as a barrier for residual  $\text{CH}_4$  emissions. On the other hand, low moisture levels can also decrease  $\text{CH}_4$  uptake due to microbial desiccation resulting in lower methanotrophic activities.  $\text{CH}_4$  consumption is substantially reduced when soil moisture content decreases below 5 % (Scheutz and Kjeldsen, 2004; Stein and Hettiaratchi, 2001; Whalen et al., 1990).  $\text{CH}_4$  oxidation will be at its maximum when there is both maximum gas phase molecular diffusion and sufficient soil moisture content (Scheutz et al., 2009) (see Figure 2.7).

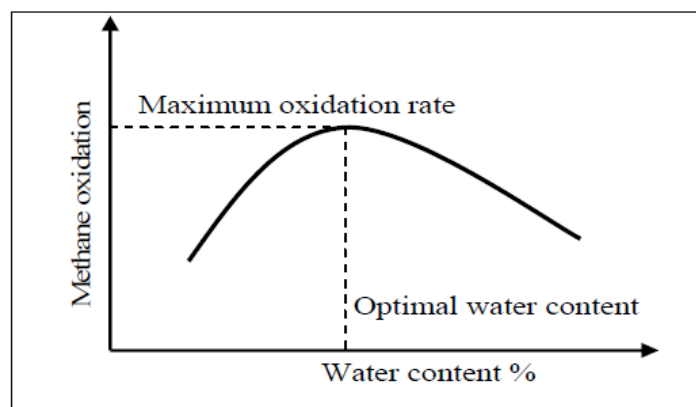


Figure 2.7: Response of CH<sub>4</sub> oxidation to soil water content (Yuan, 2006)

The optimum moisture content varies for different soil types and is further dependent on temperature (see section 2.4.5) and other environmental factors. Experimental studies showed that optimal moisture content in mineral landfill covers ranged between 10 % and 20 % w/w (Boeckx and Van Cleemput, 1996; Boeckx et al., 1996; Whalen et al., 1990) whereas higher optimum water contents have been observed in organic landfill cover materials (Albanna et al., 2007; Mor et al., 2006; Humer-Huber, 2004; Scheutz and Kjeldsen, 2004; Humer and Lechner, 1999b). A high CH<sub>4</sub> oxidation was reported at a moisture content of 45 % w/w in composted municipal solid waste (24 % organic matter) used as landfill cover (Humer and Lechner, 1999b). Mor et al. (2006) found optimum moisture content of garden waste compost (31 % and 52 % organic matter) ranging from 45 to 110 % on a dry weight basis. However, under wet conditions, the high water level may block gas transport causing a reduction of CH<sub>4</sub> consumption or even contributing to CH<sub>4</sub> production in extreme cases (Scheutz et al., 2009). Hence, a balanced grain size distribution is a prerequisite for sufficient gas permeability at high moisture contents (Scheutz et al., 2009). Humer-Huber (2004) measured oxidation rates up to 100 % even at water saturation (57 % w/w) in mature sewage sludge compost mixed with roughly shredded wood chips (1:1 w/w). The author concluded that high performance could be maintained at high moisture contents because of the adequate porosity and high water-holding capacity (WHC) of the waste material. As a consequence, compost covers control water infiltration into the landfill which enhances waste degradation and establishes optimal conditions for the methanotrophic community (Scheutz et al., 2009; Stern et al., 2007; Huber-Humer, 2004).

In numerous studies, soil moisture has been suggested as a major controlling factor of CH<sub>4</sub> oxidation (Jugnia et al., 2008; Albanna et al., 2007; Gebert et al., 2003; Boeckx et al., 1996; Castro et al., 1994). Boeckx et al. (1996) stated that the moisture content largely controls methanotrophic activity. A multiple linear regression analysis indicated that moisture content has more influence than temperature explaining much of the variation of CH<sub>4</sub> emission (Boeckx et al., 1996). The authors concluded that the optimal moisture content is situated at about 50 % of the WHC supporting both activity of methanotrophic bacteria and gas permeability. In forest soils, the soil moisture content was a key variable influencing CH<sub>4</sub> oxidation rates, where 78 % of the variability in CH<sub>4</sub> oxidation rates has been correlated to soil moisture variability (Castro et al., 1994).

Investigations by Spokas and Bogner (2010) indicated that soil moisture potential, which expresses soil moisture in terms of the physical force with which water is held

in soil, provides a more robust parameter than gravimetric or volumetric moisture for examining the dependency of landfill CH<sub>4</sub> oxidation rates on soil moisture. The authors found that optimal soil moisture potential in landfill cover soils was close to field capacity (50 kPa) whereas the minimum threshold soil moisture potential was at approximately 1500 kPa (soil wilting point). Using soil moisture potential, differences in soil texture and structure could be normalised and, thus, a direct comparability to other soils would be achieved.

#### 2.4.2 Organic content and nutrient supply

The organic matter in substrates strongly influences the CH<sub>4</sub> oxidation capacity. On the one hand, organic matter improves the substrate properties such as soil structure and aggregation, WHC and aeration and it further serves as a carrier for microorganisms (Sparks, 1995). On the other hand organic matter provides nutrient supply which is a prerequisite for cellular metabolism of methanotrophic bacteria and other bacteria in a substrate.

In general, CH<sub>4</sub> oxidation rate increases with increasing content of organic matter in a substrate. Composted materials with high organic content (of up to 35 % w/w) show 10 to 100 fold higher CH<sub>4</sub> oxidation potentials compared to soils with organic contents of 1 to 10 % (He et al., 2008; Zeiss, 2006). Huber-Humer (2004) reported 100 % steady-state CH<sub>4</sub> oxidation in fully matured sewage sludge compost used as landfill cover exceeding performance of conventional soil covers (40 – 45 % uptake). Compost maturity (7-day oxygen demand < 8 mg O<sub>2</sub>/g DM) proved to be important to ensure minimum competition for O<sub>2</sub> from other microorganisms as it provides biochemically stable organic matter (Scheutz et al., 2009; Kettunen et al., 2006; Humer and Lechner, 1999). Compared to conventional soils, composts have considerably higher content of long-term available nutrients (Nitrogen: 0.85 – 1.25 % DM; Phosphorus: 0.43 – 3.06 % DM) (Huber-Humer, 2004). Therefore, several studies have suggested that high organic materials such as composted sewage sludge, biowaste, yard waste, municipal solid waste or mechanically-biologically pretreated municipal solid waste and biowaste may be used as a support medium for CH<sub>4</sub> oxidation in landfill covers to mitigate greenhouse gas emissions from landfills (Bohn and Jager, 2009; Einola et al., 2008; Jugnia et al., 2008; Stern et al., 2007; Mor et al., 2006; Humer-Huber, 2004; Cossu et al., 2003).

The addition of nutrients such as sewage sludge, phosphate, lime and commercial fertiliser has also been found to enhance CH<sub>4</sub> oxidation rates in landfill cover soils (Albanna et al., 2007; Hilger et al., 2000; Börjesson et al., 1998; Boeckx and Van Cleemput, 1996; Kightley et al., 1995). Kightley et al. (1995) documented that amendment of the coarse sand with sewage sludge (2.5 g/kg of soil) raised CH<sub>4</sub> oxidation rate by 26% whereas mineral fertiliser showed no increase or even a decrease in the CH<sub>4</sub> uptake rate. According to investigations by Albanna et al. (2007), commercial fertiliser (0.3 g N<sub>2</sub>:0.13 g P:0.249 g K for each kg soil) could be a source of nutrients for methanotrophs when the moisture in the landfill cover is adequate. Adding fertiliser to a landfill cover soil (1.5 g/kg of soil) that contained 30 % moisture content increased the CH<sub>4</sub> oxidation rate from 38 % to 81 % while adding nutrient to the soil with low level of moisture content (15 %) negatively affected the methanotrophic performance. Based on a statistical design model higher maximum CH<sub>4</sub> oxidation rates could be achieved when nutrients are added to the cover soil with 45 % moisture content, as can be seen in Figure 2.8. These results again confirmed the importance of moisture content for CH<sub>4</sub> oxidation in landfill cover soil.

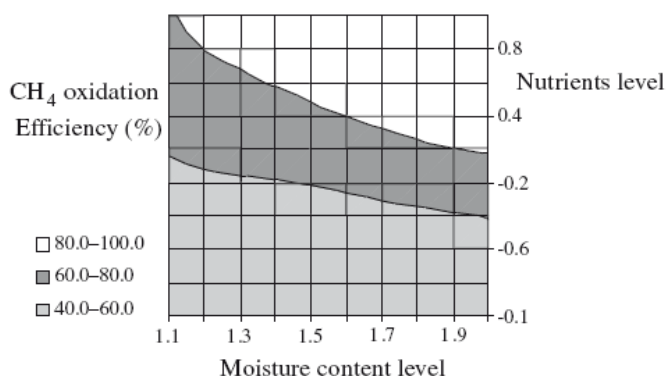


Figure 2.8: Plot of the modelled maximum CH<sub>4</sub> oxidation efficiency (%) for moisture content levels between 30 % and 45 % and nutrient additions (-1 code represents no nutrients added, +1 code, added nutrients of 1.5 g fertiliser/kg of soil dry weight) (Albanna et al., 2007)

Also the C:N ratio of the nutrient amendment or the used substrate seems to considerably affect CH<sub>4</sub> oxidation in landfill cover soil. Addition of crop residues (potato and sugar beet leaves) with low C:N ratios (13.5 and 11.3) produced high and inhibitory concentrations of ammonium (NH<sub>4</sub>) and nitrite (NO<sub>2</sub><sup>-</sup>) due to nitrogen mineralisation resulting in a strong inhibition of the CH<sub>4</sub> oxidation (Boeckx and Van Cleemput, 1996) (see also section 2.4.7). In contrast, crop residues (wheat and maize straw) with a high C:N ratio (97.2 and 24.8) stimulated N-immobilisation and, thus, minor amounts of ammonium and nitrite were produced, which caused no inhibition.

### 2.4.3 pH-value

Methanotrophs are mainly neutrophilic and their optimum growth and activity rate lies between pH 5.5 and 8.5 (Figuerola, 1993). Some methanotrophs, however, tolerate high pH values up to 9 and oxidation activity seems to continue down to pH 2 (Islam et al., 2007; Huber-Humer, 2004). Two genera of moderately acidophilic methanotrophs have been characterised and the optimal value for their development is between pH 5.0 – 5.5 (Islam et al., 2007). Hilger et al. (2000) observed enhanced CH<sub>4</sub> oxidation when adding lime to landfill covers which raised pH from 6.3 to 7.4. According to Scheutz and Kjeldsen (2004), it is questionable whether significant pH gradients will develop in “natural” landfill covers as they often have high buffer capacities. Hence, the CH<sub>4</sub> oxidation capacity will be less affected by pH.

### 2.4.4 Particle size and porosity

Substrate physical properties such as soil texture and porosity can influence the supply of essential elements (O<sub>2</sub>, water, nutrients) to methanotrophic microorganisms and thereby control microbial CH<sub>4</sub> oxidation. In general, a topsoil of coarse texture shows a higher CH<sub>4</sub> removal rate than a fine one due to its better aeration property (Mor et al., 2006; Watzinger et al., 2005; Boeckx et al., 1997). For instance, higher oxidation rates have been found in coarse sand (61 %) than in fine sand or clay (40 – 41 %) (Kightley et al., 1995). In addition, coarse substrates have shown a better insulation from atmospheric temperature fluctuations than fine substrate (Humer-Huber and Lechner, 2009; Humer-Huber, 2004). Huber-Humer (2004) observed that during winter months the temperature produced by microbial activity was retained much better in coarse sewage sludge compost mixed with big wood chips than the fine-sieved MSW-compost. Moreover, materials with a high air-filled pore space such as composts (on the order of 50 % compared to 20 – 30 % for soils) will allow more O<sub>2</sub> to penetrate into the cover material creating a more extended aerobic zone (Mor

et al., 2006). Humer and Lechner (1999a) and Börjesson et al. (2004) observed that soils with high porosity and large particle size distribution can retain  $\text{CH}_4$  and  $\text{O}_2$  for a longer period of time, leading to higher oxidation rates. However, Pawlowska et al. (2003) measured slightly higher  $\text{CH}_4$  uptake rates in coarse sand (0.5 – 1 mm) compared to coarse gravel (2 – 4 mm). The lower specific surface area of the gravel fraction might have led to lower potential contact between  $\text{CH}_4$  molecules and microorganisms. Gebert and Gröngroft (2009) suggested a gas-filled porosity of at least 17.5 vol. % to ensure a rate of diffusive oxygen ingress that enables complete oxidation of a  $\text{CH}_4$  flux of  $0.5 \text{ l CH}_4/\text{m}^2\text{h}$ .

Since substrate physical properties constitute the main design variables of engineered covers, they should be optimised to allow almost the full cover depth to support  $\text{CH}_4$  oxidisers.

#### **2.4.5 Soil temperature**

The activity of methanotrophic microorganisms is dependent on the soil temperature. Typically,  $\text{CH}_4$  uptake rates increase with rising temperatures while at low temperatures inhibition effects have been observed (De Visscher et al., 2001; Visvanathan et al., 1999; Börjesson and Svensson, 1997; Whalen et al., 1990). As of most methanotrophs are mesophilic, the optimum temperature for  $\text{CH}_4$  oxidation is usually in the range of 20 – 38 °C (see Figure 2.9) (Gebert et al., 2003; Visvanathan et al., 1999; Börjesson and Svensson, 1997; Boeckx et al., 1996; Figueroa, 1993; Whalen et al., 1990).

Huber-Humer (2004) reported that a complete  $\text{CH}_4$  oxidation was achieved between 8 °C and 30 °C in mature sewage sludge. At a lower temperature of 4 °C  $\text{CH}_4$  oxidation decreased to about 70 – 80 %. Experiments by Zeiss (2006) showed even a halt in biological  $\text{CH}_4$  oxidation in yard waste compost during the winter period. Similar observations have been made by Börjesson et al. (2001) in a Swedish landfill cover soil. Boeckx and Van Cleemput (2000) suggested that  $\text{CH}_4$  oxidation does not occur at temperatures below 0 °C. The susceptibility of methanotrophs to extreme temperatures may be associated with a small cover depth (Börjesson et al., 2001; Chanton and Liptay, 2000). In contrast,  $\text{CH}_4$  oxidation continued in biofilters during winter (Streese and Stegmann, 2003). Methanotrophic bacteria in the cover soil of a boreal landfill were also able to oxidise  $\text{CH}_4$  at increasing rates even at temperature below 10 °C and close to freezing point (Einola et al., 2007). The increase in  $\text{CH}_4$  uptake rates was due to growth or activation of psychrophilic (cold tolerant) methane oxidisers. According to Hanson and Hanson (1996) and Börjesson et al. (2004) psychrophilic methanotrophs are mainly of type I which prefer low  $\text{CH}_4$  and high  $\text{O}_2$  concentrations. Results from Börjesson et al. (2004) and Gebert et al. (2003) showed that enriching methanotrophic mixed cultures at low temperatures (3 – 10 °C) lead to a shift towards more methanotrophs of type I indicating that temperature response in support media such as soil or compost might be time-dependent.

Spokas and Bogner (2010) demonstrated that  $\text{CH}_4$  uptake was amplified by increasing temperatures up to 30 °C and then rates declined and dropped to zero by 55 °C, as can be seen in Figure 2.9. However, there have been also some instances, where thermophilic species have been detected with optimal growth temperature at 55 – 59 °C (Islam et al., 2007).

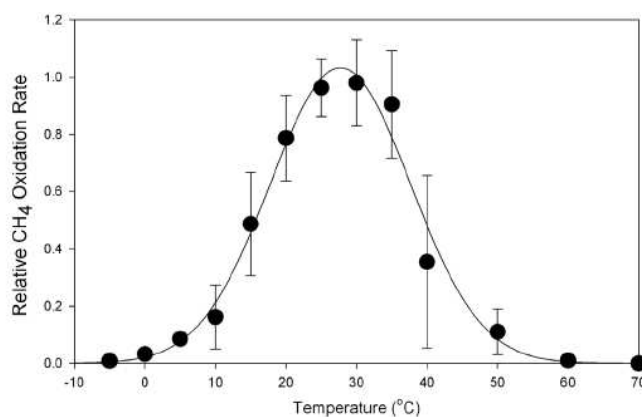


Figure 2.9: Effects of temperature on relative rates of CH<sub>4</sub> oxidation with associated standard deviations and fit to a 3-parameter Gaussian curve ( $n = 3456$ ) (Spokas and Bogner, 2010)

In order to quantify the temperature influence on microbial CH<sub>4</sub> oxidation, the so called  $Q_{10}$  value might be used which gives the number of times the oxidation rate increases when the temperature is increased 10 °C at temperatures below the optimum temperature. The  $Q_{10}$  value between the temperatures of 10 °C and 30 °C was found to be in the range of 1.7 – 4.1 (Börjesson et al., 2004; De Visscher et al., 2001; Boeckx et al., 1996; Whalen et al., 1990). Also  $Q_{10}$  values as high as 4.10 – 7.26 have been observed in landfill cover soils (Christophersen et al., 2000). In general, a high  $Q_{10}$  value indicates that temperature controls the CH<sub>4</sub> oxidation more than other factors. Börjesson and Svensson (1997) stated that soil temperature is the key factor of CH<sub>4</sub> oxidation, explaining 85 % of the variation in measured CH<sub>4</sub> oxidation. According to De Visscher et al. (2001), the influence of temperature on CH<sub>4</sub> oxidation is more pronounced at high CH<sub>4</sub> concentrations than at low concentrations. In contrast, experiments by Boeckx et al. (1996) indicated that temperatures had only slight effects on CH<sub>4</sub> oxidation. As can be seen in Table 2.1, the optimum incubation temperature (30 – 20 °C) declined with increasing moisture contents. The authors concluded that equilibrium exists between CH<sub>4</sub> diffusion rate at certain moisture content and the uptake rate by the methanotrophic bacteria. Hence, at high moisture contents substrate supply of CH<sub>4</sub> and O<sub>2</sub> becomes the limiting factor and not the bacterial activity due to temperature.

Table 2.1: Correlation between moisture content and optimum temperature (Boeckx et al., 1996)

Moisture content (%)	Optimum temperature (°C)
5	30.0
10	27.1
15	24.9
20	23.5
25	21.2
30	20.1

Not surprisingly, air temperature has direct effects on soil temperature, especially in the upper horizons, leading to seasonal temperature dependence of methanotrophic activity in the landfill cover (Mor et al., 2006; Hilger and Humer, 2003). However, methanotrophic bacteria themselves can influence ambient temperature as CH<sub>4</sub> oxidation is an exothermic process releasing 210.8 kCal per mol of converted CH<sub>4</sub>. Berger et al. (2005) reported that methanotrophic activity in the landfill cover (mixture of compost and sand, 0.3 m, over a layer of loamy sand, 0.9 m) was able to produce temperatures which were 5 – 8 °C higher than the ambient temperature. Field

experiments by Humer and Lechner (2001) showed that temperatures  $> 15\text{ }^{\circ}\text{C}$  were maintained in a biocover made of compost materials at a depth of 0.5 m downwards even during winter months. However, heat generation also occurs inside the waste due to anaerobic exothermic reactions leading a temperature range from  $30 - 60\text{ }^{\circ}\text{C}$  (Williams, 2005).

#### **2.4.6 Air pressure**

As already mentioned, a rapid drop in atmospheric pressure could pull  $\text{CH}_4$  through a  $\text{CH}_4$  oxidation zone faster than methanotrophs can consume it resulting in occasional peaks of  $\text{CH}_4$  emissions. On the other hand, a rapid atmospheric pressure rise enables  $\text{O}_2$  to penetrate into deeper regions of relatively inactive sites where it could oxidise  $\text{CH}_4$  (Latham and Young, 1993).

In addition, aerobic methane oxidation itself shows a “gas-pumping-effect”. The resulting volume reduction during aerobic  $\text{CH}_4$  oxidation (according to equation 2.1) creates a low pressure enhancing the ability of air to diffuse deeper into the oxidation layer (Huber-Humer 2004; De Visscher, 2001; Kjeldsen, 1996). While the oxygen from the air-flow is immediately consumed by the methanotrophs, nitrogen accumulates. Moreover, the upward gas flow slows down as it reaches the top (DeVisscher, 2001). This “gas-pumping-effect” may increase oxygen transport into the substrate, thereby enhancing methane oxidation. Experiments by Huber-Humer (2004) confirmed this phenomenon using Ar as a tracer in an actively methane oxidising column and a sterile column without methanotrophic activity. The tracer-component clearly accumulated in the “active column” compared to the “sterile” one, indicating that the “pumping effect” triggered a higher flow of air into the cover.

#### **2.4.7 Inorganic Nitrogen**

As already stated nutrient supply, especially nitrogen, is a prerequisite to build up methanotrophic biomass. Methanotrophs take up nitrogen in the form of ammonium or nitrate. In addition, some strains (type II) even can fix atmospheric nitrogen. According to Anthony (1982; cited in Scheutz et al., 2009) 0.25 mole of nitrogen is required for every mole of assimilated carbon. The effect of inorganic nitrogen (ammonium, nitrate) on  $\text{CH}_4$  oxidation is quite complex and can be both stimulatory and inhibitive depending on several factors including the species and concentration of nitrogen, soil pH and type of methanotrophs present (Scheutz et al., 2009; Bodelier and Laanbroek, 2004). Several studies have shown that elevated  $\text{NH}_4^+$  concentrations in soil may inhibit  $\text{CH}_4$  oxidation rates due to possible substrate competition between  $\text{CH}_4$  and  $\text{NH}_4^+$  at the level of MMO enzymes (Reay and Nedwell, 2004; Boeckx and Van Cleemput, 1996; King and Schnell, 1994). For instance, Boeckx and Van Cleemput (1996) observed inhibition of the  $\text{CH}_4$  oxidation rate in landfill cover soil upon  $\text{NH}_4^+$  addition (25 mg N/kg). Scheutz and Kjeldsen (2004) found unaltered  $\text{CH}_4$  oxidation rates with the addition of  $\text{NH}_4^+$  up to 14 mg N/kg (added as  $\text{NH}_4\text{Cl}$ ) to landfill cover soil, whereas soil showed decreasing  $\text{CH}_4$  oxidation rate upon increasing  $\text{NH}_4^+$  application rates (see Figure 2.10).



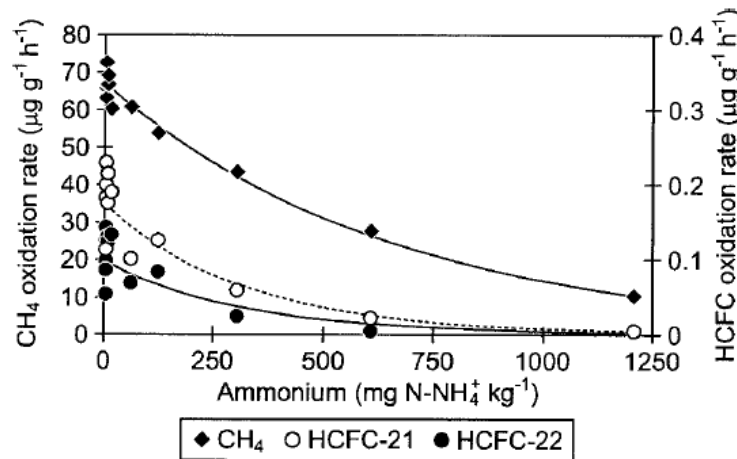


Figure 2.10: Influence of ammonium on the oxidation rate of methane and hydrochlorofluorocarbons (HCFCs) (Scheutz and Kjeldsen, 2004)

In contrast, stimulation of CH<sub>4</sub> oxidation following NH<sub>4</sub><sup>+</sup>-based fertilisation has been reported (Lee et al., 2009; Hilger et al., 2000). As can be seen in Figure 2.11, the CH<sub>4</sub> uptake rate in landfill cover soils amended with 100 mg-N NH<sub>4</sub>/kg soil (added as NH<sub>4</sub>Cl) was increased by about 60 % (Lee et al., 2009). However, the ammonium application also led to N<sub>2</sub>O production, which increased over 16-fold compared to the baseline conditions.

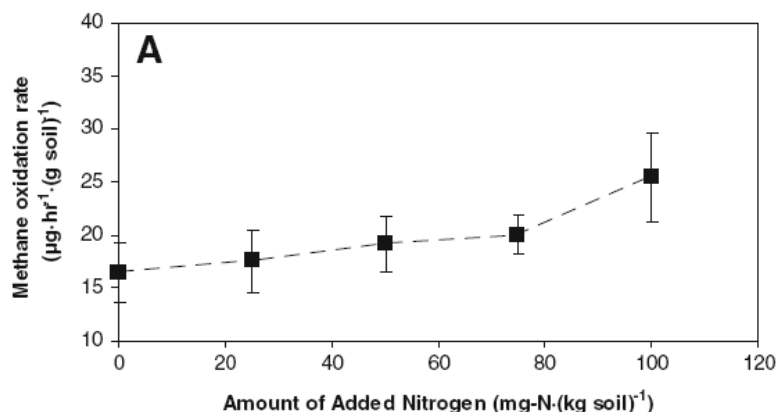


Figure 2.11: Effect of ammonium on the rate of methane consumption (Lee et al., 2009)

De Visscher et al. (1999) concluded that NH<sub>4</sub><sup>+</sup> inhibition becomes more pronounced at low atmospheric CH<sub>4</sub> concentrations in soil. De Visscher and Van Cleemput (2003) reported that at steady state NH<sub>4</sub><sup>+</sup> inhibited CH<sub>4</sub> oxidation at low CH<sub>4</sub> concentration while at high concentrations CH<sub>4</sub> oxidation was stimulated. The authors further observed that after an initial peak of methanotrophic activity, growth rates, probably of type I bacteria, declined considerably as a response to N shortage and later on methanotrophic activity, probably of type II methanotrophs, increased in spite of the low inorganic N content, due to their independence from inorganic N.

Boeckx et al. (1996) and Boeckx and Van Cleemput (1996) suggested that the inhibition by NH<sub>4</sub><sup>+</sup> seems to be considerably related to its nitrification rate or N-turnover rate rather than to its actual concentration. In addition, the authors found that ammonium has a greater effect on CH<sub>4</sub> oxidation than the soil moisture content.

Experiments by Wilshusen et al. (2004) indicated that high NH<sub>4</sub><sup>+</sup> concentrations within fresh compost materials were negatively correlated with CH<sub>4</sub> oxidation

suggesting that highest methane oxidation efficiency could be achieved with a low  $\text{NH}_4^+$  content in compost. Similar observations have been made with fresh sewage sludge showing inefficient  $\text{CH}_4$  oxidation rates, most probably because nitrification suppressed  $\text{CH}_4$  oxidation (Huber-Humer, 2004).

Nitrate showed inhibition of  $\text{CH}_4$  oxidation only at very high concentrations through osmotic effects which usually do not occur in landfill environments (Scheutz et al., 2009; Bodelier and Laanbroek, 2004).

#### 2.4.8 Vegetation

Previous studies have indicated that vegetation may have an important effect on the growth and activity of methanotrophs as well as on the soil physical properties (Xiaoli et al., 2010; Bohn and Jager, 2009; Wang et al., 2008; Hilger and Humer, 2003). The influence on soil physical properties is still not well known. Generally, vegetation stabilises and protects the top cover from erosion by spreading roots and controls moisture infiltration due to its evapotranspiration capacity (Bohn and Jager, 2009). Since roots provide a channel for oxygen penetration and build up different pore sizes, gas diffusion of  $\text{O}_2$  and  $\text{CH}_4$  to the methanotrophs is improved and, therefore,  $\text{CH}_4$  oxidation is likely to be enhanced. In addition, the root system of vegetation releases exudates into the surrounding soil and induces a more suitable environment for methanotrophs. Stralis-Pavese et al. (2004) observed that all tested plant covers on sewage sludge compost considerably enhanced  $\text{CH}_4$  oxidation potential compared to bare substrate covers. These results were related to a higher diversity of methanotrophs in the vegetated landfill covers than in the substrate alone. Similar observations have been made by Hilger et al. (2000) and Bohn and Jager (2009). There is evidence that the efficiency of  $\text{CH}_4$  oxidation also depends on plant species and physiological status of the vegetation (Xiaoli et al., 2010; Reay et al., 2005; Stralis-Pavese et al., 2004). On the other hand vegetation is closely related to the depth and moisture retentiveness of the cover material (Xiaoli et al., 2010). Experiments by Huber-Humer and Lechner (2003) and Huber-Humer (2004) indicated that the evapotranspirative capacity of vegetation combined with the high water retention capacity of compost material used in a biocover (1 m high) will be sufficient to prevent high levels of infiltration and leachate production. The tests revealed a water infiltration rate of < 10 % annual precipitation (700 mm) within the first years of operation.

Various studies have indicated that the plant growth in landfill covers is severely inhibited by the presence of high landfill gas concentrations in the root zone (Xiaoli et al., 2010; Chan et al., 1997). High  $\text{CO}_2$  concentrations are directly toxic to the roots even at sufficient  $\text{O}_2$  availability (Nagendran et al., 2006). In addition, the root penetration depth might be shortened due to a possible suppression of  $\text{O}_2$  as a result of high  $\text{O}_2$  demand by methanotrophs (Gerzabek and Reichenauer, 2006). Consequently, the plant growth will be inhibited resulting in reduced transpiration. However, some tree, shrub and grass species seem to tolerate high landfill gas concentrations (Wang et al., 2008; Chan et al., 1997). Wang et al. (2008) studied the effect of a LFG tolerant plant (*Chenopodium album* L.) on  $\text{CH}_4$  oxidation in landfill cover soil. Methanotrophs population and  $\text{CH}_4$  oxidation activity in soils exposed to landfill gas were significantly higher than in soils without landfill gas.

Some plant species (e.g. Typhaceae) are even able to obtain aerenchyma (air-filled spaces in stem and roots), which facilitates the movement of  $\text{O}_2$  to the roots (Bosse and Frenzel, 1997). In addition to their own consumption, roots release  $\text{O}_2$  to the

surrounding anoxic soil. In this way, plants may provide  $O_2$  for methanotrophic bacteria, thus reducing  $CH_4$  emissions (Zak, 2008; Mainiero and Kazda, 2004; Bosse and Frenzel, 1997). However, the complex interactions between these plants and methane-oxidising bacteria should be subjected to further investigation due their relevance for  $CH_4$  oxidation.

In contrast to the above mentioned advantages, plant-mediated transport mechanisms may also affect localised fluxes (Chanton, 2005). There is also the potential for increased gas emissions through the creation of preferential channels by plant roots (Scheutz et al., 2009). In addition, vegetation could also compete with  $CH_4$  oxidisers for nutrients and water resulting in decreased  $CH_4$  oxidation (Hilger and Humer, 2003; De Visscher et al., 1999).

#### 2.4.9 Methane and oxygen supply

Methanotrophic activity depends on the presence of sufficient concentrations of both  $CH_4$  and  $O_2$  at the same time. Hence, their habitat is confined to a narrow layer limited by the downward diffusion of atmospheric  $O_2$  and the upward diffusion of  $CH_4$  from the underlying waste (Scheutz et al., 2009). In addition advective forces (see section 2.2) and the “gas pumping effect” (see section 2.4.6) may be important as well.

Previous studies have indicated that  $CH_4$  removal rates are sensitive to  $CH_4$  concentrations. Higher  $CH_4$  supply increased  $CH_4$  oxidation in investigations of Visvanathan et al. (1999). However, the effect was not proportional to the supply rates and at certain rates a limiting  $CH_4$  oxidation capacity of the soil was observed. Jones and Nedwell (1993) found that counts of methanotrophs increased with rising  $CH_4$  concentrations suggesting that the methanotrophic community had adapted to the presence of elevated  $CH_4$  concentration. In contrast, high  $CH_4$  flow rate from underlying waste can hinder the diffusion of  $O_2$  in the landfill cover resulting in reduced  $CH_4$  oxidation (Abichou et al., 2004). In general, microbial  $CH_4$  oxidation occurs in a zone, where optimal conditions for methanotrophs growth,  $O_2:CH_4$  ratio, retention time and suitable environmental conditions exist. Kightley et al. (1995) and Jones and Nedwell (1993) reported that  $CH_4$  oxidation potentials were greatest where the vertical profiles of  $O_2$  and  $CH_4$  overlapped.

Huber-Humer (2004) investigated the  $CH_4$  oxidation capacity in mature sewage sludge compost exposed to a range of  $CH_4$  supplies. The results showed an upward displacement of the  $CH_4$  oxidation zone in the soil column upon increasing  $CH_4$  loads. At low  $CH_4$  supply (25 l/m<sup>2</sup>d) the main  $CH_4$  oxidation zone was situated at a depth between 40 and 60 cm whereas at high  $CH_4$  loads (355 l/m<sup>2</sup>d) all of the  $O_2$  was already depleted between 0 – 15 cm.

Several researches reported different optimum  $CH_4$  oxidation zones at different depths depending on the moisture content, temperature,  $O_2$  availability and  $CH_4$  supply. For instance, Kightley et al. (1995) found that the maximum zone of  $CH_4$  oxidation is situated between 20 and 30 cm in coarse sand, while Visvanathan et al. (1999) located it between 15 and 40 cm in different landfill cover soils. Generally, the methanotrophic active zone is situated in the upper part of the soil profile (30 – 40 cm), with maximum  $CH_4$  oxidation between 15 – 20 cm below the surface (Scheutz et al., 2004, 2009; He et al., 2008). Figure 2.12 depicts a gas concentration profile measured at a Danish landfill compared with maximum  $CH_4$  oxidation rates obtained in batch incubation experiments, indicating that the optimal  $CH_4$  oxidation zone is located in the upper part of the soil profile.

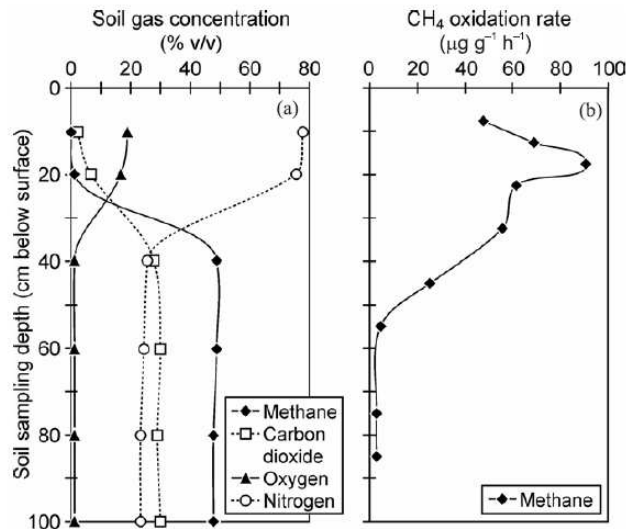


Figure 2.12: Soil gas concentration profiles measured at a Danish landfill and maximum methane oxidation rates obtained in batch incubation experiments vs sampling depth (Scheutz et al., 2009)

Scheutz et al. (2009) concluded that methanotrophic activity is limited by low O<sub>2</sub> concentrations at depth below 60 cm (Scheutz et al., 2009). However, Humer and Lechner (2001c) found in their field experiment that the optimum zone extended from 40 and 90 cm in sewage sludge and MSW compost.

Czepiel et al. (1996, cited in De Visscher et al., 1999) found that O<sub>2</sub> concentration of about 3 % v/v is a threshold for CH<sub>4</sub> oxidation to occur, which means changing O<sub>2</sub> concentration above 3 % v/v will have very little influence on CH<sub>4</sub> oxidation but oxidation will decrease dramatically when the concentration is lower 3 % v/v. The insensitivity to O<sub>2</sub> concentration above 3% v/v can be the reason for the sharp slope in CH<sub>4</sub> concentration profiles with depth.

Based on the assumption that no carbon is converted into biomass, the required O<sub>2</sub>:CH<sub>4</sub> ratio for CH<sub>4</sub> oxidation is 2:1 (see stoichiometric equation 2.3), which is equivalent to 2 l O<sub>2</sub>: 1 l CH<sub>4</sub> and 4 g O<sub>2</sub>:1 g CH<sub>4</sub>, respectively (Huber-Humer, 2004). This would suggest that O<sub>2</sub> availability in landfill cover soils is most likely the limiting factor. However, as biomass is accumulated, generally less O<sub>2</sub> is required for CH<sub>4</sub> oxidation (Huber-Humer, 2004).

## 2.5 Construction and design of a biocover

A biologically active cover (biocover) or methane oxidation layer is a landfill cover system that has been designed to mitigate CH<sub>4</sub> emissions by optimising conditions for microbial CH<sub>4</sub> oxidation. In general, a biocover is a layered system with different layers serving different purposes. Typically, the cover consists of a basal gas distribution layer with high gas permeability to homogenise and distribute landfill gas to the overlying oxidation and vegetative layer, where living conditions for methanotrophic populations are optimised. An appropriate vegetative layer may also serve as an evapotranspiration cap to reduce leachate. Figure 2.13 shows a simple conceptual scheme of a methane oxidation layer.

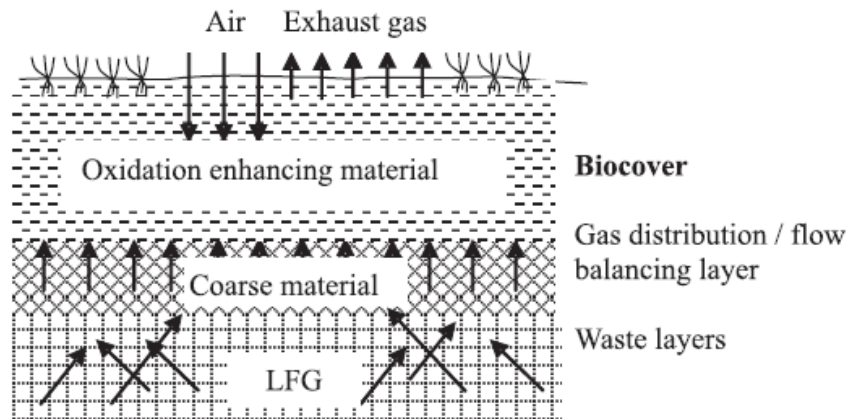


Figure 2.13: Schematic illustration of a methane oxidation layer (Huber-Humer et al., 2008b)

Generally, the biocover effectiveness relies on the use of a suitable carrier and a proper technical design and dimensioning of the landfill cover in order to oxidise variable rates of  $\text{CH}_4$  (Bogner et al., 2010; Huber-Humer et al., 2008a; Humer and Lechner, 2001). As  $\text{CH}_4$  fluxes typically exhibit high spatial inhomogeneity across a landfill site, the installation of a coarse gas distribution layer is crucial to achieve a more uniform supply and a slowing down of gas fluxes. Importantly, biocovers must maintain high  $\text{CH}_4$  oxidation rates even during different seasonal climatic situations leading to changes in soil moisture, temperature and other variables (Bogner et al., 2010; Humer and Lechner, 2001a). According to recommendations from laboratory and field studies an efficient biocover will provide a long-term nutrient supply (N, P), a high temperature-insulating capacity, good porosity and gas permeability as well as a high water holding capacity (Scheutz et al., 2009; Kettunen et al., 2006; Humer and Lechner, 2001a). The design of the biocover system should accommodate a methane load of at least  $4 \text{ l CH}_4/\text{m}^2\text{d}$  to ensure efficient mitigation (Huber-Humer et al., 2008a). Moreover, a biocover should be structurally stable and settle very little to maintain its porous cover (Scheutz et al., 2009; Humer and Lechner, 1999a). As already mentioned mature or specific compost materials have proven to be a suitable substrate carrier enhancing microbial  $\text{CH}_4$  uptake (see section 2.4.2).

### Engineered approaches

Currently designed biocover systems engineered to optimise  $\text{CH}_4$  oxidation have demonstrated huge potential to mitigate residual landfill  $\text{CH}_4$  emissions (Stern et al., 2007; Barlaz et al., 2004; Huber-Humer, 2004). In general, biocover design and dimensioning can vary in order to meet local site-specific conditions and depend on the respective climate conditions, expected gas fluxes, the purpose of the cover (final or temporary), characteristics of substrate carrier (settling behaviour, oxygen-penetration depth) and the intended after-use of the site (Humer-Huber et al., 2008b). Biocovers can be potentially applied in the absence of a gas collecting system or as a complementary method to an active system to capture escaping emissions. In addition, such systems can be used to mitigate residual emission from MBT waste or closed old landfills as emissions are too low for energetic use. Natural attenuation could be especially favourable at smaller landfill sites where the installation of a gas collection system is not technically and economically feasible. Moreover, biocovers can also be combined with forced in-situ aeration as a postclosure remediation method for old landfills. Figure 2.14 shows various specialised landfill biocovers using compost materials as a substrate carrier during different climatic conditions.

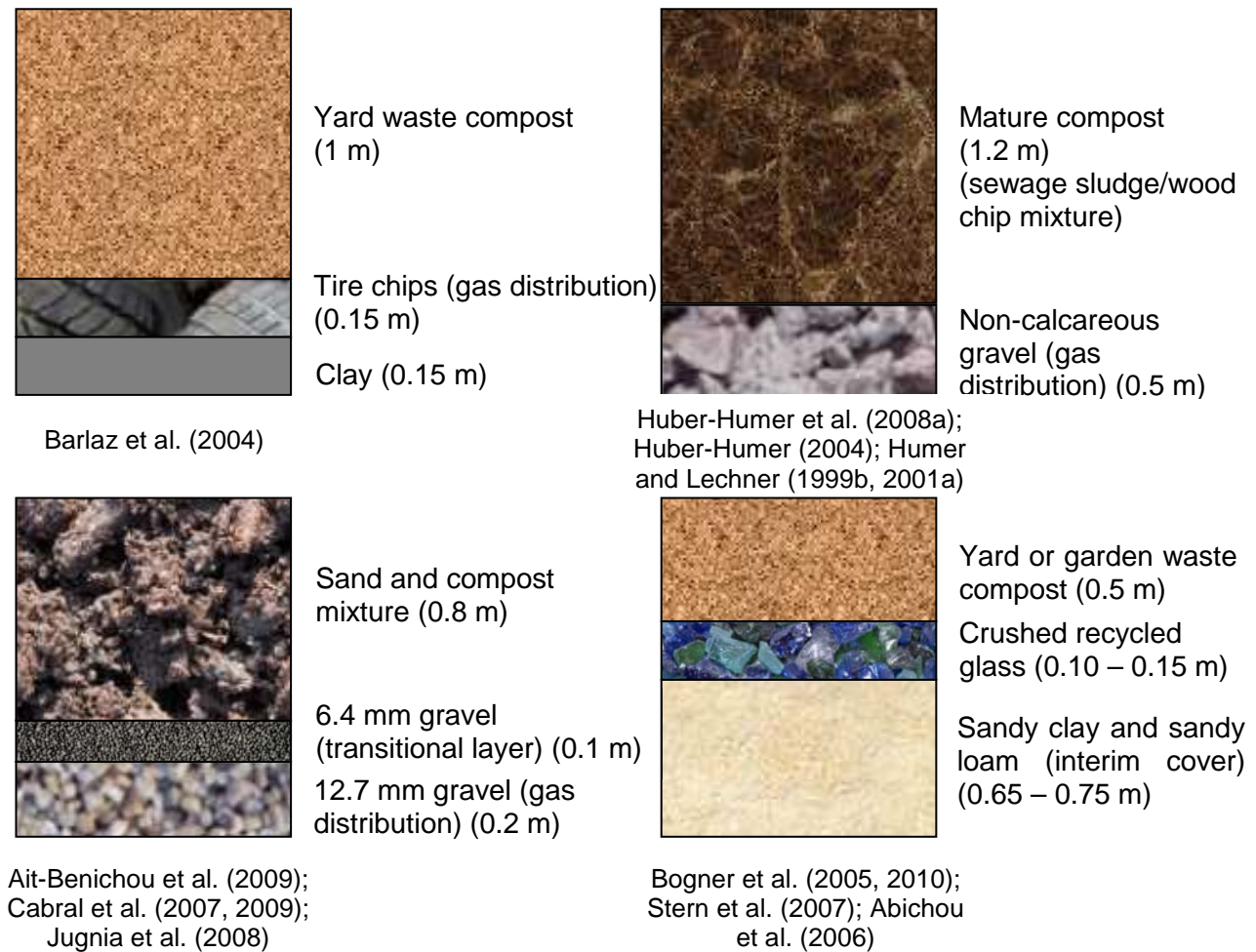


Figure 2.14: Engineered biocover designs

A field trial was conducted on the Outer Loop Landfill in Louisville, Kentucky, to assess a biocover system in parallel with 1 m clay soil cover for mitigating  $\text{CH}_4$  emissions (Barlaz et al., 2004). The biocover system consisted of 0.15 m thick base layer of clay over the waste, followed by a 0.15 m gas distribution layer of tyre chips and 1 m ground yard waste compost (35 – 50 % brush) previously windrow-composted for three months. A variety of broad leaf vegetation became spontaneously established on both covers.  $\text{CH}_4$  measurements using static chambers (see section 2.6.1) were performed while the gas collection system was operating and after the system was turned off. The biocover consumed atmospheric  $\text{CH}_4$  or had almost zero  $\text{CH}_4$  emissions ( $-1.73 - 1.33 \text{ g/m}^2\text{d}$ ) during the 14 months measuring campaign even when the gas collection system was turned off.  $\text{CH}_4$  emissions from the soil cover were more variable with relatively high fluxes ( $> 15 \text{ g/m}^2\text{d}$ ) due to deactivation of the gas collection system and were mainly associated with desiccation cracks. Both covers showed atmospheric  $\text{CH}_4$  uptake half of the time when the the gas collection system was operational. With the gas collection system off, the biocover continued to consume atmospheric  $\text{CH}_4$  about 60 % of the time compared to only 12 % on the conventional soil cover. When positive emissions were measured, stable isotope measurements (see section 3.3.6) showed that an average of 55 % of  $\text{CH}_4$  was oxidised in the biocover but only 21 % in the soil cover. In addition, measurements of trace organic compounds indicated that the biocover reduced these emissions to a greater extent than the soil cover. Barlaz



et al. (2004) concluded that compost-made biocovers serve both to reduce emissions through biotic mitigation and as deterrents to soil cracking.

Humer and Lechner (1999b, 2001a) developed some of the first prototype biocovers using various composted materials. The field trial assessment of the CH<sub>4</sub> removal effectiveness of compost as a landfill cover for middle-European climatic conditions was carried out on two different Austrian MSW landfills between spring 1999 and winter 2002 (Humer and Lechner, 2001a; Humer-Huber, 2004). Based on these experiences a simple two-part cover system proved to be most effective. The biocover system consisted of a 0.3 – 0.5 m coarse gravel layer (minimum particle size 16/32 mm) overlain by a layer of up to 1.2 m of mature, well-structured compost. Results showed almost 100 % CH<sub>4</sub> capture when 1.2 m compost was used in conjunction with a gravel under bed, and 68 – 74 % CH<sub>4</sub> capture when only compost (about 30 – 40 cm) was used (Humer and Lechner, 2001a). In general, the coarse mixture of sewage sludge compost with wood chips had a much better temperature insulating capacity during winter than a fine-sieved MSW-compost (Huber-Humer, 2004). The authors concluded that high CH<sub>4</sub> oxidation rate was mostly associated with the installation of a coarse gas distribution layer for uniform CH<sub>4</sub> fluxes and the good temperature insulation effect due to the sufficient dimensioning of the cover and the adequate porosity of the used material. Presently, at least five closed MSW-landfills or section of landfills in Austria are fitted with this biocover design, which has been officially approved as an acceptable interim MSW landfill cover for a period of about 20 years. These biocovers are serving either as the sole means to mitigate CH<sub>4</sub> emissions or complement an operating gas extraction system (Huber-Humer et al., 2008b).

Two field-scale biocovers (PMOB-1 and PMOB-3B) composed of a mixture of sand and compost have been installed at the St-Nicéphore landfill in Quebec in order to assess the efficiency of the covers in reducing CH<sub>4</sub> emissions (Ait-Benichou et al., 2009; Cabral et al., 2007, 2009; Jugnia et al., 2008). The biocover PMOB-1 included a 0.8 m thick substrate layer consisting of a sand/compost mixture (1:5 v/v) underlain by a 0.1 m thick transitional layer of 6.4 mm net gravel and 0.2 m thick gas distribution layer of 12.7 mm net gravel above a 3.5 year old buried waste mass. The compost was composted from a mix of municipal sewage sludge and sludge from the pulp of paper and agri-food industries. The biocover PMOB-3B consisted only of a 0.3 m thick substrate layer above 0.1 m of 6.4 mm net gravel as a transitional layer and 0.8 m of 12.7 mm gravel layer. For the substrate layer a coarser material was used that resulted from mixing one volume of the same material used as substrate in PMOB-1 with one volume of 6.4 mm gravel. In contrast to PMOB-1, PMOB-3B was fed with biogas coming from a well. In order to isolate the biocover from the existing silty cover and the waste mass, PMOB-3B was lined with a 1 mm thick HDPE geomembrane. During the first four months of monitoring (Jul. – Oct. 2006), CH<sub>4</sub> emissions from PMOB-1 remained low and varied between 2.5 to 30 g CH<sub>4</sub>/m<sup>2</sup>d (Jugnia et al., 2008) following the closed dynamic chamber method (see section 2.6.2). A high CH<sub>4</sub> emission flux of 210 g CH<sub>4</sub>/m<sup>2</sup>d was recorded once when the barometric pressure was the lowest during this study. The higher water content (51 – 64 % v/v) through the depth profile acted as a physical barrier to O<sub>2</sub> penetration and therefore the uppermost 0 – 10 cm layer appeared to be the most important layer for CH<sub>4</sub> oxidation. Stable isotope analyses were performed for both biocovers on selected dates from July to September 2007 (Cabral et al., 2009). The results showed that the substrates used in the two biocovers were able to promote CH<sub>4</sub> oxidation which ranged from 2.9 % to 89.7 % (at a depth of 0.1 m) in PMOB-1 and

was equal to 88.7 % for a representative profile in a relatively dry period in PMOB-3B. Quite high  $\text{CH}_4$  emission fluxes (21.4 – 458.2 l/m<sup>2</sup>h) were measured on five occasions while the surface flux from PMOB-3B was relatively low (4.3 l/m<sup>2</sup>h). Only on two selected dates PMOB-1 had almost zero fluxes (not detectable – 1 l/m<sup>2</sup>h). In some cases, poor aeration of the substrate in PMOB-1 was observed leading to quite low efficiencies which may be linked to the degree of water saturation, the magnitude of the biogas flux and the substrate temperature.

Experiments were performed at the Leon County Landfill in Florida to study biocover effectiveness in a subtropical environment and the use of multiple recycled materials for biocover construction (Bogner et al., 2005, 2010; Stern et al., 2007; Abichou et al., 2006). Two test areas were established. On the first site (S1) a 0.5 m substrate layer of a 3 year old composted garden waste was placed over 0.1 – 0.15 m of crushed recycled glass distribution layer overlying a 0.65 – 0.75 m existing clay cover (sandy clay and sandy loam). Test area S4 included a shallow (0.3 m) or deep (0.6 m) substrate layer consisting of freshly ground garden waste (to compost in-situ). Both substrate layers were laid on top of 0.15 m of crushed fluorescent glass tubes as a gas distribution layer and a very thin interim cover consisting of 0.15 m of compacted sandy clay. On both test areas (S1 and S4) the interim covers acted as a control. Based on stable isotope analysis the biocover at site S1 consumed twice as much  $\text{CH}_4$  (64 %) as the control cells (30 %) (see Figure 2.15).

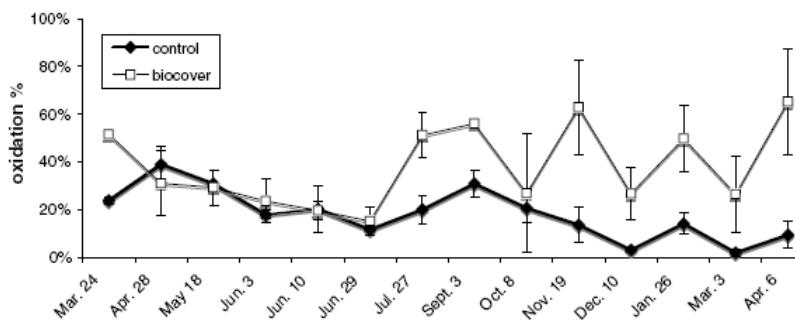


Figure 2.15: Percent oxidation of  $\text{CH}_4$  in the control and the biocovers cells, calculated from isotope data only (Stern et al., 2007)

Following static chamber measurements, mean  $\text{CH}_4$  emission rates from the control cells (10.6 g/m<sup>2</sup>d) were significantly greater than flux from the biocover (2 g/m<sup>2</sup>d) (see Figure 2.16) leading the authors to conclude that the thickness and moisture-holding capacity of the biocover increased the retention times for transported  $\text{CH}_4$  and provided better protection against desiccation (Stern et al., 2007). As a consequence a higher fraction of  $\text{CH}_4$  could be oxidised.



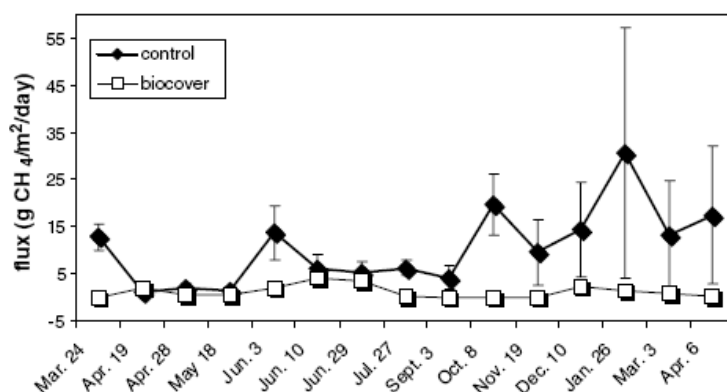


Figure 2.16: Mean CH<sub>4</sub> emission rates (g CH<sub>4</sub>/m<sup>2</sup>d) from control and biocover cells (Stern et al., 2007)

At the second test area (S4) it was determined that CH<sub>4</sub> uptake of the deep biocover varied between 20 % and 70 %, with lower but overlapping values for the shallow biocover and the control cells (0 – 40 %) (Bogner et al., 2010). Gross CH<sub>4</sub> fluxes from the shallow biocover and the control cells (up to 300 – 700 g/m<sup>2</sup>d) were 10-fold higher than the 30 – 70 g CH<sub>4</sub>/m<sup>2</sup>d calculated for the deep biocover. These results indicate that low-dimensioned and poorly designed covers do not achieve as high CH<sub>4</sub> oxidation capacities as properly designed biocovers, as also demonstrated by Humer and Lechner (2001a).

## 2.6 Determination of landfill gas emissions

To deal with the difficulties of surface gas flux measurements, a number of methods have been designed, each with associated strengths and weaknesses. Several methods are used to quantify the emission rate for small areas, while others are used for large surface areas (e.g. for the entire landfill). For small area measurements, techniques are used, such as the chamber method and the concentration-gradient method based on Fick's law, whereas for large area measurements, micrometeorological methods, the trace method and infrared spectroscopy are applied (Abushammala et al., 2009; Fowler, 1999; Tregoures et al., 1999). Up to now, no single technique is recognised as 'the standard' methodology. In the available literature the different methods used for measuring landfill gas emission rate from landfill sites have been reviewed (Tregoures et al., 1999), however each technique has unique advantages and disadvantages. The choice will depend on economic constraints, measurement objectives and the sampling site.

Following this, the research in this study concentrates on the chamber methods which are the most common technique for the measurement of gas fluxes from different ecosystems. Chamber techniques have been widely used in wetlands (Van Huissteden et al., 2008), agricultural and semi-natural soils (Kusa et al., 2008), landfills (Chen et al., 2008; Stern et al., 2007; Barlaz et al., 2004), forest ecosystems (Von Arnold et al., 2005), but also active volcanic areas (Carmada et al., 2009, Cardellini et al., 2003, Chiodini et al., 1998) measuring various gaseous emissions, including CO<sub>2</sub>, N<sub>2</sub>O, NO<sub>x</sub>, CH<sub>4</sub>, selenium (Se) and volatile organic compounds (VOCs) such as volatile pesticides (Reichmann and Rolston, 2002; Laville et al., 1997; Hutchinson and Mosier, 1981).

Chamber techniques are based on the principle to restrict the volume of air with which gas exchange occurs so as to magnify changes in gas concentration in the

headspace. The aim is to have a linear increase indicating a constant flux from the surface. In general, chamber techniques can either measure (i) the rate of gas accumulation (closed system) or (ii) the instantaneous gas flux (open system). Two types of closed accumulation chambers have been developed, which are characterised by the absence (static) or presence (dynamic) of airflow inside the chamber. In Figure 2.17, the different types of chamber methods are depicted.

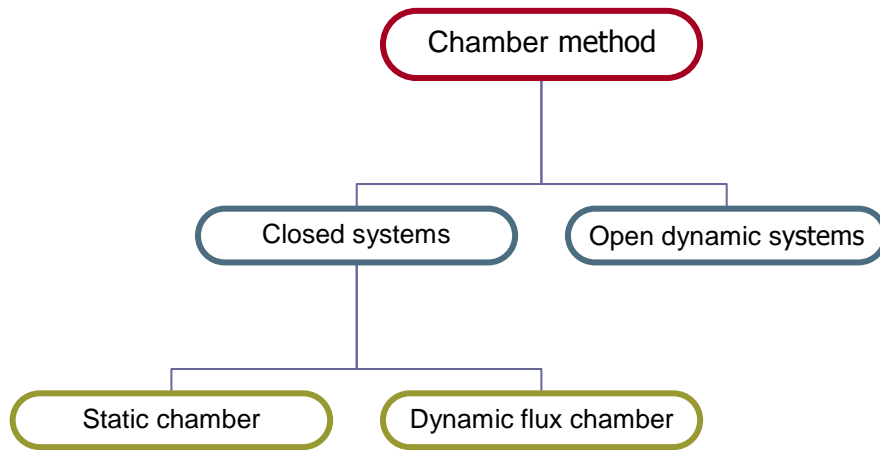


Figure 2.17: Different systems of chamber methods

In previous research studies chamber systems have been either:

- Process orientated (investigation of production, emission or mechanisms of exchange),
- Designed for long-term flux measurement (traceability of the seasonal or annual time course in fluxes of trace gases which show large temporal cycles); and
- Complementary flux measurements (Fowler, 1999).

The advantages, limitations, assumptions and computation of exchange rate differ greatly among the different designs.

Current types of chamber used usually vary in basal sampling area, from 0.1 to 100 m<sup>2</sup>. The upper limit represents the megachamber approach such as that used by Smith et al. (1994) and Galle et al. (1994). Currently, chambers < 1 m<sup>2</sup> are the most common tools since larger ones are difficult to transport. However, considerations of the basal area depend on the practicality of installing chambers as determined by the terrain of the sampling site (Livingston and Hutchinson, 1995). Closed systems have been used more often than open dynamic chambers as they are mechanically simpler and small gas fluxes are easier to detect. In addition to the choice of general enclosure type, decisions are required concerning the deployment, chamber geometry, fabrication materials, temperature control and measurement equipment to monitor concentration changes within the enclosure (Livingston and Hutchinson, 1995).

Results obtained by chamber methods, both closed and open, can be statistically evaluated to determine whole landfill fluxes by establishing statistically-based sampling schemes. However, a large number of samples ( $\geq 100$ ) are needed for the descriptive statistics of spatial data based on the arithmetic mean as only point emissions can be measured (Bogner et al., 1997a). Given the high spatial and temporal variability in LFG fluxes, it might be helpful to use several enclosures

simultaneously or a higher number of replicates in order to obtain a representative estimate. General errors in gas flux measurements are related to the chamber effects on modifications of the microclimate (e.g. temperature, humidity), physical disruption of the surface, pressure-induced gas flows in open chamber designs, inhibition of fluxes through concentration and pressure build-up in closed chambers and perturbations of the natural conditions (Denmead, 2008; Asman et al., 1999).

### 2.6.1 Closed static chamber

Kunz and Lu (1980, cited in Bogner et al., 1997a) reported the first use of a closed static chamber measuring CH<sub>4</sub> fluxes from a New York site. Nowadays, this enclosure method has been recognised as the simplest technique to measure gas emission fluxes at selected points on the surface of a landfill (Bogner et al., 1997a; Livingston and Hutchinson, 1995). Generally, an open-bottom chamber (cylindrical or rectangular) made, for instance, of plastic or metal is placed over a small area of soil surface so that the gas concentrations increase continually in the head space. A sealing has to be done, otherwise air can infiltrate into the chamber compromising the sample. The gas emission flux ( $F$ ) at the surface is calculated from the relationship:

$$F = \frac{V}{A} \cdot \frac{dC}{dt} \quad (2.4)$$

where  $V$  is the volume of the head space,  $A$  the surface covered by the chamber,  $C$  the change in gas concentration and  $t$  is the sampling time. In addition to the gas concentration, temperature ( $T$ ) in the chamber and barometric pressure ( $P$ ) have to be measured to convert the change in volumetric concentration to a mass flux (g/m<sup>2</sup>d). When temperature and pressure correction are applied, the following equation results:

$$F_{cor} = F \cdot \frac{273.15}{273.15 + T} \cdot \frac{P}{1013} \quad (2.5)$$

Typically, air samples are taken periodically from the head space with a gas syringe and the gas concentrations are measured later in the laboratory, for example, using a gas chromatograph (Chanton and Liptay, 2000; Bogner et al., 1997b). The enclosure period should be no longer than 40 min and at least three time points are required for flux calculation (e.g. 0, 20, 40 min) (Livingston and Hutchinson, 1995). Fans are often used to obtain better mixing of the air inside the chamber ensuring representative sampling. Two closed static chambers are shown in Figure 2.18.

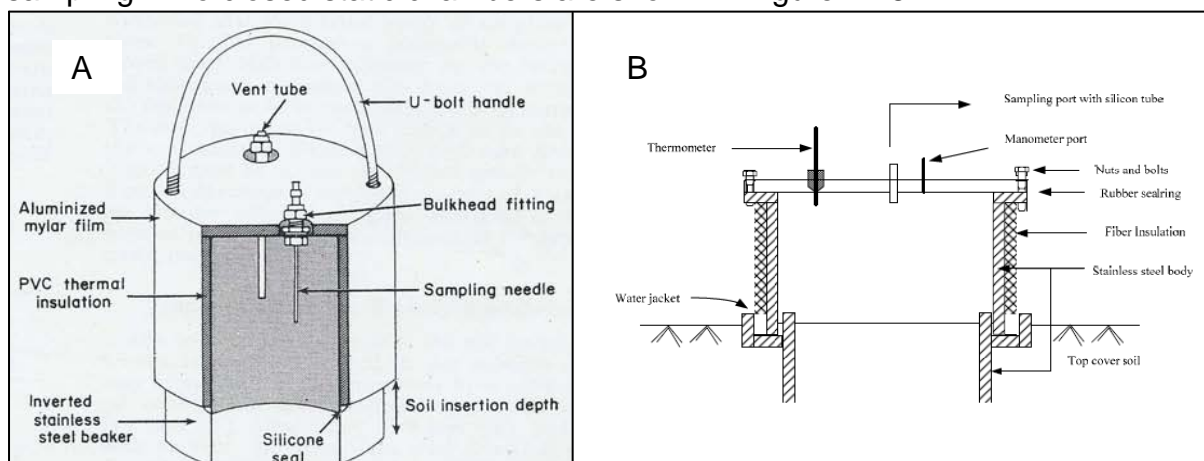


Figure 2.18: Closed static chamber A: (Hutchinson and Mosier, 1981), B: Visvanathan et al. (2004)

### Strengths and weaknesses

The static chamber technique is low in cost, simple in concept and operation and requires no power in the field. It is especially useful for addressing research objectives needing spatial and temporal variability of fluxes at a small scale (Abichou et al., 2006; Bogner et al., 1997a). Further advantages include portability and adaptability to a wide variety of field conditions. Since gas collected inside the static chamber is not being continually diluted with external air (as is the case with open dynamic chambers), this method can be highly sensitive detecting small fluxes or even net uptake of atmospheric CH<sub>4</sub> (Denmead, 2008; Perera et al., 2002; Whalen and Reeburgh, 1990).

However, the static chamber may not give reliable results as the chamber itself influences actual emission rates from the soil surface. Once the chamber is placed on top of a landfill cover during a measurement, the gas concentration gradient is altered reducing diffusional flux and thereby actual emission rate is suspected to be underestimated (see Figure 2.20). Matthias et al. (1978, cited in Perera et al., 2002) reported that the actual flux could be as much as 55 % higher than the calculated flux from the chamber measurement. If the flux into the chamber is controlled by pressure difference, a build-up of pressure above barometric pressure will occur in the chamber decreasing the gas flux with time (Rolston, 1986, cited in Bogner et al., 1997a). Due to these reasons, the chamber can only be used for relative comparison of fluxes from place to place. In addition, natural wind conditions, an increase of temperature inside the chamber and gas sampling can induce pressure differences between the chamber and outside air creating artefact gas fluxes. Denmead (1979), for example, demonstrated that a pressure deficit of only 100 Pa between the atmosphere inside and outside the chamber could alter the gas emission rate by a factor of 10. Moreover, closed chambers may cause a by-pass and enhanced emission fluxes outside the measuring chamber when placing it on top of a coarse-textured cover material with high gas permeability (Huber-Humer et al., 2009). Therefore, installation of a vent tube in a chamber wall to permit pressure equilibration is a wise precaution. Hutchinsons and Mosier (1981) provided guidelines for appropriate vent dimensions for effectively transmitting ambient pressure fluctuations to the enclosed space (for wind speeds up to 4 m/s) while minimising loss of the accumulating gas by diffusion to the outside (< 1 % diffusion loss). Optimum vent tube diameter and length for selected wind speeds and enclosure volume are summarised in Figure 2.19.

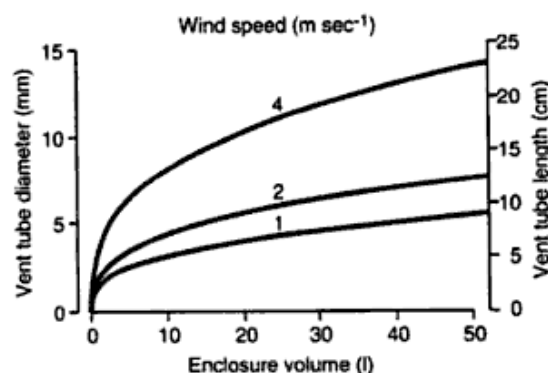


Figure 2.19: Optimum vent tube diameter and length for selected wind speeds and enclosure volume as described by Hutchinson and Mosier (1981)

The errors in flux measurement by closed chambers largely depend on the configuration of the chamber and the gas flux rate. Based on experiments by Perera et al. (2002), the smaller the chamber and higher the flux rate, the higher the percentage error in measurement. Most of the artefacts introduced by the chamber itself can be overcome with sophistication of the equipment or quantified by experiment using a test bench (Fowler, 1999).

### 2.6.2 Closed dynamic chamber

Compared to static chamber, closed dynamic chambers are more sophisticated. Their operating principle involves air circulation in a closed loop between the head space and a gas analyser. The rate of gas enrichment of the recirculated mixture can be monitored continuously and, thus, any inhibition of the flux through a build-up in head space concentration can be detected. The increase of gas concentration over time will be calculated with the equation used for static chamber systems.

#### Strengths and weaknesses

As air is pumped between chamber and gas analyser the gas inside the chamber is mixed minimising negative effects of a possible temperature increase inside the chamber. However, this configuration can also cause pressure effects. Apart from that, the closed dynamic chamber has the same advantages and disadvantages as static chambers except that power is needed for air pumping.

Figure 2.20 shows the simultaneous time course of the concentrations of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O in the head space of a closed dynamic chamber installed in a fertilised field (Breuer et al., 2000, cited in Denmead, 2008). The plot indicates that towards the end of the 18-min measurement period the gas fluxes (CO<sub>2</sub>, N<sub>2</sub>O) are inhibited, even over this short enclosure period, due to high gas concentration in the head space. This is a particular problem for closed chamber methods. Hence, the time series of the concentration change should be investigated in order to establish an appropriate sampling period (Denmead, 2008).

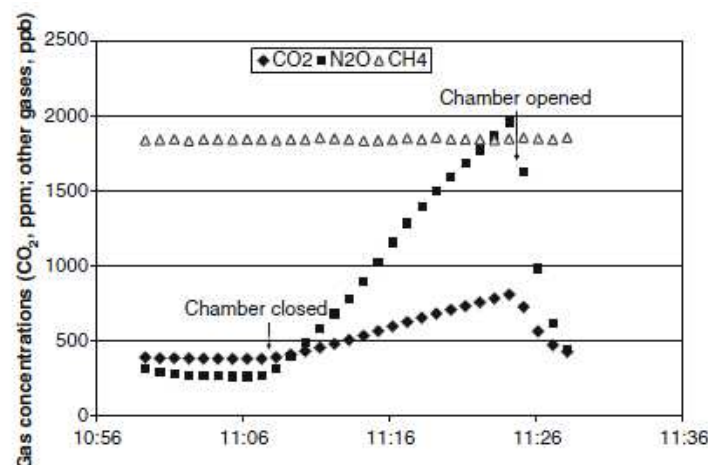


Figure 2.20: Time course of gas concentrations in the head space of a closed dynamic chamber (Breuer et al., 2000, cited in Denmead, 2008)

### 2.6.3 Open dynamic systems

An alternative flux measurement method is the open dynamic system in which a constant flow of outside air is maintained through the head space of the chamber and the difference in concentration between inlet and outlet is measured. Usually, air is

blown or pumped into the enclosure at a fixed rate and gas concentration is measured using appropriate detection instruments. The gas emission flux is calculated as follows:

$$F = \frac{Q}{A} \cdot (C_{out} - C_{in}) \quad (2.6)$$

where  $F$  and  $A$  are as defined previously,  $Q$  is the volume flow rate,  $C_{out}$  is the gas concentration in the air leaving the chamber and  $C_{in}$  the gas concentration in the air entering the chamber. Various possible configurations and operating features of open dynamic chambers have been reported in the literature (Müller et al., 2009; Röder et al., 2004; Gao et al., 1997; Pokryszka et al., 1995), as can be seen in Figure 2.21.

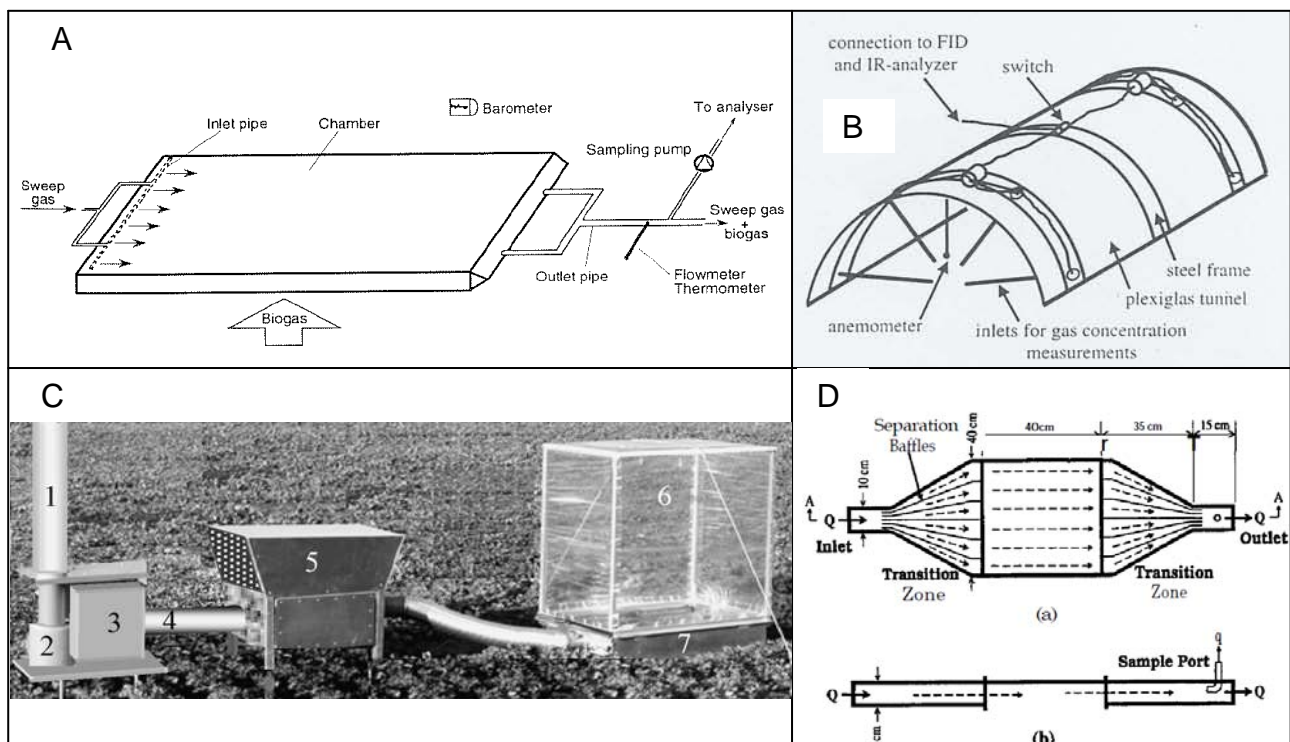


Figure 2.21: A: Dynamic flux chamber (Pokryszka et al., 1995); Open wind tunnel (Röder et al.; 2004), C: Canopy chamber system (Müller et al., 2009); Top-view and cross-section of the flow-through chamber system (Gao et al., 1997)

In the dynamic flux chamber introduced by Pokryszka et al. (1995) a sweep gas flow (inert gas) is injected via a perforated pipe at the entry of the measuring chamber. At the outlet the sweep gas/biogas mixture is collected passively and the flow rate is measured by a hot-wire anemometer. Gas samples from the outlet pipe are pumped to the gas analyser where concentrations of biogas constituents are measured.

An open wind tunnel was developed by Röder et al. (2004), in which the natural wind velocity is used instead of a pump or blower to carry the air and the emitted gases through the tunnel. In the rear and the front section of the tunnel gas concentrations are detected while at the outlet wind speed is measured.

The canopy chamber is an automated system which consists of a tube for supplying ambient air to the system, a radial blower, a central measuring and controlling unit and the canopy enclosure (Müller et al., 2009). Air and soil temperature as well as soil moisture can be measured within the chamber.



The flow-through chamber system consists of an inlet transition zone, a square main body, and an outlet transition zone (Gao et al., 1997). In order to conduct and spread the flowing air uniformly across the soil surface, six equally-spaced channels are installed in both inlet and outlet transition zones. Gas samples can be taken at the inlet and outlet of the chamber.

### Strengths and weaknesses

In recent studies, open dynamic chambers have been preferred as these minimise the difference between ambient conditions and those within the enclosure since the ambient air is continuously flowing through the chamber (Fowler, 1999; Gao et al., 1997). The increase in gas concentration above background can be controlled by the volume flow rate and, thus, the risk of inhibiting the surface flux can be reduced. In addition, open systems can detect a wider range of gas concentration (Pokryszka et al., 1995).

However, because of the small magnitude of the concentration increase (emitted gas is diluted with air), open dynamic chambers may not be able to detect small fluxes. Besides, the air flowing in the chamber may change the pressure gradient between soil-gas phase and the chamber interior (Gao et al., 1997) depending on the degree to which the chamber is open to the atmosphere (Fang and Moncrieff, 1998). A change in the pressure gradient may create an additional advective mass flow of the target gas or an influx of gases from outside the enclosure, in particular, when the air in the enclosure flows relatively fast compared to the wind outside the chamber. This pressure deficit may cause an overestimation of the emission rate (Fang and Moncrieff, 1998; Gao et al., 1997). Another potential problem with open dynamic chambers is associated with the air-flow pattern within the enclosure (Gao et al., 1997). If the inlet and outlet of the chamber are not designed properly, the air flowing through the enclosure may not sweep over the entire covered surface. As a result, local stagnant air zones are created inside the chamber which can cause a spatially variable emission flux. This is also true for large closed dynamic chambers.

In practice, both closed and open chamber methods are subject to a wide range of errors, including modification of environmental conditions within the chamber relative to those in the field (Fowler, 1999). Anomalous pressure effects seem to pose major challenges for accurate chamber-based measurements given their difficulty to measure or control them (Perera et al., 2002).

## **2.7 Existing standards and legal ordinances**

Currently, there are no specific technical standards or legal specifications on the setup of biocovers in Austria or in the European Union. In spite of the numbers of research studies regarding the applicability of biocovers, standardised implementation and construction criteria within the European Union are still missing. However, a national “Technical Guideline for Biocovers” has been drafted under the auspices of the “Austrian Association for Management of Contaminated Sites” (see Huber-Humer et al., 2008a).

In Austria, implementation of biocovers may be affected by some specific rules and standards including the “Austrian law on remediation of inherited waste (“Altlastensanierungsgesetz”), the “Austrian Ordinance on Composting” and the “Austrian Landfill Ordinance”.

According to paragraph § 2 in the “Austrian law on remediation of inherited waste” (ALSAG, BGBl. Nr. 299/1989, amended by BGBl. I Nr. 40/2008), waste materials (including waste composts) used for constructing landfill covers, including methane oxidation layers and reclamation layers, are subject to “contaminated site liability” (Altlastenbeitragspflicht). The “contaminated site liability” is a tax for each ton of waste material put on a landfill site, which has to be paid by the landfill operator. However, reclamation layers and temporary covers (which also serve the purpose of oxidising methane) will be excluded from the “contaminated site liability”, if they are implemented in full compliance with criteria defined in Appendix 3, sections 4.5 and 6.1 of the “New Landfill Ordinance 2008” (BGBl. II 39/2008).

In certain circumstances, the use of compost for constructing a biocover is subject to the “Austrian Ordinance on Composting” (BGBl. II 292/2001), in particular paragraph § 6 (3) which limits the quantities of compost applied for the construction of a reclamation layer. This is the case, if high-quality compost is used meeting the quality criteria of the Compost Ordinance. As this compost is no longer classified as waste, the “contaminated site liability” according to ALSAG can be disregarded.

The “Austrian Landfill Ordinance” (BGBl. II 39/2008) provides requirements for intermediate covers as well as for temporary permeable covers (acting as a biocover for a max. period of 20 years), which can be used to minimise methane emissions from landfills (or compartments) with high amounts of biodegradable waste. According to paragraph § 29 (4), the use of compost for constructing an intermediate cover is prohibited whereas it is allowed for temporary covers, except compost made of municipal solid waste. The use of compost for constructing a final reclamation layer is only allowed in accordance with regulations in the “Austrian Ordinance on Composting”. Appendix 3, section 6.1 provides specifications regarding biocover effectiveness and a monitoring program for temporary covers. Gaseous emissions from a whole landfill site must not exceed an average of 5 kg CH<sub>4</sub>/m<sup>2</sup>year and single values occurring at “hot spots” may exceed this up to a maximum of 10 kg CH<sub>4</sub>/m<sup>2</sup>year. According to the monitoring program, the temporary cover surface has to be surveyed every quarter for possible methane emissions using a grid-based FID-mapping (Flame-Ionisation-Detector). Emission rates (CO<sub>2</sub> and CH<sub>4</sub>) have to be quantified at least twice a year taking into account the grid-based FID-survey. In addition, landfill operators are committed to submit a project to the local authorities concerning the covering of the landfill site after the temporary period of 20 years. (Huber-Humer et al., 2008a)



### 3 MATERIALS AND METHODS

#### 3.1 Development of a measuring system

The primary considerations in the choice of method for measuring gas fluxes include design of the lysimeters under study (see section 3.3.1), gas species in question ( $\text{CO}_2$  and  $\text{CH}_4$ ), availability of analytical instruments and cost. After extensive literature research, it was decided to develop a measuring system based on the closed dynamic chamber technique. Specific efforts included design and development of the measuring system and testing of this system under different known  $\text{CH}_4$  and  $\text{CO}_2$  fluxes to demonstrate the feasibility of the measurement technique.

##### Prototype

The technique used in this work was inspired by the accumulation chamber method developed by INERSIS (Institut National de l'Environnement et des Risques) for estimating  $\text{CH}_4$  emissions from MSW landfills (Savanne et al., 1995; Tregoures et al., 1999). In the first instance a prototype was developed to test the operating principle only for  $\text{CH}_4$  fluxes, as illustrated in Figure 3.1.

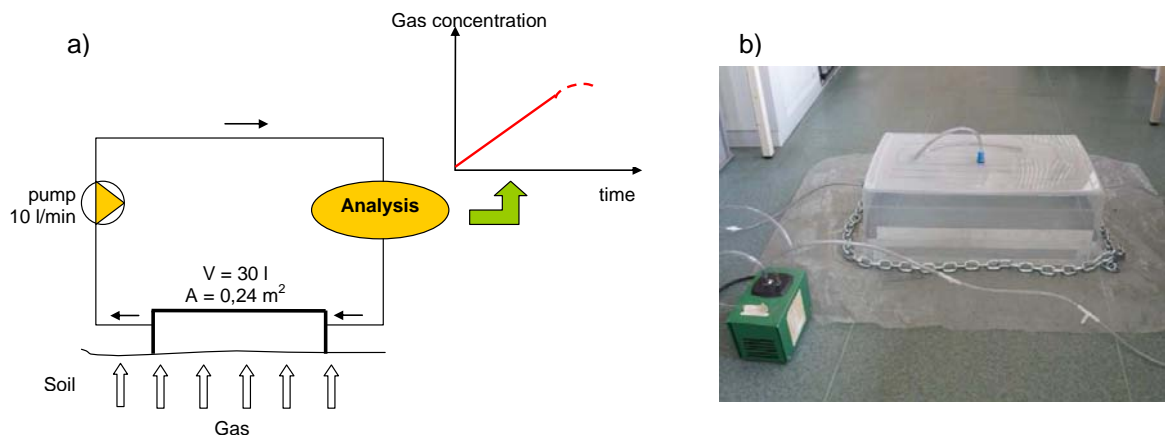


Figure 3.1: Schematic of the measurement system (a) and illustration of the developed prototype (b)

The prototype was a rectangular plastic box covering an area of  $0.24 \text{ m}^2$  with an enclosed volume of  $30 \text{ l}$ . The internal atmosphere was renewed using an external pump with a flow rate of  $10 \text{ l/min}$ . The methane flux obtained outside the chamber was analysed using a portable Flame Ionisation Detector (FID) equipped with a  $1 \text{ l/min}$  internal pump.

The chamber had an attached plastic skirt held in place on the soil surface with a chain. In order to avoid physical disruption of the cover materials upon chamber installation, the use of an anchor or collar for sealing was avoided. Three holes, to accommodate fittings for vent tube and gas sampling ports for gas recirculation, were drilled in the chamber top and sidewalls, respectively.

First test runs were performed in the laboratory and in the field revealing the feasibility of the chamber. Given the high spatial variability in LFG fluxes, a small chamber ( $0.24 \text{ m}^2$ ) may not give representative estimates of the gas fluxes emitted from the experimental biocovers in the landfill lysimeters ( $4 \text{ m}^2$ ) unless several

chambers are used simultaneously or the number of replications is increased. Kaiser et al. (1996) reported higher variability from small boxes ( $0.0078 \text{ m}^2$ ) than from larger boxes ( $5.76 \text{ m}^2$ ). Therefore, it was decided to build a large chamber with the same operating principle covering almost the whole area of the lysimeter chamber. The challenge was to include the aeration pipe ( $D = 6 \text{ cm}$ ) of the lysimeter chamber into the design of the accumulation chamber.

#### Further development

The chamber was constructed by covering a square wooden base frame with a plastic tarp which had a hole in the middle reserved for the aeration pipe of the lysimeter chamber (see Figure 3.2). It had an internal length of 190 cm and therefore covered an area of  $3.61 \text{ m}^2$ . The chamber was equipped with one gas sampling port at the inlet and one at the outlet. The sampling port at the outlet was connected to a portable FID via an external pump with a flow rate of  $55 \text{ l/min}$ . The large chamber also had an attached plastic skirt.



Figure 3.2: Illustration of the constructed accumulation chamber

Laboratory tests were conducted to study the effect of chamber behaviour on flux measurement with a constant emission source of pure  $\text{CH}_4$ , which was introduced directly into the chamber base via a hose. The  $\text{CH}_4$  fluxes were regulated by a flow controller based on the rotameter principle. The rotameter was carefully calibrated by a bubble flow meter. For the calculation of the  $\text{CH}_4$  flux (see equation 2.4) it is essential to know the exact chamber volume. In this case, the use of a plastic tarp was not suitable as the chamber volume could only be estimated.

#### Final chamber

Instead of the plastic tarp, plexiglas was used to cover the wooden base frame (see Figure 3.3). A hole in the middle of the chamber top ( $15 \times 18 \text{ cm}$ ) was fitted with a plastic foil which could be mounted on the aeration pipe after installation of the chamber on the surface of the lysimeter. At the inlet and outlet side of the chamber four sampling ports were installed and connected by using tubes in cascade.

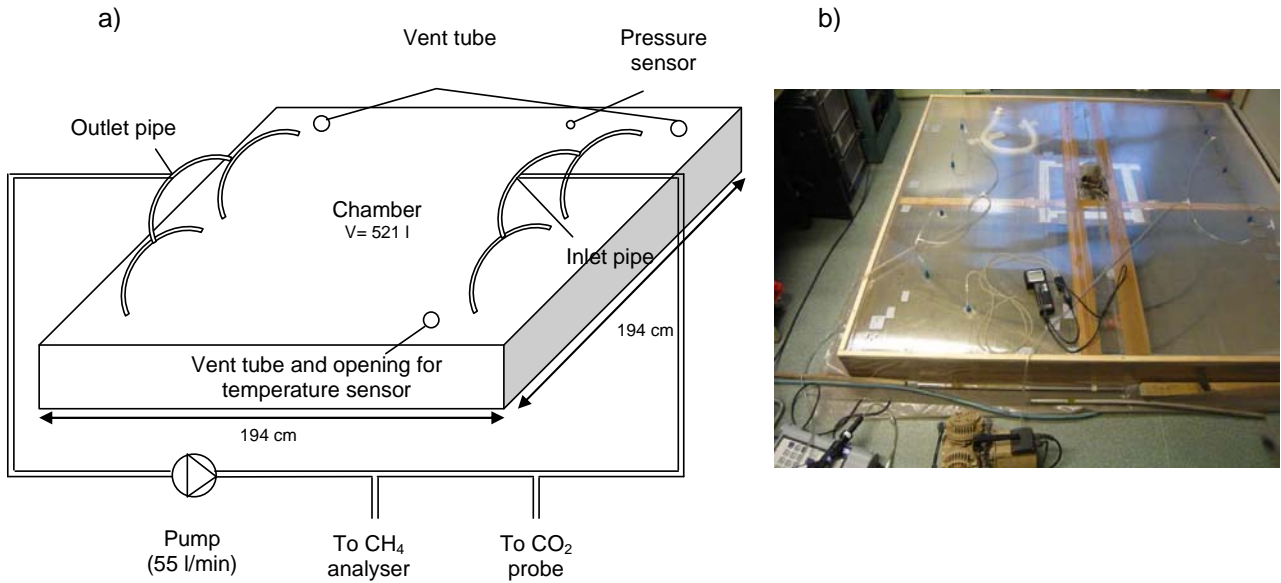


Figure 3.3: Schematic drawing (a) and illustration (b) of the developed chamber

The outlet tube of the chamber was connected to a portable FID (Thermo Scientific FID, TVA-100) and CO<sub>2</sub> detector (Vaisala CARBOCAP™ MI70 + GMP22) via an external pump (55 l/min) (see also Table 3.3). The FID was connected to a three-way-valve, sucking ambient air via an internal pump. Reading of CH<sub>4</sub> concentrations were taken every minute. 15 seconds before CH<sub>4</sub> concentrations were recorded, the open tube was closed and the valve was opened towards the chamber. The CO<sub>2</sub> detector is based on diffusive sampling. Therefore, the CO<sub>2</sub> probe was placed into a gas bag where CO<sub>2</sub> concentrations were measured simultaneously each minute.

To prevent pressure buildup, the chamber was equipped with a vent tube according to Hutchinson and Mosier (1981). The dimensions of the vent tube should allow a pressure loss not greater than 0.1 µbar. The internal tube volume should be five times greater than the volume of enclosed air displaced by the largest anticipated pressure wave. Table 3.1 gives the computed pressure wave amplitude for a range of windspeeds.

Table 3.1: Amplitudes of wind-caused air pressure waves at selected frequencies as a function of wind speed (Hutchinson and Mosier, 1981)

	Log Frequency (Hz)				
	-3.0	-2.0	-1.0	0.0	1.0
Wind speed (cm/s)	Pressure wave amplitude (µbar)				
100	3.3	1.1	0.33	0.11	0.03
200	8.9	2.8	0.89	0.28	0.09
400	59.0	19.0	5.9	1.9	0.59

Based on the values given in Table 3.1 the maximum rate of air displacement ( $v$ ) from the cover caused by each pressure oscillation and the total volume of enclosed air displaced per pressure cycle ( $b$ ) can be calculated using the following equations 3.1 and 3.2, respectively. Results are presented in Table 3.2 assuming an average barometric pressure of 1000 mbar and a chamber volume of 521.5 l.

$$v = \frac{2\pi f a V}{p_m} \quad (3.1)$$

$$b = \frac{2aV}{p_m} . \quad (3.2)$$

v	(μl/s)	volumetric air flow rate
b	(μl/cycle)	displaced volume
f	(Hz)	frequency
a	(μbar)	pressure wave amplitude
V	(μl)	chamber volume
p <sub>m</sub>	(μbar)	average barometric pressure

Table 3.2: Maximum air displacement rate and total air volume displaced per cycle for a 521.5 l chamber volume and an average barometric pressure of 1000 mbar

	Log Frequency (Hz)				
	-3.0	-2.0	-1.0	0.0	1.0
Wind speed (cm/s)	Air displacement rate (μl/sec)				
100	10.85	36.18	108.53	361.78	986.68
200	29.27	92.09	292.72	920.90	2960.04
400	194.05	624.90	1940.47	6248.97	19404.71
	Air volume displaced (μl/cycle)				
100	3454.77	1151.59	345.48	115.16	31.41
200	9317.41	2931.32	931.74	293.13	94.22
400	61767.10	19891.10	6176.71	1989.11	617.67

Equations 3.3 and 3.4 predict the optimum vent tube dimensions based on the largest values of v and b from Table 3.2.

$$D = \frac{51200\mu r b v}{(\pi^2 g_c \Delta p)^{\frac{1}{16}}} . \quad (3.3)$$

$$L = \frac{4rb}{\pi D^2} . \quad (3.4)$$

D	(mm)	tube diameter
L	(mm)	tube length
μ	(Pa s)	air viscosity (1.7*10 <sup>-5</sup> )
r	(-)	the volume ratio 5:1
g <sub>c</sub>	(m/s <sup>2</sup> )	gravity (9.81)
Δp	(μbar)	pressure drop (0.1)

The optimum vent tube dimensions of the final chamber are a diameter of 2.9 cm and a length of 48.3 cm. Therefore, the chamber was equipped with a 50 cm open tube (D = 3 cm), that connected the chamber volume with the outside atmosphere and thus allowed for a maximum diffusion loss of 1 %. However, the formulas provided by Hutchinson and Mosier (1981) are usually applied for small chambers up to 157 l (Bauer, 1996) and, thus, the applicability for large chambers is not confirmed.

To monitor the pressure in the chamber with respect to atmosphere, a fitting for a pressure sensor was drilled in the chamber top. The temperature inside the chamber was measured using a temperature probe inserted into the vent tube. Carrying straps were installed on the chamber for better handling.

In Table 3.3 the main characteristics of the measurement system used are summarised.

Table 3.3: Main characteristics of the used measurement system

Chamber internal length	190 cm
Chamber height	14.5 cm
Vent tube length	50 cm
Vent tube internal diameter	3 cm
Pumping flux	55 l/min
Dead volume	1,96 l
CO <sub>2</sub> detector and accuracy	Vaisala CARBOCAP™ MI70 + GMP222 ±1.5 % in the range of 0 to 10,000 ppm or ±2 % of reading
CH <sub>4</sub> detector and accuracy	Thermo Scientific FID, TVA-100 ±25 % of reading or ±2.5 ppm in the range of 1.0 to 10,000 ppm
Pressure sensor and accuracy	Testo 452 ± 10 Pa (0...20 hPa) and ±0.5 % of reading (20...100hPa)
Temperature probe	Testo 720, PT100 measuring range -50 to 240°C

### Reliability testing

Laboratory tests were carried out to become familiar with the measurement system, to improve the measurement procedure and to quantify the accuracy of the system. In order to verify the performance and accuracy of the measurement system, different known LFG fluxes were introduced into the chamber under constant barometric pressure and temperature. It was also important to check the time course of concentration change in order to establish an appropriate sampling period.

The LFG fluxes were regulated by a flow control based on the rotameter principle. The rotameter was carefully calibrated by a bubble flow meter at the beginning of each test run.

The first laboratory tests using only the CH<sub>4</sub> analyser (portable FID) and a constant emission source of pure CH<sub>4</sub> have shown good accuracy and repeatability in the emissions measurement capability of the system. Figure 3.4 shows a comparison between actual methane emission rate and collected methane measured during the laboratory experiments. Methane emission rate was measured three times for each known gas flux rate of pure CH<sub>4</sub>. Results deviate from imposed CH<sub>4</sub> flux values by 10 % or less. The dispersion coefficients of the results obtained under similar conditions were in the range of 1.3 – 8.5 %, thus the method does, under laboratory conditions, ensure a good measurement repeatability rate. However, when the applied CH<sub>4</sub> emission was increased (> 35 g/m<sup>2</sup>d), the measured CH<sub>4</sub> rates were all significantly higher than those applied, indicating that this measurement system overestimates higher fluxes and, thus, it is only able to detect small gas fluxes.

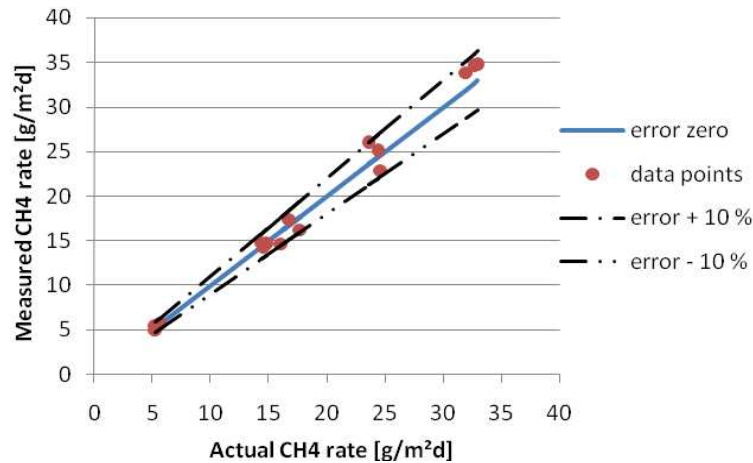
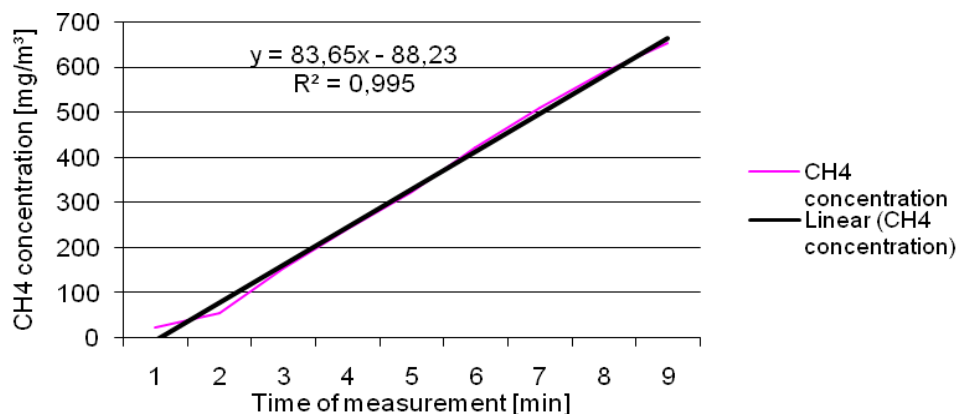


Figure 3.4: Laboratory results of method reliability test

Figure 3.5 shows methane concentrations changing with time measured during the first laboratory experiment. The  $\text{CH}_4$  flux was calculated from the linear regression of the  $\text{CH}_4$  concentrations versus time. It was found, that the appropriate time curve for gas sampling by this accumulation chamber should be about 6 – 9 min. The increase of  $\text{CH}_4$  concentrations proved to be linear for all measurements. The coefficients of determination ( $R^2$ ) ranged between 0.9506 – 0.9953.

Figure 3.5:  $\text{CH}_4$  enrichment in the chamber over 9 minutes

In the second series of laboratory experiments, a constant source of synthetic landfill gas (60 %  $\text{CH}_4$ , 40 %  $\text{CO}_2$ ) was introduced into the chamber.  $\text{CH}_4$  and  $\text{CO}_2$  emission rates were measured simultaneously in the external loop using the  $\text{CH}_4$  and  $\text{CO}_2$  detector. Three to five replicates were performed for each LFG flux. It could be shown that the uncertainty margin of the modified measurement system was about  $\pm 30$  % (see Figure 3.6). This is a reasonable result considering the limitations of the experiment. The used flow controller is based on the rotameter principle, itself introducing an uncertainty of 3 – 10 % due to imprecise scaling.

The dispersion coefficients of the measured  $\text{CH}_4$  and  $\text{CO}_2$  fluxes ranged between 1 % and 16.5 % and 1.9 – 10.6 %, respectively. The coefficients of determination ( $R^2$ ) were found in the range of 0.95 – 0.993. The chamber tended to underestimate  $\text{CO}_2$  emission rate over the tested range of LFG fluxes, as can be seen in Figure 3.6. At higher fluxes the deviation from actual  $\text{CO}_2$  and  $\text{CH}_4$  values increased markedly and, thus, the measurement system gives only reliable results for small LFG fluxes.

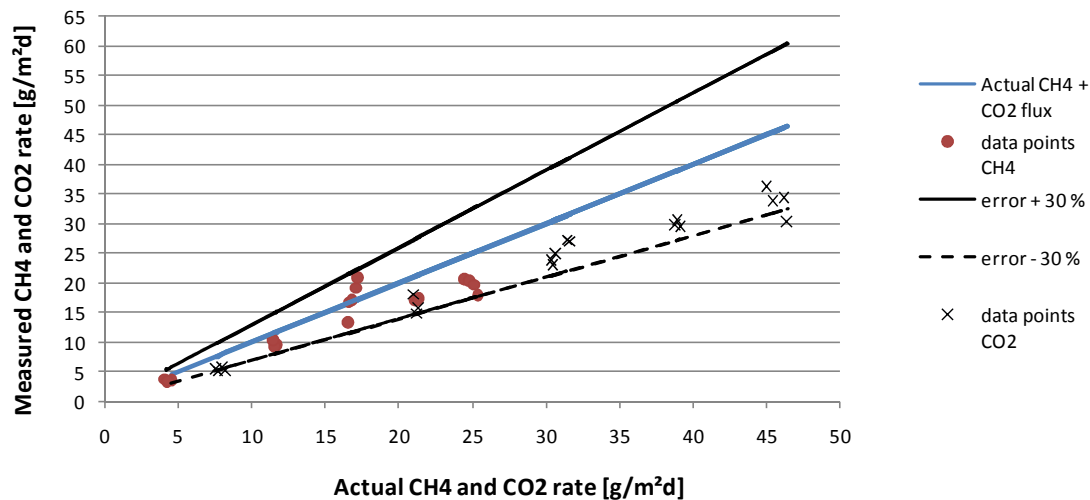


Figure 3.6: Laboratory results of second method reliability test

Following the laboratory validation phase, the chamber was fine tuned under natural conditions. Based on the measurements of the pressure difference, the installation of another two vent tubes at the accumulation chamber appeared necessary in order to compensate pressure differences during the measurements.

It can be concluded that reliable  $\text{CH}_4$  and  $\text{CO}_2$  emission rates are obtained in the range of 0.01 g  $\text{CH}_4/\text{m}^2\text{d}$  (lower detection limit) to 35 g  $\text{CH}_4/\text{m}^2\text{d}$  and 1 – 47 g  $\text{CO}_2/\text{m}^2\text{d}$ , respectively. Due to inhibition of fluxes through concentration and pressure build-up in closed chambers, the chamber can only be used for relative comparison of fluxes from the different biocovers and, thus, absolute data should be regarded with caution. Because of the size of the chamber and the need of power supply, portability is limited.

## 3.2 Suitability of substrates to enhance methane oxidation

### 3.2.1 Potential methane oxidation activity

Prior to biocover construction and implementation, laboratory experiments should be conducted for assessing potential  $\text{CH}_4$  oxidation activity of possible cover materials. Common approaches are incubation experiments (batch tests) or flow-column set-ups (undisturbed or packed column experiments) under controlled laboratory conditions (Huber-Humer et al., 2009; Gerbert and Streese-Kleeberg, 2008). For batch tests, defined quantities (usually about 10 – 30 g) of cover material samples are placed in incubation jars. In the presence of methane and oxygen, the jars are incubated and the decrease in methane concentration is continuously measured over time which allows the determination of kinetic properties of methane oxidation and short-term responses to environmental factors (Scheutz et al., 2009; Maurice and Lagerkvist, 2004; Boeckx et al., 1996; Kightley et al., 1995). In general, batch tests are a fast method to characterise the methane oxidation potential of soil materials and to evaluate their suitability for biocover construction as batch conditions can be manipulated easily (Huber-Humer et al., 2009). However, due to small sample quantities, this approach may only be suitable for homogeneous materials whereas heterogeneous substances like composted waste materials are not considered to be representative, unless they are coupled with continuously charged column tests (Huber-Humer et al., 2009). Column experiments simulate a landfill cover material matrix through which gas is transported and therefore closely resemble the dynamic



behaviour in landfill settings (Scheutz et al., 2009). An undisturbed soil column, where a  $\text{CH}_4/\text{CO}_2$ -mixture is applied at known rate to the bottom, and the top is flushed with air, is considered to be the most realistic laboratory microcosm (Chanton et al., 2009). Due to considerable variability between undisturbed columns, some research is conducted with repacked columns which allow better control of soil composition (He et al., 2008). In general, column tests allow higher mass input, coarser particle size, and longer test runs in order to reveal some of the long-term phenomena that can occur, such as microbial exopolymers (EPS) formation influencing gas exchange and oxidation efficiency (Huber-Humer et al., 2009; Scheutz et al., 2009; Huber-Humer, 2004).

Before the lysimeter experiment was conducted, pre-investigations in the laboratory have been carried out to check material suitability and oxidation capacity. At the Institute of Waste Management in Vienna continuously charged soil columns (repacked) were used to test various cover materials, both mineral soils and compost materials, concerning their potential methane oxidation activity. Table 3.4 lists the substrates investigated in these experiments.

Table 3.4: Various substrates investigated in column experiments

Substrate	Abbr.	Source
Sewage sludge compost	SSC	Hollabrunn
Biowaste compost	BWC	Wien - Lobau
Sand 2/4	Sand-Fischa	Fischamend
Topsoil (silt)	TS-Fischa	Fischamend
Topsoil (loess)	TS-Zister	Zistersdorf
Subsoil (loess)	SS-Zister	Zistersdorf
Sand-compost mixture (60:40 vol. %)	SSC-Mix	Fischamend / Hollabrunn

Based on the column experiments (see Figure 3.7) the most suitable cover materials were selected for long-term observations in the landfill lysimeters under natural conditions.

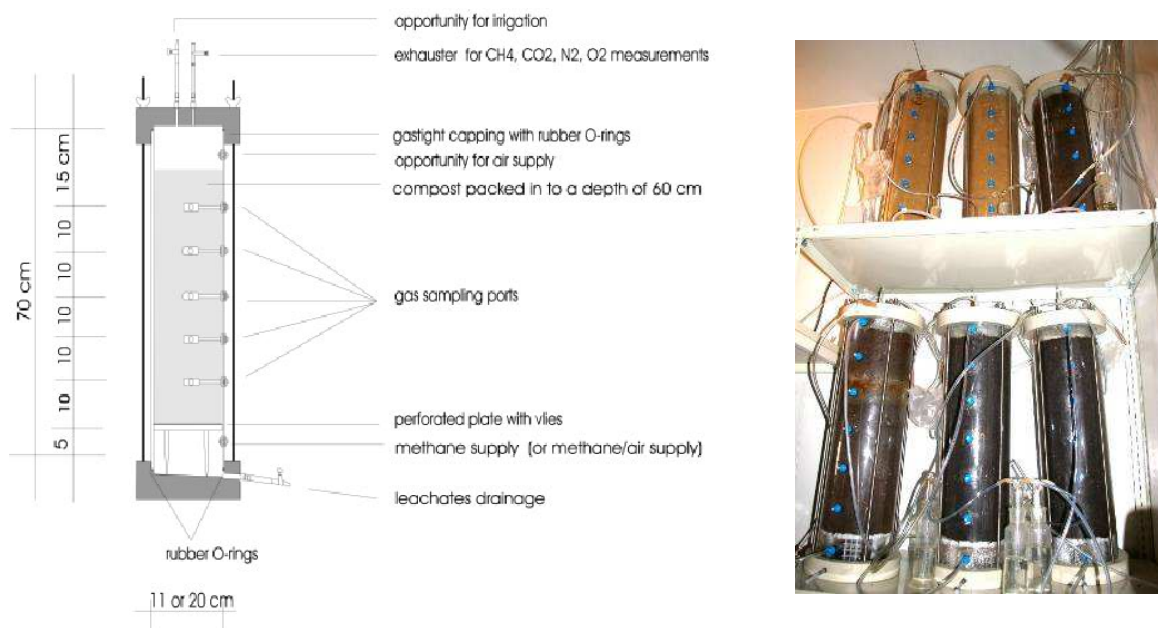


Figure 3.7: Laboratory test column to investigate methane oxidation capacity; continuous methane supply to the bottom; air supply to the top; lateral measuring holes to establish gas concentration and temperature profiles (Humer and Lechner, 1999a; Internal Report Nutzraum, 2008)



In Figure 3.8 the percents of oxidised methane load in all investigated substrates are pictured as time curves. While the mineral soils (TS-Fischa, TS-Zister, Sand-Fischa) showed only moderate methane oxidation potential (medial oxidation rate 38 %, 15 % and 20 %, respectively), the highest methane turnover rate was observed in mature sewage sludge compost (SSC). The mature compost material mitigated loadings up to 350 l CH<sub>4</sub>/m<sup>2</sup>d indicating a constant CH<sub>4</sub> degradation rate of more than 95 %. The lowest methane oxidation potential was observed in the quite fresh biowaste compost (BWC, medial oxidation rate 0 %) and the mineral subsoil (SS-Zister, medial oxidation rate 4 %). Due to the low degree of maturity of the biowaste compost, other decomposition processes probably suppressed CH<sub>4</sub> oxidation. There was a high CO<sub>2</sub> production resulting from the endogenous respiration of the fresh compost material itself. Also the low organic content in the mineral material was not a beneficial property for methanotrophic activity.

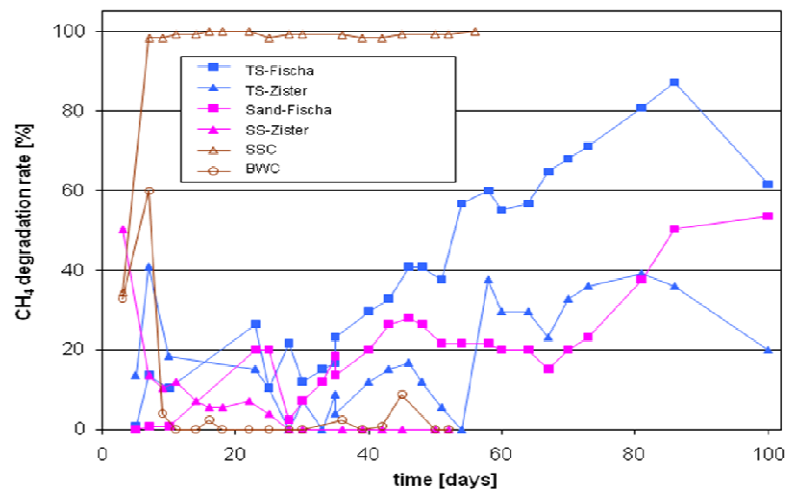


Figure 3.8: Methane degradation rates of tested substrates over 100 days under laboratory conditions (Gamperling et al., 2008)

### 3.2.2 Air Permeability

In general, the transport mechanism in porous media is controlled by both diffusive and advective forces (see section 2.2). In the context of landfills, the primary driving force for gas transport, especially through cover systems, is pressure gradients induced by natural fluctuations in barometric pressure and gas generation in waste and, thus, advective flux dominate diffusional flux (Stern et al., 2007, De Visscher et al., 2004; Czepiel et al., 2003). The air permeability of a soil, a measure of advective flux, is defined as its ability to conduct air by the mass flow of gas in the ease of response to a pressure gradient. The most important factor that controls the air permeability of soil is the water content and the pore structure of soil or substrate (Hamamoto et al., 2009). Since water content could vary significantly after the biocover system is constructed, it is necessary to determine the air permeability of possible cover materials at various water contents. The air permeability shows generally inverse correlation with water content. Lower water content leads to higher gas permeability whereas higher water content leads to lower gas permeability.

At the Austrian Institute of Technology in Seibersdorf a series of gas permeability tests were performed to investigate the variation of air permeability of the substrates listed in Table 3.4 at four different water contents (25 %, 50 %, 75 % and 100 % of WHC) by flowing air through a soil column. The air permeability test apparatus is shown in Figure 3.9.

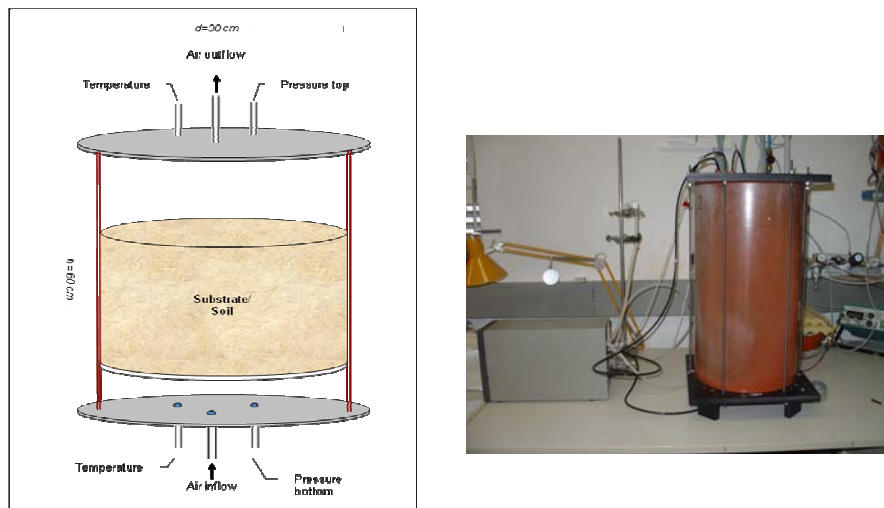


Figure 3.9: Schematic illustration of the air permeability test apparatus (Internal Report Nutzraum, 2008 and AIT Seibersdorf, 2008)

Different air flow rates (10 – 50 ml/min) were applied continuously to the bottom of the column and gas pressure and temperature at the column base and top were monitored during a test run. From this laboratory experiment the intrinsic permeability of the different substrate materials at various water contents was calculated using Darcy's Law:

$$q_A = \frac{k_{int}}{\eta} * \frac{\Delta p}{\Delta z} \quad (3.5)$$

$q_A$  (m<sup>3</sup>/m<sup>2</sup>s) air mass flow

$k_{int}$  (m<sup>2</sup>) intrinsic permeability of the porous media

$\eta$  (Pa s) dynamic air viscosity

$\frac{\Delta p}{\Delta z}$

$\Delta z$  (Pa/m) pressure gradient along a flow distance in the soil column

It is assumed that the intrinsic permeability is a function of the properties of the porous material, not the permeating fluid or gas. In general, the substrate used in a biocover should ensure sufficient air permeability even at 100 % WHC. Huber-Humer et al. (2008a) recommended that the intrinsic permeability of the material used as a landfill cover should not go below a value of  $5 \cdot 10^{-13}$  m<sup>2</sup> at 100 % WHC (equivalent to field capacity). This would mean an air flow rate of 10 l/m<sup>2</sup>h at a pressure gradient of 1 hPa at a soil depth of 1 m. Figure 3.10 shows the pressure at the bottom and top of the column of SSC-Mix at 34 % WHC with varying air flow rate.

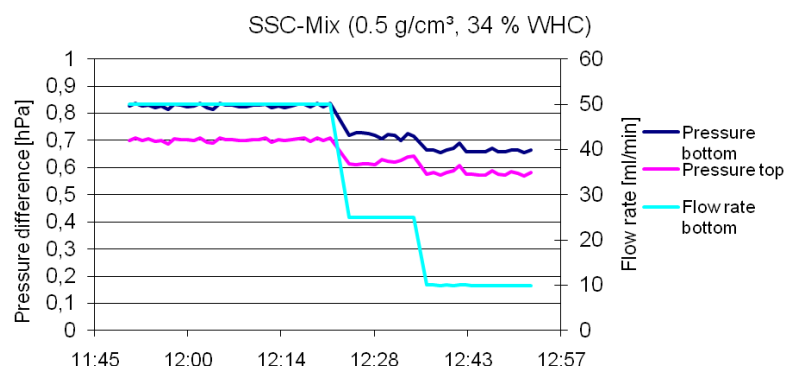


Figure 3.10: Pressure curve at the base and top of the column with varying flow rate (based on data from AIT Seibersdorf)

It could be shown that all tested substrates had sufficient intrinsic permeability up to 100 % WHC. Only sand (Sand-Fischa) showed limited air permeability ( $k_{\text{int}} = 3.5 \cdot 10^{-13} \text{ m}^2$ ) at 75 % and 100 % WHC, with pressure differences up to 90 Pa between base and top of the soil column.

### 3.3 Lysimeter experiment

A lysimeter experiment was set up at the Austrian Institute of Technology in Seibersdorf, Austria in February 2008 to investigate three biocovers using different substrates as well as one control cover regarding water infiltration and methane emission mitigation in combination with in-situ aeration. However, this diploma thesis presents only data concerning the relative emission reduction effect of the passive biocovers. Therefore a monitoring program has been developed to verify biocover performances including

- monitoring of qualitative (gas and temperature profiles, surface FID-screening) and quantitative (stable isotope analysis) indications for in situ methane oxidation in the biocovers and
- measurement of remaining  $\text{CH}_4$  and  $\text{CO}_2$  surface emissions using a closed dynamic chamber in order to see the relative difference in landfill gas flux through the different ground surfaces.

The measurement campaign was conducted from August 2009 to April 2010.

#### 3.3.1 Lysimeter setup

In a lysimeter facility four chambers (A, B, C and D), each measuring 2 m x 2 m x 3 m (width x length x depth), are filled with different substrates according to Table 3.5 and Figure 3.12. As can be seen in Figure 3.11, the lysimeter facility is a reinforced concrete structure with a basement level 1.2 m below the surroundings level and it is thermally shielded from the outside environment by a mound.



Figure 3.11: Lysimeter facility at the Austrian Institute of Technology in Seibersdorf, Austria

Chamber A, B and C included a 1 m thick substrate layer, whereas the control chamber (D) consisted of a 0.5 m layer. All substrate layers were underlain by a 0.2 m gas distribution layer consisting of 10/40 mm coarse gravel. A 0.2 m drainage layer was installed at the lowest point to evacuate infiltrating waters. Only the control cover included a 0.7 m drainage system to compensate for the smaller substrate layer.

Each of the lysimeter chambers were fed directly by biogas coming from fresh municipal solid waste, sieved with a 80 mm sieve. The municipal solid waste was

provided by a mechanical biological treatment plant (“Wiener Neustädter Stadtwerke und Kommunal Service GmbH”) in Steinabrückl, Lower Austria, which was only mechanically pretreated. As a result, it was not possible to control the upward flux of biogas. Table 3.6 shows the solid matter and eluate analyses of the buried waste mass. A perforated pipe was installed in the middle of each chamber to aerate the waste body for rapid waste stabilisation (forced in-situ aeration) (see Figure 3.12 and Figure 3.17) as soon as the gas generation is stable and the evaluation of the passive biocovers is completed.

Table 3.5: Designs of the lysimeter chambers

<p><b>Chamber A</b> “classic methane oxidation layer”</p> <p>100 cm sewage sludge compost 20 cm coarse gravel (gas distribution layer) 150 cm municipal solid waste 20 cm coarse gravel (drainage layer)</p>	<p><b>Chamber B</b> “Sand-compost mixture”</p> <p>2 cm topsoil Fischamend 48 cm sand 50 cm sand-compost mixture (60:40 % v/v) 20 cm coarse gravel (gas distribution layer) 150 cm municipal solid waste 20 cm coarse gravel (drainage layer)</p>
<p><b>Chamber C</b> “evapotranspiration cover”</p> <p>50 cm topsoil Fischamend 50 cm subsoil Zisterdorf 20 cm coarse gravel (gas distribution layer) 150 cm municipal solid waste 20 cm coarse gravel (drainage layer)</p>	<p><b>Chamber D</b> “control cover”</p> <p>10 cm topsoil Fischamend 40 cm subsoil Zisterdorf 20 cm coarse gravel (gas distribution layer) 150 cm municipal solid waste 70 cm coarse gravel (drainage layer)</p>

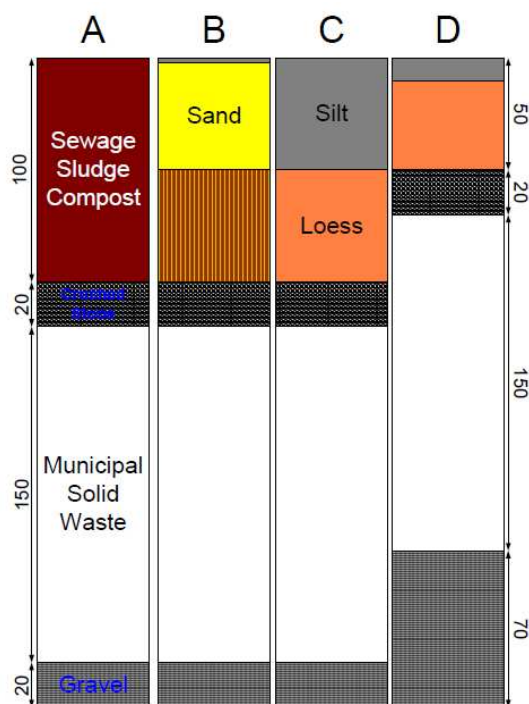


Figure 3.12: Lateral cross-section of the installed lysimeters (2 m x 2 m x 3 m) and view of the top of the lysimeter facility (AIT Seibersdorf 2008)

Table 3.6: Data of the municipal solid waste at the beginning of the lysimeter experiment (a – solid matter analyses, b – eluate analyses); RA = respiration activity; DM = dry matter; LOI = loss on ignition at 550 °C (organic content); TOC = total organic carbon; TC = total carbon; GS = potential of gas formation; COD = chemical oxygen demand; BOD = biochemical oxygen demand

a) Parameter	Unit	Charge 1	Charge 2	Method
LOI	% DM	48.1	48.5	ÖNORM EN 15169
TC	% DM	28.3	28.1	ÖNORM EN 13137
TOC	% DM	27.3	27.5	ÖNORM EN 13137
N <sub>total</sub> (Dumas)	% DM	0.98	1.13	ÖNORM EN 13654
C/N	-	28	24	
TOC/LOI	-	0.57	0.57	
RA <sub>4</sub>	mg O <sub>2</sub> /g DM	48.7	68.8	ÖNORM S 2027-1
GS <sub>21</sub>	NI/kg DM	40.8	39.2	ÖNORM S 2027-2

b) Parameter	Unit	Charge 1 and 2	Method
pH	-	6.8	ÖNORM M 6244
Conductivity	mS/cm	6.7	ÖNORM EN 27888
NH <sub>4</sub> <sup>+</sup> N	mg/kg DM	429	ÖNORM EN ISO 11732
NO <sub>3</sub> <sup>-</sup> N	mg/kg DM	2	ÖNORM EN ISO 10304-2
COD	mg O <sub>2</sub> /kg DM	61,071	ÖNORM M 6265
BOD	mg O <sub>2</sub> /kg DM	30,400	ÖNORM EN 1899-2

### 3.3.2 Substrates used in the landfill lysimeters

In order to characterise the various substrates, standard physical and chemical parameters were analysed and soil grain analyses were performed.

The standard parameters were carried out in full agreement with the “Austrian Ordinance on Composting” (BGBl. II 292/2001). The water holding capacity was determined according to an institute specific technique described in Huber-Humer (2004).

Particle size distributions were analysed with two different methods. Sand and gravel fractions of the substrates were measured according to the sieving method of DIN ISO 3310 (1992), whereas the smaller fractions (sand, silt and clay) were measured by the conventional pipette method (ÖNORM L 1061). The sand fractions obtained by these methods are not directly comparable, as a dispersion agent was used only for the pipette method.

The following table presents some relevant substrate characteristics (see Table 3.7) of the cover materials placed in the landfill lysimeters (see section 3.3.1). Not surprisingly, the organic content (loss on ignition) of the compost materials was quite high compared to the natural soils. In addition, the sewage sludge compost was initially mature which was confirmed by the low respiration activity of 0.8 mg O<sub>2</sub>/g DM. Importantly, low ammonium concentrations were present in all cover materials avoiding possible inhibition effects. The nitrate concentration was remarkably higher in the compost materials than in the mineral soil, revealing that the compost materials provide microorganisms with adequate nutrient supply. The mineral materials (TS-Fischa, Sand-Fischa, SS-Zister) exhibited a similar total pore volume (42.8 – 48.7 % v/v), whereas the sand-compost mixture (SSC-Mix) offers a higher porosity. Consequently, both compost materials (SSC and SSC-Mix) have a higher water-holding capacity.



Table 3.7: Data of the cover materials at the beginning of the lysimeter experiment; WHC = water holding capacity; RA = respiration activity; DM = dry matter; n.d. = not detectable; LOI = loss on ignition at 550 °C (organic content); TOC = total organic carbon, TC = total carbon

Parameter	Unit	SSC	SSC-Mix	TS-Fischa	Sand-Fischa	SS-Zister
Moisture content	% on a dry weight basis	64.7	21.9	21.8	7.7	18.5
WHC	% DM	117.1	43.2	41	28	37
Bulk density	kg/l	0.6	1.1	1.4	1.5	1.3
Total pore volume	% v/v	72.8	55.8	47.9	44.5	52.8
RA <sub>4</sub>	mg O <sub>2</sub> /g DM	0.7	0.1	-	-	-
LOI	% DM	24.9	4.0	5.2	1.1	2.9
TOC	% DM	11.8	1.5	1.5	0.1	0.4
TC	% DM	13.6	3.0	3.3	1.6	2.2
N <sub>total</sub> (Dumas)	% DM	1.5	0.6	0.1	n.d.	n.d
CaCO <sub>3</sub>	% DM	14.4	12.2	14.5	12.4	14.9
pH	-	6.9	7.1	8.0	8.0	8.3
Conductivity	mS/cm	2.8	0.5	0.1	0.1	0.1
NO <sub>3</sub> <sup>-</sup> N	mg/kg DM	1750	200	31	3	n.d.
NH <sub>4</sub> <sup>+</sup> N	mg/kg DM	n.d.	n.d	2	2	2

Regarding the particle size distribution, both soils, TS-Fischa and SS-Zister (Figure 3.14 and Figure 3.16), show quite similar curves with a higher content of finer fraction (< 2 mm), whereas the compost material SSC exhibits a balanced grain size distribution (approx. 50 % < 2 mm and 50 % > 2 mm) (Figure 3.13). Compared to the pure compost material SSC, the SSC-Mix exhibits a particle size distribution with a higher amount of sand-like fractions (< 2 mm) (Figure 3.13).

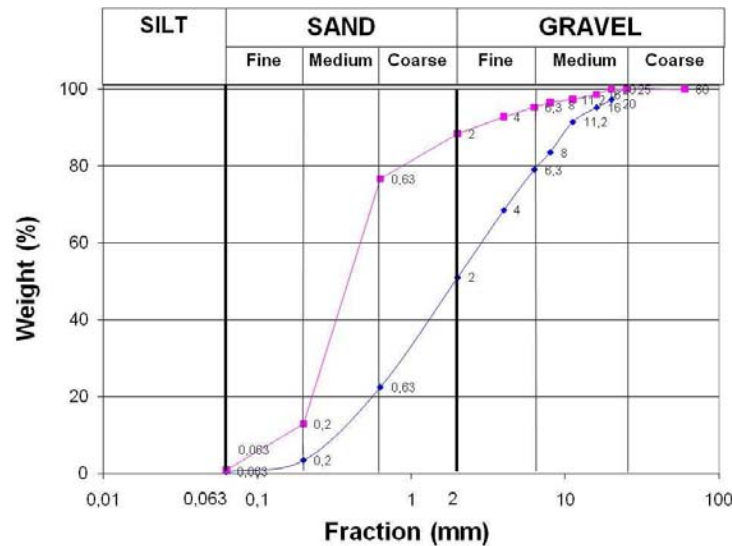


Figure 3.13: Particle size distribution of SSC and SSC-Mix according to DIN ISO 3310

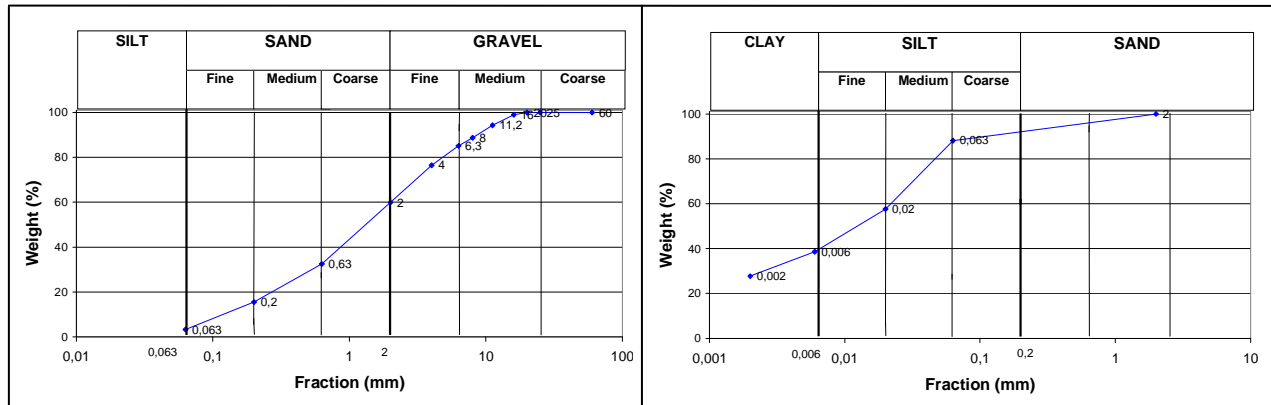


Figure 3.14: Particle size distributions of the topsoil Fischamend (TS-Fischa) according to DIN ISO 3310 (left side) and ÖNORM L 1061 (right side)

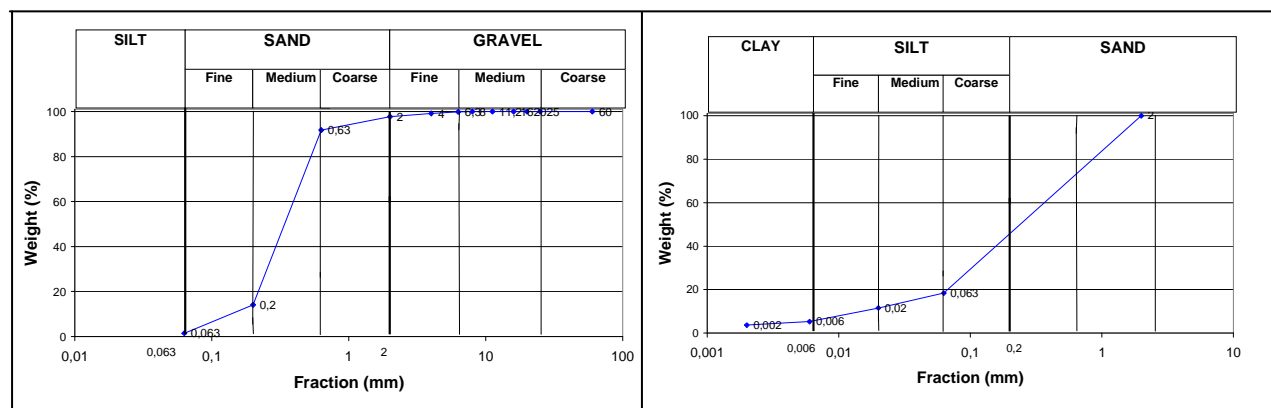


Figure 3.15: Particle size distributions of the sand Fischamend according to DIN ISO 3310 (left side) and ÖNORM L 1061 (right side)

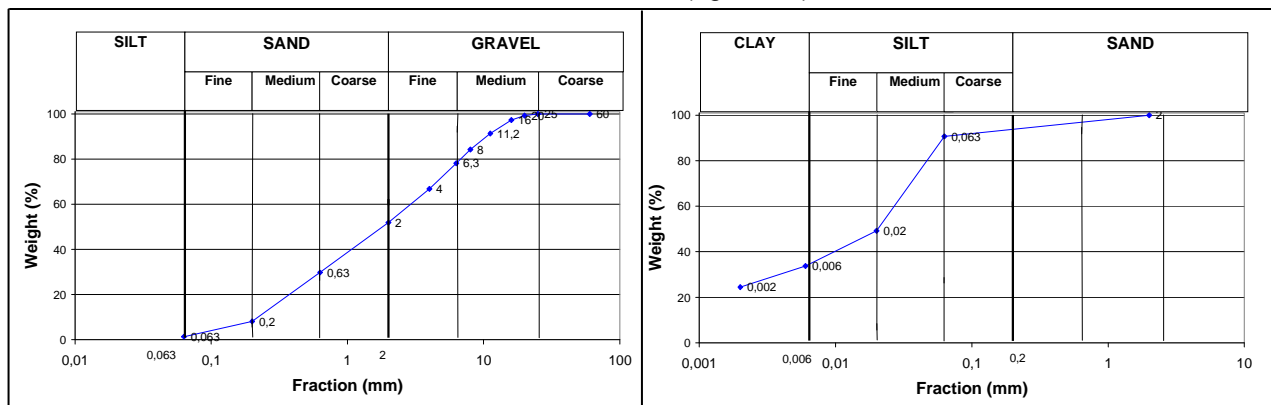


Figure 3.16: Particle size distributions of the subsoil Zistersdorf (SS-Zister) according to DIN ISO 3310 (left side) and ÖNORM L 1061 (right side)

### 3.3.3 Measurement of pore gas composition, temperature and water content

Gas probes, consisting of an open polyethylene (PE) pipe with an inner diameter of 5 mm, were installed permanently in six depths (25, 50, 75, 110, 160, 210 cm) of the lysimeter chambers A, B and C. The top end of the pipes was closed with rubber stoppers. Temperature sensors and water content sensors, connected to a data acquisition system and data loggers, were installed at four separate depths (25, 50, 75, 190 cm) and three depths (25, 75, 190 cm), respectively. Figure 3.17 depicts the measuring probes installed in the chambers A, B and C, making up for a total of 8 gas probes, 8 temperature sensors and 6 water content sensors for each chamber.

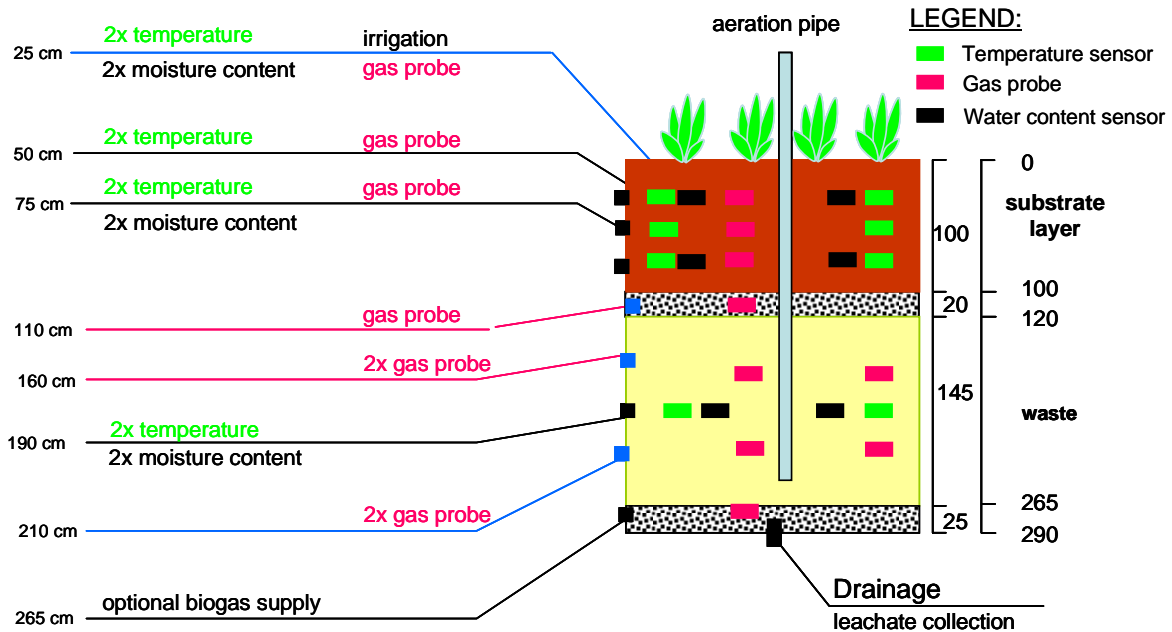


Figure 3.17: Design of landfill lysimeter and measuring probes installed in chambers A, B and C (Internal Report Nutzraum, 2008)

As lysimeter chamber D had a less thick substrate layer (0.5 m), the location of the measuring probes differed slightly from those installed in chambers A, B and C. As illustrated in Figure 3.18, in total 6 gas probes, 6 temperature sensors and 6 water content sensors were installed in chamber D.

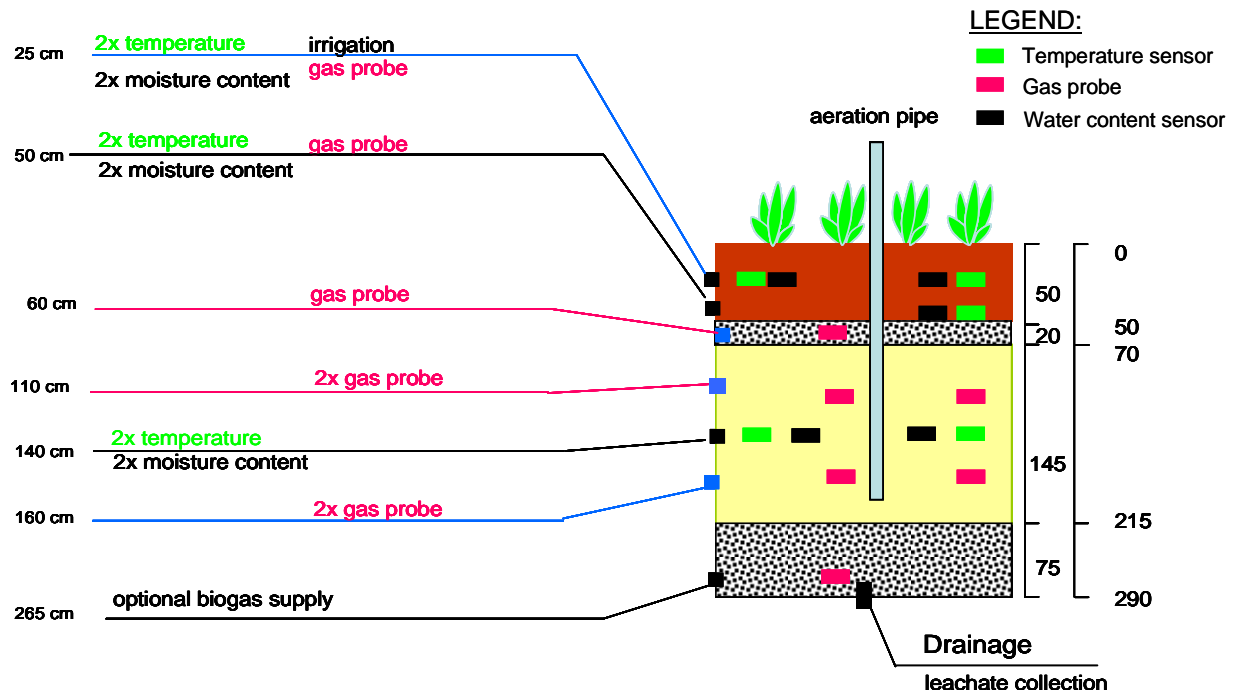


Figure 3.18: Design of landfill lysimeter and measuring probes installed at chamber D (Internal Report Nutzraum, 2008)

A sharp decline in  $O_2$  and  $CH_4$  concentrations with depth in the soil profile provides a qualitative indication of the start of microbial methane oxidation (Huber-Humer et al., 2009b). Also the temperature profile may be an indicator of the location of the



oxidation layer. From August 2009 to April 2010 measurements of gas concentration profiles were carried out on a weekly basis. The volumetric concentrations of CH<sub>4</sub>, CO<sub>2</sub> and O<sub>2</sub> were measured using a mobile gas analyser (Multigas analyser LMSx) equipped with an infrared methane and carbon dioxide detector (precision of measurement:  $\pm 0.1$  % in the range of 0–10 vol.%, and  $\pm 1$  % in the range of 10–80 vol.%) and an electrochemical sensor able to detect O<sub>2</sub> (precision of measurement:  $\pm 0.5$  % in the range of 0–25 vol. %). Nitrogen gas is a good indicator for the oxidation level as it is neither consumed by oxidation near the surface nor by soil respiration. In order to draw concentration profiles, N<sub>2</sub> was calculated as the difference between 100 % and the sum of the CH<sub>4</sub>, CO<sub>2</sub> and O<sub>2</sub> concentrations for each depth within the soil profiles. Temperature and water contents within the chambers were measured by the employees of the Austrian Institute of Technology in Seibersdorf (Internal Report Nutzraum, 2008).

### 3.3.4 Measurement of gas fluxes

In order to assess biocover performance and functionality a series of surface emission measurements was made in December 2009 as well as in March and April 2010. Grass and other plants developed on the lysimeter chambers were cut away to ensure minimum disturbance. Measurement of the CH<sub>4</sub> and CO<sub>2</sub> concentrations inside the closed dynamic chamber (see Figure 3.19) was done using a portable Flame Ionisation Detector (Thermo Scientific FID, TVA-100; (precision of measurement:  $\pm 25$  % of reading or  $\pm 2.5$  ppm in the range of 1.0 to 10,000 ppm) and a portable carbon dioxide detector (Vaisala CARBOCAP™ GM70; precision of measurement:  $\pm 20$  ppm + 2 % of reading), respectively. At the time of measurement, the chamber was placed on the ground of the lysimeter chamber, and the air from the chamber was pumped to the CH<sub>4</sub> and CO<sub>2</sub> analyser by the external pump. Gas composition in the chamber was analysed immediately after installation. The duration of each measurement was approximately 6 – 9 minutes, depending on the definiteness of the increase. As already described in chapter 3, the linear increase in CH<sub>4</sub> and CO<sub>2</sub> concentrations ( $R^2 > 0.9$ ) in the chamber over time was used to calculate the LFG emission rates. Correlations less than  $R^2 = 0.7$  were treated as zero fluxes. For each lysimeter chamber, three reliable measurements were made and then a mean flux rate and standard deviation were calculated. Before start of measurement, the Flame-Ionisation Detector was calibrated with a 1-point calibration using standard test gas (Linde, 500 ppm CH<sub>4</sub> in N<sub>2</sub>) at the temperature and humidity of the calibration. In addition, weather conditions, in particular wind speed, barometric pressure and air temperature, were monitored.



Figure 3.19: Developed chamber placed on the lysimeter

### 3.3.5 Measurement of gas emissions (FID-measurement)

Repeated screenings of the surface methane concentrations represent a qualitative survey of the overall CH<sub>4</sub> emissions pattern from the different biocovers. Surface methane concentrations were scanned once a week (or every second week) using a portable FID (Thermo Scientific FID, TVA-100 Toxic Vapour Analyser) according to a predefined grid. These investigations provide a qualitative check of the biocover functionality under specific environmental and weather conditions. The FID was calibrated before start of measurement and weather conditions were monitored. As can be seen in Figure 3.20, a funnel-shaped probe was placed directly on the cover surface and the emitted methane was pumped to the FID via an internal pump.

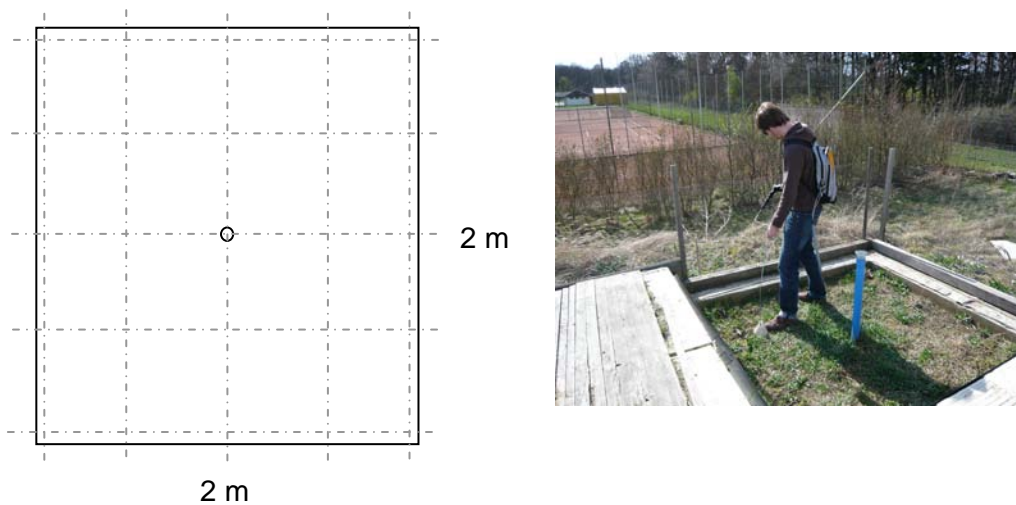


Figure 3.20: Screening of the surface emission concentration on a 50 x 50 cm grid

### 3.3.6 Stable Isotope Analyses

#### Background

The stable isotope technique is a promising field method for quantifying methane oxidation efficiency in landfill cover materials based on changes in the ratio of two stable carbon isotopes, namely <sup>13</sup>C and <sup>12</sup>C, which comprise 1 % and 99 % of carbon atoms, respectively. Methanotrophic bacteria preferentially oxidise the lighter isotope <sup>12</sup>C over <sup>13</sup>C in methane, leaving residual methane enriched in the heavier isotope <sup>13</sup>C (Barker and Fritz, 1981; De Visscher et al., 2004). As a result, a shift in the stable carbon isotope composition occur when methane is oxidised, changing the isotope ratio or fractionation. During the passage of landfill gas from anaerobic zones through zones of oxidation, methane becomes heavier and carbon dioxide becomes lighter. The carbon stable isotope composition is defined as follows:

$$\delta^{13}C_A\text{‰} = \left( \frac{R_{\text{sample}}}{R_{\text{st}}} - 1 \right) * 1000 \quad (3.6)$$

with  $R_{\text{sample}}$  the <sup>13</sup>C/<sup>12</sup>C ratio of the sample, and  $R_{\text{st}}$  the <sup>13</sup>C/<sup>12</sup>C ratio of the reference standard VPDB (Vienna Pee Dee Belemnite;  $R_{\text{st}}=0.01124$ ).

Typically, microbial CH<sub>4</sub> is produced at values between -50 ‰ and -60 ‰ while CH<sub>4</sub> actually emitted has <sup>13</sup>C-enriched values of -30 to -50 ‰ following oxidation (Chanton et al., 1999). Barker and Fritz (1981) found by experiment that CO<sub>2</sub> produced by methanotrophic bacteria was enriched in <sup>12</sup>C by 5.0–29.6 ‰, relative to the residual

methane. The negative  $\delta$  value indicates a  $^{13}\text{C}$  depletion relative to the carbonate standard whereas more positive values indicate a  $\delta^{13}\text{C}$  enrichment.

Carbon isotope fractionation has been widely used to obtain in situ estimates of methane oxidation in cover materials by measuring the change in the isotope signature of methane produced inside the landfill and methane being emitted from the cover surface or in the upper part of soil profile (Chanton et al., 2008; De Visscher et al., 2004; Chanton and Liptay, 2000). Recently, isotope fractionation has been employed to quantify methane oxidation combined with flux chamber measurements (Abichou et al., 2006; Barlaz et al., 2004; Chanton and Liptay, 2000; Börjesson et al., 2001) but also with tracer techniques (comparison between  $\delta^{13}\text{C}$  of atmospheric  $\text{CH}_4$  in an upwind transect to a downwind transect) (Börjesson et al., 2007; Chanton et al., 1999). The latter was used to measure the average methane oxidation rates of an entire landfill.

Knowledge of the  $\delta^{13}\text{C}$  of emitted  $\text{CH}_4$  compared to the  $\delta^{13}\text{C}$  of unoxidised  $\text{CH}_4$  in the anaerobic zone (determined from soil probe data) allows calculation of the fraction of methane that is oxidised during transport through the landfill cover materials. The percentage of oxidised  $\text{CH}_4$  ( $f_{\text{ox}}$ ) can be determined according to two approaches. The first approach is based on the assumption that  $\text{CH}_4$  moves as a closed system between source and sampling point and does not mix with other  $\text{CH}_4$  ("simplified Rayleigh approach") using the following equation (Mahieu et al., 2006):

$$f_{\text{ox}, z} = 1 - \left( \frac{\delta_z + 1000}{\delta_{\text{anox}} + 1000} \right)^{\frac{\alpha_{\text{ox}}}{1 - \alpha_{\text{ox}}}} \quad (3.7)$$

where  $f_{\text{ox}, z}$  is the fraction oxidised at depth  $z$ ,  $\delta_z$  and  $\delta_{\text{anox}}$  are standard  $\delta^{13}\text{C}$  isotope ratios for sampling depth  $z$  or emitted  $\text{CH}_4$  of the anoxic zone, and  $\alpha_{\text{ox}}$  is the isotopic fractionation factor for microbial oxidation. The second approach assumes that  $\text{CH}_4$  in the porous media is well mixed ("open system approach"). The fraction oxidised ( $f_{\text{oxo}, z}$ ) is calculated with the equation according to Chanton et al. (1999):

$$f_{\text{oxo}, z} = 1 - \left( \frac{\delta_z - \delta_{\text{anox}}}{1000(\alpha_{\text{ox}} - \alpha_{\text{trans}})} \right) \quad (3.8)$$

where  $\alpha_{\text{trans}}$  is the isotope fraction factor from transport ( $\alpha_{\text{trans}} = 1$  for purely advective transport and  $\alpha_{\text{trans}} = 1.014$  where diffusion is important) (De Visscher et al., 2004). The fractionation factor for microbial oxidation ( $\alpha_{\text{ox}}$ ), an estimate of the preference of the methanotrophs for the lighter isotope, can be obtained empirically by controlled field or laboratory incubations of cover materials with known  $\text{CH}_4$  headspace concentrations in order to derive site- and time-specific fractionation factors (De Visscher et al., 2004). Different values of  $\alpha_{\text{ox}}$  have been reported in the literature, some of which are presented in Table 3.8.

However, to date the main disadvantage of the stable isotope analysis is the variations in methane stable isotope ratios due to an extremely specific fractionation influenced by the individual characteristics of methanotrophic bacteria and growth condition (Nozhevnikova et al., 2003 cited in Huber-Humer et al., 2009) and fractionation processes during gas transport (De Visscher et al., 2004). Consequently, the fractionation factor must be determined specifically for each landfill site, a process which is very laborious and costly and may constrain practical applicability (Huber-Humer et al., 2009). Stable isotope analysis is not applicable in situations where flux chambers yield negative fluxes, indicating that landfill covers

consume rather than emit methane (Scheutz et al., 2009). According to De Visscher et al. (2004) previous application of the conservative stable isotope analysis may have led to an underestimation of CH<sub>4</sub> oxidation. Also CH<sub>4</sub> flow through macro pores may contribute to underestimations as more emitted methane is sampled at the cover surface whereas completely oxidised methane is not represented (Huber-Humer and Lechner, 2007).

### Present study

Gas samples for stable isotope analysis were taken at selected dates (21.10.2009 and 09.12.2009) to study methane oxidation efficiency of the biocovers. Samples were taken from each depth of the soil profile including waste body, gas distribution layer and substrate layer. Samples were extracted through a gas sampling tube (100 and 150 ml) and directly measured with a mobile gas analyser detecting CH<sub>4</sub>, CO<sub>2</sub> and O<sub>2</sub>. For the last date gas samples were also taken during surface flux measurements.

Isotope analyses were performed at the Austrian Institute of Technology in Seibersdorf. The gas chromatography combustion isotope ratio mass spectrometry system (GC/C/IRMS) consisted of a HP5859 Series II gas chromatograph (GC) coupled with a isotopic mass spectrometer (Finnigan MAT Delta S) via a combustion interface (Finnigan MAT combustion interface). The GC column was a CP PoraPlot Q fused silica column (25 m, 0.32 mm). The used gas standards had a precision and accuracy better than <0.05 ‰.

The isotopic fractionation factor for bacterial oxidation ( $\alpha_{ox}$ ) was determined according to literature values, as illustrated in Table 3.8. Only those values in the temperature range found during gas sampling dates were considered.

Table 3.8: Literature values of  $\alpha_{ox}$  with associated soil temperature (adapted from Cabral et al., 2009)

$\alpha_{ox}$	Soil temperature [°C]	Reference
1.0140	25	Liptay et al. 1998
1.0220	25	Liptay et al. 1998
1.0300	25	Liptay et al. 1998
1.0235	22	Chanton et al. (1999)
1.0288	9	Chanton et al. (1999)
1.0307	5	Chanton et al. (1999)
1.0311	4	Chanton et al. (1999)
1.0315	3	Chanton et al. (1999)
1.0304	6	Chanton et al. (1999)
1.0240	20	Chanton et al. (1999)
1.0266	15	Chanton et al. (1999)
1.0269	13	Chanton et al. (1999)
1.0240	21	Chanton et al. (1999)
1.0241	20	Chanton et al. (1999)
1.0316	3	Chanton et al. (1999)
1.0244	-	Chanton et al. (2008)
1.0250	35	Chanton and Liptay (2000)
1.0490	8	Chanton and Liptay (2000)

Previous studies have demonstrated that the fractionation factor depends on soil temperature modifying the calculated methane oxidation efficiency (Chanton and Liptay, 2000; Chanton et al. 1999). As a consequence, the fractionation factor was

corrected for temperature (see Table 4.3) based on Tyler et al.'s (1994; cited in Cabral et al., 2009) temperature dependence relationship:

$$\alpha_{ox} = \alpha_{oxaverage} + 0.00046(20 - T[^{\circ}C]) \quad (3.9)$$

Due to the high variability of the fractionation factor ( $\alpha_{ox}$ ), a sensitivity analysis for the calculation of  $f_{oxc}$  and  $f_{oxo}$  was carried out relative to changes in  $\alpha_{ox}$ . The oxidised fractions ( $f_{oxc}$  and  $f_{oxo}$ ) were calculated for closed and open systems using equations 3.7 and 3.8, respectively. The isotope fraction factor from transport ( $\alpha_{trans}$ ) was set to be 1 for purely advective systems (see section 4.4).

### **3.3.7 Measurement of meteorological data**

Meteorological data, including air temperature, precipitation, atmospheric pressure and wind speed were continuously recorded by a weather station of the Central Institute for Meteorology and Geodynamics installed near the lysimeter facility.

## 4 RESULTS AND DISCUSSIONS

Each of the lysimeters was fed directly by biogas coming from fresh municipal solid waste (see section 3.3.1) and, thus, the magnitude of the  $\text{CH}_4$  loading could not be controlled. Methane production in the respective solid waste layers did not start until summer 2009, although the lysimeter experiment was set up in February 2008. In order to activate and intensify the LFG production, the waste layers were irrigated additionally in summer and autumn 2009. Following the irrigation, the  $\text{CH}_4$  concentration increased slightly in all waste layers and reached a peak in lysimeters A, B and D in autumn 2009. Though all the four lysimeters contained the same type of waste, methane development in the solid waste layers of lysimeters A and B did not indicate the same range as other two. The gas composition by volume in the waste layer of lysimeters C and D was close to or above 50 vol.% for  $\text{CO}_2$  and  $\text{CH}_4$  except for lysimeters A and B (30 vol.% and 8 vol.%  $\text{CH}_4$ ). In winter 2009/2010  $\text{CH}_4$  concentration started to decrease in all waste layers, most probably due to the low temperatures. However,  $\text{CH}_4$  concentration increased again in spring 2010. As can be seen in Figure 4.1, in spring 2010  $\text{CH}_4$  concentration in the waste layers of lysimeters C and D ranged between 40 vol.% and 50 vol.% whereas it was still low in the lysimeters A and B (15 vol.% and 2 vol.%  $\text{CH}_4$ ) indicating a low LFG production rate.

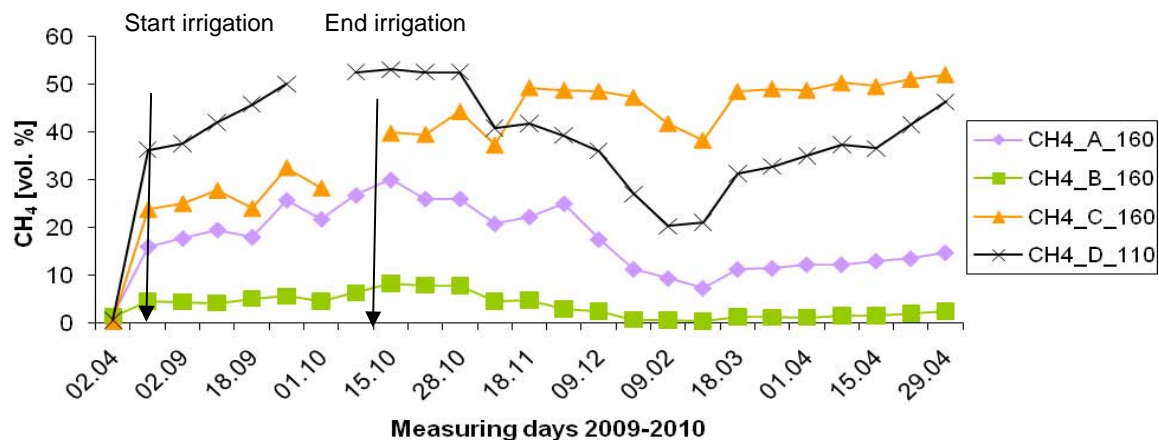


Figure 4.1: Methane development in the solid waste layers of the lysimeters A - C (at 160 cm depth) and D (at 110 cm depth)

### 4.1 Pore gas composition, temperature and water content

The gas concentration profiles of the lysimeters were determined as described above. During the monitoring period none of the solid waste layers reached a stable methane production phase, except for lysimeter C. Because of these circumstances, a systematic comparison of the different biocovers concerning their relative emission reduction effect was challenging. In general, the interpretation of gas concentration profiles in biocovers is quite difficult as gas composition in the landfill covers is controlled by numerous microbial processes (e.g. uptake and release of gases), biochemical processes (e.g.  $\text{CO}_2$  fixation) and physical conditions as well as meteorological processes (see section 2.2). Although gas concentration profiles represent a snapshot of a single measuring event, they can also give a comprehensive insight into the zoning of processes taking place in the cover layers, particularly when combined with other data. As a first step those gas concentration



profiles were compared, where the concentration of  $\text{CH}_4$  in the waste layers showed its highest value. The highest concentration of  $\text{CH}_4$  in the waste body of lysimeters A, B, and D was observed in the middle of October 2009, while the  $\text{CH}_4$  level in the waste layer of lysimeter C reached a peak at the end of April 2010. Figure 4.2 presents the gas composition ( $\text{CH}_4$ ,  $\text{CO}_2$ ,  $\text{O}_2$  and  $\text{N}_2$ ) and the temperature along the solid waste layers and the top covers of all lysimeters at selected dates. As already mentioned, the design of lysimeters C and D differed only in the thickness of the substrate layer. Although the gas and temperature profiles of lysimeters C and D are shown at different dates, the similar LFG composition at these dates allows a better comparison. The volumetric moisture contents in the upper part of the substrate layers were also in the same range (17 – 21 vol.% based on dry weight). The location of the methane oxidation zone inside the four biocovers was mainly indicated by a sharp decline in methane and oxygen.

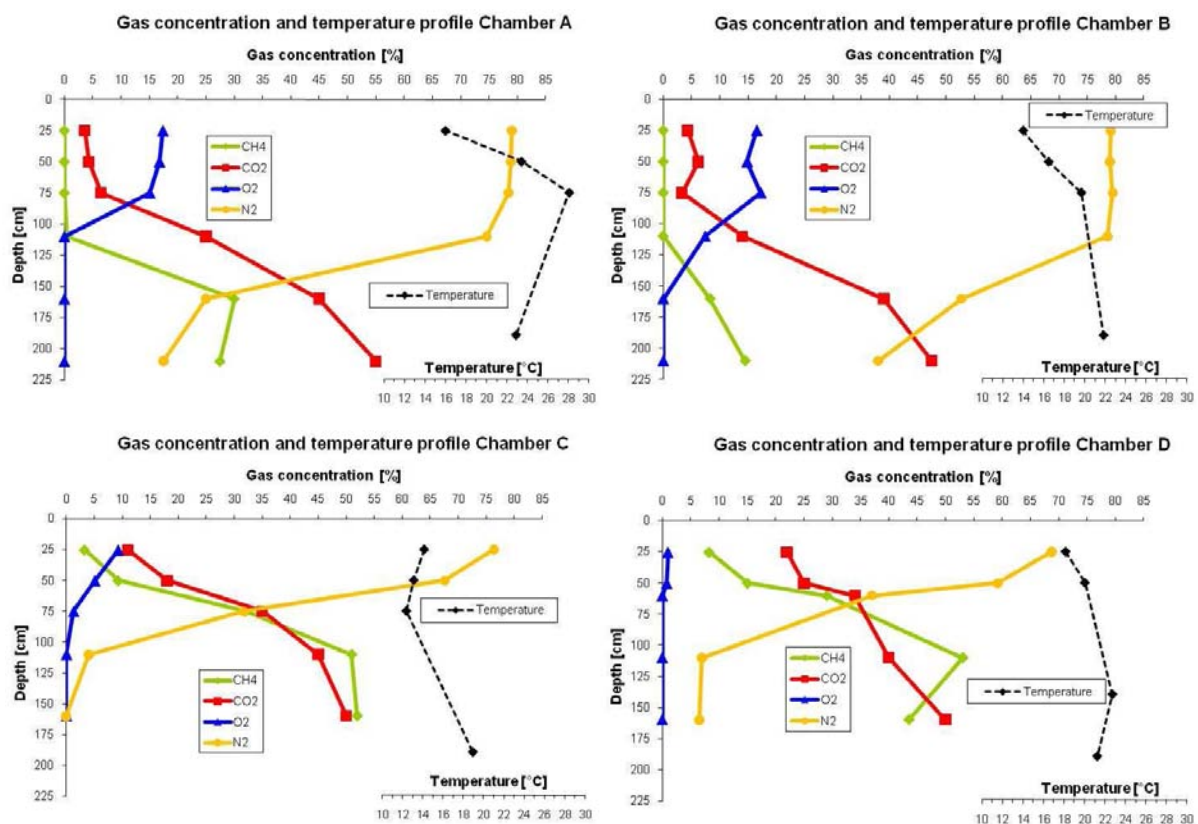


Figure 4.2: Typical soil gas concentration and temperature profiles of all lysimeters (A, B, D: 15.10.2009, C: 29.04.2010)

### Lysimeter A

The gas composition in the solid waste layer of lysimeter A contained approx. 30 %  $\text{CH}_4$ , 55 %  $\text{CO}_2$  and 15 %  $\text{N}_2$  (v/v).  $\text{O}_2$  has never been detected in the waste layer. The shift in  $\text{CH}_4$  and  $\text{CO}_2$  concentrations between 110 and 160 cm indicates that all the methane was reduced in the waste layer and the gas distribution layer beneath the compost layer. This finding was supported by the stable isotope analyses (see section 4.4). In addition, the peak in the temperature profile also indicates  $\text{CH}_4$  uptake as well as the location of the oxidation zone (note that between 75 and 190 cm no temperature sensors were installed). There was no  $\text{CH}_4$  detectable in the substrate layer as well as on the surface of the lysimeter (see appendix A.2). Similar observations have been made by Cabral et al. (2009) and Scheutz et al. (2009b), where some oxidation occurred within the gas distribution layer. A biofilter

experiment by Gebert and Gröngroft (2006b) also showed high CH<sub>4</sub> oxidation rates in coarse, purely mineral material. In the present case, it is assumed that nutrients of the sewage sludge compost might have been washed into the gas distribution layer by precipitation, promoting the methane oxidation processes in this zone. The high temperature insulation capacity of the compost material (see Figure 4.3) might have additionally supported the activity of methane oxidising bacteria within the gas distribution layer. In addition, the coarse compost substrate enabled a good aeration down to the gas distribution layer as indicated by the uniformly high N<sub>2</sub> concentration through the base of the cover. In contrast to oxygen, N<sub>2</sub> might be a better indicator of air penetration as it is neither used for CH<sub>4</sub> oxidation near the surface nor by soil respiration. However, also the CH<sub>4</sub> loading might have been quite low allowing for a large diffusive intrusion of atmospheric air.

### Lysimeter B

The gas profile of lysimeter B revealed a similar pattern, but with a lower level of CH<sub>4</sub> concentration in the solid waste layer (14.5 vol.% CH<sub>4</sub>, 47.5 vol.% CO<sub>2</sub>, 38 vol.% N<sub>2</sub>). The methane depletion also occurred at a depth of 110 to 160 cm, where the CH<sub>4</sub> concentration decreased to zero (see also stable isotope analyses, section 4.4). There was no CH<sub>4</sub> detectable in the substrate layer up to 110 cm. The nearly vertical N<sub>2</sub> concentration profile clearly indicates that the substrate (SSC-Mix) was well aerated throughout the 1 m cover depth. The O<sub>2</sub> peak at a depth of 75 cm may have resulted from a leakage of the gas probe. In the SSC-Mix layer, O<sub>2</sub> penetrated deeper into the cover layer than in the SSC, most probably due to the lower CH<sub>4</sub> loading.

### Lysimeter C

The gas concentrations in the solid waste layer of lysimeter C showed a typical landfill gas composition during the methanogenic phase (52 vol.% CH<sub>4</sub>, 45 vol.% CO<sub>2</sub>). A steady decrease in CH<sub>4</sub> concentration was observed throughout the substrate cover (TS-Fischa, SS-Zister). The concentration of CH<sub>4</sub> at a depth of 25 cm was quite low (<1 %). However, dilution of the pore gas also contributes to the decrease in CH<sub>4</sub> concentration. CH<sub>4</sub> concentrations indicate that the main part of the oxidation process has taken place at a depth between 75 cm and 50 cm from the surface since about half of the inlet concentration is reduced in this layer. The concentrations of O<sub>2</sub> and N<sub>2</sub> confirm that the soil was well aerated. Oxygen could be detected till a depth of 50 cm and deeper and N<sub>2</sub> reached down to the gas distribution layer. On the basis of the emission measurements (FID-surface screenings and surface flux measurement), the substrate cover was not efficient in oxidising the entire CH<sub>4</sub> loading. Although, the substrate cover was well aerated throughout the upper part of the cover, preferential flows along the edges of the lysimeter chamber have been observed (see section 4.2). Despite the observed CH<sub>4</sub> emissions, a significant portion of CH<sub>4</sub> was oxidised according to stable isotope analysis (see section 4.4). When the LFG in the waste layer showed a similar composition to those in lysimeter A (approx. 30 vol.% CH<sub>4</sub>, 55 vol.% CO<sub>2</sub> and 15 vol.% N<sub>2</sub>) in August/Sept. 2009, no CH<sub>4</sub> emissions could be detected at the surface by FID screening (see appendix A.2). These results indicate that the biocover system does have an ability to reduce the methane emission to a certain extent. It is currently discussed in the literature that, in addition to climate and soil type, the magnitude of methane flux into the cover material has a significant influence on oxidation. For design purposes it is not only useful to quantify the maximum methane oxidation capacity of a cover



system under specific climatic conditions but also to know the flux at which the cover can oxidise 100 % of the methane coming from below (Abichou et al., 2010).

### Lysimeter D

The LFG composition in the waste layer of lysimeter D consisted of approx. 53 % CH<sub>4</sub> and 45 % CO<sub>2</sub> (v/v). The gas profile shows a sharp decline in CH<sub>4</sub> almost throughout the substrate (TS-Fischa, SS-Zister), although the concentrations of CH<sub>4</sub> at a depth of 25 cm were still not negligible (approx. 8 vol.%). It is possible, that further oxidation may have taken place above a depth of 25 cm. However, low oxygen concentration as well as a sharp decrease in N<sub>2</sub> near the surface indicates restriction of aeration. In contrast, from December 2009 onwards higher O<sub>2</sub> concentration was measured in the upper part of the substrate cover.

According to the emission measurements (FID-surface screenings, see appendix A.2A.1A.2), the control cover showed a poor CH<sub>4</sub> oxidation efficiency, as high CH<sub>4</sub> concentrations (> 1000 ppm) could be detected along the edges of the lysimeter. The substrate cover was not even efficient in oxidising the entire CH<sub>4</sub> loading, when the gas composition was similar to those in lysimeter A (approx. 37 vol.% CH<sub>4</sub>, 55 vol.% CO<sub>2</sub> and 8 vol.% N<sub>2</sub>) (see appendix A.2 on 27.08.2009). On the other hand, a considerable percentage of CH<sub>4</sub> was oxidised based on the stable isotope analyses (see section 4.4). However, the percentage that is emitted in a more homogeneous way determines methane oxidation (Oonk, 2010). Since a significant portion of the methane load was most probably emitted through preferential flows along the edges of the lysimeter (see FID screenings, section 4.2), the oxidation based on stable isotope analyses may have been overestimated. The improvement of quantification of methane oxidation considering both homogeneous and preferential flows is currently discussed in the literature (Oonk, 2010).

### Temperature profiles

Figure 4.3 compares the seasonal temperature profiles measured in all lysimeters. The temperature profiles exhibit variations with depth and time and clearly show the different temperature performances and insulation capacities of the used substrates. The heat produced during waste degradation (and to a much lesser extent during microbial CH<sub>4</sub> oxidation) could be better maintained in the coarse sewage sludge compost (lysimeter A) than in the finer soil and compost materials (lysimeters B-D) throughout the whole monitoring period. As already mentioned, the CH<sub>4</sub> loading was always higher in lysimeters C and D compared to the others assuming a greater heat release. Nevertheless, the higher porosity (and thus higher amount of air-filled space) of the sewage sludge compost makes CH<sub>4</sub> oxidation more independent of low or varying external temperatures due to the lower thermal conductivity of the coarse substrate (Scheffer and Schachtschabel, 1992). As already described in section 2.4.5, the optimum temperature for CH<sub>4</sub> oxidation appears to be in the range of 20 – 38 °C (Visvanathan et al., 1999; Börjesson and Svensson, 1997; Figueroa 1993; Whalen et al., 1990). In summer and autumn 2009, the temperatures within the SSC-cover were about 10 to 15°C higher than in the other biocovers and remained in the vicinity of 20 – 25°C at a depth of 50 cm downwards. The shape of the temperature profile of lysimeter A, therefore, indicates more favourable conditions for CH<sub>4</sub> uptake during this period compared to the other substrate covers. During winter 2009/2010 the temperatures within all cover materials were relatively low (< 10°C) which might have reduced CH<sub>4</sub> oxidation. However, also waste decomposition in all lysimeters, except for lysimeter C, decreased during this time as indicated by decreasing CH<sub>4</sub> concentration levels in the waste layers (see Figure 4.1).

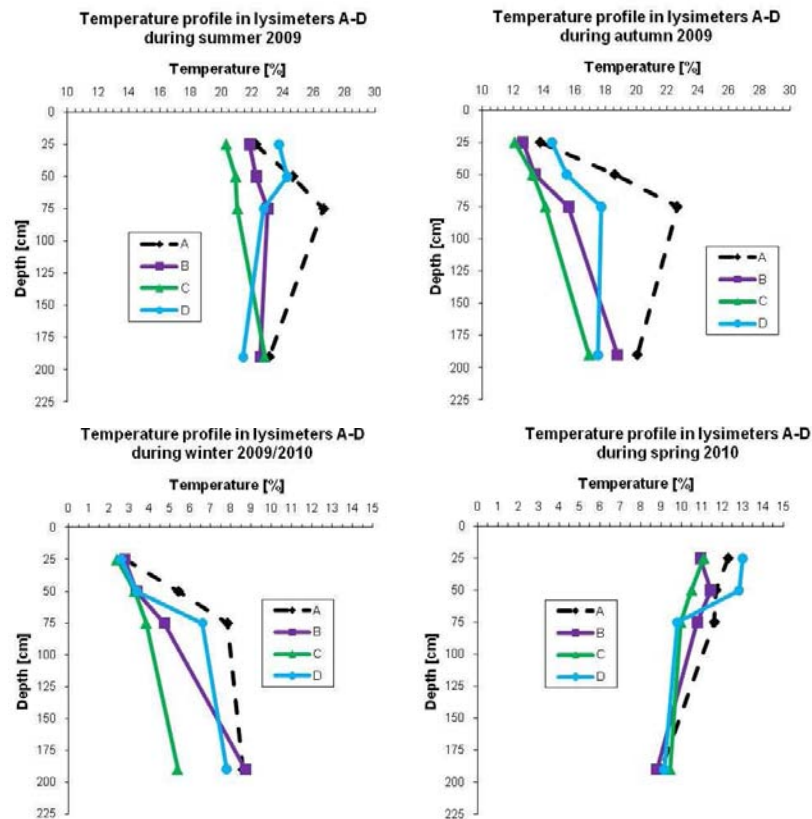


Figure 4.3: Comparison of seasonal, average temperature profiles in all lysimeters during the monitoring period from August 2009 to April 2010 (SD in summer: 0.4 – 1.7; SD in autumn: 3.2 – 5.3; SD in winter: 1.5 – 3.3; SD in spring: 1.4 – 5.9)

### Moisture content profiles

As discussed previously, the moisture content is a crucial factor influencing  $\text{CH}_4$  oxidation. In Figure 4.4 the seasonal moisture contents measured at different depths of the four lysimeters are depicted. The water content depended mainly on precipitation and temperature as well as on the capacity of the substrates to retain or release moisture. The average moisture content in all waste layers remained stable at approx. 50 vol.% throughout the entire monitoring period. In general, the volumetric moisture content was lower in the upper layers than in the lower ones, except for lysimeter B. Moisture content in the soil layers of lysimeters C and D (TS Fische, SS Zister) was not significantly influenced by the season. During the investigation period the moisture content in the top layer of lysimeter C (25 cm) remained almost stable at about 20 % (based on dry weight) which correspond to 50 % WHC in that material (TS-Fische). This is in the optimum range for methane oxidation (Boeckx et al., 1996; Figueroa, 1993). In the lower layer (75 cm) of the substrate (SS-Zister) higher moisture contents were found (approx. 40 vol.%) corresponding to more than 100 % WHC. In the top layer of lysimeter D (25 cm) the actual moisture content ranged between 24 – 32 vol.% (60 – 80 % WHC), while in the lower layer (50 cm) higher moisture contents were detected varying from 31 – 41 vol.% (85 – 100 % WHC). The high water levels in the lower layers of lysimeter C and D may have blocked gas transport causing bypass flows along the edges of the lysimeters (see section 4.2) and/or a reduction of methane consumption. Near the surface of lysimeter B (sand) the greatest fluctuations occurred with the lowest moisture content in summer (6.4 vol.%, 25 % WHC) and the highest in winter (22 vol.%, 80 % WHC). Lower moisture levels were found in the deeper layer (SSC-

Mix) varying from 6 – 13 vol.% (15 – 30 % WHC) due to the higher water holding capacity of SSC-Mix. In the top layer of lysimeter A (SSC) the moisture content was close to the lower range (< 10 %) while in deeper regions higher moisture content occurred at approx. 36 vol.% (30 % WHC). According to Huber-Humer (2004), the threshold value for microbial activity in compost materials was found at a moisture content of about 25 % WHC under laboratory conditions. As can be seen in Figure 4.4, the moisture content in the lower layer of lysimeter A never fell below that threshold while the moisture level in the deeper region of lysimeters C and D may have caused a decline in methane oxidation rates.

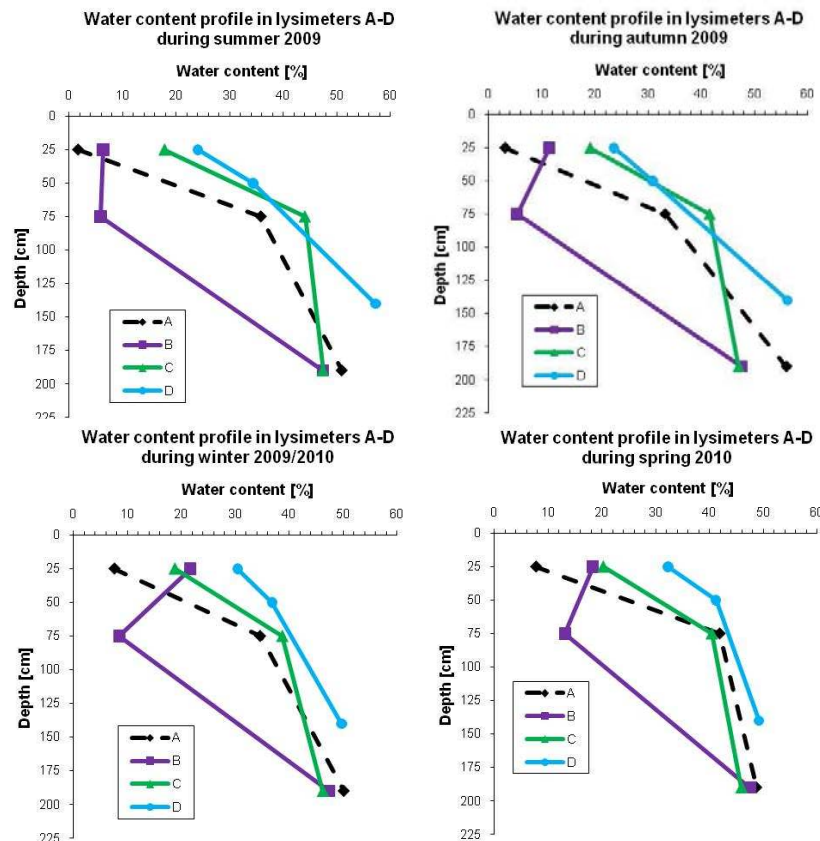


Figure 4.4: Comparison of seasonal, average water content profiles in all lysimeters during the monitoring period from August 2009 to April 2010 (SD in summer: 0.1 – 2.9; SD in autumn: 0.2 – 6.5; SD in winter: 0.1 – 3.0; SD in spring: 0.1 – 3.3)

## 4.2 Screening of methane surface concentration (FID-measurements)

To qualitatively survey the overall CH<sub>4</sub> emission pattern from the different biocovers and the expected escape routes, repeated FID surface screenings were performed. Figure 4.5 shows examples of surface CH<sub>4</sub> concentration patterns of lysimeter C and D at selected dates. During the monitoring period from August 2009 to April 2010 no methane emissions above background level were detected on the biocover surfaces of lysimeters A and B (see appendix A.2). The data gathered from FID screenings for CH<sub>4</sub> surface concentrations indicates that CH<sub>4</sub> emissions varied significantly between the different measuring events and suggests that landfill gas was distributed unevenly to the substrate layers.

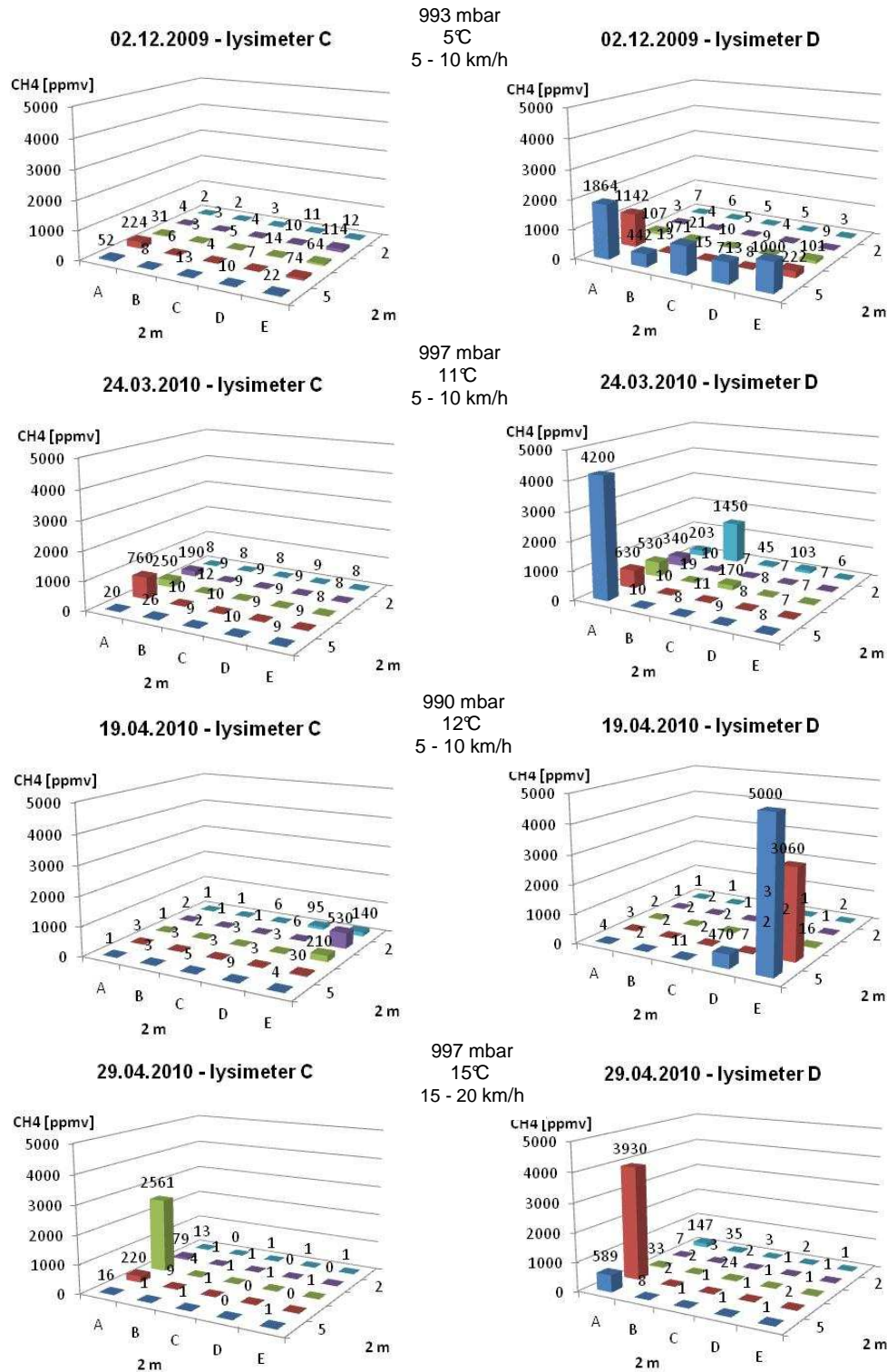


Figure 4.5: Spatial and temporal variability of surface CH<sub>4</sub> concentrations within a 50 cm grid on lysimeters C and D for selected measuring events (02.12.2009, 24.03.2010, 19.04.2010, 29.04.2010)

In general, higher surface CH<sub>4</sub> concentrations were observed along the edges of the lysimeters C and D. This indicates that a main part of the load was coming from the sides of the lysimeters through the gas distribution layer, although the walls of the lysimeter chambers were sealed with framed rubber lips at the upper part of the substrate layers. It can be hypothesised that the high moisture content in the soil layers of lysimeters C and D may have blocked the gas transport. However, the preferential flows may also be an artefact of the lysimeter facility itself. In contrast,

stable isotope analyses show that a significant portion of CH<sub>4</sub> could be oxidised in the substrate covers (see section 4.4). As already mentioned the preferential flows along the edges of the lysimeters had a significant influence on the actual oxidation. In general, loamy soils tend to develop crack formations additionally supporting the preferential channels along the edges.

Methane emission through the substrate layer of lysimeter C seemed to be relatively low as also indicated by the surface flux measurements (0 – 1.1 g/m<sup>2</sup>d, see section 4.3). The surface screening showed ambient CH<sub>4</sub> concentrations ranging from background concentrations up to 760 ppmv. Only on 29.04.2010 were CH<sub>4</sub> concentrations higher than 2,500 ppmv detected. The general picture was that methane escaped preferentially from the eastern and western edges of lysimeter C.

Even though the CH<sub>4</sub> loading was always higher in lysimeter C than in lysimeter D (as indicated by higher CH<sub>4</sub> concentration in the LFG, see Figure 4.1), higher CH<sub>4</sub> concentrations were found at the cover surface of lysimeter D (control cover). Highest CH<sub>4</sub> emissions escaped along the edges, suggesting that the retention time might have been too low for CH<sub>4</sub> oxidation to occur. The CH<sub>4</sub>/CO<sub>2</sub> ratio curve of lysimeter D further indicates the occurrence of preferential flows (see Figure 4.19). Following the surface flux measurements (see section 4.3), CH<sub>4</sub> emissions from lysimeter D (0.9 – 8.2 g/m<sup>2</sup>d) were significantly greater than fluxes from lysimeter C (0 – 1.1 g/m<sup>2</sup>d).

### 4.3 Flux measurements

Table 4.1 lists the average CH<sub>4</sub> and CO<sub>2</sub> emissions from the 6 – 8 measurement campaigns conducted on the four lysimeters, together with the temperature and wind speed of the measuring day as well as the pressure change 24h prior to the beginning of the measuring day (9am to 9am). In addition, the pressure change over the measuring day (9am to 5pm) is presented.

As can be seen in Figure 4.6 and Table 4.1, all measurement campaigns were done during stable pressure conditions (during measuring day: -0.4 to 0.3 hPa/h; 24 h prior to the measurement campaigns: -0.3 to 0.3 hPa/h) and wind speeds < 4.2 m/s. Consequently, no indication was obtained that atmospheric pressure influences landfill gas emissions. Only the measurement campaigns on 10.12.2009 and 29.04.2010 were done during decreasing pressure ( $\Delta p > 0.6$  hPa/h). Between the middle of December and end of March no surface emission measurements were performed.



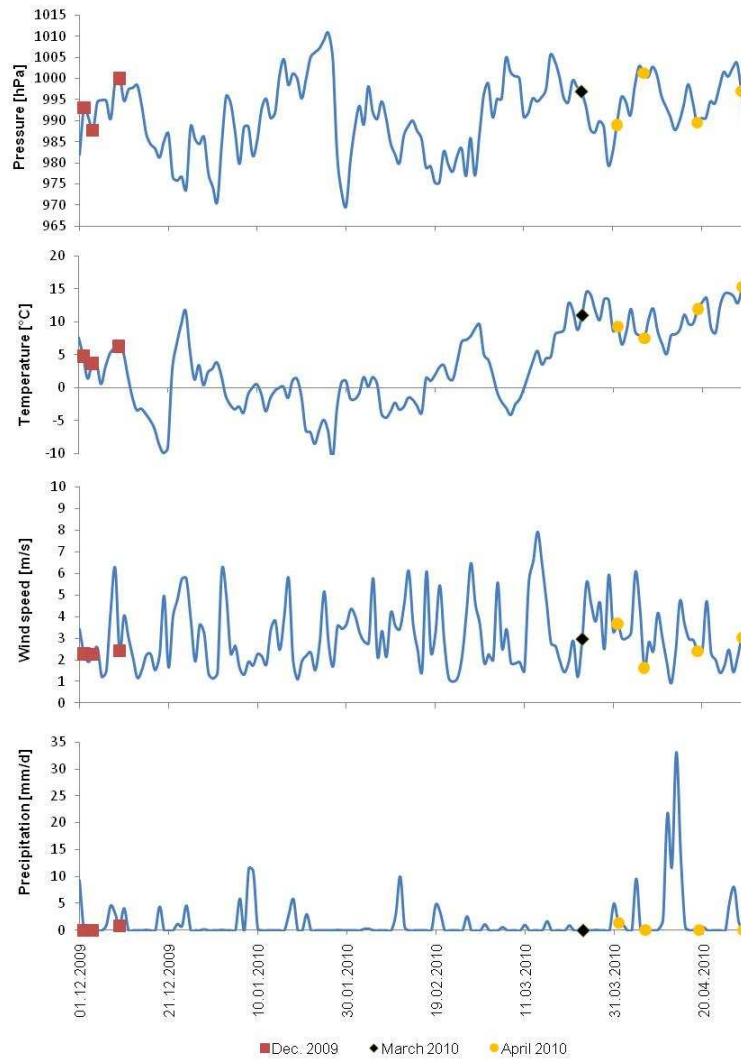


Figure 4.6: Weather conditions under measurement campaigns: atmospheric pressure, temperature, wind speed and precipitation

The average methane emissions from lysimeter D ranged from 0.9 – 8.2 g/m<sup>2</sup>d whereas lysimeter C had almost zero CH<sub>4</sub> emissions (0 – 1.1 g/m<sup>2</sup>d) during the measurement campaigns. In contrast, no CH<sub>4</sub> emissions were detected from the cover surface of lysimeters A and B. Carbon dioxide emissions were 2.9 – 21.9 g/m<sup>2</sup>d from lysimeter A, -4.7 (uptake) to 16 g/m<sup>2</sup>d from lysimeter B, 5.8 – 9 g/m<sup>2</sup>d from lysimeter C and 6.1 – 22 g/m<sup>2</sup>d from lysimeter D. It should be noted that the ability to compare emission data between lysimeters is limited due to the varying methane development in the solid waste layers of the lysimeters (see Figure 4.1).

Table 4.1: Results of the measurement campaigns

Lys.	Date	$\Delta p^a$ [hPa/h]	$\Delta p^b$ [hPa/h]	T [°C]	Wind speed [m/s]	Emissions [g/m <sup>2</sup> d]		Std. error	
						CH <sub>4</sub>	CO <sub>2</sub>	CH <sub>4</sub>	CO <sub>2</sub>
D	02.12.09	0.6	0.0	7.2	1.6	2.0	10.0	0.6	2.6
D	04.12.09	-0.26	0.34	3.5	1.5	8.2	22.0	1.0	2.4
A	10.12.09	0.22	-0.71	7.4	1.8	0.0	21.9	-	2.3
B						0.0	16.0	-	1.9
C						1.1	5.8	0.1	0.3
D						2.3	9.0	2.1	4.5
A	24.03.10	-0.06	-0.35	16.1	4.2	0.0	8.8	-	0.3
B						0.0	5.5	-	1.2
C						0.5	6.9	0.1	1.4
D						2.9	11.1	0.2	0.8
A	01.04.10	0.27	-0.19	15.3	3.8	0.0	2.9	-	3.0
B						0.0	5.5	-	0.8
C						0.0	7.5	-	0.6
D						0.5	6.1	0.2	2.5
A	07.04.10	-0.09	-0.33	12.8	1.8	0.0	13.5	-	1.8
B						0.0	6.5	-	0.5
C						1.0	8.2	0.3	1.0
D						0.9	7.8	0.1	0.7
A	19.04.10	-0.31	-0.10	18.2	3.7	0.0	10.0	-	0.5
B						0.0	-4.7	-	0.9
C						0.7	7.7	0.2	0.4
D						1.5	9.0	1.1	2.5
A	29.04.10	-0.26	-0.69	20.8	4.1	0.0	11.3	-	5.9
B						0.0	8.3	-	0.3
C						0.6	9.0	0.1	0.4
D						1.4	11.4	0.5	1.8

<sup>a</sup> The pressure gradient 24 h prior to the measurement campaign; <sup>b</sup> The pressure gradient is the pressure difference over the measuring day from 9 am to 5 pm; Temperature (T) and wind speed are the average between 9 am and 5 pm

In general, methane emission fluxes from landfill show high spatial and temporal variability. Table 4.2 gives a summary of methane emissions released from different landfills and large field trials. The measured surface emissions correspond with the results reported in Barlaz et al. (2004) and Maurice and Lagerkvist (2003). However, due to the use of different measuring methods, surface emission rates are hardly comparable.

Table 4.2: Summary of methane emissions rates from landfills and large field trials (adapted from Scheutz et al., 2009)

Reference	Country	Cover material	GES	Approach	CH <sub>4</sub> emission rate [g/m <sup>2</sup> d]		
					Average (n)	Min.	Max.
Maurice & Lagerkvist (2003)	Sweden	120 cm silty soil	no	SCT	0.35 (3-12)	<0.04	2
Scheutz et al. (2008)	France	30 cm top soil on 70 cm clay	yes	SCT	-0.001 (12)	-2.5	29
Scheutz et al. (2003)	France	80 cm loam on 40 cm coarse sand	yes	SCT	2 (23)	-0.01	10
		40 cm coarse sand	yes	SCT	37.8		49.9
Börjesson and Svensson (1997)	Sweden	10-80 cm sandy loam	-	SCT	0.01-7.7 (72)	-0.3	18.4
Barlaz et al. (2004)	USA	100 cm clay soil cover	no	SCT			>15
		100 cm yard waste compost underlain by 15 cm gravel and 15 cm clay	no	SCT		-1.73	1.33
Jugnia et al. (2008)	Canada	80 cm sand/compost	no	CDC	2.5 – 30		210
Humer & Lechner (2001a,c)	Austria	90 cm compost underlain by 30 cm gravel	yes	DFT	0-0.3 (27)	-0.5	2.1
		90 cm compost underlain by 30 cm gravel	yes	DFT	0-1.9 (27)	-3.3	5.6
		40 cm compost cover	yes	DFT	0-248.9 (20)	0	706.8
Stern et al. (2007)	USA	50 cm compost underlain by 15 cm crushed glass and 65-75 cm clay cover	-	SCT	2 (44)		
		65-75 cm (control cover)	-	SCT	10.6 (45)		
		30 cm compost underlain by 15 cm crushed glass and 15 cm clay	-	SCT	300 - 700		
		60 cm compost underlain by 15 cm crushed glass and 15 cm clay	-	SCT	30 - 70		

SCT, static chamber method; DFT, dynamic flux tunnel; CDC; closed dynamic chamber; GES, gas extraction system



In Figure 4.7, a graphical presentation of the emission data in Table 4.1 can be seen.

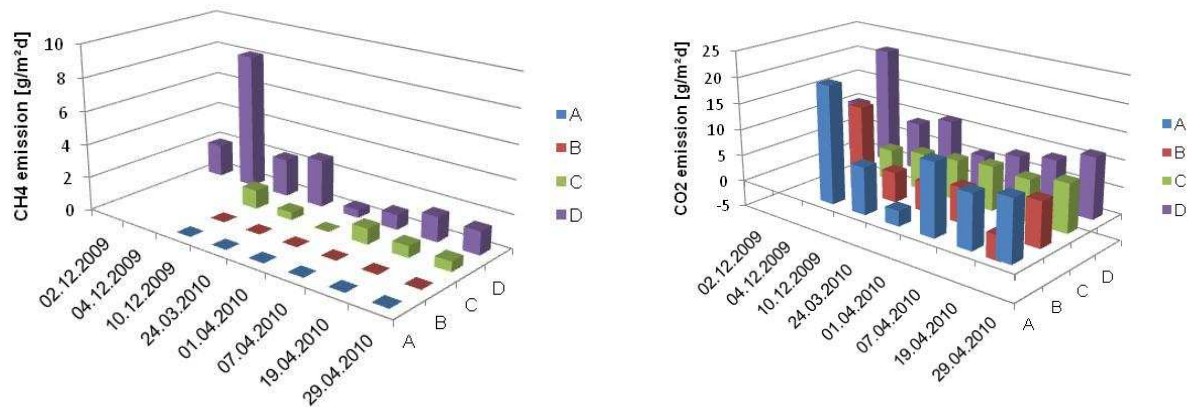


Figure 4.7: Graphical presentation of the emission data in Table 4.1

The individual emission data of the lysimeters are addressed particularly in the following.

#### Lysimeter A

In Figure 4.8 the development of CO<sub>2</sub> emissions of lysimeter A over six measuring campaigns are depicted together with the air and soil temperatures of the respective measuring days. In general, results of surface emission measurements showed high CO<sub>2</sub> emissions and no CH<sub>4</sub> emissions indicating significant methane oxidation in lysimeter A. However, respiration of compost may also contribute to the overall CO<sub>2</sub> emissions, though the compost material placed in lysimeter A had a very low oxygen consumption ( $AT_4 = 0.7 \text{ mg O}_2/\text{g DM}$ ) under laboratory conditions. Experiments by Scheutz et al. (2009b, c) showed that respiration of compost (placed in a biowindow) generated significant CO<sub>2</sub> emissions even though the compost has been originally characterised as mature and stable. With the exception of the CH<sub>4</sub> and CO<sub>2</sub> concentrations measured on 10.12.2009, the CH<sub>4</sub> and CO<sub>2</sub> concentrations at the bottom of the waste layer remained at approx. 17 vol.% and 35.5 vol.%, respectively, as can be seen in Figure 4.9. On 10.12.2009, CH<sub>4</sub> and CO<sub>2</sub> concentrations were slightly higher (22 vol.% CH<sub>4</sub>, 45 vol.% CO<sub>2</sub>) explaining the high observed CO<sub>2</sub> emission rate on this day.

Compared to the other lysimeters, almost higher CO<sub>2</sub> emissions were observed at the biocover surface of lysimeter A but with lower CO<sub>2</sub> concentrations in the upper part of the substrate cover. This indicates a higher dilution in the porous sewage sludge than in the finer soil and compost materials placed in lysimeters B, C and D. In general, materials with a high air-filled pore space such as composts enable higher diffusive or advective intrusion of atmospheric air (Mor et al., 2006). It is also hypothesised that CO<sub>2</sub> emission rates (and also CH<sub>4</sub> emissions) were underestimated with lysimeter C and D since high volumetric moisture contents in the upper part of the soil layers may have blocked gas transport causing bypass flows along the edges of the lysimeters (see the discussions below).

Several studies have shown that CH<sub>4</sub> uptake by methanotrophs increases with rising temperatures (Gebert et al., 2003; Boeckx et al., 1996; Figueroa, 1993; Whalen et al., 1990) when the CH<sub>4</sub> concentration, among other factors, is not limiting (Einola et al., 2007). Although LFG concentration remained almost stable during spring measurement campaigns, no significant relation between increasing air temperature

and CO<sub>2</sub> emissions has been found. It is hypothesised that either methane was a limiting factor or vegetation had a great influence on CO<sub>2</sub> emission measurement.

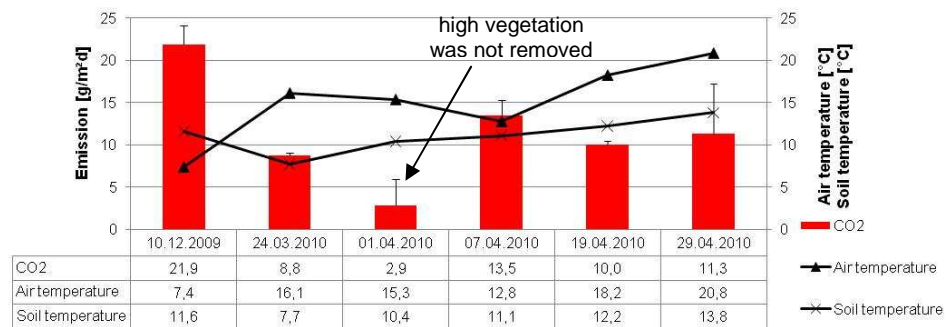


Figure 4.8: Development of CO<sub>2</sub> emissions of lysimeter A and the corresponding soil temperature at 50 cm depth and air temperature. The chamber data are shown as the mean of 3 measurements with error bar representing the positive standard deviation.

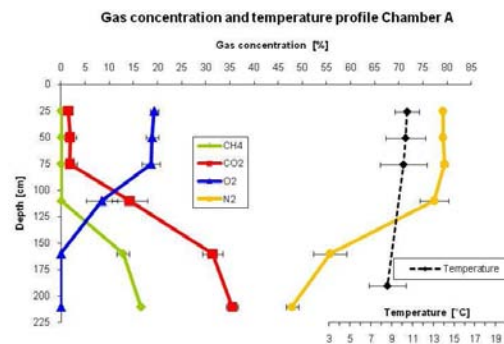


Figure 4.9: Average soil gas concentration and temperature profile of lysimeter A during spring 2010.

Except for 10.12.2009, all surface emission measurements were conducted during the growing season. In the presence of landfill gas, the vegetation developed differently on lysimeters A-D, as can be seen in Figure 4.10. Although specific plant species were sown (see Internal Report Nutzraum 2008), spontaneous vegetation developed on the surfaces of the lysimeters. The vegetation coverage was about 30 - 50 % at lysimeters C and D whereas lysimeter A was fully covered. At the end of April the vegetation coverage of lysimeter B was close to 100 %. It seems that the compost material placed in lysimeter A provided a better nutrient supply for vegetation than mineral soils.



Figure 4.10: Vegetation condition of the lysimeters A, B, C and D (from left to right) on 01.04.2010 (top) and 29.04.2010 (bottom)

Prior to surface flux measurements, grass and small plants were moved from the cover soil to avoid plant respiration and photosynthesis. However, due to the small chamber height (14.5 cm), the vegetation's condition had an effect on the CO<sub>2</sub> emission rate even when the vegetation height was low. As can be seen in Figure 4.8, the CO<sub>2</sub> emission rate on 01.04.2010 was relatively low compared to the other measurement campaigns. The vegetation was not cut on this day, as the main part of the vegetation was short (< 5 cm) and only some plant species were higher than 10 cm. To investigate the influence of vegetation on the CO<sub>2</sub> emission measurement in the chamber, surface emission measurements were performed before and after vegetation was cut. As shown in Figure 4.11, CO<sub>2</sub> emission rate increased 4.5-fold after high vegetation was removed indicating the photosynthetic use of CO<sub>2</sub> by vegetation. On 29.04.2010 even a higher increase (7.5-fold) was observed. However, no plant physiological investigations have been conducted so far in order to verify these observations. But several studies have demonstrated that the net photosynthetic rate of a wide range of plant species is stimulated when plants are exposed to an elevated CO<sub>2</sub> environment (Lewis et al., 2002; Ziska and Bunce, 1997; Arp, 1991). In order to improve the CO<sub>2</sub> emission measurement, the chamber could be equipped with both a transparent and an opaque cover. Measuring the CO<sub>2</sub> flux under both light and dark conditions will result in estimates of the assimilation by the vegetation.

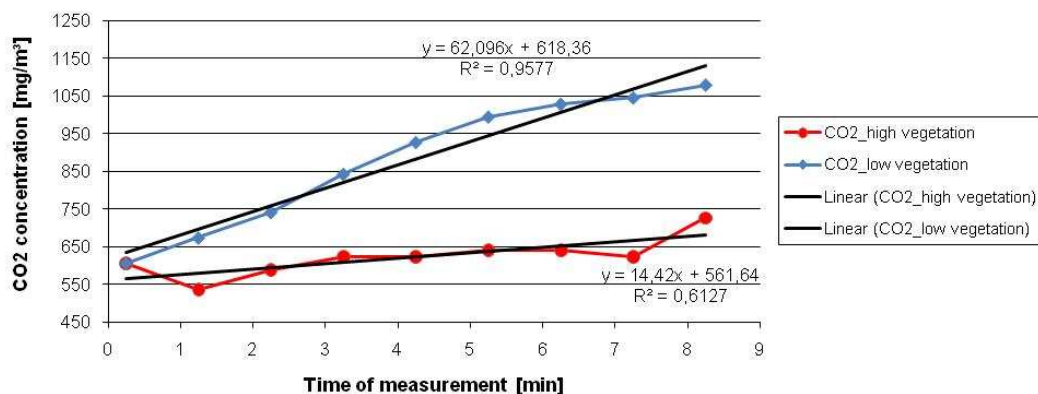


Figure 4.11: Influence of vegetation height on CO<sub>2</sub> emission measurement (07.04.2010)

### Lysimeter B

The surface emissions of lysimeter B revealed a similar pattern, but with a lower load of LFG in the solid waste layer (4 vol. % CH<sub>4</sub>, 30 vol. % CO<sub>2</sub>, see Figure 4.13). Elevated CO<sub>2</sub> emissions and no CH<sub>4</sub> emissions were detected during surface emission measurements (see Figure 4.12). The higher CO<sub>2</sub> emission rate observed on 10.12.2009 might be a result of the higher LFG concentration measured at the bottom of the waste layer (9 vol. % CH<sub>4</sub>, 45 vol. % CO<sub>2</sub>). Similar to lysimeter A, no significant relation between increasing air temperature and CO<sub>2</sub> emissions has been found. It seems that methane was a limiting factor due to low concentrations observed in the waste layer. Besides, vegetation also influenced CO<sub>2</sub> emission measurements. On 19.04.2010 CO<sub>2</sub> emission was negative indicating that much of the carbon dioxide emitted from the biocover surface can be rapidly assimilated by plant photosynthetic processes. The main height of the vegetations was < 5 cm, only some plant species were higher than 10 cm. CO<sub>2</sub> uptake has also been reported by Börjesson and Svensson (1997) and Chen et al. (2008).

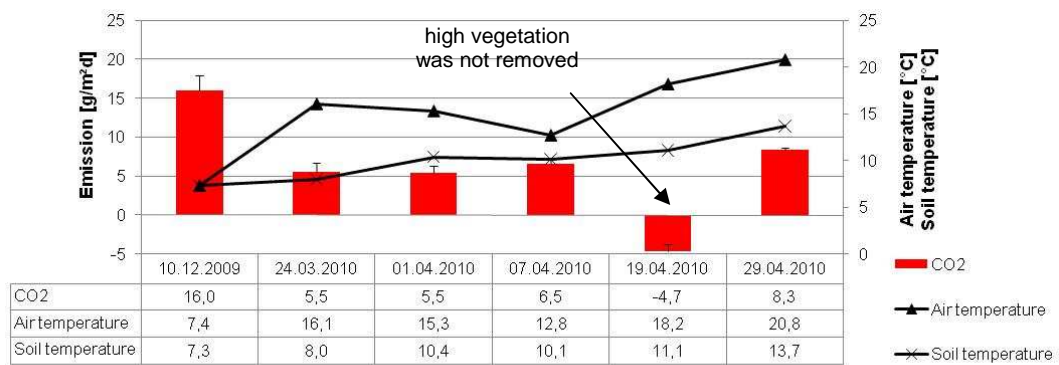


Figure 4.12: Development of CO<sub>2</sub> emissions of lysimeter B and the corresponding soil temperature at 50 cm depth and air temperature. The chamber data are shown as the mean of 3 measurements with error bar representing the positive standard deviation.

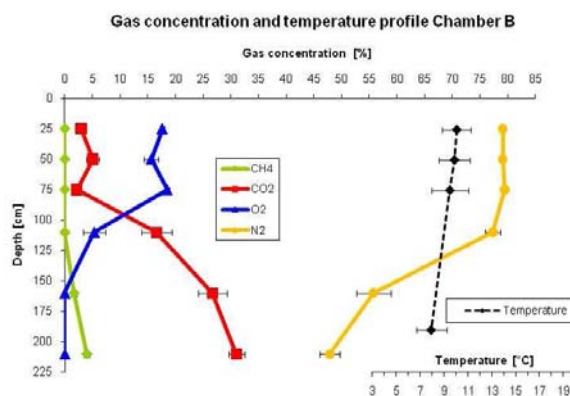


Figure 4.13: Average soil gas concentration and temperature profile of lysimeter B during spring 2010

The influence of vegetation was also investigated during surface emission measurement at lysimeter B on 29.04.2010. Similar to the investigations at lysimeter A (see Figure 4.11) CO<sub>2</sub> emissions increased approx. 4-fold after higher vegetation (> 10 cm) was removed. The measured CO<sub>2</sub> emission rates should be interpreted with caution, as they represent only snapshots of a single measuring event. In general, CO<sub>2</sub> emissions are low in the daytime due to photosynthesis and increase at night. In addition, CO<sub>2</sub> emissions can vary during a day. Park and Shin (2001) reported maximum CO<sub>2</sub> fluxes when the temperature was at its peak. Although CO<sub>2</sub> emissions were corrected for the temperature measured inside the enclosed chamber, humidity also influences CO<sub>2</sub> emissions. However, relative humidity inside the chamber was not monitored during the sampling period. In order to crosscheck the disturbance effect of relative humidity change on CO<sub>2</sub> concentration, a background measurement and alignment should be done in future studies using the closed dynamic chamber.

### Lysimeter C

The concentrations of CH<sub>4</sub> and CO<sub>2</sub> in the biogas sampled at the base of the waste layer were around 50 vol.% during all surface flux measurements (see Figure 4.1). This indicates that the methane supply was not a limiting factor during this period. As can be seen in Figure 4.14, methane emissions remained low during these measurement campaigns, while carbon dioxide emissions were increasing over time as the temperature rose.



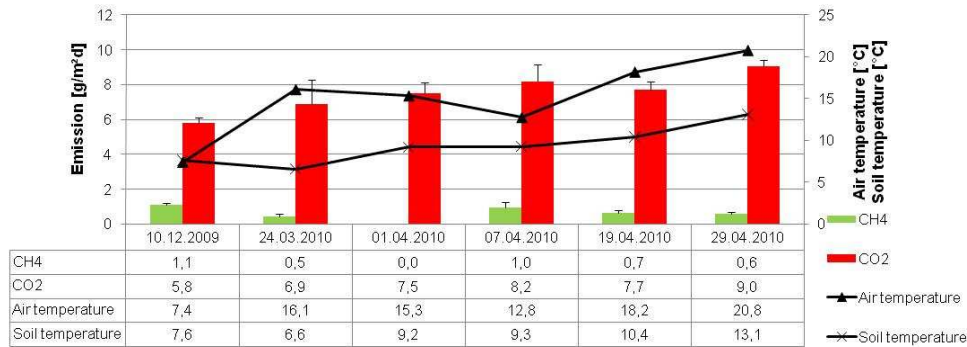


Figure 4.14: Development of CH<sub>4</sub> and CO<sub>2</sub> emissions of lysimeter C and the corresponding soil temperature at 50 cm depth and air temperature. The chamber data are shown as the mean of 3 measurements with error bars representing the positive standard deviation.

Figure 4.15 presents the average soil gas concentration and temperature profile during spring 2010.

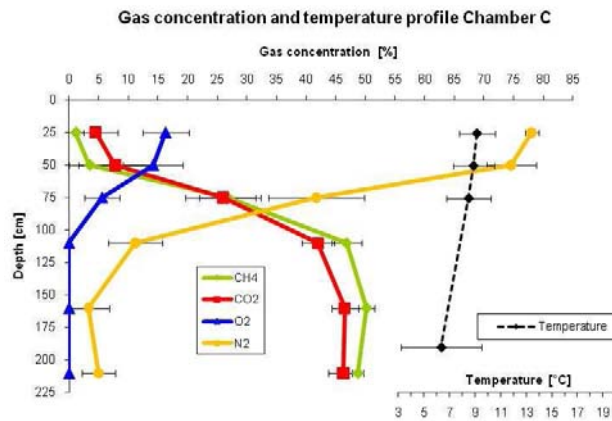


Figure 4.15: Average soil gas concentration and temperature profile of lysimeter C during spring 2010

In Figure 4.16 CO<sub>2</sub> emissions from lysimeter C related to atmospheric pressure and air temperature are presented. The trend line indicates a significant relation between change of air temperature and CO<sub>2</sub> emissions ( $R^2=0.79$ ,  $n=18$ ). Fluxes of CO<sub>2</sub> were also positively correlated with soil temperatures at 25 cm and 50 cm depth ( $R^2=0.67$  and  $R^2=0.52$ ). In contrast, CO<sub>2</sub> emissions were only weakly correlated with soil temperature at 75 cm depth ( $R^2=0.33$ ). No correlations were obtained between CO<sub>2</sub> emissions and atmospheric pressure change ( $R^2 = 0.02$ ). Similar observations have been made by Scheutz et al. (2009b), where CO<sub>2</sub> emission depended more on temperature ( $R^2=0.62$ ) than on barometric pressure ( $R^2=0.03$ ). By excluding the day 01.04.2010, CH<sub>4</sub> emissions showed a negative correlation with air temperature ( $R^2=0.57$ ,  $n=15$ ). Since CH<sub>4</sub> concentrations were not limiting, these observations strongly suggest that the increasing CO<sub>2</sub> emissions and decreasing CH<sub>4</sub> emissions over time were most likely due to the higher temperatures resulting in higher methane oxidation rates.

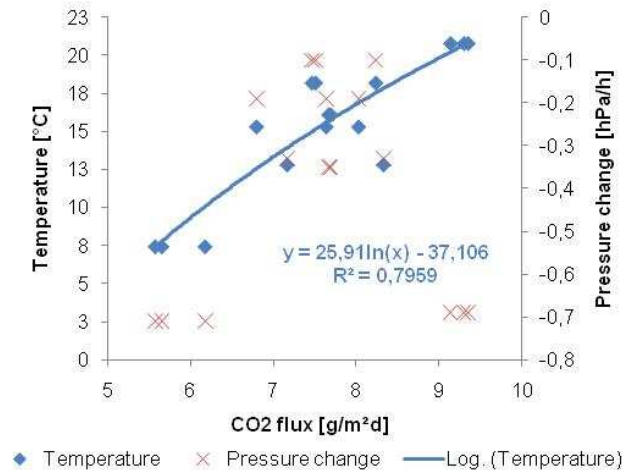


Figure 4.16: CO<sub>2</sub> emissions from lysimeter chamber C related to temperature and atmospheric pressure change (9am – 5 pm on the measuring day)

However, results obtained by the surface flux measurements should be interpreted with caution, not only for CH<sub>4</sub> but also CO<sub>2</sub>. As can be seen in Figure 4.4, the high volumetric moisture content in lysimeter C, especially in the upper part of the substrate cover may have blocked gas transport causing bypass flows along the edges of the lysimeters. In contrast, gas permeability tests conducted at the Austrian Institute of Technology in Seibersdorf revealed that all substrates used in this study should ensure sufficient air permeability even at 100 % WHC. Despite the use of a plastic skirt attached on the closed chamber for sealing purposes, a great portion of the methane and carbon dioxide flux may have bypassed leading to an underestimation of CH<sub>4</sub> and CO<sub>2</sub> emissions.

#### Lysimeter D

The development of CH<sub>4</sub> and CO<sub>2</sub> emissions of lysimeter D over eight measuring campaigns are presented in Figure 4.17 together with the air and soil temperatures of the respective measuring days. As can be seen in Figure 4.1, methane concentration in the biogas sampled at the base of the waste layer started to decrease after the end of October 2009. During the surface emissions measurement, CH<sub>4</sub> concentrations were around 40 vol.% in the waste layer (see Figure 4.18). Clear differences in CH<sub>4</sub> emissions were observed between both the growing and the winter season. Compared to lysimeter C, CH<sub>4</sub> and CO<sub>2</sub> fluxes were weakly, negatively correlated with temperature ( $R^2=0.30$  and  $R^2=0.36$ , respectively). There are no specific reasons for the relatively high CH<sub>4</sub> emission rate recorded on 04.12.2009. However, several studies have demonstrated diurnal variation in the release of CH<sub>4</sub> from landfills (Börjesson and Svensson, 1997). There was almost no CH<sub>4</sub> detectable in the substrate layer up to 50 cm. Although a significant portion of CH<sub>4</sub> could be oxidised (see stable isotope analyses, section 4.4), the main part of the CH<sub>4</sub> load most probably came from the side of the lysimeter through the gas distribution layer beneath the substrate layer (see Figure 4.5). The high volumetric moisture content throughout the soil profile (see Figure 4.4) remained high during surface flux measurements (approx. 32 vol.% based on dry weight, 80 – 100 % WHC) and probably caused a blockage for gas transport. As a consequence, the absolute data should be interpreted with caution. As already discussed, preferential flows decrease the overall oxidation efficiency of the biocover. Consequently, the oxidation based on stable isotope analyses may have been overestimated since only

the methane load that is emitted in a more homogeneous way determines methane oxidation.

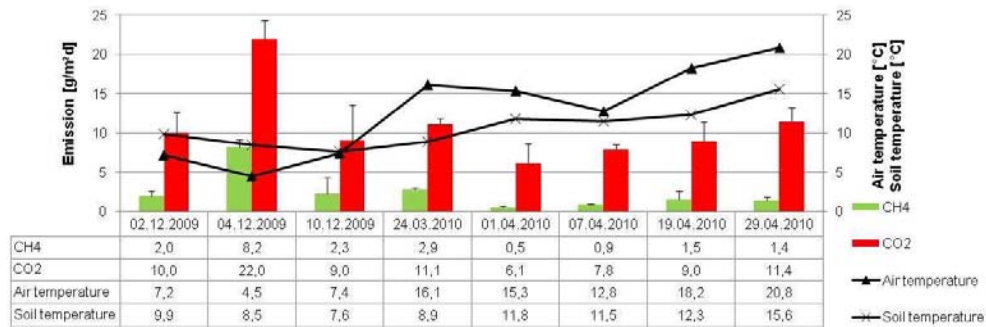


Figure 4.17: Development of CO<sub>2</sub> emissions of lysimeter D and the corresponding soil temperature at 50 cm depth and air temperature. The chamber data are shown as the mean of 3 measurements with error bars representing the positive standard deviation.

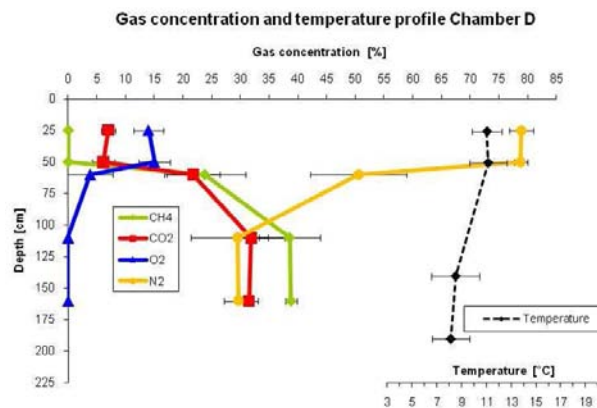


Figure 4.18: Average soil gas concentration and temperature profile of lysimeter D during spring 2010

In general, CH<sub>4</sub> emissions from lysimeter D (0.9 – 8.2 g/m<sup>2</sup>d) were significantly higher than fluxes from lysimeter C (0 – 1.1 g/m<sup>2</sup>d). This result indicates that low-dimensional covers do not achieve as high CH<sub>4</sub> oxidation capacities as properly designed biocovers.

Figure 4.19 presents typical CH<sub>4</sub>/CO<sub>2</sub> ratio curves for all lysimeters. In general, a decreasing ratio is a strong indication for methane consumption. The shape of the curves further confirms the location of the oxidation zone, which is indicated by the sharpest decline in the curves.

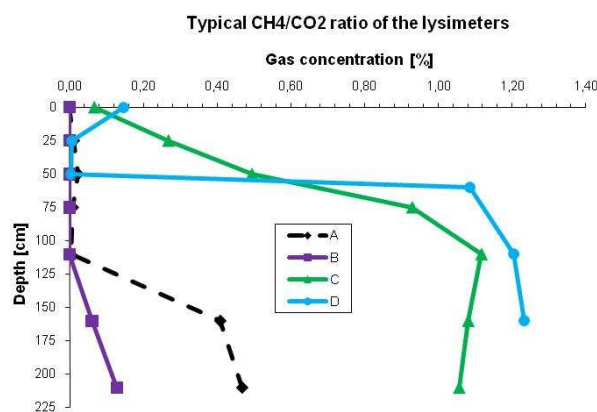


Figure 4.19: Typical CH<sub>4</sub>/CO<sub>2</sub> ratio of the lysimeters during the study period



For lysimeters C and D the ratios changed from about 1.1 - 1.2 to very low ratios, even approaching to zero. However, for lysimeter D the CH<sub>4</sub>/CO<sub>2</sub> ratio of the emitted landfill gas captured within the closed chamber (depth 0 cm) was higher than in the subsurface. This finding is a further indication for preferential flows along the edges of lysimeter D.

#### 4.4 Isotope fractionation

Two sets of samples (21.10.2009 and 09.12.2009) from each depth of the cover profile were subjected to an analysis for stable isotopes. Stable isotope analyses were used as a first indication to what extent the biogas had been oxidised within the different biocovers. Table 4.3 presents the  $\delta^{13}\text{C}$  values of both sampling dates as well as the percentage of oxidised CH<sub>4</sub> for open and closed systems (equation 3.7 and 3.8, see section 3.3.6) and the fractionation factor for bacterial oxidation,  $\alpha_{\text{ox}}$ , corrected for temperature using Eq. 3.9 (see section 3.3.6). The value used for average  $\alpha_{\text{ox}}$  (1.027) was determined according to literature values (Table 3.8) in the temperature range of 6 – 26°C. This approach was also conducted by Cabral et al. (2009). Samples from the deepest waste layers had the lowest CH<sub>4</sub>- $\delta^{13}\text{C}$  values representing landfill gas before oxidation. These values ranged from -54.8 to -44.1 ‰ which are higher than those found in the literature (-50 to -60 ‰; Chanton et al., 1999).

Table 4.3: Summary of stable isotope analyses and estimated fraction of CH<sub>4</sub> oxidised by closed system ( $f_{\text{oxc}}$ ) and open system ( $f_{\text{oxo}}$ )

lysimeter and depth	material	$\delta^{13}\text{C}$ [‰] 21.10.		$\delta^{13}\text{C}$ [‰] 09.12.		$f_{\text{oxc}}$ [%]	$f_{\text{oxo}}$ [%]	$f_{\text{oxc}}$ [%]	$f_{\text{oxo}}$ [%]	$\alpha_{\text{ox}}^{\text{a}}$ (corr. f. temp) <sup>c</sup>	
[cm]		CH <sub>4</sub>	CO <sub>2</sub>	CH <sub>4</sub>	CO <sub>2</sub>	21.10.		09.12.		21.10	09.12
<b>A</b>											
25	SSC		-24.1		-24.5						
50	SSC		-25.0		-24.1						
75	SSC		-24.1		-24.9						
110	GDL	-35.2	-24.4		-24.1	58.3	81.6			1.024	
160	waste	-50.4	-12.5	-52.3	-13.8	17.0	17.2	0.7	0.7	1.026	1.030
210	waste	-54.8	-13.5	-52.5	-13.5	0	0	0	0	1.026	1.030
<b>B</b>											
25	sand		-20.2		-21.4						
50	SSC-Mix		-21.3		-21.8						
75	SSC-Mix		-20.4		-21.8						
110	GDL		-21.9		-21.8						
160	waste	-51.3	-13.6	-45.1	-14.6	1.2	1.1	3.2	3.0	1.026	1.030
210	waste	-51.6	-11.3	-46.0	-10.8	0	0	0	0	1.026	1.030
<b>C</b>											
25	TS-Fischa	-28.4	-21.1		-18.7	59.7	84.5			1.030	
50	TS-Fischa	-27.4	-20.4		-16.6	61.9	89.9			1.030	
75	SS-Fischa	-40.8	-15.2	-42.3	-14.3	38.9	45.6	19.3	19.8	1.029	1.032
110	GDL	-49.2	-10.4	-45.7	-9.9	16.6	16.7	9.6	9.3	1.029	1.032
160	waste	-51.8	-11.2	-45.1	-11.9	8.5	8.2	11.7	11.5	1.028	1.031
210	waste	-54.1	-10.2	-48.7	-11.1	0	0	0	0	1.028	1.031

lysimeter and depth	material	$\delta^{13}\text{C}$ [‰]		$\delta^{13}\text{C}$ [‰]		$f_{\text{oxc}}$	$f_{\text{oxo}}$	$f_{\text{oxc}}$	$f_{\text{oxo}}$	$\alpha_{\text{ox}}^{\text{a}}$	
		21.10.		09.12.		[%]	[%]	[%]	[%]	(corr. f. temp) <sup>c</sup>	
[cm]		CH <sub>4</sub>	CO <sub>2</sub>	CH <sub>4</sub>	CO <sub>2</sub>	21.10.		09.12.		21.10	09.12
D											
25	TS-Fischa	-22.4	-23.7	-26.2	-30.4	54.7	74.4	45.3	56.4	1.029	1.033
50	SS-Fischa	-26.3	-22.2	-31.5	-30.4	48.6	62.4	35.4	40.7	1.029	1.033
60	GDL	-37.8	-13.4	-43.3	-13.2	21.1	22.1	4.8	4.6	1.029	1.033
110	waste	-42.7	-11.0	-43.2	-9.2	5.5	5.3	5.5	5.2	1.027	1.031
160	waste	-44.1	-12.7	-44.8	-8.6	0	0	0	0	1.027	1.030

<sup>a</sup> the average  $\alpha_{\text{ox}}$  (1.027) was adopted from literature values (Table 3.8) and corrected for temperature (see eq. 3.9 in section 3.3.6)

The CH<sub>4</sub>- $\delta^{13}\text{C}$  values usually increased at more shallow depths indicating a  $^{13}\text{C}$  enrichment whereas CO<sub>2</sub>- $\delta^{13}\text{C}$  values decreased. The  $^{12}\text{C}$  enrichment of CO<sub>2</sub> and the corresponding  $^{12}\text{C}$  depletion of CH<sub>4</sub> at more shallow depths is strong evidence that methane oxidation is microbially mediated (Figure 4.20).

On 09.12.2009 gas samples for isotope analysis were also taken during surface flux measurement. However, the isotope ratios obtained during the chamber measurements are not comparable to the isotope composition measured in the gas profiles of the lysimeters due to the different volume injection for the GC. While injection volumes of gas sampled at the different depths of the lysimeters ranged between 3 and 30  $\mu\text{l}$ , a large sample volume (5,000  $\mu\text{l}$ ) of gas sampled during chamber measurement was injected in order to obtain appropriate  $\delta^{13}\text{C}$  values. Therefore, the  $\delta^{13}\text{C}$  values of CH<sub>4</sub> and CO<sub>2</sub> obtained in the closed chamber were rejected.

The individual stable isotope analyses are addressed in the following.

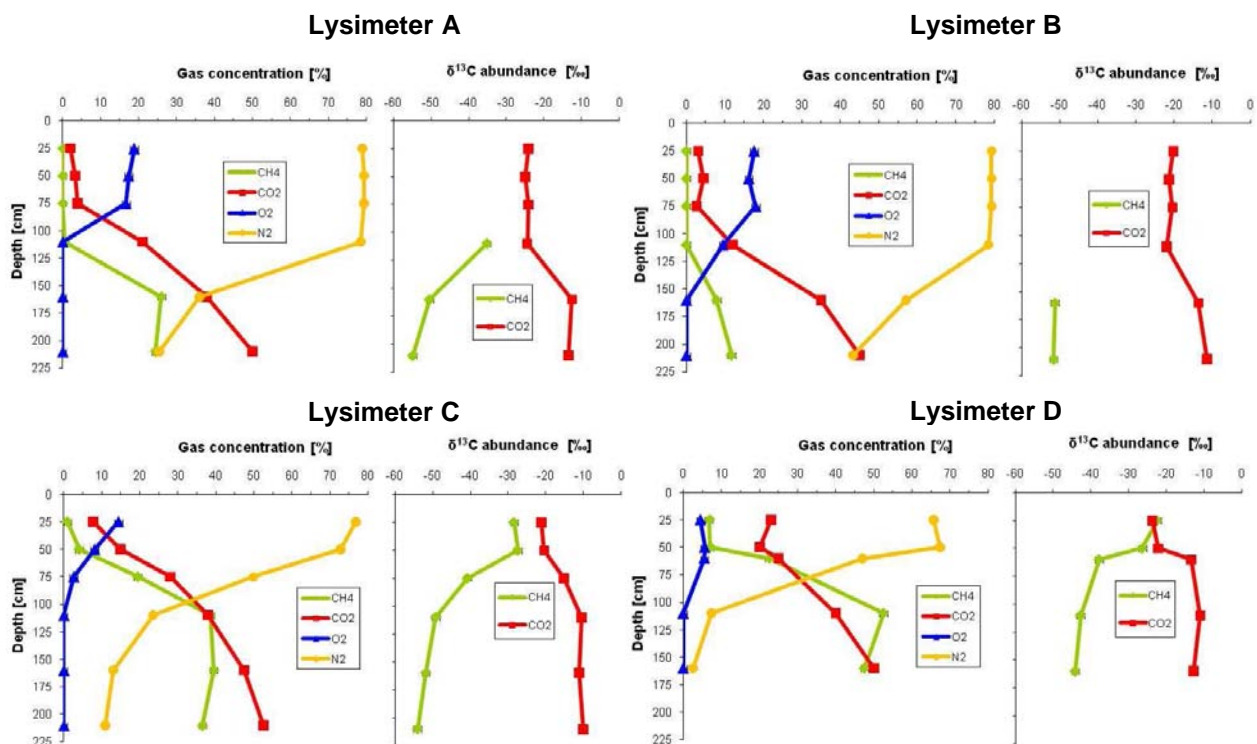


Figure 4.20: Gas concentration profiles and stable isotope results for all lysimeters on 21.10.2009

### Lysimeter A

As can be seen in Table 4.3, the October analyses (21.10.2009) revealed that methane in lysimeter A had already been oxidised by an average of 58 % (closed system interpretation, eq. 3.7) or 82 % (open system interpretation, eq. 3.8) before reaching the substrate layer. Between 25 cm and 75 cm no methane could be detected and  $\text{CO}_2\text{-}\delta^{13}\text{C}$  remained at approx. -24 ‰ indicating no further  $^{12}\text{C}$  enrichment (see Figure 4.20). This supports the assumption that  $\text{CH}_4$  oxidation was going on deeper in the gas distribution layer and in the waste itself (see section 4.1).

In contrast, on 09.12.2009 no indication for  $\text{CH}_4$  oxidation in the deeper layers has been found. However, there was also no methane detectable in the substrate layer and  $\text{CO}_2\text{-}\delta^{13}\text{C}$  value decreased slightly from 160 cm to 110 cm indicating a possible  $\text{CH}_4$  oxidation within the gas distribution layer. Although the  $\text{CH}_4$  concentration decreased slightly in the waste layers during both sampling dates, the very low fraction of oxidised methane may have resulted from a measurement error.

### Lysimeter B

The LFG production in lysimeter B was probably too low in order to see any evidence for microbial  $\text{CH}_4$  via stable isotope analysis. Only gas samples of the waste layer contained methane. The  $\delta^{13}\text{C}$  of  $\text{CH}_4$  ranged from -51.6 to 45.1 ‰ between both sampling dates. A slight decrease of  $\text{CO}_2\text{-}\delta^{13}\text{C}$  value from 160 cm to 110 cm indicate that  $\text{CH}_4$  oxidation was possibly occurring within the gas distribution layer and the waste body itself.

### Lysimeter C

Based on the stable isotope analyses conducted in October 2009, the oxidation zone in lysimeter C seems to be located between 50 cm and 25 cm from the surface. In contrast, gas composition and temperature profiles (see section 4.1) indicate that the main part of the oxidation process took place in a depth between 75 cm and 50 cm from the surface. The  $f_{\text{ox}}$  values obtained at 25 cm and 50 cm ranged between an average of 61 % (closed system interpretation) or 87 % (open system interpretation). These estimated fractions of oxidised  $\text{CH}_4$  correspond well with the results obtained by column experiment under laboratory conditions (see Figure 3.8). Oxidation activity was already detectable in the gas distribution layer, where  $f_{\text{oxc,o}}=16.6$  %. As can be seen in Figure 4.20, the  $\text{CH}_4$  concentration decreased steadily throughout the substrate layer, with a quite low concentration of  $\text{CH}_4$  (1 %) at the uppermost sampling point (25 cm).

The analysis conducted in December 2009 revealed different results. Although the  $\text{CH}_4$  concentration in the solid waste layer increased from 40 vol.% to 49 vol.% between October and December 2009, no methane was measured in the upper part of the substrate layer (25 – 50 cm). As a consequence, oxidation was only detectable at the base of the 1 m thick soil cover (at 75 cm), where  $f_{\text{oxc,o}}=19$  %. It is assumed that the high moisture content in the lower layer may have blocked gas transport causing preferential flows along the edges of the lysimeter. In addition, the temperature within the profile of lysimeter C was relatively low (< 10°C) which might have reduced  $\text{CH}_4$  oxidation.

### Lysimeter D

On 21.10.2009 the most enrichment in  $\text{CH}_4\text{-}\delta^{13}\text{C}$  (-44.1 to 22.4 ‰) was observed at 25 cm depth indicating that the oxidation zone is located between 50 cm and 25 cm from the surface. Compared to lysimeter C, the oxidation efficiency was slightly lower

and reached 55 % ( $f_{oxc}$ ) and 74 % ( $f_{oxo}$ ) at 25 cm. This is also in the range of the potential  $CH_4$  degradation capacity measured in column experiments under laboratory conditions (see Figure 3.8). However, the concentrations of  $CH_4$  near the surface were still not negligible (approx. 7 vol.%). On 09.12.2009, stable isotope analysis revealed slightly lower  $f_{ox}$  values ( $f_{oxc}=45\%$  and  $f_{oxo}=56\%$ ) most probably due to the lower temperatures within the soil profile ( $<10\text{ }^{\circ}\text{C}$ ) and lower  $CH_4$  concentration in the solid waste layer compared to the October analysis.

#### Considerations concerning $\alpha_{trans}$

The fraction of methane oxidised in an open system ( $f_{oxo}$ ) is calculated with the transport fractionation factor ( $\alpha_{trans}$ ) according to Eq. 3.8 (see section 3.3.6). In general,  $\alpha_{trans}$  is difficult to assess as it depends on the relative importance of diffusion relative to advection in the transport of  $CH_4$ . In the present case, the transport fractionation factor was set to be 1 for purely advective systems. Based on the potential of gas formation ( $GS_{21}$ ) of the waste bodies (potential  $CH_4$  load =  $130\text{ l/m}^2\text{h}$ ), it was determined that gas transport is dominated by advection with the diffusive flux representing 5 – 15 % of this potential methane load. The diffusive flux was determined by using Fick's first law:

$$J = -D \frac{dC}{dz} \quad (4.1)$$

J (mol/m<sup>2</sup>s) diffusion flux

D (m<sup>2</sup>/s) diffusion coefficient

dC/dz (mol/m<sup>4</sup>) concentration gradient

The calculated diffusive flux is based on a typical concentration gradient between the waste layer (160 cm) and the top of the substrate layer (25 cm) of lysimeters C and D ( $dC/dz = 15 - 22\text{ mol/m}^4$ ), the diffusion coefficient of methane in air ( $0.16\text{ cm}^2/\text{s}$ ) and a substrate porosity of 20 %. However, the assumption of  $\alpha_{trans}=1$  may have lead to an underestimation of the oxidation efficiency of the lysimeters (assuming an open system,  $f_{oxo}$ ) since the calculations are based on potential methane loading (LFG loading data were not available).

#### Sensitivity Analysis

Because the isotopic fraction factor for microbial oxidation ( $\alpha_{ox}$ ) was calculated based on previous laboratory and field studies (Table 3.8), a sensitivity analysis of  $f_{oxc}$  (closed system) and  $f_{oxo}$  (open system) (presented in Table 4.3) to changes in  $\alpha_{ox} \pm$  standard deviation ( $\sigma$ ) has been carried out. The data given in Table 4.4 show that  $f_{oxc}$  decreases by an average of 19 % when adopting  $\alpha_{ox}+\sigma$  while it increases by an average of 31 % with  $\alpha_{ox}-\sigma$ . The sensitivity analysis for  $f_{oxo}$  showed slightly greater variations. When  $\alpha_{ox}+\sigma$  is adopted,  $f_{oxo}$  decreased by 23 % and increased by 36 % with  $\alpha_{ox}-\sigma$ . In the case of samples of the shallow subsurface of lysimeters C and D as well as the gas distribution layer of lysimeter A, the adoption of  $\alpha_{ox}-\sigma$  resulted in oxidation efficiencies higher than 100 %, suggesting that the  $\alpha_{ox}-\sigma$  value (1.019) is too low. In general, the closed system interpretation ( $f_{ox}$ ) was almost always less than the open system interpretation ( $f_{oxo}$ ) with the highest deviation of 45 % (see Table 4.3 and Table 4.4). It can be concluded that the closed system approach yields a lower limit appraisal of methane oxidation (Chanton et al., 2009).

Although  $\alpha_{ox} \pm \sigma$  represents a slight variation in the adopted  $\alpha_{ox}$  (0.8 %), it has a significant influence on the oxidation efficiency of the different biocovers. This result suggests a certain degree of uncertainty using isotope analyses to quantify microbial

CH<sub>4</sub> oxidation of landfill covers. Therefore, oxidation efficiency analyses should be interpreted with caution. Another source of concern is the natural variability in  $\alpha_{ox}$ . In the literature the isotopic fractionation factor varies considerably (Templeton et al., 2006; Chanton and Liptay, 2000; Chanton et al., 1999) depending on soil temperature and other environmental factors. Consequently,  $\alpha_{ox}$  should be determined at each landfill site and within each soil type at a site to minimise this source of uncertainty (Chanton et al., 2008b).

Table 4.4: Results of sensitivity analysis of  $f_{oxc}$  (closed system) and  $f_{oxo}$  (open system) to changes in  $\alpha_{ox}$  for the sampling date 21.10.2009

lysimeter and depth [cm]	closed system					open system				
	mean $f_{oxc}$ [%]	$f_{oxc}$ [%] (with $\alpha_{ox}+\sigma$ )	% difference in relation to $f_{oxc}$	$f_{oxc}$ [%] (with $\alpha_{ox}-\sigma$ )	% difference in relation to $f_{oxc}$	mean $f_{oxo}$ [%]	$f_{oxo}$ [%] (with $\alpha_{ox}+\sigma$ )	% difference in relation to $f_{oxo}$	$f_{oxo}$ [%] (with $\alpha_{ox}-\sigma$ )	% difference in relation to $f_{oxo}$
lysimeter A										
110	58.3	48.2	17.4	73.2	-25.6	81.6	60.8	25.5	100 (123.9)	-22.6
160	17.0	13.3	22.0	23.8	-40.2	17.2	13	24.3	25.4	-47.3
210	0	0		0		0	0		0	
lysimeter B										
160	1.2	0.9	23.1	1.8	-43.9	1.1	0.9	23.8	1.7	-45.5
210	0	0		0		0	0		0	
lysimeter C										
25	59.7	51.4	13.9	70.9	-18.8	84.5	66.5	21.3	100 (115.8)	-18.4
50	61.9	53.3	13.9	73.4	-18.5	89.9	70.4	21.7	100 (124.2)	-11.3
75	38.9	32.1	17.5	49.4	-27.0	45.6	35.6	22.0	63.5	-39.3
110	16.6	13.3	19.9	22.1	-33.7	16.7	13.0	22.0	23.3	-39.3
160	8.5	6.7	21.6	11.8	-38.8	8.2	6.3	23.0	11.6	-42.4
210	0	0		0		0	0		0	
lysimeter D										
25	54.7	46.4	15.2	66.5	-21.5	74.4	58.1	22.0	100 (103.5)	-34.4
50	48.6	40.6	16.5	60.4	-24.3	62.4	48.5	22.3	87.6	-40.4
60	21.1	16.9	19.8	28.1	-33.2	22.1	17.2	22.3	31.0	-40.4
110	5.5	4.3	22.5	7.8	-41.9	5.3	4.0	23.6	7.6	-44.8
160	0	0		0		0	0		0	

Average  $\alpha_{ox}=1.027$ ; ( $\alpha_{ox}+\sigma$ )=1.035; ( $\alpha_{ox}-\sigma$ )=1.019

## 5 CONCLUSIONS

A main part of the present thesis was to develop a measuring system to quantify remaining CH<sub>4</sub> and CO<sub>2</sub> surface fluxes from the different biocovers. It was decided to build an accumulation chamber based on the closed dynamic chamber technique covering almost the whole area of the lysimeter (4 m<sup>2</sup>). The challenge was to include the aeration pipe (D = 6 cm) of the lysimeter into the design of the accumulation chamber without disturbing the measurement procedure. In contrast, chambers with a basal sampling area of < 1 m<sup>2</sup> are the most common tools. In order to verify the performance and accuracy of the measuring system, laboratory tests were conducted under different known CH<sub>4</sub> and CO<sub>2</sub> fluxes. Following the laboratory validation phase, it can be concluded that reliable CH<sub>4</sub> and CO<sub>2</sub> emission rates can be obtained in the range of 0.01 g CH<sub>4</sub>/m<sup>2</sup>d (lower detection limit) to 35 g CH<sub>4</sub>/m<sup>2</sup>d and 1 – 47 g CO<sub>2</sub>/m<sup>2</sup>d, respectively. However, absolute emission data should be regarded with caution due to inhibition of fluxes through concentration and pressure build-up in closed chambers. In the present case, the measurement of surface fluxes both under open and closed vent tubes revealed high sensitivity to small pressure gradients between the chamber headspace and the external environment. In addition, altering emission rates have been observed in response to wind events. Therefore, measurement campaigns were done during periods with wind speed < 4 m/s.

Among the four biocovers studied, CH<sub>4</sub> emission fluxes could only be detected from the biocovers with mineral soil covers (lysimeters C and D), whereas no CH<sub>4</sub> emissions were measured on the biocovers with mature sewage sludge compost and sand/compost mixture, respectively (lysimeters A and B). It should be noted that methane production developed differently in the solid waste layers and methane load of lysimeters A and B did not indicate the same range as in lysimeters C and D. According to stable isotope analyses and gas composition profiles of lysimeters A and B, most of the methane oxidation processes were going on below the profile in the gas distribution layer and in the waste. There was no methane detectable in the substrate layer as well as on the surface of lysimeters A and B. However, the mature sewage sludge compost (SSC) placed in lysimeter A showed, in principle very promising results regarding the optimal ambient conditions for methanotrophic bacteria. Beside the nutritional factors, the physical properties such as high porosity and water-holding capacity as well as low thermal conductivity of the compost material provided an adequate methane oxidation milieu. High nitrogen and oxygen concentrations suggest that the substrate cover was well aerated. Moreover, the SSC-cover was capable of retaining the moisture content and the temperature profiles at the optimum level as suggested by other research studies. Compared to the other substrates investigated in this study, sewage sludge compost provided also better conditions to foster the growth of plants, especially during spring.

Following the surface flux measurements, methane emissions from lysimeter D (0.9 – 8.2 g/m<sup>2</sup>d) were significantly greater than fluxes from lysimeter C (0 – 1.1 g/m<sup>2</sup>d), although the methane load was probably lower (as indicated by lower CH<sub>4</sub> concentrations in the solid waste layer). This result indicates that low-dimensioned biocovers do not achieve as high CH<sub>4</sub> oxidation capacities as properly designed biocovers. However, a relative comparison of the emission data is limited due to the varying methane development in the solid waste layers of the lysimeters. In addition, results obtained by the surface flux measurements should be interpreted with caution, not only for CH<sub>4</sub> but also CO<sub>2</sub>. From FID screenings it is known that higher



surface CH<sub>4</sub> concentrations are seen along the edges of the lysimeters C and D. Although the closed dynamic chamber had an attached plastic skirt for sealing purposes, a great portion of methane and carbon dioxide fluxes may have bypassed leading to an underestimation of CH<sub>4</sub> and CO<sub>2</sub> emissions. In general, it is assumed that the chamber area of 3.61 m<sup>2</sup> allowed small point sources, such as cracks and fissures to be addressed more reliably.

It is hypothesised that the high moisture content in the mineral soil layers may have blocked gas transport causing preferential flows along the edges of the lysimeters. In contrast, gas permeability tests conducted at the Austrian Institute of Technology in Seibersdorf revealed that all substrates used in this study should ensure sufficient air permeability even at 100 % WHC. However, the preferential flows may also be an artefact of the lysimeter facility itself. In general, loamy soils tend to crack formation, which may have additionally supported the preferential channels.

The study revealed that the magnitude of methane flux into the cover material is a main controlling factor on methane oxidation. When the landfill gas composition in the solid waste layer of lysimeter C was similar to those in lysimeter A, no CH<sub>4</sub> emissions could be measured. For design purposes, it is not only useful to quantify the maximum methane oxidation capacity of a cover system under specific climatic conditions but also to know the flux at which the cover can oxidise 100 % of the methane coming from below (Abichou et al., 2010).

Since already a small vegetation height had an influence on CO<sub>2</sub> emissions measurements, in future the chamber height should be adaptable on vegetation's condition. In order to compensate the resulting increased chamber volume, four fans could be spaced uniformly near the bottom of the enclosed chamber, with air flow directed diagonally upward. In addition, the chamber could be equipped with both a transparent and an opaque cover to investigate the influence of vegetation on CO<sub>2</sub> emission measurement. The use of a combined dark-light measurement enables an estimation of the CO<sub>2</sub> assimilation of the vegetation enclosed in the chamber. Moreover, a background measurement and alignment of relative humidity change on CO<sub>2</sub> concentration in the closed dynamic chamber should be done in order to crosscheck the disturbance.

Stable isotope analyses performed at the four lysimeters indicate that almost all biocover systems were able to reduce CH<sub>4</sub> emissions. The LFG production in lysimeter B was probably too low in order to see an evidence for microbial methane oxidation via stable isotope analysis. The estimated fractions of oxidised CH<sub>4</sub> correspond well with the results obtained by column experiment under laboratory conditions. A considerable percentage of CH<sub>4</sub> was oxidised in the mineral soil covers placed in lysimeters C and D. However, the percentage that is emitted in a more homogeneous way determines methane oxidation (Oonk, 2010). Since a significant portion of the methane load was most probably emitted through preferential flows along the edges of the lysimeters, the oxidation based on stable isotope analyses may be overestimated. Due to the probably low methane load in lysimeter A, methane has already been reduced at a great portion in the gas distribution layer before reaching the SSC-cover. However, under laboratory conditions, the mature compost material mitigated loadings up to 350 l CH<sub>4</sub>/m<sup>2</sup>d indicating a CH<sub>4</sub> degradation rate of more than 95 %. Because the actual fractionation factor ( $\alpha_{ox}$ ) for microbial oxidation was not determined specifically for this study, a sensitivity analysis of  $f_{oxc}$  (closed system interpretation) and  $f_{oxo}$  (open system interpretation) to changes in  $\alpha_{ox} \pm$  standard deviation ( $\sigma$ ) has been carried. Since small differences in

the adopted  $\alpha_{ox}$  had measurable effects on the oxidation efficiency of the different biocovers, results should be interpreted with caution, however, not only in this study.

It can be concluded that a mature, well-structured compost material underlain by a coarse gas distribution layer represent an attractive alternative of the reduction of methane coming out from landfills, particularly for mechanically and biologically pre-treated (MBT) waste or old and small landfills with low  $CH_4$  production potential.

The next challenge will be to explore the efficiencies of the different biocovers regarding water infiltration and methane mitigation in combination with in-situ aeration.

Future studies should also focus on the complex interaction of methanotrophic bacteria with plants species obtaining aerenchyma in anoxic soils (e.g. Typhaceae) in order to investigate their relevance for  $CH_4$  oxidation.

## 6 INDEXES

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## 6.4 Formula Symbols, Indices, Abbreviations

### 6.4.1 Glossary

Some letters were multiple allocated due to the various literatures.

A	m <sup>2</sup>	Surface area covered by the chamber
a	μbar	Pressure wave amplitude
b	μl/cycle	Displaced volume
D	mm	Diameter
D	m <sup>2</sup> /s	Diffusion coefficient
dC	ppm, mg/m <sup>3</sup>	Change in gas concentration
dt	min, sec	time
dz	cm	Change in sampling depth
F	g/m <sup>2</sup> d	Gas emission flux
f	Hz	Frequency
f	%	Oxidised fraction
g	m/s <sup>2</sup>	gravity
J	mol/m <sup>2</sup> s	Diffusive flux
L	mm	Tube length
p	μbar	Barometric pressure
Q	m <sup>3</sup> /h, l/s	Volume flow rate
r	-	Volume ratio
T	°C, K	Temperature
V	m <sup>3</sup>	Head space, chamber volume
v	μl/s	Volumetric air flow rate

### 6.4.2 Greek Symbols

Δ	-	Delta, difference
μ	Pa s	Air viscosity
α	-	Isotopic fractionation factor
δ	-	Isotope ratio
σ	-	Standard deviation

### 6.4.3 Indices

anox	anoxic
c	Closed system approach
int	intrinsic
m	mean
o	Open system approach
ox	oxidised
trans	transport
z	Sampling deth

#### 6.4.4 Abbreviations

<i>a</i> .....	<i>annual</i>
<i>ALSAG</i> .....	<i>Altlastensanierungsgesetz</i>
<i>AIT</i> .....	<i>Austrian Institute of Technology</i>
<i>BGBI</i> .....	<i>Bundesgesetzblatt</i>
<i>BOD</i> .....	<i>biological oxygen demand</i>
<i>BWC</i> .....	<i>biowase compost</i>
<i>C</i> .....	<i>carbon</i>
<i>CDC</i> .....	<i>closed dynamic chamber</i>
<i>CH<sub>4</sub></i> .....	<i>methane</i>
<i>C:N</i> .....	<i>carbon/nitrogen ratio</i>
<i>CO<sub>2</sub></i> .....	<i>carbon dioxide</i>
<i>COD</i> .....	<i>chemical oxygen demand</i>
<i>d</i> .....	<i>day</i>
<i>DFT</i> .....	<i>dynamic flux tunnel</i>
<i>DM</i> .....	<i>dry matter</i>
<i>e.g.</i> .....	<i>exempli gratia (= for example)</i>
<i>EPS</i> .....	<i>Exopolimeric substances</i>
<i>FID</i> .....	<i>Flame-Ionisation-Detector</i>
<i>GC</i> .....	<i>gas chromatography</i>
<i>GES</i> .....	<i>gas extraction system</i>
<i>GHG</i> .....	<i>greenhouse gas</i>
<i>h</i> .....	<i>hour</i>
<i>H<sub>2</sub></i> .....	<i>hydrogen</i>
<i>INERSIS</i> .....	<i>Institut National de l'Environnement et des Risques</i>
<i>IPCC</i> .....	<i>Intergovernmental Panel on Climate Change</i>
<i>kCal</i> .....	<i>kilocalorie</i>
<i>Kg</i> .....	<i>kilogram</i>
<i>l</i> .....	<i>litre</i>
<i>LFG</i> .....	<i>landfill gas</i>
<i>LOI</i> .....	<i>loss on ignition</i>
<i>n.d.</i> .....	<i>not detectable</i>
<i>m</i> .....	<i>meter</i>
<i>MBT</i> .....	<i>mechanical-biological pretreatment</i>
<i>MMO</i> .....	<i>Methane Mono-oxygenase</i>
<i>n</i> .....	<i>number of samples</i>
<i>N<sub>2</sub></i> .....	<i>nitrogen</i>
<i>O<sub>2</sub></i> .....	<i>oxygen</i>
<i>ppm</i> .....	<i>parts per million</i>
<i>pMMO</i> .....	<i>particulate Methane Mono-oxygenase</i>



<i>RA</i> .....	<i>respiration activity</i>
<i>SCT</i> .....	<i>static dynamic chamber</i>
<i>SD</i> .....	<i>standard deviation</i>
<i>sMMO</i> .....	<i>soluble Methane Mono-oxygenase</i>
<i>SS</i> .....	<i>subsoil</i>
<i>SSC</i> .....	<i>Sewage sludge compost</i>
<i>Tg</i> .....	<i>Tera gram (=10<sup>12</sup> g)</i>
<i>TOC</i> .....	<i>total organic carbon</i>
<i>TS</i> .....	<i>topsoil</i>
<i>VOC</i> .....	<i>volatile organic compounds</i>
<i>v/v</i> .....	<i>volume per volume</i>
<i>w/w</i> .....	<i>weight per weight</i>
<i>WHC</i> .....	<i>water holding capacity</i>

# APPENDIX

## A.1 Gas composition and temperature profiles

02.04.2008						
Mittelwert	CH4	CO2	O2	Mittelwert	Temperature	
[cm]	%	%	%	[cm]	°C	
Kammer A						
25	0,1	4,8	16,3	25	0,00	
50	0	9,2	13,2	50	0,00	
75	0	14	9,1	75	0,00	
110	0	0	0	190	0,00	
160	1,5	65				
210	2,1	82,5	0			
Kammer B						
25	0	6,8	16,7	25	0,00	
50	0,1	10	14	50	0,00	
75	0	17	9,9	75	0,00	
110	0,1	30	3,8	190	0,00	
160	1,4	70	0			
210	1,8	85	0,05			
Kammer C						
25	0	6,5	16,7	25	0,00	
50	0	12	12,6	50	0,00	
75	0	17,5	9,4	75	0,00	
110	0,1	31	2	190		
160	0,3	60	0			
210	0,4	75	0			
Kammer D						
25	0	9,8	14,3	25	0,00	
50	0	15	11,2	50	0,00	
60	0,2	38	3	140	0,00	
110	0,55	62,5	0	190	0,00	
160	0,65	75	0			

09.09.2009						
Mittelwert	CH4	CO2	O2	Mittelwert	Temperature	
[cm]	%	%		[cm]	°C	
Kammer A						
25	0	2,1	19,2	25	20,42	
50	0	2,8	18,4	50	23,72	
75	0	4,6	16,7	75	26,49	
110	0	19	4,2	190	22,62	
160	19,5	50	0			
210	16,25	60	0			
Kammer B						
25	0	4,2	16,7	25	20,62	
50	0	5,8	15,2	50	21,20	
75	0	16	19,2	75	22,48	
110	0	16	5,6	190	22,20	
160	4,2	42,5	0			
210	6,75	55	0			
Kammer C						
25	0	4,8	17,1	25	18,89	
50	0	14	11	50	19,95	
75	12	37	3,9	75	20,37	
110	32,5	55	0	190		
160	27,75	60	0			
210	26	65	0			
Kammer D						
25	0	16	7	25	22,30	
50	0	15	7,4	50	23,38	
60	22	39	0	140	22,77	
110	42	50	0	190	21,43	
160	36,75	55	0			

01.10.2009						
Mittelwert [cm]	CH4 %	CO2 %	O2 %	Mittelwert [cm]	Temperature °C	
Kammer A						
25	0	2,6	18,5	25	21,4	
50	0	2,9	18	50	24,3	
75	0	4,7	16,2	75	27,1	
110	0,1	25	0	190	26,6	
160	21,75	38,5	1,6			
210	18	45	1,8			
Kammer B						
25	0	2,7	17,8	25	23,2	
50	0	3,9	16,6	50	21,1	
75	0	1,6	19,3	75	22,4	
110	0	11	9,7	190	23,1	
160	4,55	31	2			
210	6,7	39	1,95			
Kammer C						
25	0	4,3	16,8	25	18,7	
50	0	11	11	50	19,6	
75	7,6	20	5,6	75	20,2	
110	29,5	36	2	190	29,3	
160	28,25	45	2,1			
210	28,25	52,5	1,05			
Kammer D						
25	was irrigated					
50						
60						
110						
160						

27.08.2009						
Mittelwert	CH4	CO2	O2	Mittelwert	Temperature	
[cm]	%	%	%	[cm]	°C	
Kammer A						
25	0	2	19,1	25	23,96	
50	0	2,6	18,4	50	25,26	
75	0	4,3	16,7	75	27,23	
110	0	17	5,2	190	26,60	
160	16	45	0			
210	11,75	55	0			
Kammer B						
25	0	3,8	17	25		
50	0	5,9	15,2	50	23,51	
75	0	1,8	19,1	75	23,86	
110	0	15	7,3	190	22,78	
160	4,55	45	0			
210	4,15	55	0			
Kammer C						
25	0	5,4	16,8	25	22,08	
50	0	14	10,9	50	22,51	
75	11	39	3,1	75	22,37	
110	28	55	0	190		
160	23,75	62,5	0			
210	22	65	0			
Kammer D						
25	0	15	7,9	25	24,33	
50	0,2	15	8,1	50	24,66	
60	22	40	0	140	23,28	
110	36,25	55	0	190	21,80	
160	31,75	55	0			

18.09.2009						
Mittelwert	CH4	CO2	O2	Mittelwert	Temperatur	
[cm]	%	%	%	[cm]	°C	
Kammer A						
25	0	2,3	18,7	25	22,04	
50	0	2,1	18,8	50	24,55	
75	0	3,6	17,2	75	26,30	
110	0	16	5,3	190	23,29	
160	18	37	2,6			
210	14	45	2,95			
Kammer B						
25	0	2,7	17,6	25	21,56	
50	0	3,6	16,8	50	21,90	
75	0	1,6	19,1	75	22,63	
110	0	11	9,8	190	23,18	
160	5,1	38	0			
210	7,05	50	0			
Kammer C						
25	0	5,9	15,7	25	20,03	
50	0	13	9,3	50	20,53	
75	8,9	27	4,1	75	20,79	
110	29,5	40	1,8	190		
160	24	45	3,7			
210	22	45	4,25			
Kammer D						
25	0	11	9,1	25	24,26	
50	0	12	8,7	50	24,71	
60	31	37	0	140	22,26	
110	45,75	45	0	190	21,08	
160	44,25	47,5	0			

02.09.2009						
Mittelwert	CH4	CO2	O2	Mittelwert	Temperature	
[cm]	%	%	%	[cm]	°C	
Kammer A						
25	0	2,2	18,9	25	22,70	
50	0	2,8	18,2	50	24,77	
75	0	4,4	16,6	75	27,05	
110	0	19	4,3	190	24,34	
160	17,75	50	0			
210	13,75	55	0			
Kammer B						
25	0	4,1	16,8	25	23,76	
50	0	5,8	15,2	50	22,65	
75	0	1,5	19,3	75	23,45	
110	0	16	6,1	190	22,45	
160	4,4	45	0			
210	5,6	55	0			
Kammer C						
25	0	4,3	18	25	20,75	
50	0	11	13,4	50	21,43	
75	8	32	5,5	75	21,60	
110	29,5	55	0	190		
160	25	60	0			
210	23	65	0			
Kammer D						
25	0	15	7,5	25	23,63	
50	0,2	15	7,7	50	24,34	
60	21	40	0	140	23,12	
110	37,5	50	0	190	21,67	
160	32,5	55	0			



09.02.2010							
Mittelwert [cm]	CH4 %	CO2 %	O2 %	N2 %	Mittelwert [cm]	Temperature °C	
<b>Kammer A</b>							
25	0	0,2	20,8	79	25	0,78	
50	0	0,3	20,6	79,1	50	3,26	
75	0	0,2	20,7	79,1	75	5,24	
110	0	4,8	17,8	77,4	190	6,72	
160	9,35	25	0	65,65			
210	14,25	30,5	0	55,25			
<b>Kammer B</b>							
25	0	4,3	15,5	80,2	25	1,06	
50	0	7,4	12,3	80,3	50	1,73	
75	0	4,5	15,1	80,4	75	2,96	
110	0	13	4,5	82,5	190	7,13	
160	0,6	21	0	78,4			
210	2,4	28	0	69,6			
<b>Kammer C</b>							
25	1,5	2,2	18,5	77,8	25	0,81	
50	2,6	4,9	17	75,5	50	1,66	
75	13	16	10,7	60,3	75	2,19	
110	35	35	0	30	190	4,19	
160	41,75	39	0	19,25			
210	42,5	40	0	17,5			
<b>Kammer D</b>							
25	0,1	2,6	17,6	79,7	25	0,81	
50	0	2,4	18,5	79,1	50	1,60	
60	1,5	6,3	12,9	79,3	140	4,84	
110	20,25	21	0	58,75	190	6,15	
160	22,25	22	0	55,75			

18.03.2010							
Mittelwert [cm]	CH4 %	CO2 %	O2 %	N2 %	Mittelwert [cm]	Temperature °C	
<b>Kammer A</b>							
25	0	0,9	19,1	80	25	5,39	
50	0	1,2	18,9	79,9	50	5,34	
75	0	2,2	18,3	79,5	75	5,92	
110	0	12	9,5	78,5	190	6,31	
160	11,25	28	0	60,75			
210	16,25	34	0	49,75			
<b>Kammer B</b>							
25	0	2,3	18,2	79,5	25	5,08	
50	0	4,2	16,2	79,6	50	4,90	
75	0	1,2	19	79,8	75	4,94	
110	0	14	7,1	78,9	190	6,37	
160	1,25	24,5	0	74,25			
210	3,65	31	0	65,35			
<b>Kammer C</b>							
25	0,6	1,8	18,8	78,8	25	4,14	
50	1,1	3,1	18,2	77,6	50	3,99	
75	12,5	12	13,7	61,8	75	3,90	
110	43	40	0	17	190	3,78	
160	48,5	45	0	6,5			
210	48	45	0	7			
<b>Kammer D</b>							
25	0	8,2	15,8	76	25	5,46	
50	0	7,5	16	76,5	50	5,38	
60	21	24	0	55	140	5,48	
110	31,75	27,5	0	40,75	190	6,15	
160	38,5	28,5	0	33			

01.04.2010							
Mittelwert [cm]	CH4 %	CO2 %	O2 %	N2 %	Mittelwert [cm]	Temperature °C	CH4/CO2
<b>Kammer A</b>							
25	0	0,5	19,8	79,7	25	10,61	0,0
50	0	0,4	19,9	79,7	50	10,44	0,0
75	0	0,7	19,1	80,2	75	9,88	0,0
110	0	11	7,2	81,8	190	7,99	0,0
160	12,25	30	0	57,75			0,4
210	16,5	35	0	48,5			0,5
<b>Kammer B</b>							
25	0	2,7	17,9	79,4	25	10,24	0,0
50	0	4	16,6	79,4	50	10,36	0,0
75	0	1,9	18,8	79,3	75	10,03	0,0
110	0	15	7	78	190	7,41	0,0
160	1,15	25	0	73,85			0,0
210	3,65	30	0	66,35			0,1
<b>Kammer C</b>							
25	0,3	2,5	18,1	79,1	25	9,35	0,1
50	2	4,3	16,2	77,5	50	9,22	0,5
75	22	22	7,6	48,4	75	8,83	1,0
110	45,5	45	0	9,5	190	4,86	1,0
160	48,75	45	0	6,25			1,1
210	48	45	0	7			1,1
<b>Kammer D</b>							
25	0,1	7,9	9,4	82,6	25	11,32	0,0
50	0,1	7,6	11,5	80,8	50	11,78	0,0
60	17,5	20	3,7	58,8	140	8,05	0,9
110	35	31	0	34	190	7,57	1,1
160	38,5	31,5	0	30			1,2

24.02.2010							
Mittelwert [cm]	CH4 %	CO2 %	O2 %	N2 %	Mittelwert [cm]	Temperature °C	
<b>Kammer A</b>							
25	0	1,8	17,8	80,4	25	0,70	
50	0	2,1	17,2	80,7	50	2,68	
75	0	2,3	17,2	80,5	75	4,28	
110	0	8,8	8,7	82,5	190	5,69	
160	7,3	23,5	0	69,2			
210	11,75	29	0	59,25			
<b>Kammer B</b>							
25	0	2,5	17,3	80,2	25	1,56	
50	0	6,9	11,7	81,4	50	1,88	
75	0	1,5	18,7	79,8	75	2,83	
110	0	14	3,4	82,6	190	6,26	
160	0,35	20	0	79,65			
210	1,7	26	0	72,3			
<b>Kammer C</b>							
25	0,4	2	19	78,6	25	0,78	
50	0,7	3,5	18,2	77,6	50	1,49	
75	8,3	12	12,9	66,8	75	1,99	
110	34	33	0	33	190	3,22	
160	38,25	38	0	23,75			
210	39	40	0	21			
<b>Kammer D</b>							
25	0,3	4	14,1	81,6	25	1,04	
50	0	3,5	16,8	79,7	50	1,61	
60	13	18	1,3	67,7	140	4,25	
110	21	21,5	0	57,5	190	5,43	
160	26,5	23	0	50,5			

24.03.2010							
Mittelwert [cm]	CH4 %	CO2 %	O2 %	N2 %	Mittelwert [cm]	Temperature °C	CH4/CO2
<b>Kammer A</b>							
25	0	0,9	20,1	79	25	8,99	0,0
50	0	0,9	20	79,1	50	7,67	0,0
75	0	0,8	20,2	79	75	7,09	0,0
110	0	10	13	77	190	6,67	0,0
160	11,5	29	0	59,5			0,4
210	16,25	34	0	49,75			0,5
<b>Kammer B</b>							
25	0	2,6	18,1	79,3	25	8,60	0,0
50	0	4,4	16,4	79,2	50	7,98	0,0
75	0	2	18,7	79,3	75	7,19	0,0
110	0	14	7,2	78,8	190	6,51	0,0
160	1,2	24,5	0	74,3			0,0
210	3,65	30	0	66,35			0,1
<b>Kammer C</b>							
25	0,7	2,6	18,3	78,4	25	7,36	0,3
50	3,3	4,7	16,7	75,3	50	6,59	0,7
75	29,5	26	6	38,5	75	6,03	1,1
110	46	40	0	14	190	3,74	1,2
160	49	45	0	6			1,1
210	48	45	0	7			1,1
<b>Kammer D</b>							
25	0	6,3	15,7	78	25	9,37	0,0
50	0	3,8	18,5	77,7	50	8,89	0,0
60	16	16	8,6	59,4	140	6,05	1,0
110	32,75	28,5	0	38,75	190	6,40	1,1
160	38	30	0	32			1,3

07.04.2010							
Mittelwert [cm]	CH4 %	CO2 %	O2 %	N2 %	Mittelwert [cm]	Temperature °C	CH4/CO2
<b>Kammer A</b>							
25	0,1	1,8	19,2	78,9	25	10,47	0,1
50	0,1	2,5	18,4	79	50	11,05	0,0
75	0,1	2,4	18,7	78,8	75	11,25	0,0
110	0,1	15	7,3	77,6	190	8,74	0,0
160	12,25	31	0	56,75			0,4
210	17	35	0	48			0,5
<b>Kammer B</b>							
25	0	3,3	17,2	79,5	25	9,87	0,0
50	0	4,6	15,8	79,6	50	10,06	0,0
75	0	2,2	18,2	79,6	75	10,10	0,0
110	0	15	5,9	79,1	190	8,30	0,0
160	1,5	26	0	72,5			0,1
210	3,85	30	0	66,15			0,1
<b>Kammer C</b>							
25	1,1	1,8	18	79,1	25	9,10	0,6
50	2,2	3	16,8	78	50	9,31	0,7
75	23,5	21	7,3	48,2	75	9,10	1,1
110	44,5	40	0	15,5	190	5,93	1,1
160	50,25	45	0	4,75			1,1
210	50	45	0	5			1,1
<b>Kammer D</b>							
25	0,1	5,3	15,7	78,9	25	11,10	0,0
50	0	5,4	15,7	78,9	50	11,54	0,0
60	25,5	26	0	48,5	140	9,36	1,0
110	37,25	31,5	0	31,25	190	8,59	1,2
160	40	33	0	27			1,2

15.04.2010							
Mittelwert	CH4	CO2	O2	N2	Mittelwert	Temperature	CH4/CO2
[cm]	%	%	%	%	[cm]	°C	
<b>Kammer A</b>							
25	0,1	2,8	17,5	79,6	25	10,88	0,0
50	0	3,5	16,6	79,9	50	11,55	0,0
75	0	3,6	16,3	80,1	75	12,20	0,0
110	0,1	18	2,8	79,1	190	9,89	0,0
160	13	31,5	0	55,5			0,4
210	16,25	35,5	0	48,25			0,5
<b>Kammer B</b>							
25	0	2,7	18	79,3	25	10,09	0,0
50	0	6,8	12	81,2	50	10,14	0,0
75	0	1,9	18,4	79,7	75	10,28	0,0
110	0	18	2,6	79,4	190	9,18	0,0
160	1,55	27	0	71,45			0,1
210	3,9	31	0	65,1			0,1
<b>Kammer C</b>							
25	3,1	5,3	15,6	76	25	9,67	0,6
50	7,2	9,5	13,3	70	50	9,55	0,8
75	22	19	8,3	50,7	75	9,41	1,2
110	51	40	0	9	190	6,61	1,3
160	49,5	50	0	0,5			1,0
210	49,5	50	0	0,5			1,0
<b>Kammer D</b>							
25	0,2	8,2	8,6	83	25	10,92	0,0
50	0,5	6,2	11,9	81,4	50	11,01	0,1
60	27,5	26	0	46,5	140	10,49	1,1
110	36,5	31	0	32,5	190	9,60	1,2
160							

19.04.2010							
Mittelwert	CH4	CO2	O2	N2	Mittelwert	Temperature	CH4/CO2
[cm]	%	%	%	%	[cm]	°C	
<b>Kammer A</b>							
25	0	2,5	18	79,5	25	11,88	0,0
50	0,1	3,3	17	79,6	50	12,18	0,0
75	0	4,2	15,5	80,3	75	12,32	0,0
110	0,1	19	4,4	76,5	190	10,90	0,0
160	13,5	32,5	0	54			0,4
210	16,75	36,5	0	46,75			0,5
<b>Kammer B</b>							
25	0	2,8	17,5	79,7	25	11,54	0,0
50	0	7	13,4	79,6	50	11,06	0,0
75	0	1,8	18,8	79,4	75	10,61	0,0
110	0	19	2,3	78,7	190	9,49	0,0
160	1,95	27	0	71,05			0,1
210	4	32	0	64			0,1
<b>Kammer C</b>							
25	0,2	4,7	17,7	77,4	25	10,77	0,0
50	0,5	8,8	16,1	74,6	50	10,41	0,1
75	6,2	13	13,9	66,9	75	10,02	0,5
110	47	40	0	13	190	10,84	1,2
160	51	47,5	0	1,5			1,1
210	49	50	0	1			1,0
<b>Kammer D</b>							
25	0	8,3	14,2	77,5	25	12,30	0,0
50	0	8,6	13,2	78,2	50	12,27	0,0
60	33	27	0	40	140	10,61	1,2
110	41,5	32	0	26,5	190	9,94	1,3
160							

29.04.2010							
Mittelwert	CH4	CO2	O2	N2	Mittelwert	Temperature	CH4/CO2
[cm]	%	%	%	%	[cm]	°C	
<b>Kammer A</b>							
25	0	1,9	19,6	78,5	25	14,0	0,0
50	0,1	2,4	19,2	78,3	50	13,8	0,0
75	0	1,2	19,9	78,9	75	13,7	0,0
110	0	16	10,3	73,7	190	11,0	0,0
160	14,75	34,5	0	50,75			0,4
210	16,5	37	0	46,5			0,4
<b>Kammer B</b>							
25	0	3,4	17,4	79,2	25	14,0	0,0
50	0	5,3	16	78,7	50	13,7	0,0
75	0	3	18	79	75	12,7	0,0
110	0	20	4,2	75,8	190	10,1	0,0
160	2,45	31	0	66,55			0,1
210	4,7	33	0	62,3			0,1
<b>Kammer C</b>							
25	3,3	11	9,3	76,4	25	14,1	0,3
50	9,3	18	5,1	67,6	50	13,1	0,5
75	32	35	1,3	31,7	75	12,4	0,9
110	51	45	0	4	190	18,9	1,1
160	52	50	0	0			1,0
210	49	50	0	0			
<b>Kammer D</b>							
25	0	7,2	15,1	77,7	25	16,1	0,0
50	0	5,2	16,4	78,4	50	15,6	0,0
60	27	20	6,9	46,1	140	11,7	1,4
110	46,25	36,5	0	17,25	190	10,7	1,3
160							

## A.2 FID-screenings

### FID-Kartierung - Lys Seibersdorf

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 29.05.08, 11h

Ausführende: OG, PPH, JT

Werte in ppm CH<sub>4</sub>

Gerät: PE-Micro-FID

Witterung (Temp, Wind, Luftdruck, etc.):

Sonnig, 25-26°C

Wind: 6.5 km/h in Bodennähe, 15 km/h in 1,5m Höhe

Luftdruck 995 mbar

Lichtlot: kein Wasser in Belüftungsrohr

Beschreibung der Oberfläche (trocken/feucht, etc.):

trocken mit Pflanzenbewuchs

Kammer A, B: Teil der Pflanzen kurz zuvor entfernt,

offener Boden

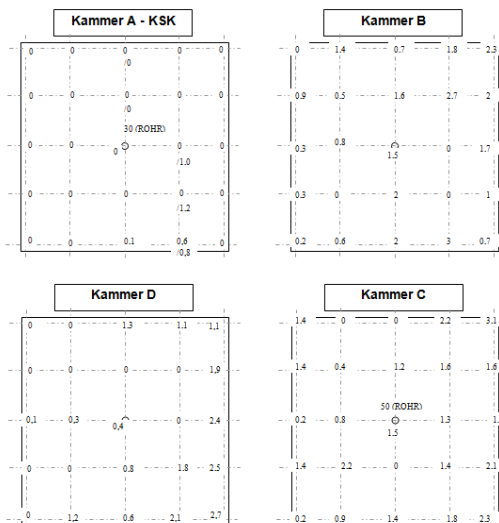
Kammer C: Teil der Pflanzen kurz zuvor entfernt,

aber noch immer geschlossene Pflanzendecke

Kammer D: geschlossene Pflanzendecke

mit hochwüchsigen Ackersenf

Hintergrund: 1 ppm in Umgebungsluft!



### FID-Kartierung - Lys Seibersdorf

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 27.08.09

Ausführende: Hrad, Huber-Humbert

Werte in ppm CH<sub>4</sub>, Umgebung: -0,1 – 0,7 ppm

Gerät: Thermo FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 12:40 0 – 2,1 km/h, sonnig, wolkenlos

Bodennähe: 0 km/h

Temp.: 32°C

Ende: 13:10 0 – 2,8 km/h

Bodennähe: 0 km/h

Temp.: 31,6°C

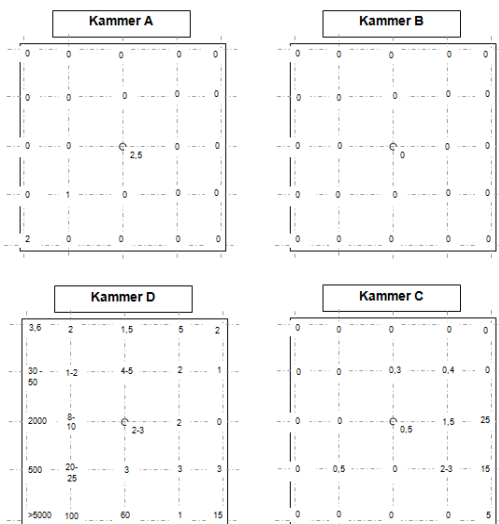
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, starker Bewuchs (bis 1,5 m)

Kammer B: trocken, starker Bewuchs (bis 2 m)

Kammer C: trocken, geschlossen Pflanzendecke (bis 1/2 m)

Kammer D: trocken, starker Bewuchs



### FID-Kartierung - Lys Seibersdorf

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 02.09.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>

Gerät: Thermo FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 14:10 0 – 2,8 km/h, bewölkt

Bodennähe: 0 km/h

Temp.: 30,8°C

Umgebung: 0,2 ppm

Ende: 15:15 0 km/h

Bodennähe: 0 km/h

Temp.: 30,8°C

Luftdruck: 985 mbar

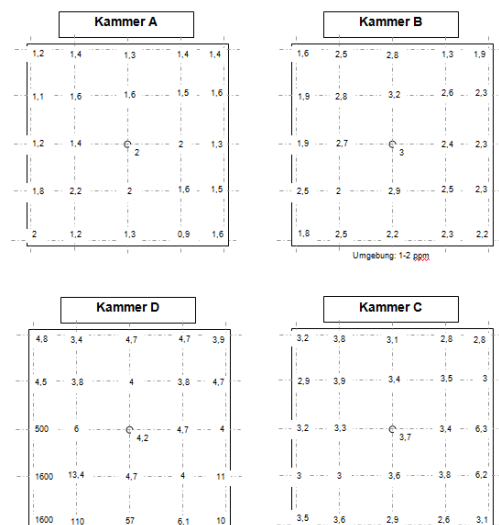
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, starker Bewuchs (bis 1,5 m)

Kammer B: trocken, starker Bewuchs (bis 2 m)

Kammer C: trocken, geschlossen Pflanzendecke (bis 1/2 m)

Kammer D: trocken, starker Bewuchs



**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 09.09.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>, Umgebung 0,08 ppm  
 Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 15:40 0 – 1 km/h, sonnig, wolkenlos

Bodennähe: 0 km/h

Temp.: 25,8°C

Luftdruck: 992 mbar

Ende: 16:40 0 km/h

Bodennähe: 0 km/h

Temp.: 26,1°C

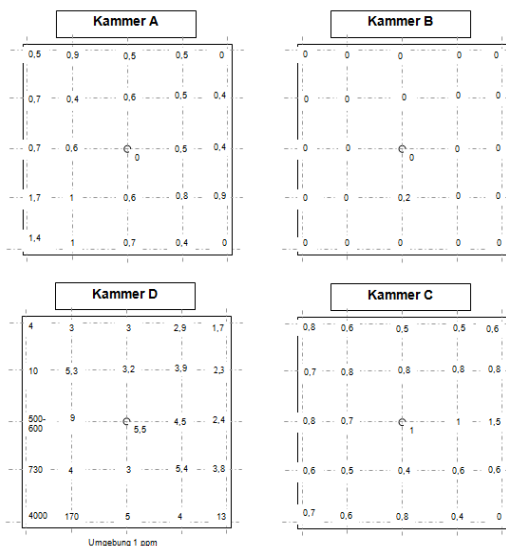
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, frisch gemäht

Kammer B: trocken, frisch gemäht

Kammer C: trocken, geschlossen Pflanzendecke (bis 1/5 m)

Kammer D: trocken, starker Bewuchs (bis 1,5 m)

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 18.09.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 14:20 0 – 1 km/h, bewölkt

Bodennähe: 0 km/h

Temp.: 24,6°C

Luftdruck: 988 mbar

Ende: 15:00 0 – 1 km/h

Bodennähe: 0 km/h

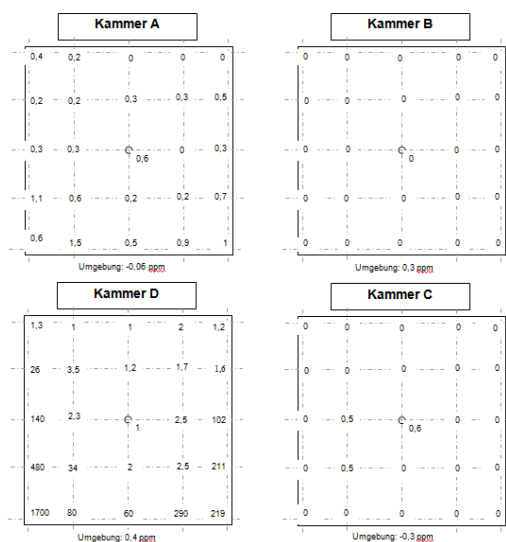
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, gemäht, geschlossene Pflanzendecke

Kammer B: trocken, gemäht, geschlossene Pflanzendecke

Kammer C: trocken, gemäht, geschlossene Pflanzendecke

Kammer D: trocken, gemäht, geschlossene Pflanzendecke

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 24.09.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 14:00 0 – 2,5 km/h, sonnig

Bodennähe: 0 km/h

Temp.: 26,2°C

Luftdruck: 991 mbar

Ende: 15:00 0 – 1,8 km/h

Bodennähe: 0 km/h

Temp.: 26,8°C

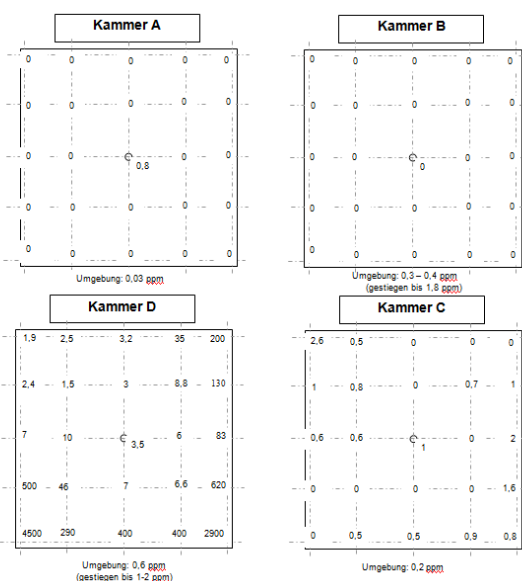
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: sehr trocken, geschlossene Pflanzendecke

Kammer B: sehr trocken, geschlossene Pflanzendecke

Kammer C: sehr trocken, geschlossene Pflanzendecke

Kammer D: sehr trocken, geschlossene Pflanzendecke





**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 01.10.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: Thermo FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 13:45 0 – 2,1 km/h, sonnig

Bodennähe: 0 km/h

Temp.: 26,6°C

Luftdruck: 981 mbar

Ende: 14:40 0 – 1,8 km/h

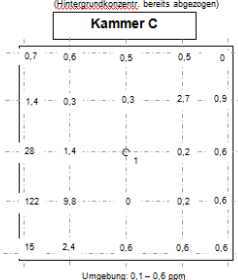
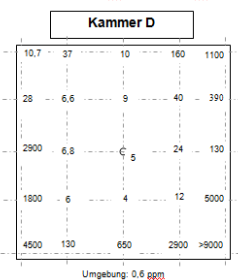
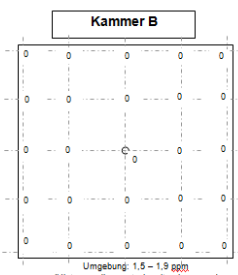
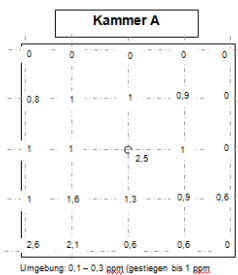
Bodennähe: 0 km/h

Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: sehr trocken, geschlossene Pflanzendecke

Kammer B: sehr trocken, geschlossene Pflanzendecke

Kammer C: sehr trocken, geschlossene Pflanzendecke

Kammer D: sehr trocken, geschlossene Pflanzendecke  
(wurde bewässert)**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 07.10.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: Thermo FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 15:40 0 – 5 km/h (Spitzen bis 6,4 km/h), sonnig

Bodennähe: 0 km/h

Temp.: 29,5°C

Luftdruck: 987 mbar

Ende: 17:00 0 – 5 km/h (Spitzen bis 6,1 km/h)

Bodennähe: 0 km/h

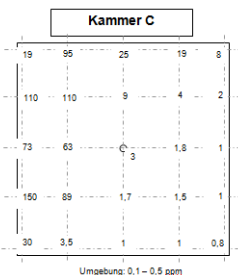
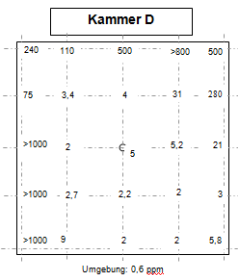
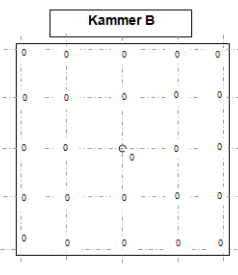
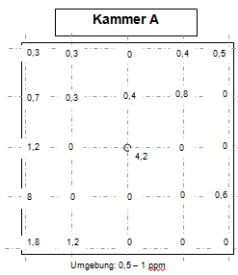
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, geschlossene Pflanzendecke

Kammer B: trocken, geschlossene Pflanzendecke

Kammer C: etwas feucht, geschlossene Pflanzendecke  
(wurde bewässert)

Kammer D: trocken, geschlossene Pflanzendecke

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 21.10.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: Thermo FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 14:05 0 – 5 km/h (Spitzen bis 6,4 km/h), sonnig

Bodennähe: 0 – 2,1 km/h

Temp.: 9,3°C

Ende: 15:05

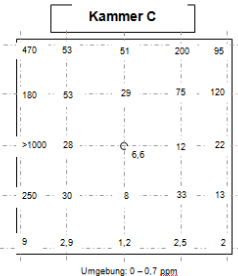
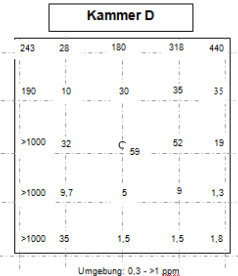
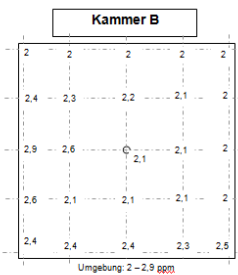
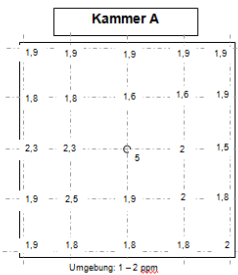
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: feucht, geschlossene Pflanzendecke

Kammer B: etwas feucht, geschlossene Pflanzendecke

Kammer C: feucht, geschlossene Pflanzendecke  
(wurde bewässert)

Kammer D: feucht, geschlossene Pflanzendecke



**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 28.10.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: Thermo FID, TVA-100Witterung (Temp., Wind, Luftdruck, etc.):

Start: 16:00 0 – 0,4 km/h, bewölkt

Bodennähe: 0 km/h

Temp.: 13,1 °C

Ende: 17:05

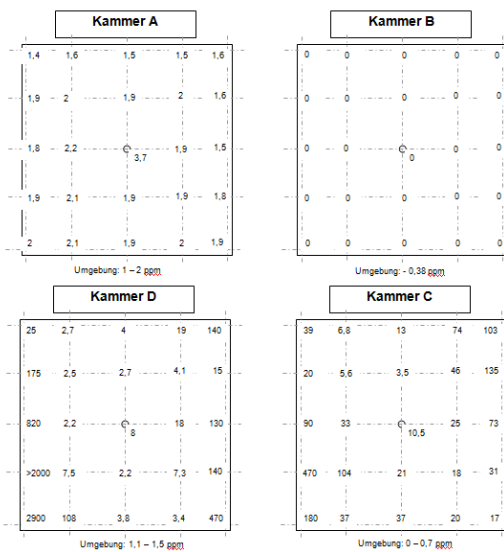
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: feucht, geschlossene Pflanzendecke

Kammer B: feucht, geschlossene Pflanzendecke

Kammer C: feucht, geschlossene Pflanzendecke

Kammer D: feucht, geschlossene Pflanzendecke

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 18.11.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: Thermo FID, TVA-100Witterung (Temp., Wind, Luftdruck, etc.):

Start: 14:05 0 – 3,2 km/h, bewölkt

Bodennähe: 0 km/h

Temp.: 13,2 °C

Ende: 15:10

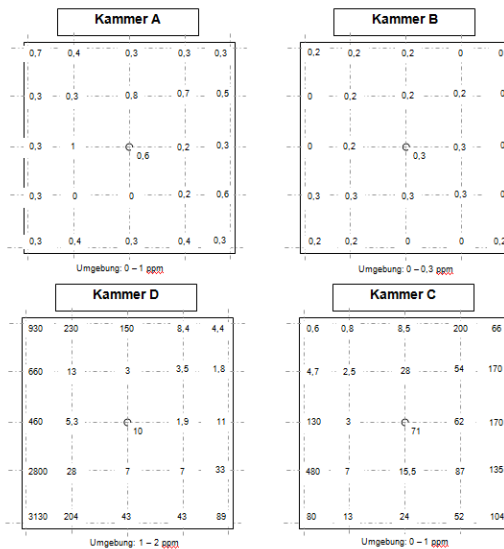
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: feucht, geschlossene Pflanzendecke

Kammer B: feucht, geschlossene Pflanzendecke

Kammer C: feucht, geschlossene Pflanzendecke

Kammer D: feucht, teilw. kahle Stellen

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 02.12.09

Ausführende: Hrad

Werte in ppm CH<sub>4</sub>Gerät: Thermo FID, TVA-100Witterung (Temp., Wind, Luftdruck, etc.):

Start: 10:15 0 – 2,1 km/h, sonnig

Bodennähe: 0 km/h

Luftdruck 984 mbar

Temp.: 8 °C

Ende: 11:10

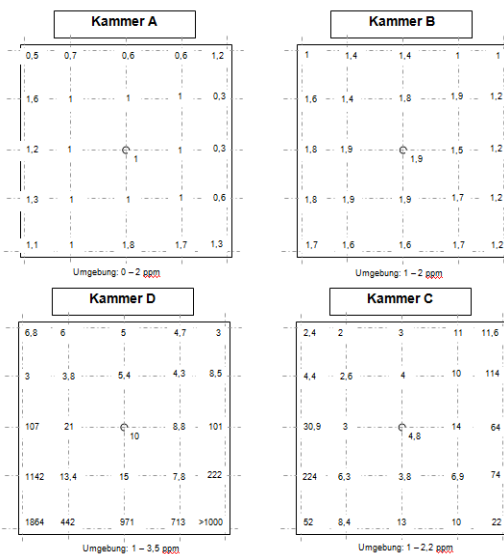
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: feucht, geschlossene Pflanzendecke

Kammer B: feucht, geschlossene Pflanzendecke

Kammer C: feucht, geschlossene Pflanzendecke

Kammer D: feucht, teilw. kahle Stellen



**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 26.01.2010

Ausführende: Hrad

Werte in ppm CH4

Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 13:00 0 – 5 km/h, bedeckt, teilw. sonnig

Bodennähe: 0 – 1 km/h

Luftdruck 1000 mbar

Temp.: -0,1 °C

Ende: 13:50

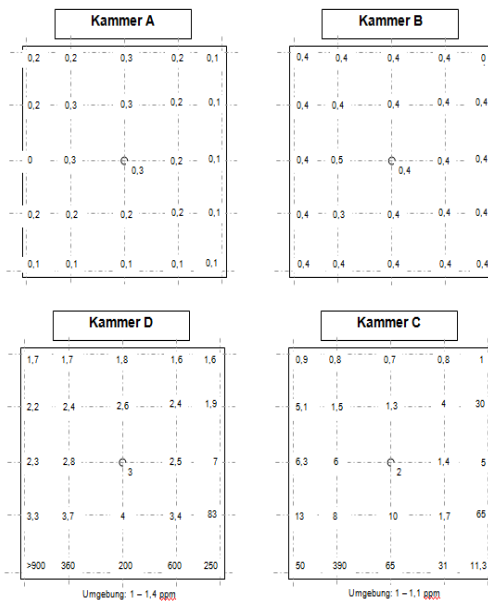
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: Schnee bedeckt (2-3 cm)

Kammer B: Schnee bedeckt (2-3 cm)

Kammer C: Schnee bedeckt (2-3 cm)

Kammer D: Schnee bedeckt (2-3 cm)

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 09.02.2010

Ausführende: Hrad

Werte in ppm CH4

Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 14:10 0 – 3,9 km/h, Spitzen 8 km/h, bewölkt

Bodennähe: 0 – 2,8 km/h

Luftdruck 974 mbar

Temp.: -0,5 °C

Ende: 15:10

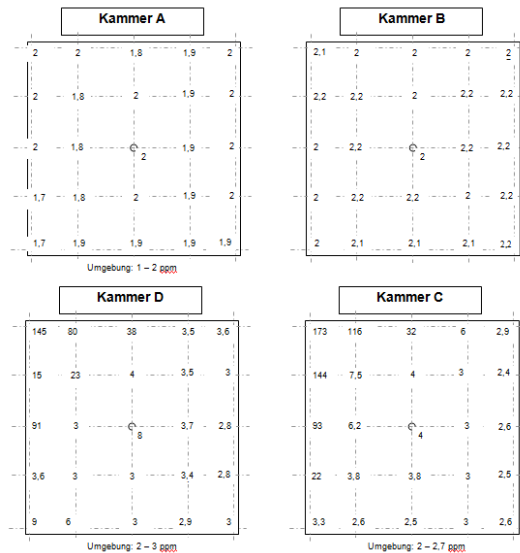
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: teilw. mit Schnee bedeckt

Kammer B: teilw. mit Schnee bedeckt

Kammer C: teilw. mit Schnee bedeckt

Kammer D: teilw. mit Schnee bedeckt

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 18.03.2010

Ausführende: Hrad

Werte in ppm CH4

Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 13:50 sonnig, windstill

Temp.: 17,7 °C

Ende: 14:40

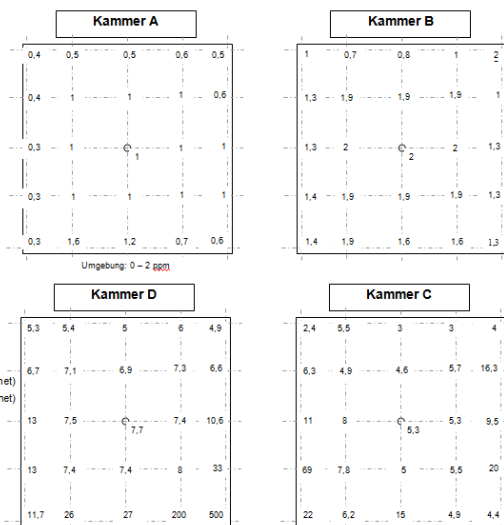
Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, geschlossene Pflanzendecke

Kammer B: trocken, geschlossene Pflanzendecke (vertrocknet)

Kammer C: trocken, geschlossene Pflanzendecke (vertrocknet)

Kammer D: trocken, teilweise geschlossene Pflanzendecke



**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 24.03.2010

Ausführende: Egle

Werte in ppm CH<sub>4</sub>Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 11:20 0 – 5,7 km/h, Spitzen bis 10 km/h

Bodennähe: 2,8 km/h

Temp.: 17,8°C

Luftdruck: 986 mbar

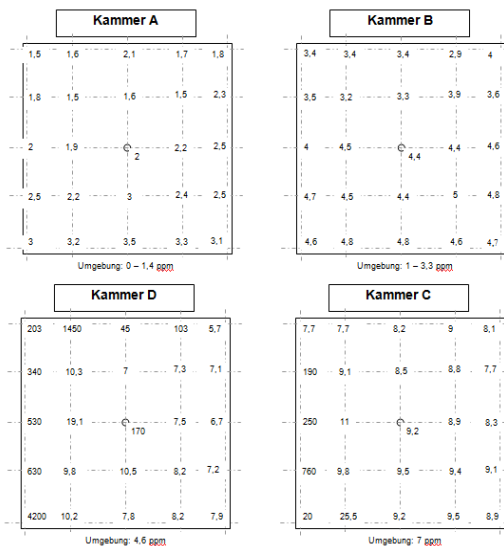
Ende: 12:30

Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, geschlossene Pflanzendecke

Kammer B: **trocken, geschlossene** PflanzendeckeKammer C: **trocken, geschlossene** Pflanzendecke

Kammer D: trocken, teilweise geschlossene Pflanzendecke

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 01.04.2010

Ausführende: Egle

Werte in ppm CH<sub>4</sub>Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 10:45 0 – 2.5 km/h, sonnig

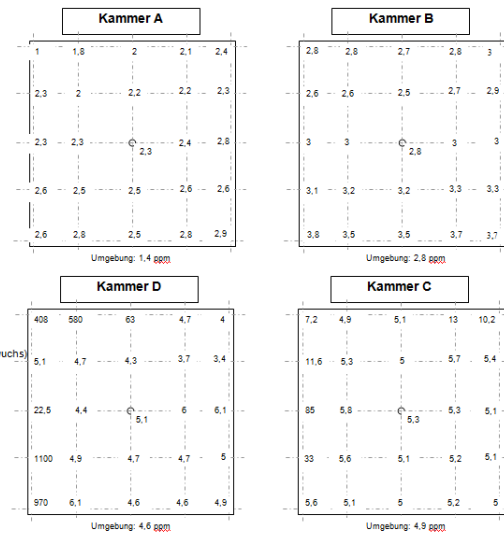
Bodennähe: 0 km/h

Temp.: 21.2 °C

Luftdruck: 980 mbar

Ende: 11:40

Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: feucht, **geschloss**, Pflanzendecke (starker Bewuchs)Kammer B: trockener, fast **geschloss**, PflanzendeckeKammer C: trockener, fast **geschloss**, PflanzendeckeKammer D: trocken, 60 % **geschloss**, Pflanzendecke**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrasterpunkt und visuelle Auffälligkeiten)

Datum: 07.04.2010

Ausführende: Andreas

Werte in ppm CH<sub>4</sub>Gerät: **Thermo** FID, TVA-100

Witterung (Temp, Wind, Luftdruck, etc.):

Start: 15:40 0 – 0,7 km/h, sonnig

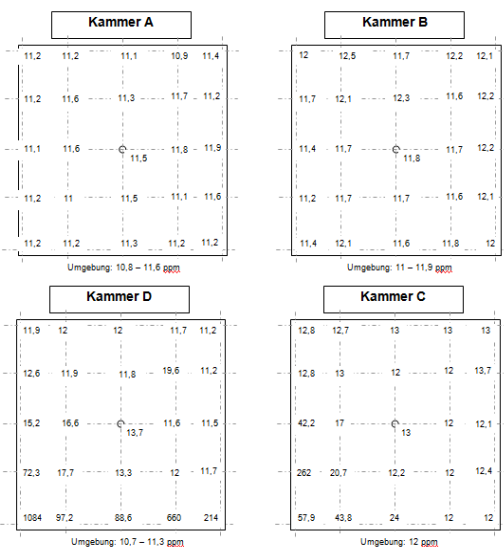
Bodennähe: 0 km/h

Temp.: 25.4 °C

Luftdruck: 990 mbar

Ende: 17:10

Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, **geschloss**, Pflanzendecke (gemäht)Kammer B: trocken, fast **geschloss**, PflanzendeckeKammer C: trocken, fast **geschloss**, Pflanzendecke (80 %)Kammer D: trocken, 50 % **geschloss**, Pflanzendecke

**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrastrasterpunkt und visuelle Auffälligkeiten)

Datum: 19.04.2010

Ausführende: LE

Werte in ppm CH4

Gerät: Thermo FID, TVA-100Witterung (T<sub>emp</sub>, Wind, Luftdruck, etc.):

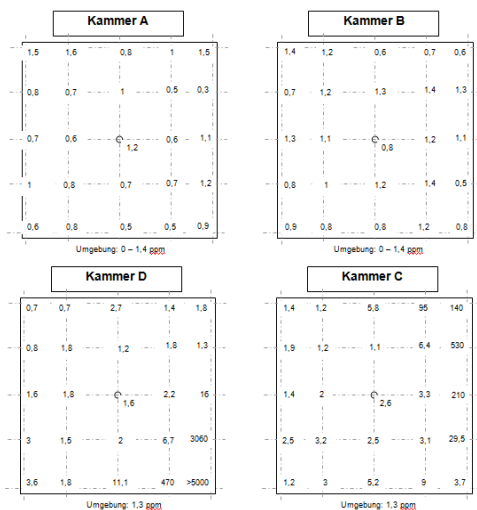
Start: 14:10 0 – 3,4 km/h, sonnig

Bodennähe: 0 – 2,1 km/h

T<sub>emp</sub>: 24,3 °CLuftdruck: 980 mbar

Ende: 15:15

Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: feucht, geschloss, PflanzendeckeKammer B: feucht, fast geschloss, PflanzendeckeKammer C: feucht, fast geschloss, Pflanzendecke (80 %)Kammer D: feucht, 50 % geschloss, Pflanzendecke**FID-Kartierung - Lys Seibersdorf**

(Aufnahme Messrastrasterpunkt und visuelle Auffälligkeiten)

Datum: 29.04.2010

Ausführende: AS

Werte in ppm CH4

Gerät: Thermo FID, TVA-100Witterung (T<sub>emp</sub>, Wind, Luftdruck, etc.):

Start: 16:45 0 – 11 km/h, sonnig

Bodennähe: 0 – 2,5 km/h

T<sub>emp</sub>: 26 °CLuftdruck: 986 mbar

Ende: 17:50

Beschreibung der Oberfläche (trocken/feucht, etc.):

Kammer A: trocken, geschloss, Pflanzendecke (geschnitten)Kammer B: trocken, fast geschloss, Pflanzendecke (geschnitten)Kammer C: trocken, geschloss, Pflanzendecke (80 %)Kammer D: trocken, 60 % geschloss, Pflanzendecke