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Technical University of Denmark

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DTU Environment
Department of Environmental Engineering

PhD Thesis
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Preface

The work reported in this PhD thesis, entitled 'Processes in a compost based landfill biocover; methane emission, transport and oxidation', was conducted at the Department of Environmental Engineering at the Technical University of Denmark with Professor Peter Kjeldsen and associate Professor Charlotte Scheutz as supervisors. The PhD project ran from August 2004 to February 2010 and was funded by the Technical University of Denmark.

The content of the PhD thesis is based on five papers prepared for scientific journals. The papers represent the many sub-projects, which were included in the PhD project and conducted in collaboration with internal and external partners. In the text, the papers are referred to by the names of the authors and their appendix number written with roman numbers.

I Pedersen, G.B., Scheutz, C., Kjeldsen, P (2010a). Mitigation of methane emission from Fakse Landfill using a biocover system II: Material availability and properties. Manuscript submitted to Waste Management. 2010a.

II Fredenslund, A.M., Pedersen, G.B., Scheutz, C., Kjeldsen, P. (2010). Mitigation of methane emission from Fakse Landfill using a biocover system III: Design and performance. Manuscript for submission to Waste Management. 2010.

III Pedersen, G.B., Scheutz, C., Kjeldsen, P. (2010b) Field study on a biocover window at Fakse Landfill, Denmark I: Short term dynamics and seasonal changes in methane emissions. Manuscript for submission to Science of the Total Environment. 2010b.

IV Pedersen, G.B., Scheutz, C., Pedicone, A., Chanton JP, Kjeldsen P (2010c). Field study on a biocover window at Fakse Landfill, Denmark II: methane oxidation. Draft manuscript for submission to Science of the Total Environment. 2010c.

V Scheutz, C., Pedersen, G. B., Costa, G., Kjeldsen, P. 2009. Biodegradation of Methane and Halocarbons in Simulated Landfill Biocover Systems Containing Compost Materials. Journal of Environmental Quality 38:1363-1371.

The papers are not included in this www-version, but can be obtained from the Library at DTU Environment
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I thank the entire *crew at Fakse Landfill*, for their help and for welcoming me and letting me use their facilities as I did indeed work there. Especially *Bente Munk* and *Jens-Erik Nielsen* have been a great help.

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I thank *my parents and my parents in law* and my brother and sister for taking care of my daughter during hectic periods and *my husband* and *daughter* for their patience and for fulfilling the need of completely different inputs than that of my study.

Last but not least special thanks are given to my *husband*, *my parents* and *my grandmother Tinne* for supporting me financially through the last months of my study.

Gitte

Summary

Methane (CH_4) is a greenhouse gas with a global warming potential (GWP_{100}) that is 25 that of carbon dioxide (CO_2). CH_4 and CO_2 (landfill gas) is produced anaerobically in landfills and emitted to the atmosphere. Landfill gas can be collected and used for energy production or flared, though the collection of landfill gas is seldom very efficient and is not feasible in low production phases and for landfills where waste with low organic content is deposited. The CH_4 emission from these landfills can be oxidized in biocovers, which are biologically active landfill top covers that promote CH_4 oxidation.

The present PhD thesis provides an insight in the parameters affecting CH_4 oxidation in a field scale biocover and the determination of CH_4 oxidation rates in passive biocover systems. The results presented is related to a full scale biocover system installed at Fakse Landfill, where parts of the low permeable clay cover was removed and replaced with compost (referred to as biocover windows). The activities included laboratory testing of locally available materials for the full scale biocover at Fakse Landfill and quantification of CH_4 oxidation rates to determine design parameters for the full scale biocover. Additionally it included the evaluation of the performance of the implemented biocover system and improvements to the system. A detailed field study was done on a selected biocover window to assess emissions and oxidation of CH4 and evaluate important variables, which affected the emission and oxidation of CH_4.

The selection of a suitable material for the full scale biocover was initiated by identifying seven different compost materials at the site. The materials were characterized and from their CH_4 oxidation potential, oxygen (O_2) demand, structure, availability and price five materials were chosen for testing in a column set-up. Similar results for three materials allowed choosing the cheaper and more easily available material, which was raw compost aged four years. The results of column test were compared to criteria given in the literature for materials suitable for CH_4 oxidizing biocovers and they only partly agreed with the criteria. Composts with high O_2 demands and low content of phosphorous did promote CH_4 oxidation (up to 195 g CH_4 m^{-2} d^{-1}). Generally it was found that criteria on the ammonium content in compost were not meaningful as the knowledge on its inhibitory affects is inconsistent. Maturity was identified as an important parameter, but the classification systems for maturity were not in agreement. A

low C/N ratio was believed to be a better parameter for evaluating the suitability of compost materials for biocover application. The five different compost materials were ranked according to their performance in both batch incubations and column incubations and it was found that batch incubations did successfully rank the materials, but it was not possible to determine realistic CH_4 oxidation from the batch incubations for sizing the biocover. Thus even the CH_4 oxidation rates determined in column incubations will be an estimate of field scale CH_4 oxidation rates as the field situation will be subject to changes in temperature, water content and flow of CH_4 to the biocover window.

The full scale biocover system at Fakse Landfill had an area of 5000 m^2 and consisted of ten biocover windows. The implementation of the system was performed by co-workers, thus the evaluation of the performance was a part of this study. Gas concentration profiles of CH_4, CO_2, O_2 and nitrogen and surface emissions of CH_4 and CO_2 in 12 random measuring points, four on each of three of the ten biocover windows were done together with measurement of the CH_4 concentrations in the air on top of the biocover windows. The results indicated that the load of CH_4 to the biocover windows was very low. Additionally FID screenings of the CH_4 concentrations revealed that the fugitive emissions of CH_4 from installations at the site (pumping station and leachate collection wells) were still major compared to the total emission of CH_4. Due to this improvements were done to the biocover system. A water lock was installed at the leachate pumping station and the area around the leachate collection wells was sealed with bentonite both to minimize fugitive emissions from these installations. Four biocover windows were improved as 2-4 m of waste mixed with clay were identified under the gas distribution layer. Trenches were dug and filled with tree roots in order to improve the load of CH_4 to these biocover windows. Despite the efforts to improve the system the CH_4 load remained low, though hotspots were identified on selected windows, where the biocover was overloaded. Additionally, the results indicated that emissions of CH_4 and CO_2 from the biocover windows were dynamic.

A detailed study was done on a biocover window with an area of 85 m^2 on Fakse Landfill. The short term dynamics in emission were assessed and it was controlled mainly by changes in atmospheric pressure. The emission of CH_4 changed within hours according to the changes in the gradient of the atmospheric pressure, but it was found that the history of the atmospheric pressure and the pressure gradient determined the magnitude of the emission. A longer negative pressure gradient would result in a lower pressure in the landfill (less storage of CH_4) and it would take a while, before the pressure in the landfill was build up by the production of landfill gas. In general decreases in atmospheric pressure shifted the CH_4 oxidation zone upwards to the upper layers of the compost. Temperature and water content was logged in three profiles over 1 year and temperatures remained high in the bottom of the biocover window (>15 ^{o}C). Therefore it was concluded that if a low load of CH_4 could be ensured the CH_4 oxidation would be unaffected by diurnal temperature fluctuations and almost unaffected by the decrease in temperature during winter. The high temperatures compared to atmospheric temperatures were mainly attributed to the degradation of organic matter in the compost and the generally low thermal conductivities of coarse compost materials compared to mineral soils. It was difficult to achieve the designed load of CH_4 to the biocover window (150 g CH_4 m^{-2} d^{-1}), and therefore it was assessed whether advective flux was diminished by high water contents in the bottom of the biocover window. Gas permeability was measured in two reference points and calculated for the entire year, with a model developed for screened garden waste compost. Based on this it was found that advective flux was not diminished by high water content above a trench filled with tree roots where drainage was good.

CH_4 oxidation rates were determined for the biocover window with a method (carbon mass balance approach) where it was assumed that the relationship between the concentration of CH_4 and CO_2 in the landfill equalled the relationship between the CH_4 and CO_2 flux into the biocover window. From this the flow of CH_4 into the window could be determined. The average CH_4 oxidation rate and efficiency of the entire biocover window was low, as the distribution of gas to the biocover window was poor. Additionally low CH_4 oxidation rates were measured during winter at pressure decreases where the CH_4 oxidation zone shifted upwards and diffusion of O_2 was limited by high water content. In the situations where the supply of CH_4 and O2 was optimum and temperatures were high, CH_4 oxidation rates higher than in the column study in the laboratory were achieved. On one measuring day the results from the carbon mass balance were compared to a better documented method, where the CH_4 oxidation rates were determined by the change in the composition of the stable isotopes of CH_4. Similar results were achieved and it was concluded that the carbon mass balance was an adequate method to evaluate the local performance of biocover systems.

Overall the efficiency of the biowindow was diminished by both high spatial and temporal variation in the load. Biocover performance can be optimized by using a design that supports a low and evenly distributed CH_4 load (deeper gas distribution layer) as such design criteria will minimize many of the effects caused by the changing environmental factors.

Dansk sammenfatning

Metan er en drivhusgas med et potentiale for global opvarmning, som er 25 gange større end kuldioxids. Metan og kuldioxid (lossepladsgas) bliver produceret under anaerobe forhold i lossepladser og udledt til atmosfæren. Lossepladsgas kan opsamles og bruges til energiproduktion eller brændes uden energiudnyttelse. Opsamlingen af lossepladsgas er sjældent særlig effektiv, og det er ikke økonomisk rentabelt for gamle lossepladser med lav gasproduktion eller lossepladser, hvor der er deponeret affald med et lavt indhold af organisk affald. Metanudledningen fra disse lossepladser kan med fordel oxideres i biologiske aktive lossepladsdæklag også kaldet ”biocover systemer”, som fremmer metanoxidationen.

Den indeværende PhD afhandling giver en gennemgang af de parametre, som påvirker metanoxidationen i biocover systemer i felten. Resultaterne, som er præsenteret i denne afhandling, er relateret til et biocover system i fuld skala, som er implementeret på Fakse Losseplads. Lossepladsen er dækket af et lerlag, som er lavpermeabelt, og dele af det blev gravet væk og erstattet med kompost (biocover vinduer). Aktiviteterne har inkluderet test af mulige, let tilgængelige materialer til biocover systemet og bestemmelse af metanoxidationsrater for disse materialer, som kunne bruges til dimensionering af biocover systemet. Virkningsgraden af biocover systemet blev undersøgt og forbedringer af systemet blev gennemført. Derudover blev der udført et mere detaljeret feltstudie på et udvalgt biocover vindue af emissionen af metan, metanoxidationen og de faktorer, som påvirker metanoxidationen.

Den første del af udvælgelsen af materialer til biocover systemet på Fakse Losseplads var at identificere mulige materialer på lossepladsen. Syv forskellige kompost materialer blev identificeret, og de blev karakteriseret og testet i laboratoriet. Fem materialer blev udvalgt til yderligere test i en kolonne opsætning, baseret på deres potentielle metanoxidationsrater i batch forsøg, deres iltforbrug, struktur, tilgængelighed og pris. I kolonne forsøget var der tre materialer, som opnåede tilsvarende metanoxidationsrater og derfor blev der valgt den kompost, som var nemmest tilgængelig og billigst, hvilket var en fire år gammel råkompost. Resultaterne fra kolonne testene blev sammenlignet med de kriterier for kompost materialer, der er i litteraturen til udvælgelse af kompost materialer til biocover systemer, og der blev kun fundet en delvis overensstemmelse. Generelt blev det fundet, at kriterier for ammonium indholdet i kompost til biocover systemer ikke giver mening, idet der er uoverensstemmelse omkrig, hvorvidt ammonium er inhiberende for metanoxidation eller ikke. Modenheden af komposten var identificeret som en vigtig parameter, men de tilgængelige klassificeringssystemer til bestemmelse af modenhed resulterede i meget forskellige resultater. Et lavt C/N ratio var formodentlig en bedre parameter at benytte til at evaluere, hvorvidt mulige kompostmaterialer er egnede til biocover systemer. Kompost materialerne blev ranglistet med resultater fra både batchforsøg og kolonneforsøg, og det kunne konkluderes at batchforsøgene var tilstrækkelige til dette formål. Men man kunne ikke bestemme realistiske metanoxidationsrater fra batchforsøgene, der kunne bruges til dimensioneringen af biocover systemet. Men metanoxidationsrater bestemt i kolonneforsøg, vil også være estimater af metanoxidationsrater i felt forsøg, da biocover systemer i felten er påvirket af forandringer i temperatur, vandindhold og tilstrømning af metan.

Implementering af biocover systemet på Fakse losseplads med et areal på 5000 m^2 bestående af 10 biocover vinduer er ikke inkluderet i dette studie, men evalueringen af systemet er. Gas koncentrationer i profiler (metan, kuldioxid, ilt og nitrogen) og overflade emissioner af metan og kuldioxid blev målt i 12 tilfældigt udvalgte punkter, fire på hvert af tre udvalgte vinduer, sammen med screeninger af metan koncentrationen i luften over biocover vinduerne. Resultaterne indikerede at tilstrømningen af gas til kompost vinduerne var meget lav. Derimod vidste screeninger omkring perkolatbrønde og pumpestationeen at emissionen herfra fortsat var betydende i forhold til the totale emission af metan fra pladsen. En vandlås blev installeret på pumpestationen og der blev forseglet rundt om perkolatbrøndene med bentonit for at minimere emissionerne fra disse installationer. Der blev gennemført forbedringer på fire af vinduerne, da der blev identificeret et 2-4 m dybt lag affald blandet med ler under gas distributionslaget. Der blev gravet render ned gennem dette lag, som blev fyldt med trærødder for at forbedre tilstrømningen af lossepladsgas. Selvom alle disse forbedringer blev gennemført var tilstrømningen af losseplads gas til biocover vinduerne fortsat lav, men der blev identificeret ”hot spots” (områder med høj emission) på udvalgte vinduer, hvor tilstrømningen af metan var for høj. Derudover indikerede resultaterne, at emissionen fra biocover vinduerne var meget dynamisk.

Et mere detaljeret felt studie blev udført på et af vinduerne (85 m^2) med et større ”hot spot”. Dynamikken i emissionerne og gas koncentrationer i kompost vinduet blev undersøgt i intensive kampagner over 3 dage og emissionerne var meget afhængige af udviklingen i det atmosfæriske tryk. Emissionerne forandredes efter få timer, når gradienten i det atmosfæriske tryk forandredes, men selve niveauet i emissionerne blev bestemt af, hvordan trykket og trykgradienten havde været i dagene op til selve kampagnen, da dette påvirker trykket inde i lossepladsen. Lave tryk og negative trykgradienter forud for kampagnen resulterer i lave tryk i lossepladsen og der vil gå flere dage før dette tryk er bygget op igen pga. produktion af lossepladsgas. Generelt blev det fundet, at et fald i det atmosfæriske tryk medførte, at metan oxidations zonen blev skubbet opad i profilet. Temperatur og vandindhold blev logget i tre profiler over et år og høje temperaturer blev målt i bunden af biocover vinduet. Det blev derfor konkluderet, at hvis en relativ lav tilstrømning af gas til biocover vinduet kunne sikres, hvilket medfører at metanoxidationen foregår in bunden af biocover vinduet eller i gasdistributionslaget, ville metan oxidationen kunne foregå året rundt, næsten uden påvirkning af fald i temperaturen om vinteren og uden påvirkning af fald i temperaturen om natten. Generelt var temperaturerne i biocover vinduet meget højere end lufttemperaturen, hvilket hovedsageligt skyldes varmeproduktion fra nedbrydning af komposten og at grove kompost materialer har lav varmeledningsevne i forhold til mineralske jorde. Det var problematisk at sikre gastilstrømningen til vinduet, derfor blev det undersøgt om den advektive tilstrømnning blev begrænset på grund af høje vandindhold i bunden af biocover vinduet. Gaspermeabiliteten blev målt i to reference punkter og gaspermeabiliteten blev udregnet udfra vandindholdet over et helt år med en model udviklet til sigtet kompost af haveaffald. Baseret på dette, kunne det konkluderes at den advektive gastilstrømning ikke var begrænset af høje vandindhold i bunden af biocover vinduet ovenover den rende, som var gravet for at sikre tilstrømningen af gas.

Metanoxidationsrater blev bestemt for biocover vinduet, med en metode (karbon masse balance), hvor man antager, at forholdet mellem koncentrationerne af metan og karbondioxid i gas prøver taget i affaldet er lig med forholdet mellem metan og karbondioxid i tilstrømningen af lossepladsgas ind i biocover vinduet. Herved kan strømmen af metan til vinduet bestemmes. De gennemsnitlige metanoxidationsrater og virkningsgraden i vinduet var lav, da lossepladsgassen ikke blev jævnt fordelt i gas distributionslaget. Derudover blev der målt meget lave metanoxidationsrater om vinteren under et trykfald, hvor metanoxidationszonen blev skubbet opad, og diffusionen af ilt var begrænset pga høje vandindhold. I de situationer, hvor tilførslen af ilt og metan var god, og temperaturen var høj, blev der målt højere metanoxidationsrater, end der blev fundet i kolonneforsøg i laboratoriet. I en feltkampagne blev karbonbalancen sammenlignet med en mere veldokumenteret metode, hvor metanoxidationen bestemmes vha. af forandringen i de stabile isotoper i metan. Lignende resultater blev opnået og det blev konkluderet, at karbon balancen var en passende metode til bestemmelse af metanoxidationsrater i biocover vinduer.

Generelt blev det konkluderet, at virkningsgraden af biovinduet blev reduceret pga. de store variationer i tilstrømningen af gas både over tid og sted. Biocover systemers virkningsgrad kan optimeres ved at designe dem, så tilstrømningen er lav og jævnt fordelt i gas distributionslaget, hvilket kan gøres med et dybere gas distributionslag, eksempelvis ½ m.

List of contents

1 Introduction and Background

1.1 CH_4; global warming potential and mitigation options

CH_4 is a green house gas with a global warming potential (GWP_{100}), which is 25 times that of CO_2 and CH_4 accounted for 14.3 % of anthropogenic greenhouse gas emissions in 2004 (Barker *et al.*, 2007). World wide the CH_4 emission from the waste sector is about 18 % of the global antropogenic CH_4 emission and the main part of this emission originated from landfills (Bogner *et al.*, 2007). Generally post consumer waste is a relatively small contributor to global green house gas emissions (<5 %), but the importance of the waste sector for reducing global green house gas emissions has been underestimated because waste management strategies are often made locally without concurrent quantification of green house gas mitigation (Bogner *et al.*, 2007).

Global CH_4 emissions from landfills are estimated to be 500-800 Mt CO_2-eq/yr and it is projected to be 2900 Mt CO_2-eq/yr in 2050 based on 2006 inventory guidelines (Bogner *et al.,* 2007). It is important to state that the emissions and projections are based on estimated CH_4 production rates applied on national statistics for landfilled refuse and not on field measurements. Landfill emission in the EU has decreased due to a decrease in the landfilling of biodegradable waste and increased recovery and flaring of landfill gas (Bogner *et al.*, 2007). However life time efficiency of flaring and recovery is estimated to be as low as 20 % (Oonk and Boom, 1995). Landfill gas is also produced at sites where little organic waste is deposited and recovery will not be beneficiary. The residual emission of CH_4 and emissions from landfills with small amounts of organic wastes can be mitigated by CH_4 oxidation in biologically active biocovers (Hilger and Humer, 2003; Barlaz *et al.*, 2004; Scheutz *et al.*, 2009a; Huber-Humer *et al.*, 2009). CH_4 emissions from landfills in developing countries are increasing due to a rising urban population, increasing economic development and to some extent replacement of open burning with engineered landfills (Barker *et al.*, 2007). Also here passively aerated low maintenance biocovers are potential mitigation solutions.

1.2 CH_4 emissions from landfills

CH_4 emissions from landfills are mainly determined by the production rate of landfill gas, as the increasing pressure inside the landfill will induce an advective flux of landfill gas towards the atmosphere (Kjeldsen, 1996). Landfill gas consists of CH_4 and CO_2, which is produced in landfills under anaerobic conditions and the concentrations are 55-60 % v/v and 40-45 % v/v respectively (Scheutz *et al.*, 2009a). Methanogenesis in landfills occur under strict anaerobic conditions by one of two mechanisms, acetate fermentation or CO_2 reduction. During acetate fermentation equal proportions of CH_4 and CO_2 is produced, but during CO_2 reduction, CO_2 is removed and CH_4 is formed, which explains the higher CH_4 concentration compared to CO_2 normally found in landfill gas (Bogner *et al.*, 1996). The production of CH_4 and CO_2 depends on several factors; the landfilled waste volume, content of organic waste fractions, waste age and environmental factors (temperature, moisture content, nutrients, inhibiting compounds, etc.) (Scheutz *et al.*, 2009a). This will influence both the spatial and temporal emission of CH_4. Often daily covering with low permeability soils is used and this will cause spatially heterogeneous gas permeabilities in the landfill (Kjeldsen, 1996). Additionally compaction of the waste together with the heterogeneity of the deposited waste will result in areas with high production of CH_4 resulting in high pressure and other areas with lower production of CH_4 resulting in lower pressure (Kjeldsen, 1996). In Fig. 1 the emission and risk of lateral gas flow is conceptualized for an unlined landfill.

High spatial and temporal variation in emissions exists and high emissions are often measured around cracks and fissures in clay covers and around installation and faulty covers (Scheutz *et al.*, 2009a). Until recently the main focus when implementing covers on landfills was minimizing the amount of leachate formed. Therefore many landfills are covered with low permeability soils, such as clay (Kjeldsen, 1996). Clay covers will, when drying out, easily crack, which will result in the formation of hotspots (areas with high CH_4 emission). Another typical high emission area are landfill slopes (Kjeldsen, 1996; Borjesson *et al.*, 2000;Fredenslund *et al.*, 2010b) as daily coverings and compaction of the waste will support lateral transport of landfill gas compared to vertical transport (Kjeldsen, 1996). With small scale chamber techniques the emission measured have ranged over more than seven orders of magnitude, from 0.0001 to 1000 g CH_4 m^{-2} d^{-1} (Scheutz *et al.*, 2009a). The temporal variation in the emission is among other things controlled by the change in atmospheric pressure (Scheutz *et al.*, 2009a). The advective flux of CH_4 will increase as the atmospheric pressure decreases. CH_4 oxidation will also result in temporal changes in the emission as it depends on temperature and precipitation.

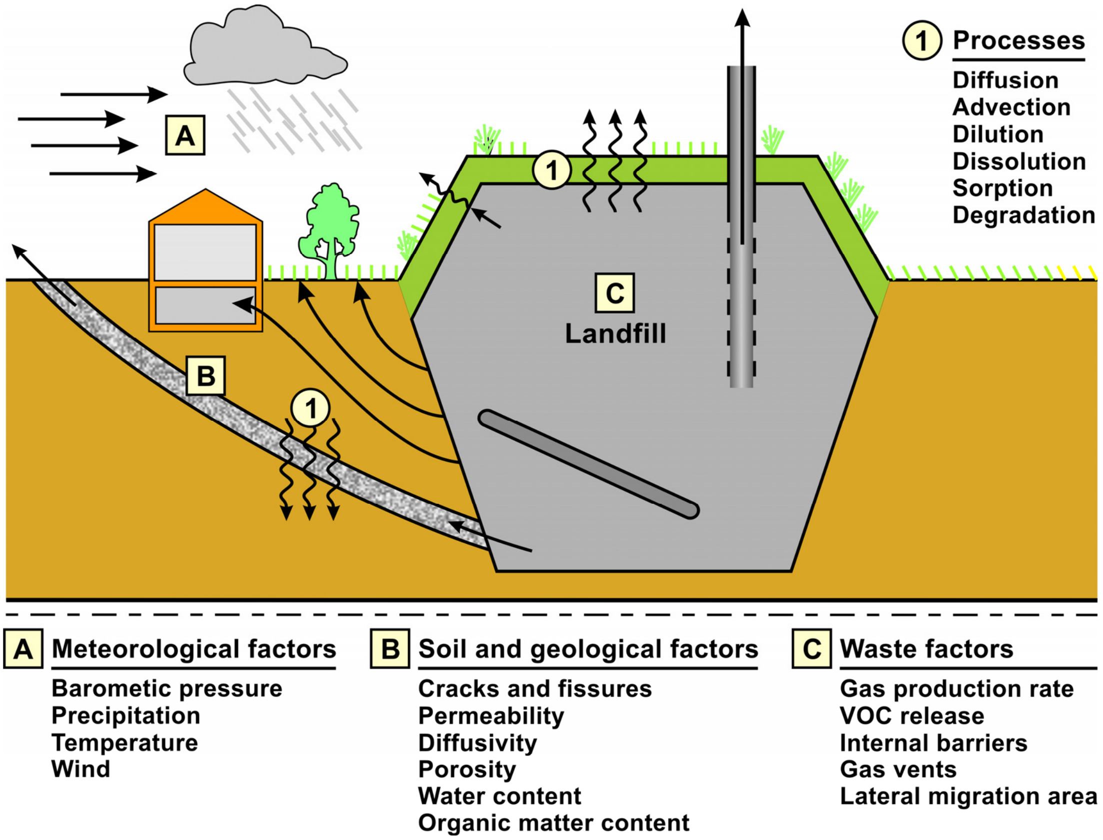

Fig. 1: Conceptual framework of landfill gas emission (Kjeldsen, 1996).

CH_4 emissions can be measured by several methods, e.g. above ground micrometeorological methods and tracer methods, and with static and dynamic chambers. The static chamber is the most common technique for measuring landfill emissions and has been field validated (Scheutz *et al.*, 2009a). Fluc chamber measurements are single point measurements, whereas the micrometeorological and tracer techniques can be used to measure total emission from landfills.

1.3 CH_4 oxidation in landfill covers

CH_4 can be oxidized by methanotrophic bacteria in landfill covers. The bacteria uses CH_4 as their carbon and energy source and CO_2 and water are produced. The methanotrophic bacteria are divided into two Types; Type I and Type II. Most Type I bacteria are not able to fixate nitrogen and they are typically associated with low CH_4 concentrations and high concentrations of O_2 and nutrients (Hanson and Hanson, 1996). Type II methanotrophs are capable of fixating nitrogen and investigations suggest that type II methanotrophs can thrive with high CH_4 concentrations and low O_2 and nutrient concentrations (Hanson and Hanson, 1996). Methanotrophic bacteria are ubiquitous soil bacteria, but long lag phases towards reaching high CH_4 oxidation rates are found especially in mineral materials (Philopoulous *et al.,* 2009; De Visscher 1999; Scheutz *et al.,* IV). Therefore the focus in recent years has been on enhancing CH_4 oxidation by using more organic materials, such as compost (Huber-Humer *et al.* 2009; Dever *et al.,* 2007; Einola *et al.,* 2009).

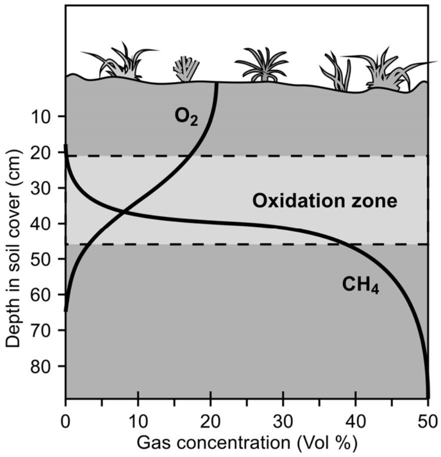

Fig. 2: Idealized gas concentration profile in landfill cover (Scheutz, 2002)

In Fig. 2 a conceptual CH_4 oxidation zone in a landfill cover soil is shown. The optimum zone for CH_4 oxidation is where the gas concentration profile of CH_4 and O_2 encounters. The transport of O_2 to the oxidation zone mainly relies on diffusion, but advective flux of atmospheric air has also been proved to be important by modelling and by measuring augmented N_2 concentrations compared to the atmosphere (Rannaud *et al.*, 2009; De Visscher *et al.*, 1999; Christophersen *et al.*, 2001). The diffusion of O_2 into the CH_4 oxidation zone will be controlled by the total and the gas filled porosity (water content) of the porous medium. Additionally the depth of the CH_4 oxidation will depend on the flow of landfill gas (LFG), when the LFG flow from the landfill increases the CH_4 oxidation zone will be pushed upwards towards the surface and becomes shallower (Scheutz and Kjeldsen, 2003; Rannaud *et al.*, 2009). Moreover a decrease in CH_4 oxidation rates has been attributed to production of exopolymeric substances (EPS), which limits the diffusion of O_2 into the CH_4 oxidation zone (Hilger *et al.*, 1999; Molins *et al.*, 2008; Scheutz *et al.*, 2009b)

Other main factors influencing CH_4 oxidation is temperature, water content and material specific parameters, such as nutrients and specific surface for methanotrophic communities. Some of these factors will be described in greater detail in the thesis. Additionally Scheutz *et al.* (2009a) and Chanton *et al.* (2009b) provide good overviews of CH_4 oxidation rates in laboratory and field studies.

1.4 Biocover Technology

CH_4 emissions from landfills can be mitigated by oxidation in biologically active landfill covers called biocovers (Barlaz et al., 2004; Hilger & Humer, 2003; Scheutz et al., 2009a; Fredenslund et al., 2010a). Biocovers are made of a filter material supporting growth of methanotrophs placed above a gas distribution layer typically made of gravel. The gas distribution layer should have a higher gas permeability and porosity than the filter material in order to evenly distribute the CH_4 coming from the landfill (Scheutz *et al.*, 2009a; Hilger and Humer, 2003). Field studies of biocovers are recently described in the literature (Huber-Humer *et al.* 2008; Jugnia *et al.,* 2008; Einola *et al.,* 2009) and they will be described in the following paragraphs. Table 6 in the thesis provides an overview of the most recent field studies described in the literature and the measured CH_4 oxidation rates.

Gebert & Groengroeft (2006a,b) evaluated the performance of a passively vented biofilter consisting of expanded clay pellets (15 m^2). The biofilter consisted of 5 layers (top to base); 15 cm of humic topsoil, 1.5 cm sand, 1.5 cm gravel, 67 cm crushed porous clay and 10-30 cm gravel for water drainage. The biofilter was embedded in the cover of the landfill and landfill gas was supplied passively through pipes and distributed via pipes embedded in the top centimetres of the drainage gravel layer. The performance of the biofilter was evaluated by various methods. The flux of CH_4 into the biofilter could easily be determined as both flow and concentration was logged in the influent. The emissions were measured in two different types of flux chambers, which were operated manually or automatically. The measured emission was assumed to be only diffusive and the advective emission was calculated. It was found that most of the time the biofilter performance was limited by the influx of CH_4 and that the capacity to degrade CH_4 was very high (up to 1286 g CH_4 m^{-2} d^{-1}). CH_4 removal was also determined from CH_4 load back flow. Analysis of the distribution of CH_4 loads over 1 year revealed that overall 62 % of the annual CH_4 load, could be oxidised (Gebert and Groengroeft, 2006a).

Barlaz *et al.* (2004) reported the results from a field scale biocover (3600 m^2) in Kentucky, USA, which was compared to a soil cover. The biocover consisted of 0.15 m clay, 0.15 m tire chips and 1 m of yard waste compost. They investigated both a flat and a sloped biocover (each covering and area of 30 m x 60 m). The biocover performance was evaluated with the stable isotopic approach, but a high amount of zero emission samples made estimation of load to the biocover impossible. They found that the biocover and the soil cover were responsible for oxidizing 55 % and 21 % of the incoming CH_4 based on 13 and 6 samples, where positive emission were measured (Barlaz *et al.*, 2004).

Stern *et al*. (2007) implemented and evaluated similar biocovers in Florida on top of a 40-100 cm interim cover, with 10-15 cm of crushed glass and 50 cm of yard waste compost. Three biocovers were investigated with each of them having and area of 7.6 x 7.6 m. This study also quantified CH_4 oxidation with the stable isotopic approach. They found that the actual CH_4 oxidation rate of 2 g m^{-2} d^{-1} was similar for the biocover and the soil cover, but a lower load to the biocover resulted in higher CH_4 oxidation efficiency for the biocover than for the soil cover (41 % compared to 14 %) (Stern *et al.*, 2007).

Huber-Humer *et al.* (2008b) tested the performance in four different cells each with an area of 25 x 25 m in an Austrian field study, which was compared with a control cell to estimate the CH_4 load. They found that the cell with a 30 cm gas distribution overlain by 90 cm of sewage sludge compost mixed with wood chips performed the best compared to the reference cell. 100 % oxidation was observed, which compared to the reference cell corresponded to a CH_4 oxidation rate of 39.5 g m^{-2} d^{-1} (Huber-Humer *et al.*, 2009). On another landfill, different biocover designs were tested and it was found, that a gas distribution layer of 0.5 m overlain with a mixture of sewage sludge and municipal solid waste gave very good results (Huber-Humer *et al.*, 2008b). Based on these results they recommended that a final compost cover should be at least 1.2 m for the climate in Middle Europe in the construction phase as they saw a 20 % compaction over 2 years of measuring (Huber-Humer *et al.*, 2008b). For landfills with high gas generation and strongly differing spatial gas fluxes, the construction of a 0.5 m gas distribution layer was recommended. Additionally they add that this should be a minimum value, as the upper part of the gas distribution layer will be clogged by finer particles from the overlying substrate, which infiltrates the upper parts of the gas distribution layer (Huber-Humer *et al.*, 2008b).

Dever *et al.* (2007) reported the implementation and set-up of a field scale biofiltration system in Australia, Sydney. The climate of the area is temperate and temperatures ranged from 18 °C – 27.9 °C (January) and 5 °C -17.1 °C (July). The biofilter was designed for a load of 576 g CH_4 m^{-2} d^{-1}. The biofiltration system was constructed partly within the existing landfill capping layer to prevent that surface water runoff from adjacent areas of the landfill should enter the filter. The gas distribution layer was 0.5 m and the compost layer was 1.2 m. Four filter beds (each 3 x 3 m) were installed and filled with four different composts. Each biofilter was supplied passively with landfill gas from 2 pipes for landfill gas drainage, which were placed in the top of the surrounding landfill in a total area of approximately 50 x 50 m. The landfill gas was supplied through parallel pipes placed in the gas distribution layer (Dever *et al.*, 2007). Dever (2009) report the load and CH_4 oxidation rates for the four biofilters, which was calculated by the carbon mass balance. The loads were 168 g CH_4 m^{-2} d^{-1}, 288 g CH_4 m^{-2} d^{-1}, 542 g CH_4 m^{-2} d^{-1} and 161 g CH_4 m^{-2} d^{-1}, which for all biofilters were lower than the anticipated CH_4 load. These rather different loads resulted in CH_4 oxidation rates of 101 g CH_4 m^{-2} d^{-1}, 94 g CH_4 m^{-2} d^{-1}, 62 g CH_4 m^{-2} d^{-1} and 108 g CH_4 m^{-2} d^{-1}, which corresponded to CH_4 oxidation rates of 60 %, 32 %, 12 % and 67 % (Dever, 2009).

Philopoulos *et al.* (2008) reported results from three Canadian pilot scale biofilters integrated into a landfill cover and sitting on top of the waste. In one of the biofilters landfill gas was supplied through a 5 m deep gas well filled with gravel. The gas distribution layer was 0.8 m and consisted of tire shreds. The gas distribution layer had a larger area than the biofilter allowing landfill gas coming from other locations, than right under the biofilter itself. A biofilter medium depth of 1.5 m was chosen. Also in this study the load of CH_4 was unknown and the performance of the biofilters was evaluated with the carbon mass balance approach. This was done in eight monthly campaigns from October to May where atmospheric temperatures ranged from about 20 °C to -20 °C. The average CH_4 loads for the eight campaigns were 54 g m^{-2} d^{-1}, 1.2 g m^{-2} d^{-1} and 37 g m^{-2} d^{-1} respectively for the three biofilters (Philopoulos *et al.,* 2008). The highest CH_4 load was achieved for the biofilter placed on top of the landfill, where a 5 m deep gas well was installed to increase the load of landfill gas. The lowest load was found for the biofilter placed on top of the landfill without a gas well, whereas the medium load was achieved for a biofilter placed on the slope of the landfill body. The calculated CH_4 oxidation rates were 36 g m^{-2} d^{-1}, 0.4 g m^{-2} d^{-1} and 28 g m^{-2} d^{-1} respectively for the three biofilters, which corresponded to CH_4 oxidation rates of 68 %, 35 % and 60 % (Philopoulos *et al.*, 2008).

Jugnia et al. (2008) reported the initial results from another Canadian study, where three experimental plots (2.75 m x 9.75 m) were installed (Cabral *et al.*, 2010). One of the plots had an 80 cm thick layer of biocover material underlain by a 10 cm thick transitional layer consisting of 6.4 mm net gravel and a 20 cm thick gas distribution layer consisting of 12.7 mm net gravel (Jugnia *et al.*, 2008). The substrate layer was a mixture of sand and compost, composed of 5 volumes of compost (before sieving) and 1 volume of coarse sand. After mixing, the material was screened (<12 mm). This plot was placed directly on top of the waste so the load could not be measured nor controlled. They determined the CH_4 oxidation rates by the stable isotopic approach during the summer in June to September and reported results in both 10 cm and in surface chambers. Using the approach suggested by Chanton *et al.* (2008b) resulted in an average CH_4 oxidation rate of 252 g m^{-2} d^{-1} for the 6 campaigns during summer (June to September), which corresponded to an efficiency of 11 %, as the average load was 2212 g m^{-2} d^{-1} (Cabral *et al.,* 2010). From these results it seemed clear that the efficiency of this plot was diminished by the high load. Another plot, was filled with the same compost, but unsieved and mixed with gravel, with a depth of 40 cm overlain a gas distribution layer of 80 cm. For this plot, the load was supplied through pipes and could be controlled with a valve (Cabral *et al.,* 2010). The same approach as above was used for calculation of the CH_4 oxidation rate, which resulted in 335 g CH_4 m^{-2} d^{-1} for the one campaign, which corresponded to an efficiency of 72 % as the load was 408 g m^{-2} d^{-1}. From this one result it seemed that the efficiency of this filter was better, as the load could be controlled and therefore was lower (Cabral *et al.,* 2010).

Einola *et al.* (2009) implemented a full scale biocover system on a 3.9 ha closed Finnish landfill. The load to the biocover system was supplied through pipes and the load could be controlled by valves. The pipes made it possible to direct the gas through the water impermeable layer and leachate formation could be kept at a minimum. On top of the waste, gas channels to supply gas to the 14 gas wells were installed. From each of the gas wells two or four perforated gas distribution pipes were installed and placed in a 50 cm deep gas distribution layer consisting of gravel. The biocover material was 50 cm of peat and sewage sludge compost (Einola *et al.,* 2009). The Finnish full scale system was evaluated during campaigns in October, November, February and June with air temperatures of 13^{o}C, 2 ^{o}C, -7 ^{o}C and 25 ^{o}C respectively. The evaluation was done by flux chambers and a carbon mass balance. This resulted in CH_4 oxidation rates of 2.1-

23 g m^{-2} d^{-1} with an average load of 2.9-27.3 g m^{-2} d^{-1}. Despite the quite high level of engineering and cost associated with this project, the gas was not distributed evenly over the landfill and they found a correlation between emission of CH_4 and the distance to the nearest gas pipe (Einola *et al.,* 2009). They found hotspots, which were very significant for the mean emission and CH_4 oxidation and 1 to 3 points accounted for 77-98 % of the CH_4 emission (Einola *et al.* 2009). It was found that the load could be successfully controlled by the valves, though some additional measurements would be needed in order to find the optimal valve setting. A thicker CH_4 oxidation layer was also suggested by the authors as a possible improvement, especially during low temperatures (Einola *et al.*, 2009).

Generally passive biocover systems should be aimed at, as they do not need operation and maintenance and therefore they are more advantageous compared to more complex and engineered systems. Landfills will keep emitting CH_4 long after their closure and biocover system should preferably work for decades without maintenance. Additionally passive systems are cheaper and they can more successfully be adopted in developing countries. The main aim of the Fakse biocover project was to install and prove the efficiency of such a full-scale, passive and low maintenance system.

1.5 Fakse Biocover Project

In September 2005 a 3 year long project with the aim of implementing a full scale biocover system at Fakse Landfill, Denmark was initiated. An overview of the methodology can be seen in Fig. 4. The project consisted of 4 major tasks, which were 1) Baseline study on the landfill and the emission of CH_4, 2) Testing of available materials for the biocovers, 3) Construction of the biocover system and 4) Evaluation of CH_4 oxidation performance (Fig. 3). The baseline study (1) and the Construction of the biocover windows (3) were not a part of this thesis and will therefore be described beneath. The remaining of the tasks will be described in the thesis.

1.5.1 Baseline study at Fakse Landfill

The baseline study consisted of four sub tasks; 1) Initial characterization of the landfill, waste characterization and evaluation of previous LFG studies on the site, 2) Calculating LFG production using models, 3) Mapping CH_4 emission at the site and 4) Measurement of the total CH_4 emission (Fig. 3).

Fakse Landfill is located in South-eastern Zealand, Denmark, Fig 3. The landfill is divided into two sections. Section I, which is the oldest section, was mainly active from 1981 until 1997. After 1997 disposal at Fakse Landfill has taken place at section II. Section I is the part of the landfill were the biocover system was installed. A total of approximately 660.000 ton of waste had been disposed of in Section I in the period 1981-2005 and the total area of the section is 103.000 m^2. The dominating waste types are soil fill (26 %), house hold refuse (23 %) and mixed waste (21 %). Fig. 5 shows a map of the units in Section I and the placement of the implemented biocovers. The disposal unit was filled one at a time. Disposal began in unit 1 in 1981 and continued in unit 2 in 1984. The order of disposal corresponds to the numbers given to the units and the last unit in use was unit 7 (1993-1997) (Fredenslund *et al.*, 2010b).

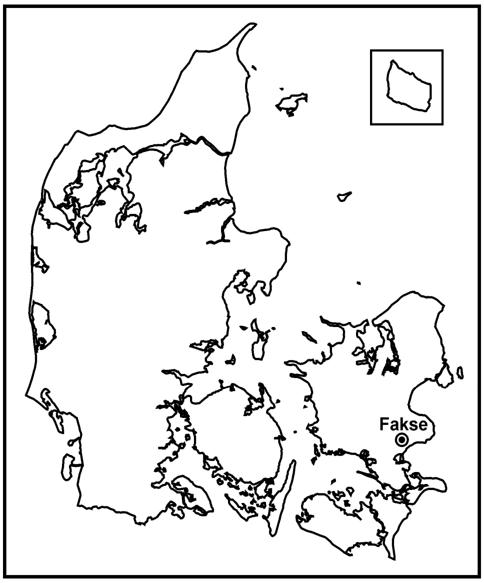

Fig. 3. Map of Denmark showing the location of the study landfill, Fakse Landfill

A natural layer of clay (15-20 m depth) functions as a bottom liner beneath the landfill. A drainage layer consisting of 0.3 m of gravel overlies the bottom liner. Leachate drainage pipes were placed in trenches of coarse gravel placed 15-20 m apart. In each of the disposal units the leachate collection system was connected to an inspection well and a collection well. Two different Danish consulting companies investigated the site in 2003 and 2004. The composition of landfill gas in samples from 10 installed gas wells was investigated. In 2003 the concentration of CH_4 and CO_2 were 61 % v/v and 36 % v/v respectively ($CH_4/CO_2=1.69$) and for 2004 the numbers were 61 % v/v and 28 % v/v respectively ($CH_4/CO_2=2.18$) (Lemming and Kjeldsen, 2006).

The CH_4 production, at the site, was modeled with four different models (Fredenslund *et al.,* 2010). The production varied from 555 kg CH_4 d^{-1} using the Avfalzorg Multi Phase Model, to 1689 kg CH_4 d^{-1} using the LandGEM model (Fredenslund et al., 2010). The production of CH_4 from unit 1 was 44 to 176 kg CH_4 d^{-1}, which corresponded to 8-10 % of the total modeled production

(Lemming & Kjeldsen, 2006). The total CH_4 emission from Fakse landfill was measured using a tracer technique in two campaigns and was 748 kg CH_4 d^{-1} and 732 kg CH_4 d^{-1} (Fredenslund *et al.*, 2010b; Scheutz *et al.*, 2010)

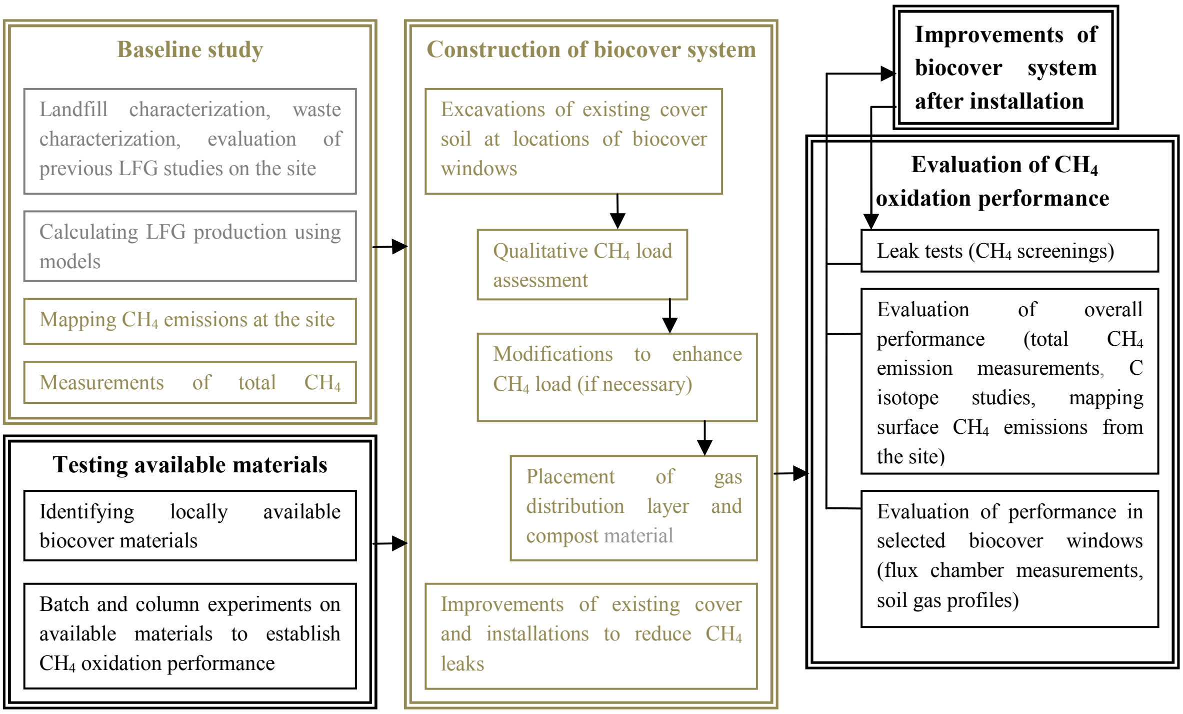

Fig. 4. Overview of the methodology used for designing, implementing and monitoring the biocover system at Fakse landfill. Actions, which are outside the focus of the study presented in this thesis, are marked in grey in this figure.

Near surface emission screenings with a portable FID showed that emission through the soil cover occurred mainly through relative small hotspots located on slopes of the temporary soil cover. Flux rates measured at these hotspots ranged from 5 to 4000 g CH_4 m^{-2} d^{-1} and the total emission of CH_4 from hotspots was calculated by determining the areas of the hotspot with the FID and multiplying by the average flux measured in the specific hotspot. This procedure was repeated four times and the total emission ranged between 73 kg CH_4 d^{-1} during increasing pressure and 336 kg CH_4 d^{-1} during decreasing pressure. The results showed that the CH_4 emission from the hotspots at Fakse Landfill was very influenced by the change in atmospheric pressure. The average CH_4 emission through hotspots was 182 ± 99 kg CH_4 d^{-1} (Fredenslund *et al.* 2010b).

A continuous tracer release method was used to assess the LFG emission through the leachate collection system (Fredenslund *et al.*, 2010a). The principle of this technique is to continuously release a gaseous tracer at a known rate near the source of the CH_4 emission, and subsequently measure the concentration of tracer and CH_4 downwind (Fredenslund *et al.* 2010a). Sixteen wells were measured and the emission ranged from 0 to 76 kg CH_4 d^{-1} for the individual wells. The sum of the emission was 351 kg CH_4 d^{-1} and therefore it was concluded that the leachate collection system was a very important route of fugitive CH_4 emission at Fakse Landfill (Fredenslund *et al.*, 2010a; Fredenslund *et al.*, 2010b).

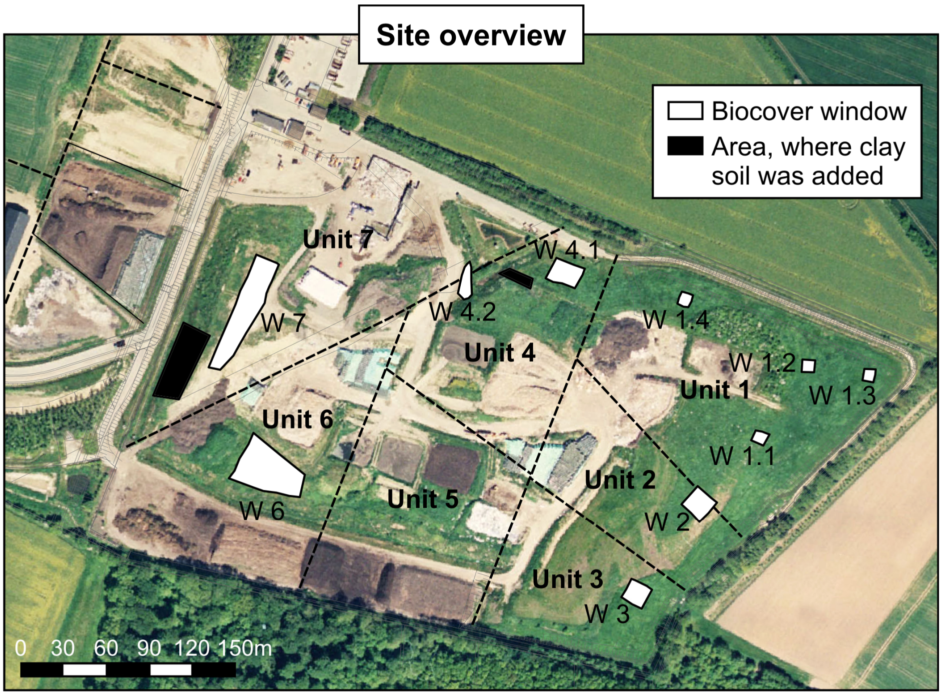

Fig. 5. Map of Fakse Landfill, with the units and the biocover windows (Fredenslund *et al.,* II)

The landfill gas composition was investigated in 10 gas probes in 4 campaigns during summer and early autumn. The CH_4 concentration ranged from 63.5±2.9 % v/v to 41.5±9.4 % v/v and the CH_4/CO_2 ratio ranged from 1.3 to 1.9. The average CH_4 concentration was 54±4.5 % v/v and 35±2.6 % v/v. This corresponded to an average CH_4/CO_2 ratio of 1.54. Differential pressure of up to 19 mbar was measured in unit 1 (Fredenslund *et al.,* 2010b).

1.5.2 Construction of the biocover system

The biocover system at Fakse Landfill was constructed as described in Fredenslund et al. (2010, II). The low permeable clay cover was removed from an area of 5,000 m^2 corresponding to 5 % of the total area of the landfill and these “windows” in the cover were filled with a gas distribution layer (15 cm) and a compost layer (100 cm). This approach was found suitable as rather low emission of CH_4 was expected from the landfill with a low content of organic matter. Additionally the impermeable clay layer would stay intact on 95 % of the surface area of the landfill, which would keep the leachate formation at a minimum.

The construction phase included four steps 1) Excavation of the existing cover soil, 2) Qualitative CH_4 load assessment, 3) Modifications to enhance CH_4 load 4) Placement of gas distribution layer and compost material. Additionally improvements of the existing cover and the installations to reduce CH_4 leaks were done (Fig. 4). The biocover system consisted of 10 biocover windows and their situation was based on the modeling of the production of CH_4 in each unit, done in the baseline study (Fig. 5). Hereby each unit was supplied with a biocover window area resulting in an average load of 150 g CH_4 m^{-2} d^{-1} to the biocover windows (Fredenslund *et al.,* II). The biocover windows were constructed from May 2007 to August 2007. During construction concerns about the permeability of the deposited waste beneath the biocover windows 2, 6 and 7 caused further measures to be taken (Fig. 5). Beneath window 2 a 4 m deep clay layer was situated and 16 holes were drilled and filled with gravel to secure the load to the biocover window. In window 6 and 7, the waste beneath the window was mixed with clay and three; 4 m deep trenches were dug in each biocover window and filled with tree roots, to improve the load to the windows.

It was found in the baseline study that the leachate collection system was an important emission route at Fakse Landfill and therefore 13 of the leachate collection wells were sealed with removable PVC caps as described in Fredenslund *et al.* (II).

1.5.3 Biocover window 1.1

A detailed study (Pedersen *et al.,* III, IV) was performed on biowindow 1.1 (Fig. 5). Biowindow 1.1 was placed on the final cover portion of the landfill, had an area of 85 m^2 (9 x 9.5 m), and sloped downwards at a grade of 13 % towards the landfill rim (south east) (Fig. 6). Preliminary investigation revealed that a large CH_4 emission hotspot was present in the upper part of the biowindow, and the hotspot appeared to be caused by poor landfill gas flow due to the occurrence of 3 to 4 meters of clay mixed with waste underneath the biowindow. In February 2008 improvements were incorporated into the biowindow design in order to increase the flux of CH_4 to the biowindow. A 1.5 m wide trench was dug and filled with tree roots to support gas collection and venting immediately underneath the biowindow (Fredenslund *et al.,* 2010b). A monitoring transect consisting of seven monitoring points (E, F, F½, G, H, I and J) was placed on top of the footprint of the back-filled trench (see Fig 6).

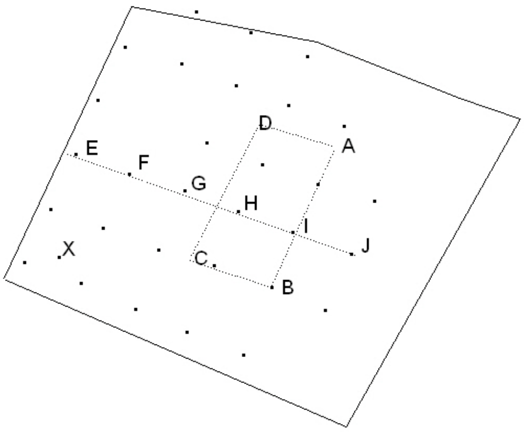

Fig. 6. Monitoring network in biowindow 1.1: original measuring points - A, B, C and D (rectangle) (Fredenslund *et al.,* II); and transect (line) - E, F, G, H, I, J. Distance between grid points was 1.5 m.

1.6 Aim of the PhD project

The overall aim of the inherent thesis was to assess the factors influencing CH_4 emission and oxidation in the field and evaluate how CH_4 oxidation can be quantified in passive systems.

The study was divided into the following partial aims:

- Determine which compost should be used for the full scale biocover at Fakse landfill. Evaluate existing criteria's for choosing composts for biocovers and to suggest a procedure for choosing materials for future biocovers.

- To assess the factors influencing the CH_4 oxidation in a full scale biocover window in particular the gas transport and how variation in load affects the efficiency of biocover systems.

- Quantify CH_4 oxidation in a passive vented field scale biocover window and evaluate methods for determination of CH_4 oxidation rates in passive systems.

- Assess the seasonal changes in compost biocovers in a temperate climate.

2 Design of biocover systems

2.1 Material specific factors influencing CH_4 oxidation.

CH_4 oxidation in biocovers is affected by several environmental variables, such as temperature and precipitation, but also the properties of the biocover materials are very important. One major factor is the physical properties, which insures that both landfill gas and O_2 can enter the biocover material; the landfill gas mainly by advective flux and the O_2 mainly by diffusion. Additionally chemical and biological factors, such as the content of nutrients and the respiratory activity of compost materials also has an impact on the CH_4 oxidation performance of possible materials (Scheutz *et al.,* 2009a; Huber-Humer *et al.,* 2009).

2.1.1 Physical parameters affecting CH_4 oxidation

The physical properties of the biocover materials, which affect CH_4 oxidation is the porosity, gas permeability, diffusivity and the particle size distribution of the biocover material. Also the water holding capacity can influence CH_4 oxidation, as it indicates the most frequently achieved gas filled porosity in the field. From knowing the gas filled porosity and the total porosity the diffusivity of the material can be estimated by different approaches. This will indicate which material is more suited for supporting the diffusion of O_2 into the CH_4 oxidation zone. This approach is believed to be adequate if a ranking of different materials are wanted and measurement of diffusivity will be too time consuming. Gas permeability gives information on the materials ability to support advective flux (see section 3.1 Gas transport). The transport of the landfill gas to the CH_4 oxidation zone is mainly advective (Kjeldsen, 1996) and therefore this is an important parameter. A material with homogeneous gas permeability will minimize the formation of hotspots and minimize the risk of the landfill gas escaping e.g. as lateral gas transport in unlined landfills (Christophersen and Kjeldsen, 2001) and through installations such as leachate collection wells in newer landfills (Fredenslund *et al.,* 2010a,b).

The determination of these parameters is generally more difficult for coarse compost materials than for soils, as coarse compost samples can include big branches and twigs, if they are not screened prior to sampling. Generally characterization methods for soils are based on small sample sizes and the sample equipment is therefore often not possible to directly adopt for coarse compost samples. Sieving or crushing the sample is not an option, because it alters the physical composition of the sample.

A different issue when working with the physical characterization of organic materials is that they may degrade over time and need replacement as the depth of the biocover will decrease and the porosity may decrease as well. Philopoulous *et al.* (2009) investigated this and found that the bulk density of a yard waste compost increased from 600 to 1000 g L^{-1} in the bottom of the profile after a 218 days long column experiment. This together with increasing water content (from CH_4 oxidation) resulted in lower gas filled porosity. They also tested a mixture with 96 % mineral materials (sand and perlite), which had a more constant bulk density and porosity. Thus both materials oxidized 100 % of the load corresponding to 134 g CH_4 m^{-2} d^{-1} (Philopoulos *et al.,* 2009). These results show that increased bulk density and decreasing porosity over time can be avoided by using more mineral mixtures.

Another important physical property is the thermal conductivity of the materials. The thermal conductivity is the capacity of a material to conduct heat and this will affect the temperature in the biocover materials compared to that of the atmosphere (Chandrakanthi *et al.*, 2005). The temperature in the CH_4 oxidation zone is of importance as Q_{10} values of CH_4 oxidation reported in the literature range from 1.7 to 4.1 from 10 ^oC to 30 ^oC (Scheutz et al., 2009a). The thermal conductivity of compost is larger than for mineral materials, because of the higher porosity and this will provide a good insulation effect (Huber-Humer et al., 2009). Additionally the specific surface area of materials has importance, as the methanotrophs live adhered to the surface. Compost has a high specific surface, which allows a high density population of methanotrophs (Huber-Humer et al., 2009; Scheutz et al., 2009).

Several studies have reported the formation of exopolymeric substances (EPS), in both soil and compost columns in the laboratory. Gas concentration profiles indicated a decrease in diffusivity due to the formation of EPS (Hilger *et al.*, 2000; Wilshusen *et al.*, 2004; Scheutz *et al.,* V). Hilger et al. (2000) investigated EPS formation in column incubation in the laboratory and found that the flux of landfill gas was not hindered in, but a model indicated that diffusion of O_2 was limited by the formation of the EPS (Hilger *et al.,* 2000).

2.1.2 Chemical and biological parameters affecting CH_4 oxidation

Nutrients are of great importance for CH_4 oxidation and especially the effects of nitrogen and ammonia is investigated in many studies and contradictory results exist in the literature (Bodelier and Laanbroek, 2004). Ammonium is a competing inhibition ion as it can be oxidized instead of CH_4. This process forms nitrite and hydroxylamine, which can be toxic to methanotrophs and lead to further inhibition (Bodelier and Laanbroek, 2004). Thus in landfill soils and compost covers it is very likely that nitrogen is a limiting nutrient as 1 mole of nitrogen should be used, when assimilating 4 moles of carbon (Anthony, 1982). The amount of carbon is very high in landfill environments compared to the available nitrogen pool (De Visscher and Van Cleemput, 2003) and therefore the addition of ammonium in very CH_4 rich environments can stimulate CH_4 oxidation more than it limits it. Some type II methanotrophs are able to fixate nitrogen (Hanson and Hanson, 1996), which will make them independent of a nitrogen source, thus fixation of nitrogen from the air does use more energy, than taking up ammonium and nitrate from the water phase (Bodelier and Laanbrook, 2004). Type I methanotrophs are not able to fixate nitrogen from the atmosphere, but it is believed that they respond to nitrogen limiting conditions by producing EPS, which has a higher C/N ratio than bacterial cell tissue (Hilger *et al.* 2000; Wilshusen *et al.* 2004).

Especially for compost materials the stability and maturity of the materials should be considered (Huber-Humer *et al.*, 2008b; Scheutz *et al.*, 2009a; Einola *et al.*, 2008a); Pedersen *et al.,* I). If the degradation of the compost consumes large amounts of O_2 this will compete with the O2 needed for the CH_4 oxidation process (Pedersen et al., I). Immature and unstable compost will also be more likely to produce CH_4 under anaerobic conditions (Barlaz et al., 2004; Scheutz et al., IV; Pedersen *et al.,* I). On the other hand compost materials can get too old and inactive. Pedersen *et al.* (I) found that a eight year old compost of garden waste had a low potential for CH_4 oxidation (4.8 μg CH_4 g DM h^{-1}) compared to a four year old compost of garden waste (161 μg CH_4 g DM h^{-1}).

2.2 Determination of CH_4 oxidation rates in batch and column.

Table 1: CH_4 oxidation rates in batch experiments from the literature with empathize on compost materials

Reference	*Material*	*Pre-exposure*	*Max. rate*	*Start konc.*	*Tem-pera-ture*	*Porosity/ grain size*	*Org. Matter*	*Water con-tent*
			$\mu g\ g_{dw}^{-1}\ h^{-1}$	% v/v	°C		%DM	%DM
Figueroa, (1993)	Mould	1-2week lab[e]	32	10	20		4.4[c]	9-18
	Humic soil	1-2week lab	86	10	20		7.1[c]	17-33
	Compost of org. Matter	1-2week lab	128	10	20		32[c]	68-135
Kightley et al. (1995)	Coarse sand	6month, lab[e]	26	5	20			
	Coarse sand with SSC[m]	6month lab[e]	19					
Borjesson *et al.* (1998)	Mineral soil, mostly sand	1yr. field[f]	9	0.6		0.35	3.2	
	Old SSC[m] (3-4yrs.)	3-4yrs. field[f]	173	5		0.69 15%c.s.[b] 12%f.s.[b]	25	28
	Fresh sewage sludge	1yr. field[f]	5	0.6		0.79	38	
De Visscher *et al.* (1999)	Agricultural soil	Yes	V_{max}= 26	3	22			15
Streese and Stegmann, (2003)	Fine GWC[g, j]	Yes	143[d]	2.5[a]	20-30	Fine	28.2	48.9
	Mixture of fine GWC, peat and wood fibers[h]	Yes	85[d]	2.5[a]	20-30	Coarse	52.1	
Wilshusen *et al.* (2004)	Leaf compost with zoo manure	6month lab	V_{max}= 480	10	22	Fine	46	123
	Compost of woodchips	6month lab	V_{max}= 124	10	22	Coarse	34	123
	Compost of MSW[k]	6month lab	V_{max}= 249	10	22	Coarse	49	123
Mor *et al.* (2006)	GWC[j], pass. aer, 6months.[i]	20days, lab	47	5	22	?	52.1	58.5
	GWC, int. aer., 3months[i]	20days, lab	105	5	22	?	31.1	49.5
Einola *et al.* (2008a)	MBT residual 22	no	28	9	25	0.49	47.4	79.1
	MBT residual 22	124d column	581	9	25	0.55	37.1	87.4
	MBT residual 57	no	20	9	25	0.47	38.9	104
	MBT residual 57	124d column	273	9	25	0.46	38.2	111
Ait-Benichou *et al.* (2009)	Fine MSW, SSC[m] and sand (5:1)	Yes field, summer	441	12.5	25	<12mm	17.8	70%[n]
		Yes field, winter	56	12.5	25	<12mm	17.8	88%[n]

[a] Constant inlet concentration in biofilter [b] Short for coarse sand and fine sand. [c] Calculated assuming that ignition loss is two times TOC (Total Organic Carbon). [d] Calculated assuming CH4 oxidation in the entire volume of the filter [e] Pre-exposed in the lab [f] Pre-exposed in the field [g] TKN=7.75g/kg, $[NH_4^+]$ =732mg/kg, $[NO_3]$=200mg/kg. [h] TKN=7.37g/kg, $[NH_4^+]$ =881mg/kg, $[NO_3]$=166mg/kg. [i] Passively and intensively aerated under production for 6 and 3 months respectively. [j] GWC: Garden waste compost. [k] MSW: Municipal solid waste, [l] MBT: Mechanically biologically treated MSW. [m] SSC Sewage Sludge Compost. [n] Degree of water saturation

The materials available for biocovers can be tested in CH_4 oxidation test to rank available materials based on their CH_4 oxidation potential. Many references are available on CH_4 oxidation rates both in batch and column studies for a large range of materials including both mineral and organic materials e.g. (Borjesson *et al.*, 1998; Wilshusen *et al.*, 2004; Mor *et al.*, 2006; Kettunen *et al.*, 2006; De Visscher *et al.*, 1999; Scheutz *et al.*, 2009b). And Scheutz *et al.* (2009a) and Chanton *et al.* (2009b) will provide good reviews.

Table 1 lists CH_4 oxidation rates in batch incubations, with focus on compost materials as these are important for the inherent study. The CH_4 oxidation rates reported will depend on pre-exposure to CH_4, CH_4 concentration, temperature and water content, though all experiments were done around 20-25°C. In Table 1 the lowest CH_4 oxidation rate reported for a compost was 5 $\mu g\ CH_4\ g\ DM^{-1}\ h^{-1}$ for a fresh sewage sludge compost after 1 year of pre-exposure in the field, but with a low start concentration (0.6 % v/v) (Borjesson *et al.*, 1998). Due to the low start concentration of CH_4, zero order kinetics cannot be expected. The second lowest CH_4 oxidation rate was 19 $\mu g\ CH_4\ g\ DM^{-1}\ h^{-1}$ and was found for a coarse sand added with sewage sludge compost (Kightley *et al.,* 1995). The highest CH_4 oxidation rate was 581 $\mu g\ CH_4\ g_{dw}^{-1}\ h^{-1}$ for mechanical biological treatment (MBT) residual after 128 days in a column experiment (Einola *et al.,* 2008a).

In Scheutz *et al.* (2009a) CH_4 oxidation rates in column experiments simulating landfill bio-covers rich in organic matter are given. Results from 11 different studies and 28 different materials are listed. The CH_4 oxidation rate in column experiments depends very much on the load of CH_4 to the column and therefore the load is mentioned here together with the results. The minimum steady state CH_4 oxidation rate reported in Scheutz *et al.* (2009a) was -31 $g\ CH_4\ m^{-2}\ d^{-1}$ for a mixture of 50 % screening residue and 50 % unsorted sand with a load of 229-254 $g\ CH_4\ m^{-2}\ d^{-1}$ (50 % v/v CH_4). This result indicated that CH_4 was produced in the column even though the material consisted of only 50 % organic material (Scheutz *et al.,* V). However, the screening residue, which was used in the mixture has higher respiratory activity and methanogenic potential than both the raw compost and the fine part of the compost (Pedersen et al., I). Therefore screening residues should not be mixed with finer materials such as sand as this will reduce the porosity of the material an increase the risk of a deep anaerobic zone, where CH_4 could be produced. The highest steady state CH_4 oxidation rate reported was 400 g CH_4 m^{-2} d^{-1} for a 60 weeks old municipal solid waste (MSW) compost with a load of 400 g CH_4 m^{-2} d^{-1} (100 % v/v) (Humer and Lechner, 2001). The second highest steady state was found by Powelson *et al.* (2006) and was 242 g CH_4 m^{-2} d^{-1} with a load of 250 to 500 g CH_4 m^{-2} d^{-1} (50 % v/v) for a mix of compost and polystyrene pellets (Powelson *et al.*, 2006).

The optimum solution, when testing possible materials for biocovers is column incubations as they do mimic the situation in the field the most. However, in future situations for full scale application, this will most likely not be an option as it is both time-consuming and costly. In the situations where the entire landfill is not covered with biocover material (biocover windows and biofilters), realistic CH_4 oxidation rates are necessary in order to dimension the biocover system (Pedersen *et al.* I). Different approaches for choosing materials for biocovers are discussed in the next section.

2.3 Choice of materials for biocovers

Different approaches can be used when deciding, which material should be used in a field-scale or full-scale passive biocover or in a passive biofilter. In the following the approaches used in the literature is attempted divided into four different approaches.

1) Choose the in theory optimum material with the optimum physical properties; high porosity, high specific surface for bacteria to adhere to and less attention is given to whether the material is available on site and feasible to use (Gebert and Groengroeft, 2006a)
2) Rank locally available materials by criteria given in the literature (Huber-Humer *et al.*, 2009)
3) Rank locally available materials by testing CH_4 oxidation rates (Pedersen *et al.,* I)
4) Use a material, which proved high capacity for CH_4 oxidation in previous studies, with a minimum of testing prior to implementation (Dever *et al.*, 2007).

Different approaches have been applied in the literature, but to date many studies have included testing of the potential CH_4 oxidation capacity in column incubations prior to selection of the optimum material for full-scale application. As the diversity and range of materials tested in both column incubations and in the field increase the fourth option 4) becomes more and more reasonable. However the production of compost is heterogeneous and standards for compost quality varies between countries and even within countries so a minimum of testing will always be necessary.

As described in the previous section, several column studies with a high range of materials and mixtures of materials have been performed, but few authors have dared to give specific guidelines for the optimal cover material for biocovers. Humer and Lechner (1999) have provided a guideline based on the testing of 25 different materials in small column incubations. They found that the optimum materials should fulfill the following criteria:

1) “matured compost with solid organic matter and low ammonium and salt concentrations”.
2) “long-term stability and an adequate pore volume even with high water content”. “
3) “provide microorganisms with a high and easily available nutrient supply to support microbial activity”.

Humer and Lechner (2001) gave the following more specific requirements: TOC > 8 % DM, C/N ratio of about 15, respiratory activity in 7 days <8 mg O_2 g_{dw}, NH_4-N < 350 ppm DM, sufficient content of bulky material and a WHC of < 80 % of wet matter. Huber-Humer *et al.* (2009) proposed a range of physical and chemical parameters, which should be investigated, when materials for biocovers are selected. Table 2 shows this table, which is based on column experiments with 41 different waste compost materials (Huber-Humer *et al.,* 2009). The authors described the process of selection of material and the pre-check of oxidation capacity. Generally Huber-Humer obtained good CH_4 oxidation capacity in both municipal solid waste compost and sewage sludge compost, mixed with woodchips, which were done in order to improve the gas permeability of the materials. They found that coarse materials would ensure that O_2 could diffuse deep into the cover and that the CH_4 oxidation thereby would be situated deeper in the profile, where the moisture conditions were more favorable and steady than in the upper layers (Huber-Humer *et al.,* 2008).

Table 2: After Huber-Humer *et al.* (2009), see the original article for comments on each of the parameters. Recommended parameters for testing suitability of compost materials for biocover construction (Huber-Humer *et al.,* 2009)

Parameter	*Unit*	*Proposed value*
Physical properties		
Bulk density	kg/L	0.8-1.1
Moisture content	% w/w	30-50
Water holding capacity	% DM	50-130
Air filled pore volume	% v/v	>25
Particle size distribution		0.063-2mm:20-30;2-6.3mm:ca.40; 6.3-20mm:20-40; >20mm:ca 10
Chemical parameters		
Conductivity	mS/cm	<4
pH value	-	6.5-8.5
SO_4^{2-}	ppm DM	>500
NH_4^+-N	ppm DM	<400
NO_2^--N	ppm DM	<0.1
NO_3^--N	ppm DM	No limit value
P_{total}	% DM	>0.3
N_{total}	% DM	>0.5
Organic content	% DM	>15
TOC	% DM	>7
Maturity parameter		

Pedersen *et al.* (I) tested five different compost materials in a column incubation and compared the results to the criteria given in Huber-Huber *et al.* (2009), Table 2. The criteria were fulfilled for some of the properties of the composts, but not for all. Between two and five criteria were not fulfilled for each of the composts. Four year old raw compost of garden waste (RC4) fulfilled all criteria except two (ammonium and total phosphorous) and one year old raw compost of garden waste (RC1) did not fulfill five of the given criteria (ammonium, C/N, total phosphorous, sulphate and O_2 consumption). None of the tested composts fulfilled the criteria on ammonium and even compost with very high ammonium content (5000 mg kg_{dw}^{-1}) achieved a high average CH_4 oxidation rate (112 g CH_4 m^{-2} d^{-1}). The same compost, which was sewage sludge compost, did not fulfil the respiratory criterion given in Humer and Lechner (2001) and it was characterized as fresh, by the maturity classification given by (Christensen and Hansen, 1998). Generally it was concluded that criteria on ammonium should be given with care, as no agreement on the method applied was available. For the maturity classification it was found that a large range of classification systems existed and that very different results were obtained with the different approaches. Therefore it will vary almost from country to country, how a mature compost is defined (Pedersen *et al*., I).

Criteria:	**RC4**, FC, RC8, *SC*, SR1, SR3, RC1
Batch 1:	**RC4**, FC, *SC*, SR1, SR3, RC1, RC8
Batch 2:	*SC*, **RC4**, FC, SR3, RC1, SR, RC8
Batch 3:	**RC4**, *SC*, SR3, FC, RC1, SR, RC8
Column av.:	FC, *SC*, **RC4**, RC1, SR
Column d. 111	FC, *SC*, SR, **RC4**, RC1

Fig. 6. Ranking of the different materials according to the criteria in Huber-Humer *et al.* (2009), the three subsequent batch incubations, the average CH_4 oxidation rate and the CH_4 oxidation rate on Day 111 (Pedersen *et al.,* I).

Fig. 6 shows the ranking of the tested materials in Pedersen *et al.* (I) according to the number of criteria, which were fulfilled, the results of three subsequent batch incubations, the average CH_4 oxidation rate in the column experiment and the CH_4 oxidation rate after 111 days of column incubation. If only the criteria in Huber-Humer *et al.* (2009) was used for the ranking of the available materials, the RC8, which was an eight year old raw compost would receive a false positive result, as it achieved low CH_4 oxidation rates in the batch incubations (3.2 to 7.0 $\mu g\ CH_4\ g_{dw}^{-1}\ h^{-1}$), compared to for example the RC4, which oxidized 53 to 161 $\mu g\ CH_4\ g_{dw}^{-1}\ h^{-1}$ in the batch incubation tests. This shows that the knowledge on the important properties of composts for biocovers are not yet broad enough to leave out testing potential CH_4 oxidation of compost materials prior to installing them in field-scale biocover systems.

In Pedersen *et al.* (I) it was chosen to use a 4 year old raw compost of garden waste as it had a satisfying average CH_4 oxidation rate (108 $g\ CH_4\ m^{-2}\ d^{-1}$) and it was available at the landfill in large amounts, without any extra expense for the landfill operator.

Dever *et al.* (2007) followed the guideline given by Humer & Lechner (2001) and tested composted garden waste and municipal solid waste. They added shredded wood to three of the four composts tested in a field-scale set based on the recommendations in Humer & Lechner (2001) and Streese & Stegmann

(2003). They added shredded wood in order to ensure that the filter material was coarse enough for the landfill gas to enter the filter and the O_2 to diffuse into the filter bed and to avoid a decline in the CH_4 oxidation rates over time (Dever *et al.,* 2007). Seven composts were tested and characterized by measuring the parameters; bulk density, porosity, water holding capacity and gas conductivity. Additionally the carbon and the nitrogen content were measured and the C/N ratio was calculated (Dever *et al.,* 2007). The C/N ratios were 20.9, 15.2 and 17.5 for the composted municipal solid waste, garden waste compost and screened garden waste compost, respectively. Based on the guidelines and characterization of seven compost mixtures Dever *et al.* (2007) tested four different composts in a field set-up; 1) Composted garden waste with 10 % shredded wood, 2) Composted municipal solid waste with 10 % shredded wood, 3) Composted garden waste and 4) Composted municipal solid waste with 20 % shredded wood. Jugnia *et al.* (2008) also tested 3 different mixtures of compost and mineral material (sand and gravel), but with different design of the biofilters and did not give any background information on why the specific mixtures were chosen (Jugnia *et al.,* 2008).

Einola *et al.* (2009) chose a mixture of a peat and sludge compost 40:60 (v/v). The mixture was chosen as it gave balanced characteristics for promoting CH_4 oxidation. For example the sludge compost had high nutrient levels compared to peat, which in turn had high organic content, low density and high water holding capacity, which indicated high porosity (Einola *et al.,* 2009). Additionally they reported that the materials were tested in a column experiment and was capable of oxidizing 72 g CH_4 m^{-2} d^{-1} at 5°C, which was high enough to oxidize the expected load of CH_4 from the landfill (Einola *et al.,* 2009). The respiratory activity of the two materials were 2.2 µg O_2 g DM h^{-1} and 16.7 µg O_2 g DM h^{-1} respectively, which were low compared to the criteria in Huber-Humer et al. (2009) (<48 µg O_2 g DM h^{-1}). It is also low compared to other values of O_2 demand reported in the literature for example values found by Pedersen et al. (I). Only a four and eight year old raw composts had such low O_2 demands (3.8 and 6.1 µg O_2 g DM h^{-1}) whereas the tested sewage sludge compost had an O_2 demand of 122 µg O_2 g DM h^{-1} (Pedersen *et al.,* I).

The production of compost is very diverse including the addition of different amounts of screening residues, straw and different time schemes for turning piles and controlling the process and even on a national level different procedures can be used (Pedersen *et al.,* I). Furthermore a very detailed classification and describing system exist for soil, but describing compost is not so straight forward. Therefore it would still be advised to test the CH_4 oxidation capacity of different available composts prior choosing the material for a full-scale biocover. Additionally local waste products e.g. MBT residuals (Einola *et al.*, 2008a) should be tested for potential use in biocovers and despite the fact that a large range of materials have been tested in the laboratory there will be a great number of waste products, which are not.

2.4 Mineral materials versus compost materials

The use of compost materials for biocovers has disadvantages. 1) The O_2 demand of the compost material itself can compete with the O_2 demand of the CH_4 oxidation (Huber-Humer *et al.,* 2009; Scheutz *et al.,* 2009a; Einola *et al.,* 2008a). 2) There is also a potential risk of CH_4 production if high loads of landfill gas or water saturation provide anaerobic conditions in the bottom of biocovers (Barlaz *et al.,* 2004; Scheutz *et al.,* IV). 3) Additionally the degradation of the compost and the limited structural stability can cause the need of exchanging the compost material or add compost to ensure that an optimum depth of the biocover material is in place at the landfill (Philopoulos *et al.,* 2009). These three issues can be avoided by working with more mineral materials. Going through the literature it seems that a very diverse range of materials (both mineral and more organic) can support growth of methanotrophs (Gebert and Groengroeft, 2006a; Scheutz and Kjeldsen, 2003; Einola *et al.*, 2008a; Wilshusen *et al.,* 2004; Kettunen *et al.,* 2006; Powelson *et al.,* 2006 Stein and Hettiaratchi, 2001) especially after a long period of pre exposure, as long as porosity and permeability are sufficiently high and no toxic compounds are present. Though for a biocover to function optimal right after installation it is believed that an addition of organic material (microorganisms) would be needed otherwise the lag phase will be unnecessarily long.

In Scheutz *et al.,* (IV) the performance of a material that was mainly mineral was compared with the CH_4 oxidation capacity of coarse compost. The coarse compost performed better over time and this was believed to be due to the material being so coarse that it did not clog by the formation of EPS (Fig. 7). On the other hand Gebert *et al.* (2006a,b) never identified EPS in a biocover made of clay pellets.

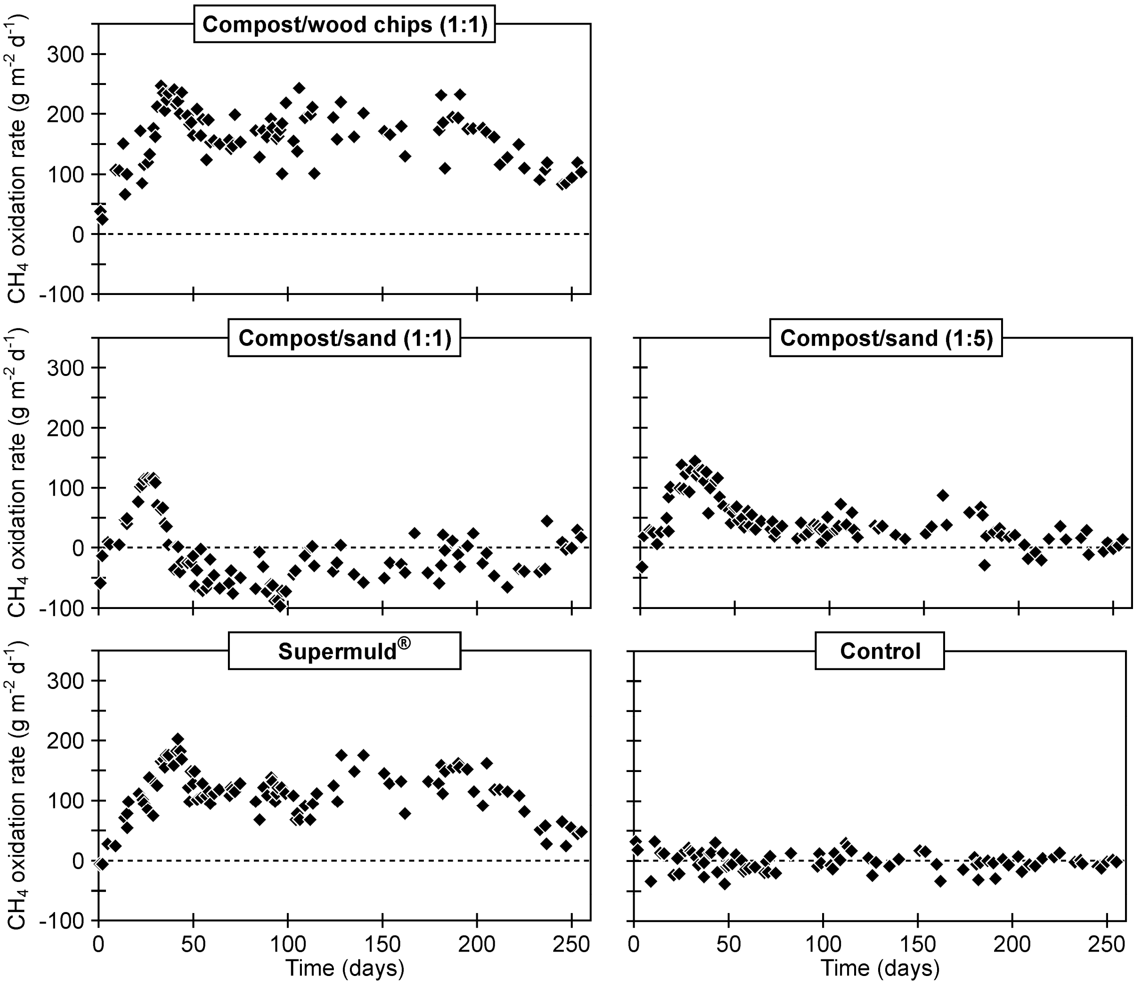

Fig. 7: CH_4 oxidation rates versus time in column incubations over 255 Days (Scheutz *et al.*, IV).

2.5 Locally available materials versus optimal materials

A large number of column incubations have been done in order to identify the optimum materials for CH_4 oxidation (Scheutz *et al.,* 2009a). Thus spending much effort and time on identifying the perfect material can be squandered, as locally available material should be used due to the economical and environmental costs associated with transporting large amounts of material to the landfill site (Pedersen *et al.,* I). Furthermore the national legislation and the intended use of the landfill area for e.g. recreational purposes should be taken into account when choosing materials. E.g. sewage sludge compost could be the ideal biocover material in Austria but Danish legislation only allows agricultural land use of such nutrient rich compost (Pedersen *et al.,* I).

In general the most advantageous solution is the use of locally available materials, though suited materials will not always be available. In the study by Pedersen *et al.* (I) no mineral material suited for use in the gas distribution layer and as structure material mixed with compost was identified at the site. In Pedersen *et al.* (I) both the economical and environmental cost associated with buying and transporting mineral materials to the landfill was assessed. It was found both economically and environmentally unsound and not within the budget frame of the project (Pedersen *et al.,* I). In Table 2 the cost of buying and transporting mineral materials to the landfill can be seen. Mixing with 50 % (v/v) sand of the poorest quality would result in an expense of 30.000 €.

Table 3. Material prices from the two nearest local gravel pits. Expenses calculated for two design of the biocover and the environmental cost associated with transport (Pedersen et al., (I)).

Inorganic materials	Size	Density	Price[a]	Price 0.2m, 1m	Price 0.5m, 1.2m	Environmental cost from transport	
	mm	kg m^{-3}	€ ton^{-1}	€	€	kg CO_2-eq[b]	eq. ton CH_4
Gas distribution (0.2 m or 0.5)							
Road gravel, partly crushed	0-16	1750	14.0	24,462	61,156	9975	249
Stable gravel (DS/EN 13285)	0-32	1750	13.3	23,286	58,216	9975	249
Gravel fill (DS/EN 13285)	0-32	1600	9.9	15,914	39,785	9120	228
Crushed stone (skærver)	32-50	1500	15.3	22,984	57,460	8550	214
Sand for biocover windows (1 m or 1.2 m)							
Sandfill Lundby (unsorted)		1600	7.5	60,215	72,258	16000	400
Washed sand 0/4	0-4	1600	11.7	93,548	112,258	45600	1140
Washed sand 0/2	0-2	1600	16.5	132,258	158,710	45600	1140
Washed sand 1/4	1 to 4	1500	20.6	154,234	185,081	42750	1069

[a] Prices are inclusive transport from Lundby gravel pit, Bårse (20 km) and Lønggårdens gravel pit, Sorø (57 km) [b] Calculated with 0.1 kg CO_2 eq. ton^{-1} km^{-1}. [c] CO_2:CH_4 of 25:1 acc. to Solomon et al. (2007).

For the gas distribution layer, only crushed tree roots were available at the site. This material is far from optimum, as it will degrade and the risk of CH_4 production is high, as the gas distribution layer must be expected to be anaerobic at the designed load (150 g CH_4 m^{-2} d^{-1}). Thus it was used, when improvements were done to the system, as filling the up to 4 m deep trenches made in the biocover windows with gravel would be very expensive. However if the landfill use and legislation do not allow for the biocover system to be dimensioned for low loads, the transport of more optimum materials to the landfill could be a plausible option.

2.6 Dimensioning biocovers

Locally available materials should be used when implementing full-scale biocovers according to section 2.5, but this can cause low (< 50 g CH_4 m^{-2} d^{-1}) CH_4 oxidation rates as there is no guarantee that materials which are optimum for CH4 oxidation will be present at the site. This is however not a problem if the biocover systems can be designed in a way that provides a load of CH_4 that can be oxidized in the available material. Therefore to be successful in using locally available materials it should be possible to design the biocovers with a low load (< 50 g CH_4 m^{-2} d^{-1}) and the intensity of hot spots should be minimized (Pedersen *et al.,* IV; Fredenslund *et al.,* II). If the layout of the landfill does not allow such conservative design other more optimum biocover materials could be considered and the design costs should be balanced towards the costs of transporting optimum materials to the landfill site. It was attempted to do a comparison of cost of the materials versus CH_4 oxidation potential, with data from the inherent study and available data in the literature, but the amount of data available was not sufficient.

In Pedersen *et al.* (I) the size of the full-scale biocover was calculated based on both the average CH_4 oxidation rates and the CH_4 oxidation rates on the last day of the experiment (Day 111) for the five materials tested in column incubations. The Day 111 CH_4 oxidation rate was used as it was believed to give an improved indication of the performance over longer time, than the average CH_4 oxidation rate. The size of the biocover ranged from 6,066 m^2 for the Day 111 CH_4 oxidation rate for the fine compost (122 g CH_4 m^{-2} d^{-1}) to 19,474 m^2 for the Day 111 CH_4 oxidation rate of the 1 year old raw compost (38 g CH_4 m^{-2} d^{-1}) (Pedersen *et al.,* I). For the average CH_4 oxidation rates the size ranged from 6167 m^2 when using the average CH_4 oxidation for the fine compost (120 g CH_4 m^{-2} d^{-1}) and to 16,444 m^2 when using the average CH4 oxidation rates for the screening residue (45 g CH_4 m^{-2} d^{-1}). All of the tested composts did oxidize CH_4 and due to this it could be argued that it does not matter, which compost was chosen, as long as the sizing and design of the biocover system ensured low and even distribution of the CH_4 load to the biocover system. Thus due to the difficulties in controlling formation of hotspots and temporal variation in the CH_4 load in reported field scale studies (Gebert and Groengroeft, 2006a,b; Einola *et al.,* 2009; Jugnia *et al.,* 2008; Cabral *et al.,* 2010; Dever, 2009) it could be argued that the compost with the highest CH_4 oxidation potential should be chosen. In this study it was found that the limitation in the CH_4 oxidation rates in the field were not caused by the chosen biocover material, but the spatial and temporal difference in the CH_4 load (Pedersen *et al.,* III, IV)

The biocover material chosen for the full scale biocover system was the four year old raw compost and it had a Day 111 CH_4 oxidation rate of 76 g CH_4 m^{-2} d^{-1}. This CH_4 oxidation rate resulted in a biocover area of 9,737 m^2, which was almost double of the actually implemented biocover area (Fredenslund *et al.,* II). It should be added that the column experiments were performed in the laboratory under close to optimum temperature and CH_4 load. However the full-scale biocover system was not generally overloaded, as there were problems with avoiding gas escaping from the leachate collection system. Some parts were heavily overloaded, but this was caused by unsuccessful gas distribution, which was identified as a bigger problem than the general overload (Pedersen *et al.,* IV). Based on this it was found that the actual design of the biocover system needed to be evaluated (Pedersen *et al.,* IV; Fredenslund *et al.,* II) and the discussion on possible improvements is presented in the next section.

2.7 Design of biocovers

Fredenslund *et al.* (II) reported the design of the biocover system at Fakse Landfill and showed that a substantial portion of the CH_4 emission escaped around the leachate collection wells despite that several initiatives where implemented in order to avoid this. Additionally both Fredenslund *et al.* (II) and Pedersen *et al.* (III, IV) showed that the spatial variation in emissions was high and it was concluded that this was due to insufficient gas distribution. In general it was found difficult to promote gas transport to the biocover windows, which was caused by the big portions of clay deposited in the landfill and on top of the waste. In general the design of the biocover system at Fakse Landfill was inadequate for a landfill with a leachate collection system and considerable amounts of clay deposited in the landfill and on top of it. Fredenslund *et al.* (II) suggested that for future biocover systems were emission of CH_4 are measured through the leachate collection system, the leachate system should be used to supply biofilters or bio windows with landfill gas. For example similar to the approach used by Einola *et al.* (2009). Fredenslund *et al.* (II) also suggested that the depth of the gas distribution layer should be increased to 30 cm as suggested by Huber-Humer *et al.* (2008). However, the use of pipes and a deeper gas distribution layer, does not guarantee that the spatial distribution of the load to biocover systems does not diminish overall efficiency. Einola *et al.* (2009) used pipes to supply landfill gas to their full-scale biocover system and they used a 50 cm drainage layer and still they found that the overall efficiency was diminished by the spatial variation in emission and few hot spots were responsible for a big portion of the emission. However the load of landfill gas was successfully controlled by valves (Einola *et al.,* 2009).

At Fakse Landfill the implementation of the biocover system was problematic, as deep clay layers (up to 4 m) were placed on top of the landfill and partly mixed with waste. Additionally it was reported by old landfill employees that it was normal to do daily coverings with clay and therefore it is likely that the waste id deposited in “clay pockets”. The clay layers on top of the landfill were partly dug out in some of the windows and trenches were formed. The entire area of the biocover windows were not dug out, as the dugout material should be deposited elsewhere on the landfill as waste, which resulted in significant costs for the landfill operator. The lost income for the landfill operator due to this and the expenses associated with digging, moving and re depositing the waste/soil would be 94 €/m^3 of waste/soil removed. This would result in an expense of more than 30,000 € for digging out the material from window 1.1 only (85 m^2 out of 5000 m^2) (Pedersen *et al.,* III). This was the main reason why only trenches were dug in order to improve the load to the biocover windows. It was difficult to assess whether the digging of the trenches had improved the load of CH_4 to the biocover windows, as the emissions were very dynamic (Pedersen *et al.,* III). Thus the results indicated this and it could be seen from the vegetation that hot spot areas were situated above the trenches. Additionally Philopoulos *et al.* (2008) found the highest load of CH_4 in a biocover filter with a gas well placed in the bottom of it. In general the first and most important issues when designing biocovers is to ensure that gas transport to the biocover system is promoted and for landfills with high amounts of clay deposited this will most likely require installation of gas wells or trenches filled with permeable material.

Huber-Humer *et al,* (2008) suggested a gas distribution layer of 0.5 m overlain by 1.2 m compost. In Table 2 the expenses associated with a 0.5 m gas distribution layer is listed. The minimum cost when using the cheapest gravel fill is almost 40,000 € and therefore the depth of the gas distribution is a cost sensitive parameter (Pedersen *et al.,* I). Due to this it is highly recommendable to investigate whether any reusable materials are available on site. Unfortunately no suitable materials were available at Fakse landfill. Ensuring an even load both over time (Pedersen *et al.,* III) and space (Pedersen *et al.,* IV) seems to be a key parameter, when designing biocover systems in the future. These two issues points at the necessity to do detailed studies on what minimum depth of gas distribution layer is needed in combination with different loads of landfill gas and CH_4 in order to achieve satisfying results and minimize costs.

From a conceptual point of view it is suggested that less permeable biocover materials compared to the gas distribution layer should be considered, as this should improve the gas distribution. A bigger "resistance" in the biocover material compared to the gas distribution layer should theoretically improve the distribution of the landfill gas in the gas distribution system prior to entering the biofilter material. Obviously this is a fine balance, as the load to the biocover systems should be ensured and the gas permeability should be higher than of alternative escape routes, such as lateral gas transport in unlined landfill (Christophersen & Kjeldsen, 2001) and escape through leachate collection systems (Fredenslund *et al.*, 2010a). Until now the focus in the research has been that materials should be very coarse and permeable (Huber-Humer *et al.,* 2008; Dever *et al.,* 2007; Gebert and Groengroeft, 2006a,b) to ensure the load of CH_4 to the biocovers and the O_2 diffusion into the biocover material, but this could also have drawbacks. Thus one study with screened composts (mixture of fine compost and sand) are reported in Canada (Jugnia *et al.,* 2008). In this study very high CH_4 oxidation rates were associated with high CH_4 loads (Cabral *et al.,* 2010).

3 Gas transport in biocover windows

3.1 Gas transport

The gas transport is important in biocover windows, as it controls the supply of O_2 and CH_4 to the CH_4 oxidation zone. The CH_4 is mainly controlled by advective flux and the O_2 by diffusion.

3.1.1 Advection

Landfill gas is produced in landfills, which results in a pressure increase in the landfill compared to the atmosphere. In this way an advective outward flux of CH_4 is induced. Previous studies have shown that the gas transport associated with landfills emission and migration mainly is due to advective flux (Kjeldsen, 1996)

Advective flux is driven by a pressure gradient and is described by Darcys law

$$J_A = -C \cdot \frac{k_a}{\mu} \cdot \frac{\delta p}{\delta x} \qquad \text{(Kjeldsen, 1996)}$$

J_A is the advective flux, C is the gas concentration of the constituent (g m^{-3}), k_a is the gas permeability of the soil, μ is the viscosity of the gas (N s m^{-2}) and δd/dx is the pressure gradient (Kjeldsen, 1996). In Kjeldsen, 1996 it is given that the gas permeability can be calculated as

$$k_a = k \cdot \frac{\varepsilon_a}{\varepsilon} \qquad \text{(Kjeldsen, 1996)}$$

where k is the hydraulic conductivity, ε is the porosity and the ε_a is the air filled porosity at the specific water content. The measurement and calculation of gas permeability will be discussed in further detail in section 3.5.

A pressure gradient over a landfill cover can be caused by a build up in the pressure inside the landfill as landfill gas is produced. The pressure gradient can also be caused by a decrease in atmospheric pressure, which will pull landfill gas out of the waste mass. Additionally Gebert & Groengroeft (2006b) stated two other reasons with generally shorter time scales. One is autooscillation of air, which is detected, at a resolution of less than 10 min and induces pressure variations less than 1 hPa. Another parameter is the warming and cooling of air caused by the daily alternation of the solarisation. When air is warmed up during the day is rises and results in a pressure decrease near the soil surface. Gebert & Groengroeft (2006b) found that this phenomena explained diurnal variations in the atmospheric pressure (Gebert & Groengroeft, 2006a). Recently the wind induced pressure differences has been investigated and was found to induce fluxes, when modeled as dispersion in a study on lateral gas transport adjacent to an old landfill in Denmark (Poulsen & Moldrup, 2006).

Advective flux can also have importance for the supply of O_2 as CH_4 oxidation removes gas molecules and thereby induces advective flux. This can be identified in gas concentration profiles by increased concentrations of nitrogen compared to the atmosphere (De Visscher et al., 1999; Christophersen et al., 2001; Molins et al., 2008) Additionally pressure increases has been reported to revert the gas flow, which will result in atmospheric air being transported into biofilters (Gebert & Groengroeft 2006a)

3.1.2 Diffusion

O_2 to the CH_4 oxidation zone is mainly supplied by diffusion. The diffusive flux in a porous medium is determined by the concentration gradient and in its simplest form it is described by Ficks law.

$$J_D = -\varepsilon_a \cdot D \frac{\delta C}{\delta x}$$

Where J_D is the diffusive flux (g m^{-2} d^{-1}), ε_a is the gas filled porosity, D is the diffusion coefficient of the soil (m^2 s^{-1}) and $\delta C/\delta x$ is the concentration gradient. According to Millington & Quirk (1961) the diffusivity of a porous medium compared to that of the compound in free air can be described by the tortuosity (τ).

$$D = D_o \cdot \tau \text{ and } \tau = \frac{\varepsilon_a^{7/3}}{\varepsilon^2} \qquad \text{(Millington and Quirk, 1961)}$$

Where ε is the total porosity and ε_a is the gasfilled porosity, at the specific water content. Several other models to determine τ, as a function of ε and ε_a and empirical factors, exist (Scheutz *et al.*, 2009a).

Ficks law describes the diffusion coefficient in binary mixtures, diffusion of trace components and in ternary mixtures with one stagnant component (Scheutz *et al.* 2009a). Other approaches, which are more appropriate for complex gas mixtures are the Wilkes model and the Stefan-Maxwell equation (Scheutz *et al.* 2009a). Transport associated with wall collision is known as Knudsen diffusion. This together with diffusion of a light gas into a heavy gas, which is hindered in a porous medium, can be described by the Dusty Gas model. It has been found that Knudsen diffusion is not important under optimum conditions for CH_4 oxidation, as the small pores where this process is relevant, will be water filled (Scheutz *et al.,* 2009). Thus it was found by Molins *et al.* (2008) that Knudsen diffusion was important, when the production of EPS was modeled and caused a reduction of the pore space (Molins *et al.*, 2008). The degradation of EPS was not modeled, and therefore the importance of the EPS it most likely over estimated. Several authors have reported increased CO_2 production from hotspot areas, where EPS was visible (Huber-Humer *et al.,* 2009; Pedersen *et al.,* IV), which indicated that the degradation of EPS is an important process in a compost based biocover.

Molins *et al.* (2008) also found that 99 % of the emitted CH_4 was diffusive flux, even when modeling high loads (up to 1336 g CH_4 m^{-2} d^{-1}). The importance of the diffusive flux became bigger the higher up in the column (Molins *et al.,* 2008). It should be stated that the high diffusive flux is created because of the advective flux, which secures the steep concentration gradient in the upper part of the column. If no advective flux was present, the concentration profile would be linear over the entire profile, which would result in much lower diffusive flux.

Molins *et al.* (2008) found that the ingress of O_2 was controlled only by diffusion when landfill gas with 50 % CH_4 was used and that the advective flux of the landfill gas counterbalances the ingress of O_2. The O_2 diffusion was diminished by 15 % in the base case, where the load was 250 g CH_4 m^{-2} d^{-1}. Different loads were also investigated in the study (77-1336 g CH_4 m^{-2} d^{-1}). The higher the load of landfill gas the higher was the diffusive flux of O_2 into the soil column, because of the steeper concentration gradient. On the other hand the higher advective flux out of the column resulted in more O_2 being “lost”. The two nearly balanced each other, but it did however result in a decrease in the net ingress of O_2 (7 to 15 mole O_2 m^{-2} d^{-1}) when the flow was increased from 77 to 250 g CH_4 m^{-2} d^{-1}. Higher loads of landfill gas resulted in a small increase in the net ingress of O_2 (Molins *et al.,* 2008). To this it should be added that the flux of O_2 might be similar, but for the high loads, it will be restricted only to the upper part of the column or the biofilter (Scheutz *et al.,* 2003; Pedersen *et al.,* III, IV), which will minimize the extend of the CH_4 oxidation and thereby limit the CH_4 oxidation rate.

Molins *et al.* (2008) also investigated the sensitivity to moisture content. It was found that at water contents above 0.34 m^3 m^{-3} void, the diffusion of O_2 into the column became inhibited. The higher water contents also resulted in a decrease in the depth of the CH_4 oxidation zone (Molins *et al.*, 2008).

Im *et al.* (2009) investigated the diffusion of O_2 into a CH_4 oxidising soil column. They found diffusion coefficients by fitting a model to three sets of column results operated with different loads (355 to 655 g CH_4 m^{-2} d^{-1}, 50 % v/v CH_4). Additionally they measured the diffusion coefficient of the soil in a diffusion test cell. Comparing the results they concluded that the diffusive coefficients fitted from the results, were generally lower than the ones measured in the diffusive cell for O_2, whereas the opposite picture was seen for the diffusion coefficient of CH_4. They suggested that the higher diffusion coefficient at higher flows were due to mechanical dispersion (Im *et al.*, 2009).

Rannaud *et al.* (2009) validated the TOUGH2-LGM model by reproducing field gas concentration profiles in a field-scale biofilter (Jugnia *et al.,* 2008) to determine CH_4 oxidation rates. They measured diffusivity and gas permeability as a function of water content and applied the best suited models (Moldrup and Corey, respectively). They fitted the model to gas concentration profiles on three measurement days in the field, to determine the load of CH_4 and the CH_4 oxidation rates for a passive biofilter. For August 15th 2006, where the saturated water content (S_w) was 87-91 %, they found that the best fit was achieved by 99 % CH_4 oxidation, and an CH_4 oxidation zone in 0.2 to 0.3 cm and a ΔP=0.05 kPa. The pressure difference was also modeled, as this number was not known. The load of CH_4 in this case was 12.2 g CH_4 m^{-2} d^{-1} (Rannaud *et al.,* 2009). The result was compared with their initial simulations, where CH_4 was not accounted for. This resulted in a relationship between the advection and diffusive flux of 0.23, meaning that the advective flux was 23 % of the diffusive flux (Rannaud *et al.,* 2009). For August 8th, 2006 (S_w=89-92 %) they found that the best fit was achieved with a lower load (4.1 g CH_4 m^{-2} d^{-1}) and a lower pressure difference

(0.03 kPa). The CH_4 oxidation zone was situated in 0.2 to 0.3 m and the efficiency was 99 %. For September 1st the best fit was found with a load of CH_4 and pressure difference similar to that on August 15th (12.6 g CH_4 m^{-2} d^{-1}, 0.05 kPa) (Rannaud *et al.*, 2009).

3.1.3 Mechanical dispersion

It has been suggested by Im *et al.* (2009) that mechanical dispersion could have importance, even at the gas loads typically found in CH_4 oxidation environments (355 to 655 g CH_4 m^{-2} d^{-1}, 50 % v/v CH_4). Mechanical dispersion is the spreading of gas phase components due to spatial variations in advective velocity and differences in the distance traveled caused by the pore system tortuosity (Sharma & Poulsen, 2009). Sharma & Poulsen (2009) investigated gas phase dispersion in compost, as a function of different water contents and airflow rates. The results showed that mechanical dispersion dominated over diffusion for all water contents and gas velocities investigated. They found that the mechanical dispersion increased with increasing velocities (Sharma and Poulsen, 2009). Though the gas velocities were generally higher than what was observed at landfills. The minimum velocity would correspond to a load of 6680 g CH_4 m^{-2} d^{-1} at 50 % v/v CH_4, which is above, what has previously been reported in landfill environments (Scheutz *et al.,* 2009a; Pedersen *et al.,* IV)

3.2 Influence of atmospheric pressure in a biocover window

The temporal variation in the emission is controlled by the change in atmospheric pressure (Scheutz *et al.*, 2009a). The advective flux of CH_4 will increase as the atmospheric pressure decreases. Especially for landfills with low production of gas this would be important as the pressure inside these landfills is lower. The relation between atmospheric pressure and CH_4 emission from landfills has been investigated (Galle *et al.*, 2001; Christophersen *et al.*, 2001; Czepiel *et al.*, 2003; Kjeldsen and Fischer, 1995), but the magnitude and effect of the changes will vary due to different landfill characteristics and will also depend on the water content of the biofilter material and the remaining escape routes for landfill gas at the landfill (Gebert and Groengroeft, 2006b). Atmospheric pressure change was also the key parameter in gas flux models developed by Young (1992), Nastev *et al.* (2001) and Poulsen *et al.* (2003). The emission of CH_4 is correlated with the development in the atmospheric pressure. Poulsen *et al.* (2003) related landfill gas migration with atmospheric pressure change and found that the fluxes, at the intermediate distances from a landfill were most sensitive to atmospheric pressure change and emitted 70 % of the total CH_4 mass emitted within 20 % of the time frame modelled (Poulsen *et al.*, 2003). The influence of atmospheric pressure change on CH_4 oxidation has been assessed by Christophersen *et al.* (2001) and Gebert and Groengroeft (2006b). Gebert and Groengroeft (2006b) found that the flow to a passively vented biofilter reversed every 20 h and that 80 % of the total CH_4 load supplied was supplied within only 33 % of the time. This reveals that the variation in the CH_4 load to the biofilter is important and this was found to have great implications for the operation of the biofilter (Gebert and Groengroeft, 2006b).

Pedersen *et al.* (III) investigated how atmospheric pressure changes affected the performance of a field scale biocover window. It was found that the emission of CH_4 was very sensitive to even small changes in atmospheric pressure. In Fig. 8 the CH_4 and CO_2 emission together with the atmospheric pressure during 3 campaigns can be seen. The first campaign A) was done under a relatively linear decrease in atmospheric pressure (constant negative pressure gradient), which resulted in high but rather constant CH_4 emissions. The second campaign B) was done during a period of a long and steep pressure decrease, which did results in very high emissions in the beginning of the campaign. In this case it was evident, how emission responded within less than 10 hours to the rather short and not very steep pressure increase. Moreover the pressure started to decrease on the Nov 19th in the morning and an almost immediate and very high increase in the emission was observed. For the third campaign C), which was performed in April 2009, the pressure up to the campaign was also decreasing, which explained the high emission in the beginning of the campaign. Thereafter as atmospheric pressure increased in the end of the campaign, the emission in all the measured points decreased, Fig. 8 (Pedersen *et al.,* III). Generally it was concluded that the emission responded immediately to changes in the atmospheric pressure. Thus the actual magnitude was determined by the history of the pressure as this was believed to control the pressure within the landfill (Pedersen *et al.,* III). This coincided with the conclusions found by Young (1992) from modeling the effect of atmospheric pressure changes on CH_4 emissions.

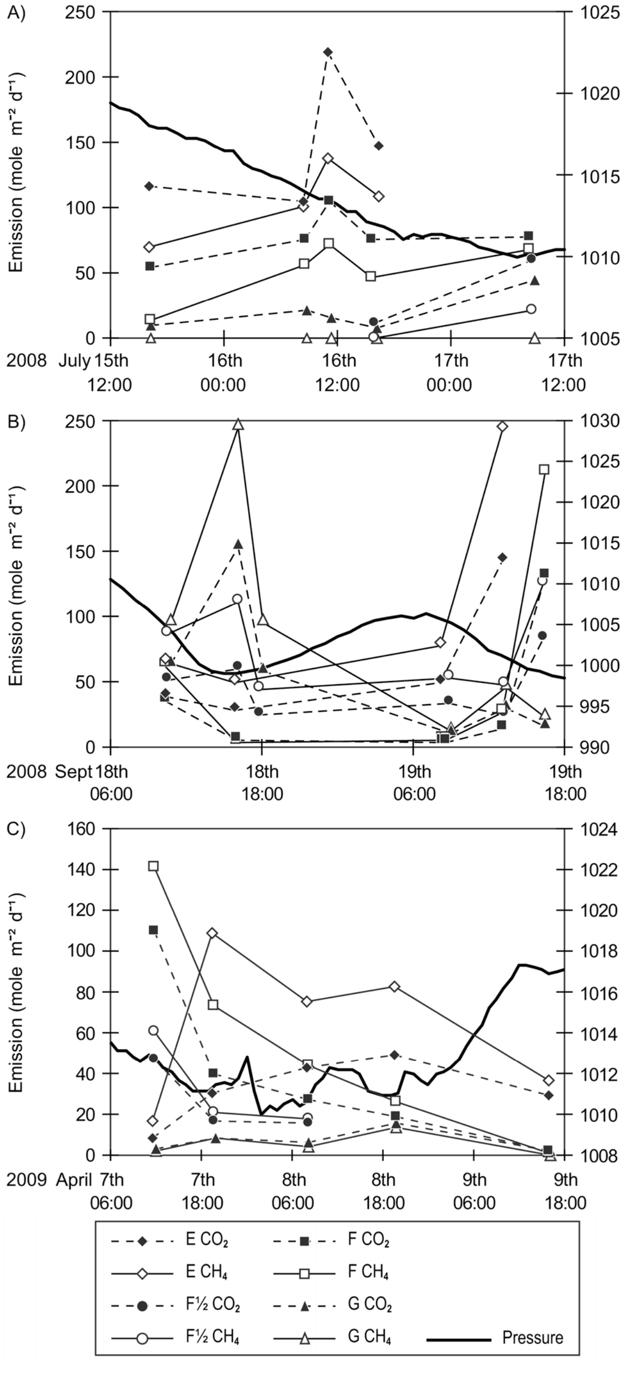

Fig. 8. Temporal evolution of the fluxes of CH4 and CO2 in the transect and the development of atmospheric pressure during 3 campaigns in A) July 2008, B) November 2008 and C) April 2009 (Pedersen *et al.,* III).

In the field study done by Pedersen et al. (III) the CH_4 and CO_2 concentration in 10 cm depth was logged, together with the temperature in the compost to assess the effect of atmospheric pressure changes in greater detail. This was done in order to improve measurement frequency and duration of measurements (Pedersen et al., III). The results from this 12 day long campaign can be seen in

Fig. 9. From the figure it can clearly be seen, how the response in the CO_2 and CH_4 concentrations to atmospheric pressure changes happens within a few hours. This shows that the CH_4 oxidation zone shifted upwards when an atmospheric pressure drop occurred. The CO_2 responded to smaller atmospheric pressure decreases than CH_4, which could be due to either CH_4 oxidation or respiration of the compost. From these investigations it can be concluded that the flux of landfill gas in the hotspot area was mainly controlled by advective flux, as a very strong effect of atmospheric pressure changes on gas concentrations was recorded. In Pedersen *et al.* (III) the temperature was also logged and the compost was almost without diurnal temperature variations below 30 cm, despite atmospheric temperature variations from 10 to 20 ^{o}C during the campaign. Based on the results it can be concluded that a CH_4 oxidation zone situated below 30 cm will be unaffected by the diurnal variations in temperature. Additionally the figure shows that temperature is not the factor controlling the changes in the gas concentration. Water content was not causing the changes either, as they were almost constant during the campaign (Pedersen *et al.,* III)

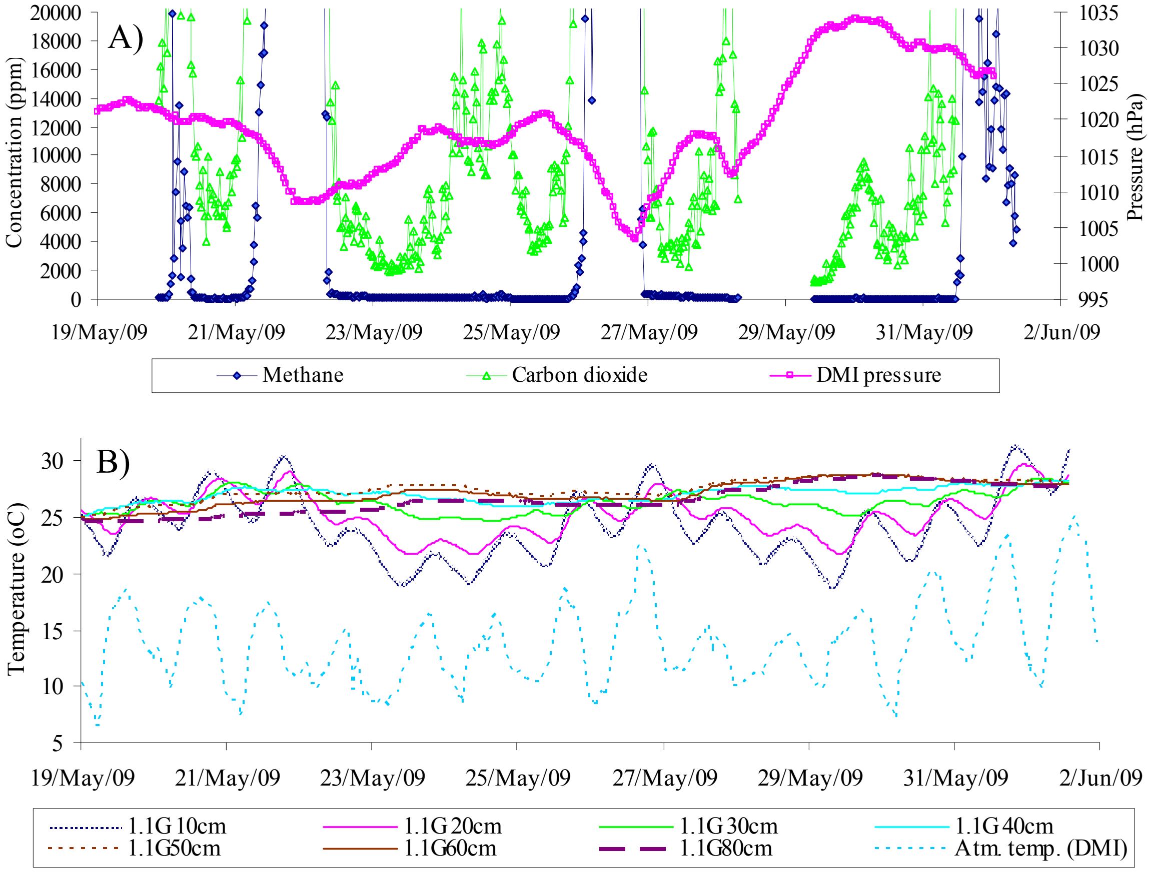

Fig. 9. A) Concentration of CO_2 and CH_4 in 10 cm depth over 12 days. B) Temperature in the compost at different depths during the logging campaign compared with atmospheric temperatures in the shadow from DMI (Køge station) (Pedersen *et al.,* III).

3.3 Influence of wind induced pressure fluctuation in a biocover window

Poulsen & Moldrup (2006) investigated the effects of wind-induced pressure fluctuations on the fluxes from the surrounding area of the Skellingsted Landfill, Denmark. They found that diffusion and wind induced gas transport were equally important with wind-induced gas transport becoming the most important at lower soil air contents (<0.22 m^3 m^{-3}) (Poulsen and Moldrup, 2006). The characteristics of the pressure fluctuations, namely the standard deviation and the power spectrum of the fluctuation depend upon wind turbulence, which depends on the wind speed and the roughness of the surface (Poulsen & Moldrup, 2006). The pressure fluctuations induced by wind turbulence causes vertical fluctuations in gas movement in the upper soil. The magnitude and depth of penetration of these fluctuations depends upon the standard deviation and the power spectrum of the pressure fluctuations, at the soil surface (Poulsen & Moldrup, 2006).

Additionally Huber-Humer *et al.* (2008) reported that compost biocovers at slopes were very influenced by wind. Due to the above, there was reason to believe that the compost biocover 1.1 at Fakse Landfill could be influenced by wind, as it was made of a coarse compost material (the total porosity was 0.7) and placed on a slope (Pedersen *et al.,* III). During the logging campaign, presented in the previous chapter, the pressure difference between 30 cm and 160 cm was also logged with a digital manometer (HM35, Thommen Instruments, 0-25 mbar). Fig. 10 shows the results together with the wind speed logged at the biocover window. It was expected that positive pressure differences could be detected in 160 cm with a reference in 30 cm, which could prove the presence of advective fluxes of landfill gas. Thus an atmospheric pressure increase (1012 to 1034 hPa) was measured on May 29th to 30th (Fig. 9), which could explain the general low pressure differences between 160 cm and 30 cm presented in Fig. 10. The reference point was situated in 30 cm in order to dampen the effect of wind induced fluctuations, but high variation was measured. Unfortunately the minimum negative pressure difference, which was measurable, was -3 mbar, which resulted in holes in the graph, when pressure was below -3 mbar (Fig. 10). Thus looking at Fig. 10 it seemed that there is a relation between wind speed and the pressure fluctuation, which showed that even in 30 cm depth the pore gas in the compost biocover was influenced by pressure variations due to wind. Additionally it can be seen from Fig. 10 that wind generally caused a decrease in the pressure in 30 cm depth if the pressure in 160 cm was assumed constant, which did indicate that wind induced fluxes were present at the site.

The wind induced fluxes are related to the variation in the pressure; the gusts and the correlation lengths. (The more variation in the wind speed, the more wind–induced gas transport) The standard deviation of all the pressure fluctuations measured was 204 Pa (average of -121 Pa). This value was compared to the results in Poulsen and Moldrup (2006), who calculated the portion of the total flux, which was wind induced (F_w) for different values of standard deviation of the wind speed, gas permeability and porosity for a sandy landfill soil from Skellingsted landfill. Comparing with the results to case 1 in Poulsen & Moldrup (2006) this would result in a relative F_w (relative wind induced gas flux) of 0.1-0.2 for a gas permeability of 5 10^{-12} m^2 and a gas filled porosity of 0.375. The F_w increased with increasing gas permeability (in the biocover window it is 10^{-10} m^2). Though gas filled porosity in the biocover window on May 18th was 0.251, which is lower and causes lower F_w. Thus the results indicate that 10 to 20 % of the emitted fluxes at the biocover during the period measured in Fig. 10 are wind induced.

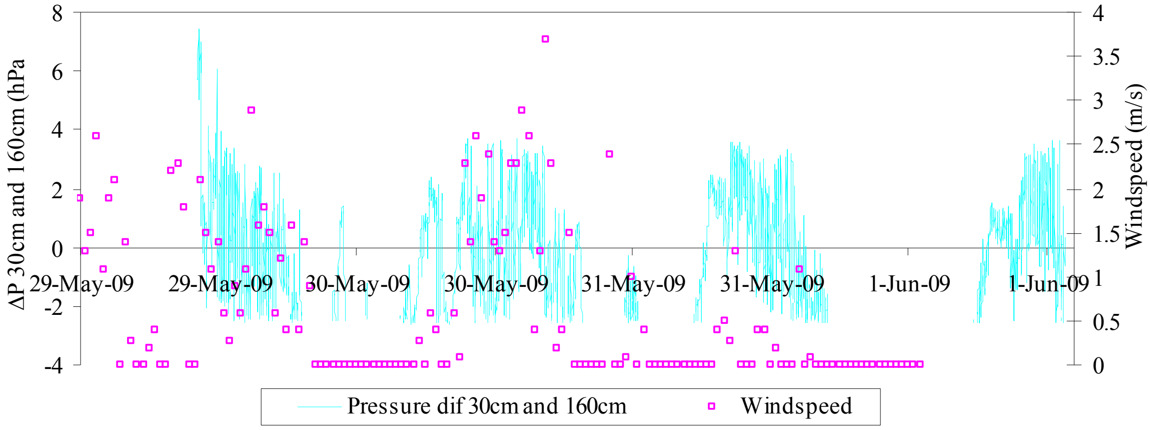

Fig. 10: Wind speed measured at the compost biocover 2 m from the situation of the logging of the pressure difference between 30 cm and 160 cm depth.

Poulsen and Moldrup (2006) found that 40 % of the CH_4 emission was wind induced at Skellingsted Landfill. In Poulsen & Moldrup 2006 it was stated that "Soil air permeability and the amplitude of the wind-induced pressure differences were the two most important parameters in controlling the magnitude of wind-induced gas transport in soil. The results further indicate that wind induced gas transport becomes the most important gas transport mechanism in the soil for higher soil water contents because the gas diffusion coefficient decreases rapidly for increasing water content compared to air permeability" (Poulsen & Moldrup, 2006). Due to this it is very likely that the wind-induced gas transport is even more significant in a compost biowindow, placed on a slope, as the gas permeability is higher than in Skellingsted and seems unchanged even at higher water content. But the actual change in the diffusion coefficient at decreasing water content is not measured in the inherent study.

Biocovers on slopes will be heavily affected by wind, but whether this is negative or positive for the efficiency in terms of CH_4 oxidation is unclear. Increased intrusion of atmospheric air has been modeled up to several meters into the vadose zone and even tens of meters during storm events (Massmann and Farrier, 1992). This can optimize CH_4 oxidation as O_2 will promote CH_4 oxidation in deeper layers (Pedersen *et al.,* IV), though increased emission of CH_4 due to wind induced pressure fluctuations can diminish the efficiency of the biocover system (Pedersen *et al.,* III, IV).

3.4 Gas permeability and water content in a biocover window

Gas permeability is a key parameter, for describing the physical properties of possible materials for biocovers. Additionally in situ gas permeabilities will allow an assessment of the advective flux in the biocover window.

3.4.1 Measuring gas permeability

In column set-up gas permeability can be relatively easily determined, as only simultaneously measured flow and pressure difference is needed for the calculation. A relatively accurate manometer is needed to avoid high flows, which can cause turbulence and leaks in the column incubation (Pedersen *et al.,* I).

$$k_g = \frac{v_g \cdot \eta_g \cdot \Delta x}{\Delta P} \quad \text{where} \quad \left[m^2\right] = \frac{\left[{}^{m}\!/_{s}\right] \cdot \left[{}^{N}\!/_{s \cdot m^2}\right] \cdot \left[m\right]}{\left[{}^{N}\!/_{m^2}\right]}$$

v_g is the Darcy velocity of the gas, and in a column incubation it can be measured with a boble flow meter.

η_g is the dynamic viscosity of the used gas. For nitrogen it is $17{,}6 \cdot 10^{-6}\ {}^{Ns}\!/_{m^2}$.

Δx is the distance between the measured pressures (m)
ΔP is the measured pressure difference (Pa)= (N/m^2)

The theory applied for in situ measurement of gas permeability is slightly more complicated as the shape of the flow lines is not known. A model has to be used to determine the relationship between the area (a_s), which is influenced by the flow and the length (L_s) of the flow lines. This relationship is normally referred as the "shape factor" (A). Iversen *et al.*, (2001) used a model by Liang *et al.*, (1995) and compared the results to gas permeability measured with known boundary conditions (a_s and L_s). They found that the results compared well and this indicated that reliable gas permeabilities could be found with the model by Liang et al. (1995) for the soils tested. The soils tested included six different Danish agricultural soils ranging from sand to clay loams. The organic matter content ranged from 0.4 to 2.9 g 100 g^{-1}. As some of the soils were structured (coarse with different particle size), this indicated that the model by Liang et al. (1995) could also be applied to structured soils in the undisturbed condition (Iversen *et al.*, 2001). The model was used for calculation of the in situ gas permeabilities in this study (2.3 10^{-10} m^2) and they compared well with the results found in the laboratory (3.0 10^{-10} m^2) (Pedersen *et al.,* III). This indicated that the use of the Liang *et al.* (1995) can be justified, as it fits with results found with known boundary conditions (a_s and L_s).

Air permeability for the in-situ measurements were calculated using a rewriting of Darcys law, where the cross sectional area (a_s) and the length of the soil sample (L_s) were replaced by the shape factor, A.

$$k_g = \frac{Q \cdot \eta_g \cdot L_s}{\Delta P \cdot a_s} = \frac{Q}{\Delta P} \cdot \frac{L_s}{a_s} \cdot \eta_g = \frac{Q \cdot \eta_g}{\Delta P \cdot A} \quad \text{(Iversen \textit{et al.}, 2001)}$$

The shape factor may be regarded as an estimate of the a_s/L_s quotient in a measuring condition where neither a_s nor L_s is well defined and was determined according to Liang et al. (1995).

$$\frac{A}{D} = 0.4862(D/L) - 0.0287(D/L)^2 - 0.1106 \quad \text{(Liang \textit{et al.}, 1995)}$$

Where D is the inside diameter of the soil core (20cm), L is the length (10cm). When the sample ring is inserted 10 cm into the ground, this corresponded to a shape factor of 0.15 m.

3.4.2 In situ measurement of gas permeability in biocover window

Pedersen *et al.* (III) measured in situ gas permeability in a compost biocover window to assess the in situ properties of the compost. As described in Pedersen *et al.* (III) five campaigns were conducted with permanent sample rings in three measuring points in the biocover window. In these three points water content was also logged in 10 cm and 20 cm. In Fig. 11 the measured gas permeabilities is plotted, as a function of water content and the relationship was poor. Little variation was measured in gas permeabilities (1.5-3.2 10^{-10} m^2) despite that variation were measured in the logged water contents (0.24-0.51 m^3 m^{-3}). In general the biocover window was well drained (Pedersen *et al.,* III). In Fig. 11 there is a tendency that gas permeabilities are increasing at increasing water content, which is surprising, but it has been found by others especially for loosely packed compost materials (Poulsen and Moldrup, 2007; Ahn *et al.,* 2008). An explanation for this behaviour could, according to Poulsen and Moldrup (2007), be that "the addition of water will cause the compost to stick together and form aggregates with the water contained in small intra-aggregate pores. This will result in larger inter aggregate air-filled pores resulting in higher gas permeability" (Poulsen and Moldrup, 2007).

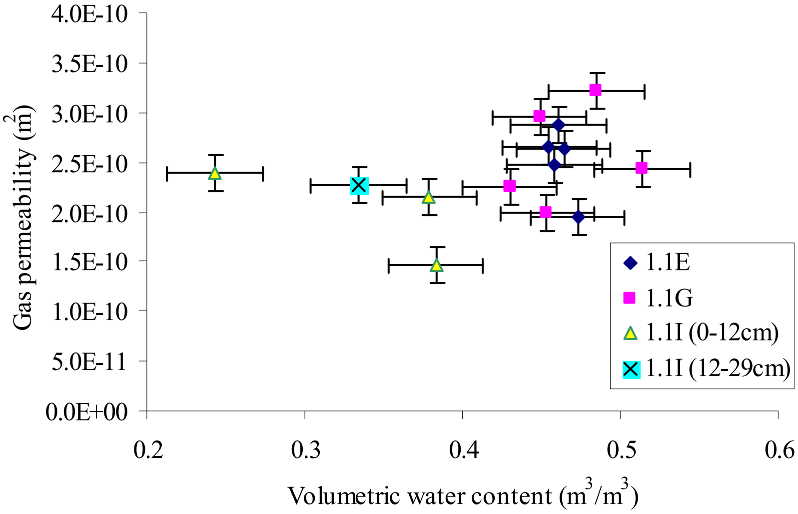

Fig. 11. Gas permeability as a function of volumetric water content for three different measuring points in the biocover window, E, G and I.

In Fig. 11 the results were also subdivided into the three measuring points in the transect E, G and I. In E, EPS was clearly visible and the compost was red and sticky and clumped together, in G this was also seen, but to a lesser extent and it was situated deeper than in E and in I, EPS was not visible and the compost did not stick together and clump (Pedersen *et al.,* III). It was hypothesized that EPS would reduce gas permeability in the biocover window, but no relationship between the situation of the sampling points and the measured gas permeability was found. This coincided with the findings of Hilger *et al.* (2000), which reported that EPS did not cause bioclogging in the investigated columns, but did decrease the diffusion of O_2 and CH_4 to the methanotrophic bacteria (Hilger *et al.*, 2000).

It was attempted to find a relationship between the in situ water contents and the in situ gas permeabilities according to the results reported by Poulsen and Moldrup (2007). Poulsen and Moldrup (2007) investigated the relationship between air permeability of compost, bulk density and volumetric air content. Two composts were investigated; a screened (< 5mm) compost of garden waste and a sewage sludge compost. They found that the relationship between gas permeability and air filled porosity could be described by the following formula;

$$\frac{k_a}{k_a^*} = \left(\frac{\varepsilon}{\varepsilon^*}\right)^{(15.1\cdot\delta_b - 11.1)} \quad \text{(Poulsen and Moldrup, 2007)}$$

Where k_a^* and ε^* are reference point values of air permeability and air filled porosity. This relation is found from the relation beneath:

$$\log(k_a) - \mu = A(\log(\varepsilon) + \alpha) \quad \text{(Poulsen and Moldrup, 2007)}$$

It was investigated whether the relationship found in Poulsen and Moldrup (2007) fitted with the results in the inherent study. For this purpose the logarithm of the gas filled porosity was plotted against the logarithm of the measured gas permeabilities. As can be seen in Fig. 12 the result was poor with an R^2 of 0.11. It could most likely be explained by macropore flow as discussed above, but there also existed uncertainties, as the water contents were not the average water content for the exact sample, where the gas permeability was measured, but a point measurement of the water content 30 to 40 cm away. This can be of importance, especially in heterogeneous and structured compost consisting of twigs and branches.

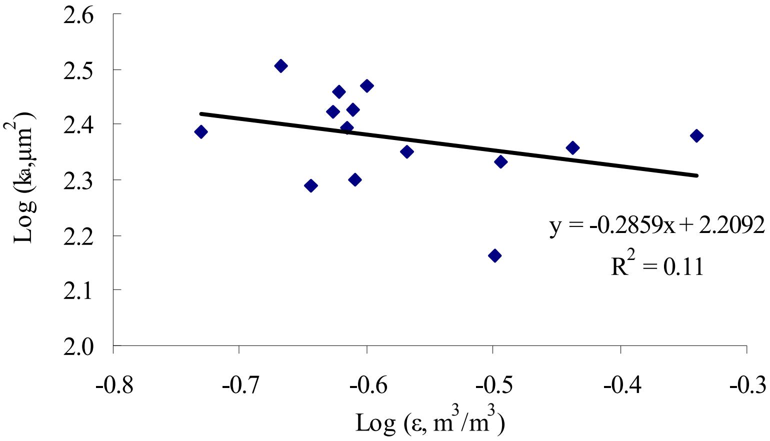

Fig. 12. Gas permeability as a function of gas filled porosity (note logarithmic scale).

The model found by Poulsen and Moldrup (2007) was applied to two data sets of gas permeability and water content determined gravimetrically for the entire soil sample in the sample ring. The bulk density of the compost was fitted with the two data sets as the compost tested in Poulsen & Moldrup (2007) had a lower particle density (less organic matter). The bulk density is in the formula to describe the level of compaction and therefore this parameter needed adjustment. The gas permeability was then calculated with one reference point of measured gas permeability and water content determined gravimetrically for the entire soil sample in the sample ring (Pedersen *et al.,* III). The gas permeability in the bottom of the biofilter was determined over the year, based on logged water contents and they were never lower than 10^{-10}, which would allow landfill gas to enter the filter over the entire year (Pedersen *et al.,* III). Therefore it could be concluded that the load of landfill gas to the biocover was not impaired by high water contents, which reduced advective flux of landfill gas through the filter.

3.5 Diffusivity and water content in a biocover window

The O_2 diffusion is of great importance as O_2 is needed for the CH_4 oxidation to occur. Therefore the seasonal change in diffusivity of the compost was assessed based on the logged water content in 10 cm, which affects the air filled porosity (Pedersen *et al.,* III). The tortuosity (τ) was calculated from the relationship presented by Millington & Quirk (1961) and all diffusive fluxes were calculated assuming that the O_2 concentration was zero in 10 cm (see section 3.1.2 Diffusion). This will only be the case at a certain advective flux, which will counteract the diffusive influx of O_2. Thus the aim of the calculations was to get an idea of the seasonal change in the diffusivity of the compost and indicate how this could affect the CH_4 oxidation. From the calculated diffusive flux of O_2 (mole m^{-2} d^{-1}) a potential CH_4 oxidation rate (mole m^{-2} d^{-1}) was calculated. An O_2/CH_4 ratio of 2 was used, which was the average found in a 111 day long column experiment (Pedersen *et al.,* I). When using this approach, it is believed that O_2 used for degradation of the compost material (respiration) was accounted for in the calculation.

Fig. 13 shows the results of the above described calculations together with the measured water contents in 10 cm depth. There is a clear seasonal variation in the water content over the year and the calculated O_2 diffusive flux and the potential CH_4 oxidation rates over the season. The water content was logged in three points; E (high load of CH_4), G (medium load of CH_4) and I (zero load of CH_4) (Pedersen *et al.,* III, IV). For the measuring point G (Pedersen *et al.,* III), where the load was optimum for CH_4 oxidation to occur, the calculated CH_4 oxidation rates ranged from 0.6 mole CH_4 m^{-2} d^{-1} (10 g CH_4 m^{-2} d^{-1}) in February and March to 9.6 mole CH_4 m^{-2} d^{-1} (154 g CH_4 m^{-2} d^{-1}) in August and September (Pedersen *et al.,* III). It can also be seen from Fig. 13 that the pattern of the changing volumetric water content over the year was different for the three measuring points. The upper layers in I starts drying out already in March and April and the decrease is more smooth and levels out in June, July. In G the decrease in the water content does not start until June, July and then it is very sudden. The same picture is seen in E, but the decrease is not as severe as in G. The difference between the three points is speculated to be caused by the emission pattern. In E no vegetation was seen as it was in the hotspot and in G only patchy vegetation was observed. In I the vegetation started using water, when the growing season started in March to April, whereas the water content in E and G was controlled by CH_4 oxidation and EPS. During CH_4 oxidation water was produced and by modeling Molins *et al.* (2007) found that this could affect the diffusivity in a CH_4 oxidizing soil column. Additionally EPS has been reported to retain water (Or *et al.*, 2007).

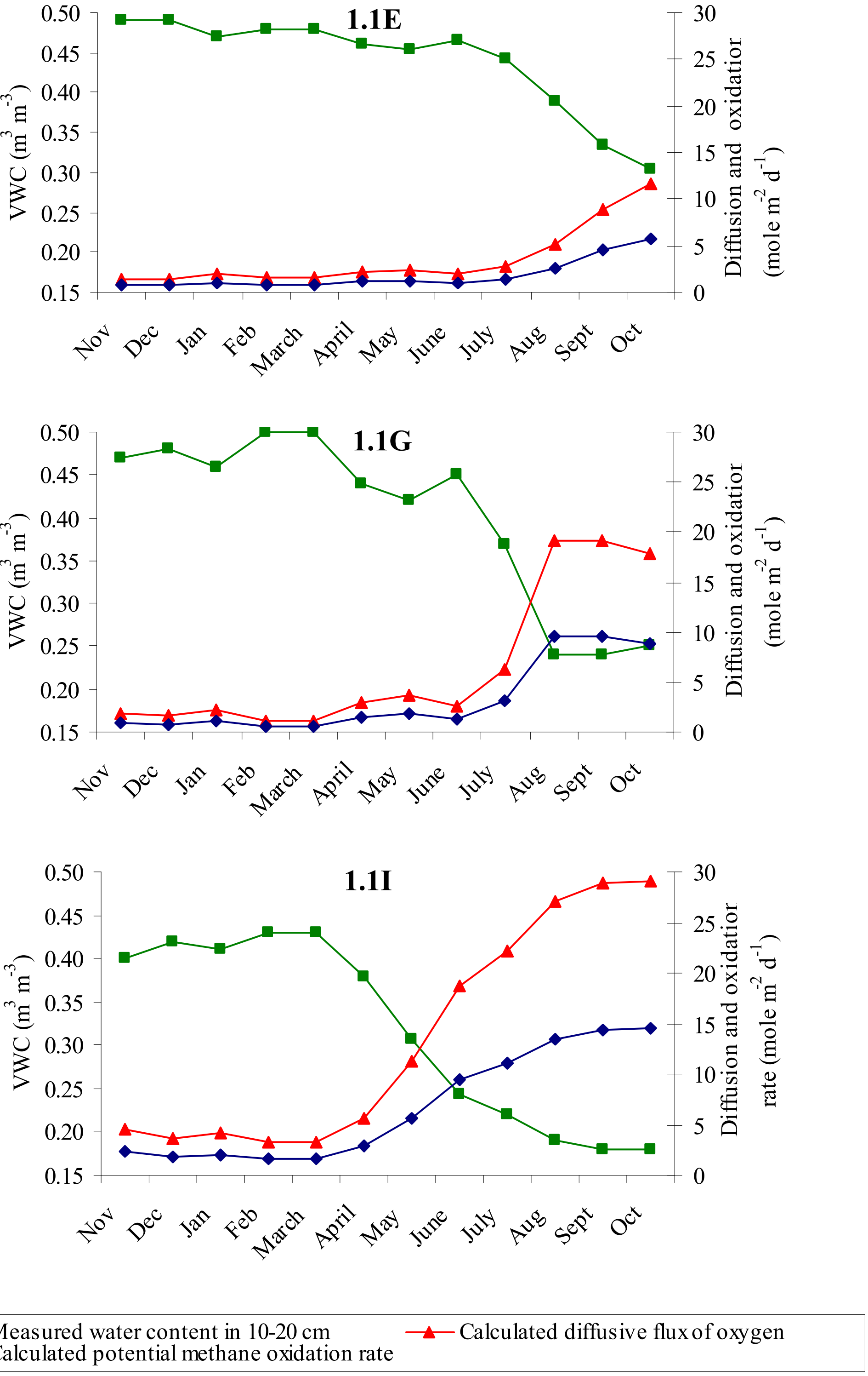

Fig. 13. Seasonal change in measured water content, 10-20 cm for the three measuring points together with calculated diffusive flux of O_2 and calculated potential CH_4 oxidation.

The calculated potential CH_4 oxidation rates were compared to measured CH_4 oxidation rates (Pedersen *et al.,* IV). On April 16th 2009 CH_4 oxidation rates were measured by two different methods and the measured CH_4 oxidation rates were as high as 238 g CH_4 m^{-2} d^{-1}. In Fig. 13 the calculated potential CH_4 oxidation rate in April was 18, 23 and 46 g CH_4 m^{-2} d^{-1} for the three points E, G and I respectively. In general the calculated CH_4 oxidation rates in Fig 13 were low compared to the CH_4 oxidation rates calculated with the carbon mass balance and measured by the stable isotopic approach (Pedersen *et al.,* IV). This was found despite that O_2 diffusion was not determined conservatively (C_{O2}=0 in 10 cm and the loss of O_2 from the advective outward flux was not accounted for). This could indicate that O_2 was also supplied by advective flux, either promoted by increasing atmospheric pressure (Gebert and Groengroeft, 2006b) or volume reduction due to CH_4 oxidation (De Visscher *et al.*, 1999; Rannaud *et al.*, 2009; Molins *et al.*, 2008)

It can be concluded that the gas transport is very important processes in field scale biocovers, as it controls the transport of substrate to the methanotrophic bacteria. The presence of methanotrophic bacteria is not a guarantee for high efficiency as both CH_4 and O_2 must be available in balanced amounts to ensure that mitigation of CH_4 emissions are successful. Additionally gas transport also affects the certainty of the methods for determining local CH_4 oxidation rates, which will be described in the next section.

4 Measurement of CH_4 oxidation in biocover windows

Emission of CH_4 from landfills can be mitigated by a different range of biocover systems (Huber-Humer *et al.*, 2008a; Einola *et al.*, 2009; Jugnia *et al.*, 2008; Dever *et al.*, 2007). To fully implement and apply biocovers it is of great importance to be able to evaluate the performance of the systems, both the total efficiency and the efficiency in individual biocover windows and smaller sub parts of the system. There is no well documented and inexpensive method to quantify CH_4 oxidation in the field in passive biocover systems, where the load of landfill gas is supplied directly from the landfill body without introducing pumps, collection pipes, regulators and meters to control and measure the landfill gas supply. Some total CH_4 oxidation measurement methods are available (Lohila *et al.*, 2007; Borjesson *et al.*, 2007a), which are still under development and needs verification and support by small scale methods. Development of biocovers technology is in progress and research tools are required in order to understand and resolve small scale problems, such as hotspot formation in the biocover (Einola *et al.,* 2009). For this purpose different methods are available and have been applied in the field and the methods will be presented in the following section.

4.1 Stable isotopic approach

The stable isotopic method is based on the fact that the bacteria prefer the C^{12} over the C^{13} carbon isotope, and by knowing the rate, they do this and the stable isotope composition for the anaerobic zone, the percent oxidation in a point z, can be determined ($f_{ox,z}$).

$$f_{oxo,z} = \frac{\delta^{13}C_z - \delta^{13}C_A}{\alpha_{ox} - \alpha_{trans}} \cdot 0.1, \_\, \alpha_{ox} = \frac{k_L}{k_H} \qquad \text{(Chanton \& Liptay, 2000)}$$

δ_E is the $\delta^{13}C$ value for the emitted CH_4 and δ_A is the value for the anoxic CH_4. The α_{ox} is the degree of isotopic fractionation done by the specific microbial population under specific conditions, where K_L and K_H are the rate constants of the light isotope ($^{12}CH_4$) and heavy ($^{13}CH_4$) isotopes, respectively. The α_{trans} value is isotope fractionation factor due to transport, which can be set to 1 when the gas transport is dominated by advection, but if the gas transport is dominated by diffusion this will cause an underestimation of the CH_4 oxidation (Chanton *et al.*, 2008a). When the CH_4 load to the biocover is very low the gas transport is most likely not dominated by advective flux.

To determine α_{ox} the simplified Rayleigh model can be used as it gives errors of only (α-1)>0.05% (Mahieu *et al.*, 2006)

$$\ln(\frac{C_{CH_4t}}{C_{CH_40}}) = \frac{\alpha_{ox}}{\alpha_{ox}-1} \cdot \ln(\frac{\delta^{13}C_t + 1000}{\delta^{13}C_0 + 1000})$$

$\delta^{13}C_0$ is the carbon isotope ratio at time 0, $\delta^{13}C_t$ is the carbon isotope at time t, $\ln(\frac{C_{CH_4t}}{C_{CH_40}})$ was plotted as a function of $\ln(\frac{\delta^{13}C_t + 1000}{\delta^{13}C_0 + 1000})$ the slope was used to calculate the fractionation factor. $\alpha_{ox} = \frac{slope}{1+slope}$ (Chanton *et al.*, 2008a)

The fractionation factor (α_{ox}) is temperature dependent. Tyler *et al.* (1994) found that the relation is -0.00046 K^{-1} in a thorough field and laboratory study. Chanton and Liptay (2000) found the following relationship between α_{ox} and temperature, $\alpha_{ox} = -0.000438 \cdot T + 1.0411$, for a mulch landfill cover soil and $\alpha_{ox} = -0.000433 \cdot T + 1.0421$, for a clay landfill cover soil. Chanton *et al.* (2008a) pooled data from the literature and added new data and got to the relationship; $\alpha_{ox} = -0.00039 \cdot (T - T_{measured}) + \alpha_{ox,measured}$.

Numerous references are available on the use of stable isotopes (Borjesson *et al.*, 2007b; Borjesson *et al.*, 2001; Abichou *et al.*, 2006; Liptay *et al.*, 1998; Chanton *et al.*, 2008b) and Chanton *et al.* (2009a) is a good review. The value and the certainty of the method have been discussed and the fractionation factor depends on temperature and potential CH4 oxidation rate (V_{max}) (Chanton *et al.*, 2008a). Templeton *et al.* (2006) found that the fractionation depends on CH_4 and O_2 concentration which in a field situation is very unlikely to be stable. Furthermore they found dependency on cell density, which coincides well with the findings of Chanton *et al.* (2008a). Borjesson *et al.* (2009) applied the stable isotopic approach to total emission measurements and did a temperature dependency study of α_{ox} for six different landfill covers. They found that the CH_4 oxidation estimates were most sensitive to variation in α_{ox} and it caused variability in CH_4 oxidation of between -40% to +72%.

Additionally the commonly used assumption that diffusion is not an important gas transport process can be questioned (De Visscher *et al.*, 2004; Chanton *et al.*, 2008b). Chanton *et al.* (2009a) stated that especially chamber measurements are "lower limit conservative estimates" and Mahieu *et al.*, (2008) found that when modeling the isotopic fractionation by diffusion the method became superior to existing isotopic approaches. By neglecting the fractionation by diffusion an underestimation of the CH_4 oxidation rate is made (Mahieu *et al.*, 2008).

In a study by Powelson *et al.* (2007) a comparison between a mass balance (with measured inflow and concentration) and the stable isotopic method was done for in an outdoor set-up. It was found that the stable isotopic method used on emitted samples gave results of approximately 45% of the mass balance approach (Powelson *et al.*, 2007). Most likely a better correlation between the two methods would have been found if the averages of emitted and 10 cm gas samples were used for the stable isotopic approach, as suggested by Chanton *et al.*, (2008b) Cabral *et al.* (2010) also compared isotopic measurements and the results from measured flows and concentrations, The stable isotopic approach resulted in a CH_4 oxidation rate of f_o=88.7% in 10 cm (578 g m^{-2} day^{-1}) and 55.6% at the surface (93 g m^{-2} day^{-1})), the average between the two is 335 g m^{-2} day^{-1} (82%). With the mass balance based on a measured influx of landfill gas f_o=70.0% (257 g m^{-2} day^{-1}), which shows that the stable isotopic approach resulted in higher CH_4 oxidation rates than the mass balance when the approach suggested by Chanton *et al.* 2008b is used. This is the opposite of previously reported results (Powelson *et al.,* 2007; Christophersen *et al.* 2001b; Chanton *et al.,* 2009a), thus in the study by Cabral *et al.* (2010) the fractionation factor was not measured, but calculated from a literature temperature dependency (α_{ox}=0.0235 at 20°C) (Cabral *et al.,* 2010). In general the underestimation of the stable isotopic method can also be caused by the fact that if no CH_4 emission is measurable the $\delta^{13}C$ in CH_4 cannot be determined and these areas will therefore not be accounted for (Powelson *et al.*, 2007).

Handling the amount of uncertainties for the stable isotopic approach is complicated by the fact, that the sample price is significant. Additionally representative sampling and determination of α_{ox} is very difficult, as the CH_4 oxidation zone will move vertically up and down in most studies (Christophersen and Kjeldsen, 2001; Cabral *et al.*, 2010; Pedersen *et al.,* III). In the study by Pedersen *et al.* (III, IV) the CH_4 oxidation zone also moved in the horizontal direction, when load of CH_4 increased or decreased. Additionally it was concluded that CH_4 was oxidized in the gas distribution layer and in the waste (Pedersen *et al.,* IV), which complicates where the sample for determination of α_{ox} should be taken even more. In other words; an in depth analysis on potential CH_4 oxidation rates determined in batch incubations is needed, to determine where the mayor amount of the CH_4 is oxidized (CH_4 oxidation zone). Using this approach the right sample for the determination of α_{ox} can be taken, but it is a very laborious approach.

4.2 Carbon mass balance

The CH_4 oxidation rate can also be estimated from the emitted CH_4 and CO_2 flux and the measured raw gas composition, as previously described in Christophersen *et al.*, (2001). It was used to determine CH_4 oxidation rates for a sandy loam, without accounting for soil respiration or assimilation of carbon into biomass. The method assumes that emitted excess CO_2 originates from CH_4 oxidation, which can be questionable in very organic materials, such as composts (Pedersen *et al.,* IV).

The method builds on the conservation of mass for carbon (Eq. 1) and assumes that the relationship between the fluxes of CH_4 and CO_2 and the concentrations of CH_4 and CO_2 is equal in the bottom of the biocover window (Eq. 2). Combining Equation 1 and 2, results in Equation 3 and the load of CH_4 into the biocover window can be calculated. Then the CH_4 oxidation rate can by determined by Equation 4. The method is based on the assumption of steady state therefore assimilation is not accounted for in the inherent study (bacterial growth equals decay). Additionally the dilution of CO_2 in percolating rainwater is assumed negligible, as was found in (Christophersen, 2000).

$$J_{LFG,bottom} = J_{CO_2,surface} + J_{CH_4,surface} - J_{CO_2,respiration} = J_{CO_2,bottom} + J_{CH_4,bottom} \qquad (1)$$

$$\frac{J_{CH_4,bottom}}{J_{CH_4,bottom} + J_{CO_2,bottom}} = \frac{C_{CH_4,bottom}}{C_{CH_4,bottom} + C_{CO_2,bottom}} \qquad (2)$$

$$J_{CH_4,bottom} = \frac{C_{CH_4,bottom}}{C_{CH_4,bottom} + C_{CO_2,bottom}} \cdot \left(J_{CO_2,surface} + J_{CH_4,surface} - J_{CO_2,respiration}\right) \qquad (3)$$

$$R_{CH_4\_oxidation} = J_{CH_4,bottom} - J_{CH_4,surface} \qquad (4)$$

$J_{LFG,bottom}$ is the load of landfill gas coming into the biocover window,

$J_{CO2,respiration}$ is the CO_2 emission originating from respiration,

$J_{CO2, surface}$ is the CO_2 emission,

$J_{CH4,surface}$ is the CH_4 emission.

$C_{CH_4,bottom}$ and $C_{CO_2,bottom}$ are the concentrations of the CH_4 and CO_2 in the bottom of the biocover window and/or the raw gas composition.

$J_{CH_4,bottom}$ is the load of CH_4 into the biocover window and

$R_{CH_4\_oxidation}$ is the CH_4 oxidation rate.

In the study by Pedersen *et al.* (IV) the CO_2 production from degradation of the compost material was subtracted, as it was found significant in a column study (Scheutz *et al.,* 2009b). The CO_2 production rate was 107 ±14 g CO_2 m^{-2} d^{-1} at room temperature and for calculation of CH_4 oxidation rates the respiration was calculated depending on temperature by using a Q_{10} value of 3 (Einola *et al.,* 2007; Einola *et al.,* 2009). There was done no adjustment for the limitation in the respiration caused by high loads of landfill gas, which will diminish the depth of the aerobic zone (Pedersen *et al.,* IV).

The carbon mass balance has recently been applied by other for determination of CH_4 oxidation rates in compost biocovers. Einola *et al.*, (2008b) used the method in an experimental landfill cover composed of mechanically-biologically treated waste where compost respiration and assimilation of carbon into biomass was assumed to be zero. In a more recent study Einola *et al.* (2009) used the method for 60 % sludge compost for determination of CH_4 oxidation rates in a full-scale biocover system. Here minimum and maximum values for the CH_4 load into the cover were calculated. For the minimum value of CH_4 load and oxidation no assimilation was assumed and respiration was accounted for. It was determined in batch incubations and a temperature dependent respiration was calculated using a Q_{10} value of 3.0. The total depth of 50 cm was assumed to be sewage sludge and the total depth was assumed to respire. If respiration was higher than the emitted CO_2, the CH_4 load was set equal to the emission of CH_4 (oxidation is zero). If no emission of CH_4 was measured a load of 0.002 g m^{-2} d^{-1} was assumed. For the maximum value of load and oxidation of CH_4, respiration was assumed to be zero and assimilation was assumed to be 70 % (Einola *et al.*, 2009).

Philopoulos *et al.* (2008) also applied the carbon mass balance method in order to determine CH_4 oxidation rates in a field scale biocover made of garden waste compost. They accounted for the production of CO_2 from degradation of the compost and calculated the respiration based on CO_2 production found in batch incubations. They assumed an aerobic zone of 20 cm and this resulted in a CO_2 production of 39 g CO_2 m^{-2} d^{-1}. Additionally they noted that the CO_2 production most likely would be lower due to biogenic competition for the O_2 from the methanotrophs (Philopoulos *et al.*, 2008). When the emission was higher than the calculated CH_4 loads, which resulted in negative CH_4 oxidation rates the CH_4 oxidation rate was assigned a zero value. This approach can be discussed, as negative CH_4 oxidation rates could be due to CH_4 being produced in the biofilter (Barlaz *et al.,* 2004; Scheutz *et al.,* V) and the approach will result in overestimating the average CH_4 oxidation rates.

The method has also been applied by Dever (2009) for a composted garden waste (60 % v/v) and shredded wood (34 % v/v) where 44 % assimilation was used based on the CH_4 oxidation reaction described in Hilger and Humer (2003). In Dever (2009) it was found that adding a measured maximum CO_2 production of 144 g CO_2 m^{-2} day^{-1}, which was varied according to the CH_4 oxidation percentage, improved the accuracy of the method. The procedure was that when the CH_4 oxidation was 100 % the respiration was also set to 100 % of 144 g CO_2 m^{-2} day^{-1} and so forth. This was realized, as the method was verified in outdoor column incubations (240 L garbage bins) with known inlet flux of landfill gas. The CO_2 production experiment was done 2 months after a 9 months field trial with loads of 14.4 and 81.6 g CH_4 m^{-2} day^{-1} respectively. The author found that the method was a good estimate for the average CH_4 oxidation rate at moderate CH_4 loading (36-480 g CH_4 m^{-2} day^{-1}) though the upper limit was believed to be caused by the rather small flux chambers (4.5 L) used in the study. The study showed that the carbon mass balance did not provide a good estimate for the individual results. The author suggested that this was caused by the change in assimilation percentage, which is plausible in a field situation with changing load, temperature, moisture etc. Additionally it was found that the method was overestimating the CH_4 oxidation rates when the filter was receiving a low load (<36 g CH_4 $m^{-2}day^{-1}$) (Dever, 2009).

4.2.1 Assimilation of carbon into biomass

Assimilation of carbon into biomass will occur when a bacterial community is building up, but a continuous growth of bacteria is in theory impossible. To evaluate the use of assimilation of carbon into biomass in the carbon mass balance it was attempted to calculate the biomass produced in the experiment performed by Dever (2009). With an average CH_4 oxidation rate of 28.5 g CH_4 $m^{-2}day^{-1}$ the total amount of assimilated carbon during the 9 months duration of the experiment, will result in 5163 mole C-biomass m^{-2} (44 % of CH_4 oxidized times the duration of the experiment). It was assumed that bacteria and exopolymeric substances (EPS) consisted of CH_2O (would result in a conservative estimate) and had a bulk density of 500 kg m^{-3}. This resulted in the formation of 0.3 m^3 biomass m^{-2}. If the gas filled porosity of the 1 m compost cover is 0.3 the entire gas filled pore space in the compost profile will be filled with micro-organisms and exopolymeric substances (EPS). The same calculation was done with the assimilation parameter of 70 % used in Einola *et al.* (2009) and the produced biomass was 0.5 m^3 biomass m^{-2}. These calculations show that the assumption of endless assimilation cannot be fulfilled, as this will result in a layer of biomass on top of the biocover. It is generally believed that both the respiration and assimilation used in Dever (2009) were too high, but that they counteracted each other and thereby more plausible results were achieved.

4.2.2 Respiration of organic matter

Huber-Humer (2004) investigated respiration in batch incubations and found that respiration increases when samples are taken from high emission areas. CO_2 emission rates were calculated based on sapromat respiratory test for 2 different composts used in a cover; sewage sludge compost and municipal solid waste compost and they were 15 g $m^{-2}day^{-1}$ and 25 g $m^{-2}day^{-1}$ respectively. Huber-Humer *et al.* (2009) reported that increased respiratory activity was associated with hotspot areas where CH_4 oxidation is high. They attributed this to decay of methanotrophic bacteria and degradation of EPS. Pedersen *et al.* (IV) also reported increased CO_2 production from a hotspot area in a compost based biocover window. This finding has implications for how the determination of the respiration should be done. It should be determined prior to installing the compost in biocover windows if the CO_2 production should be true basal respiration. If it is determined for compost samples, which has been exposed to CH_4 it will include carbon that originates from CH_4, which has been oxidized previously and assimilated into biomass. This will cause an overestimation of the respiration and an under estimation of the CH_4 oxidation rates if the respiration is used in a carbon mass balance.

4.3 Other methods

Some other methods to determine load and CH_4 oxidation rates in passive field scale biocover systems have been applied. Huber-Humer *et al.* (2009) used a reference cell without a biocover, where emissions were measured and then this number was used as the load of CH_4 for other similar cells, with the same waste mass and composition (Huber-Humer *et al.*, 2009). This approach can be questioned by the fact that waste and emission are very heterogeneous spatially in most landfills and CH_4 can be oxidized in the top of the waste. In another study the load was found by digging out the compost and measure the emission (Boeckx et al., 1996), which can be very uncertain in very dynamic systems and if the permeability of the dug out cover material is lower than the material below. Furthermore both methods are difficult to apply in a full scale system. Additional methods are available where potential CH_4 oxidation rates are measured, such as the push pull test (Urmann *et al.*, 2008), which has also been combined with stable isotopes. Furthermore intact core samples can be taken to the lab and investigated both in batch experiments and in column experiments. This will give an adequate indication of what the maximum CH_4 oxidation rate is, thus it will most likely be higher than the year round mean CH_4 oxidation rate, which varies with CH_4 load, temperature, water content etc. Fredenslund (2010) also evaluated a deep flux chamber to determine the load of CH_4 to the biocover windows. This method resulted in unreasonable high CH_4 loads, which could not be correlated with the rather low emissions reported (Fredenslund, 2010). CH_4 oxidation rate can also be determined by modeling from fitting gas concentration profiles to measured gas concentration profiles in the field, as was done by Rannaud *et al.* (2008). The physical parameters of the compost (gas permeability and diffusivity) should be measured as a function of water content and other important input parameters are water content and temperature.

4.4 CH_4 oxidation rates for the Fakse Biocover

Pedersen *et al.* (IV) determined CH_4 oxidation rates in a compost biocover at Fakse landfill by the carbon mass balance approach (described in section 4.2) using field data from 13 campaigns over the year, which resulted in a total of 303 flux measurements. The CH_4 oxidation rates calculating by two approaches 1) by assuming that the compost respiration was zero and 2) by accounting for temperature dependent compost respiration (Pedersen *et al.,* IV). In Table 4 the results from the 13 campaigns are listed and the results by the two approaches are presented as 1) / 2). Nine of the campaigns represented the CH_4 oxidation in the entire window and in these campaigns the average CH_4 load was 195 / 165 g m^{-2} day^{-1}, which resulted in CH_4 oxidation rates of 67 / 37 g m^{-2} day^{-1}. Additionally four transect campaigns were performed in the hotspot of the biocover window (Pedersen et al., III). For these four campaigns the CH_4 oxidation rate was 205 / 145 g m^{-2} day^{-1}, which was caused by the higher CH_4 load in this area (Table 4).

Table 4: Average emission of CH_4 and CO_2 and the range of average CH_4 oxidation in 13 campaigns on window 1.1 (Pedersen *et al.,* IV).

Date	**$T_{compost}$**[a]	**ΔP**[b]	**n**	**CH_4 emission**	**CH_4 load**	**CH_4 oxidation**	
	C°	hPa h^{-1}		g m^{-2} day^{-1}	g m^{-2} day^{-1}	g m^{-2} day^{-1}	%
Total CH_4 oxidation campaigns[c]							
15/04/2008	17.1	0.2	13	73	115 / 97	42 / 24	37 / 25
23/04/2008	17.1	0.15	9	58	90 / 72	32 / 14	36 / 19
24/04/2008	17.1	-0.12	11	105	139 /121	34 / 16	25 / 13
08/05/2008	24.6	-0.08	17	102	178 / 144	76 / 42	43 / 29
04/06/2008	28.9	0.13	15	96	163 / 114	67 / 18	41 / 16
25/06/2008	28.9	-0.50	14	74	223 / 174	149 / 100	67 / 57
30/06/2008	28.9	0.43	24	133	264 / 215	131 / 82	50 / 38
01/12/2008	14.0	-0.38	37	373	404 / 390	32 / 17	8 / 4
09/04/2009	14.6	0.16	44	145	173 / 157	34 / 19	20 / 12
Average			**184**	**129**	**195 / 165**	**67 / 37**	**34 / 22**
Transect campaigns[e]							
Jul-08	37.8	-0.20	19	593	1091 / 995	498 / 401	46 / 40
Aug-08	38.5	-0.37	19	57	231 / 118	174 / 61	75 / 52
Nov-08	16.2	-0.20	33	854	944 / 925	90 / 71	10 / 8
Apr-09	13.7	0.08	48	222	412/ 398	59 / 45	14 / 11
Average transect			**119**	**432**	**669/ 609**	**205 / 145**	**36 / 28**
Average all			**303**	**222**	**341 / 301**	**109 / 70**	**32 / 23**

[a]Average compost temperature for the respective months presented in Pedersen et al. 2010b. [b]Pressure change over the measuring day (8 am to 4 pm) or during the campaign

The carbon mass balance is very sensitive to changes in the raw gas composition and big variation was seen at Fakse Landfill. Primarily it was wanted to use the gas ratios found in the bottom of each of the profiles. But as CH_4 was oxidized prior to reaching the gas distribution layer and gas concentration profiles indicated vertical gas transport, this would underestimate the total CH_4 oxidation (Pedersen *et al.,* III). Therefore it was decided to use the raw gas composition measured in the waste to ensure that an anaerobic raw gas composition was used and to include the CH4 oxidation in the waste and in the gas distribution layer in the calculated CH4 oxidation rates. It was believed that the installation of the biocover windows at Fakse Landfill had improved the aeration of the landfill and therefore enhanced CH_4 oxidation in the waste body.

4.5 Comparison of the mass balance approach and the stable isotopic approach

Pedersen *et al.* (IV) compared the carbon mass balance approach and the stable isotopic approach on a single measuring day in a compost biocover window and the results are listed in Table 5. The load and the CH_4 oxidation for the stable isotopic approach were calculated as an average between the surface $\delta^{13}C$-CH_4 and the highest $\delta^{13}C$-CH_4 in the profile (Chanton *et al.,* 2008b). For the stable isotopes the average CH_4 oxidation rate was 162 g CH_4 m^{-2} d^{-1} corresponding to percent removal of 20 %. For the carbon balance the average CH_4 oxidation rate was lower 77 / 65 g CH_4 m^{-2} d^{-1} corresponding to 11 / 9 %. The CH_4 oxidation rates determined by the carbon balance were lower than for the stable isotopic approach. This was the opposite of, what had been reported earlier. Christophersen *et al.* (2001) found that the carbon mass balance resulted in higher CH_4 oxidation rates than the stable isotopic approach. The lower results of the carbon balance could be caused by the fact that assimilation was not accounted for as a steady state was assumed (bacterial growth equals decay). Ait-Benichou *et al.* (2009) found a decrease in methanotrophic counts from November to December in a Canadian biocover, which they attributed to decreasing temperature. Based on this it was likely that April will be a period of regrowth after winter and therefore the assumption of zero assimilation was not fulfilled for this measuring day. To evaluate the importance of the assimilation the average CH_4 oxidation was tentatively calculated with 70 % assimilation, which was used for calculation of the maximum CH_4 oxidation rate in Einola *et al.* (2009). This resulted in an increase in the maximum average CH_4 oxidation rates from 65 g CH_4 m^{-2} d^{-1} to 112 g CH_4 m^{-2} d^{-1} corresponding to an increase in the CH_4 oxidation efficiency from 9 % to 14 %. This resulted in a maximum rate closer to the maximum rate found by the stable isotopic campaign (162 g CH_4 m^{-2} d^{-1} corresponding to 20 %). Adding assimilation to the carbon balance resulted in CH_4 oxidation rates, which was more comparable with the ones, found with the stable isotopic approach for the one measuring day in April.

Table 5: CH$_4$ oxidation rates calculated with the stable isotopic approach compared with CH$_4$ oxidation rates calculated with the carbon mass balance approach initially with an assuming zero respiration and accounting for respiration.

	Stable isotopes[a]			**Carbon balance**		
	CH$_4$ load	CH$_4$ oxidation		CH$_4$ load	CH$_4$ oxidation	
	g CH$_4$ m^{-2} day^{-1}	g CH$_4$ m^{-2} day^{-1}	%	g CH$_4$ m^{-2} day^{-1}	g CH$_4$ m^{-2} day^{-1}	%
E	1206	61	2	1093[b] / 1081[c]	-52 / -169	-5 / -6
F	987	238	23	874 / 862	124 / 112	14 / 13
F½	253	188	74	220 / 212	159 / 147	71 / 69
G	-	-	62	59 / 47	60 / 47	101 /101
H	-	-	-	32 / 20	32 / 20	100 /100
I	-	-	-	10 / 0	10 / 0	100 /100
Average	**815**	**162**	**20**	**730 / 718**	**77 / 65**	**11 / 9**

[a] CH$_4$ oxidation rate was calculated based on averages between surface emission and probe measurement as given by Chanton et al. (2008b) and measured α_{ox}. [b]Maximum CH$_4$ oxidation was calculated with a respiration of zero [c]Minimum CH$_4$ oxidation was subtracted temperature dependent respiration. [e]

For the stable isotopic approach the diffusion affects the C^{13} enrichment of samples taken in the flux chamber as ^{12}C diffuses faster than ^{13}C. Therefore the samples in the flux chamber underestimate the CH$_4$ oxidation rate. The same affect should theoretically exist for the carbon mass balance, as CH$_4$ diffuses faster than CO$_2$ and therefore the ratio in the flux chamber underestimates the CH$_4$ oxidation rate. Therefore it could be argued that the CH$_4$/CO$_2$ ratio in the subsurface should be used together with the total flux in the flux chamber to calculate a “diffusion correlated” CH$_4$ and CO$_2$ emission. Thus this approach was not tested in the inherent study.

4.6 Field scale CH$_4$ oxidation rates in the literature

Table 6 shows the performance of the most recent field scale biocover studies in the literature. Different designs have been tested and both evaluation methods and load of CH$_4$ to the systems were very different. The study done by Cabral *et al.* (2010) stands out with very high loads of CH$_4$ (1212 and 408 g CH$_4$ m^{-2} d^{-1}), but also very high CH$_4$ oxidation rates (252 and 335 g CH$_4$ m^{-2} d^{-1}). One full-scale study was reported (Einola *et al.,* 2009), but the load of CH$_4$ was lower (0.7-23 g CH$_4$ m^{-2} d^{-1}) than what was found in the study by Pedersen *et al.* (IV) and therefore the CH$_4$ oxidation rates were also lower (2.7-27.3 g CH$_4$ m^{-2} d^{-1}), but the efficiency was higher (25-84 %). Dever (2009) and Pedersen *et al.* (IV) worked with similar loads; 168 and 160 g CH$_4$ m^{-2} d^{-1} (Dever, 2009) and 195 /

165 g CH_4 m^{-2} d^{-1} (Pedersen *et al.,* IV). Dever (2009) reported CH_4 oxidation rates of 101 and 108 g CH_4 m^{-2} d^{-1} respectively (Dever, 2009), which was significantly higher than found in Pedersen *et al.* (IV) (67 / 37 g CH_4 m^{-2} d^{-1}). This could mainly is attributed by the uneven distribution in the load of the landfill gas in the study by Pedersen *et al.,* (IV). The remaining of the studies in Table 6 worked with lower loads than in Pedersen et al., (IV) and the results were therefore not directly comparable.

Table 6: Summary of field scale results in biocover studies.

Reference	*Scale and size*	*Location*	*Biocover material*	*Biocover depth*	*Sieving diameter/ Porosity*	*Depth of gas distribution layer*	*Method for determination of CH_4 oxidation*	*Season/ seasonal*	*Load*	*CH_4 oxidation rate*	*CH_4 oxidation efficiency*
				m	m^3 m^{-3}	m			g m^{-2} d^{-1}	g m^{-2} d^{-1}	%
Huber-Humer et al. 2009	25 x 25 m	Austria	50 % SS compost and 50 % woodchips	0.9	Coarse	0.3 coarse gravel	Reference cell	Seasonal	39.5	34	85
	-	-	MSW compost	0.9	<25 mm	0.3 coarse gravel	-	Seasonal	39.5	39	99
	-	-	50 % SS compost and 50 % woodchips	0.4		-	-	Seasonal	39.5	27	68
	-	-	50 % SS compost and 50 % woodchips	0.3		0.3 compacted loam	-	Seasonal	39.5	29	74
Cabral et al. 2010[a]	2.75 x 9.75 m	Canada	Compost of SS and industrial sludge (5) and sand (1)	0.9	<12 mm 0.63	0.2	Stable isotopes	June-September	2212[a]	252	11
			Same compost (1) and gravel (1)	0.4		0.8		June-September	408[a]	335	72
Einola et al. 2009	3.9 ha	Finland	Peat and sludge compost (40:60 v/v)	0.5		0.5	Carbon mass balance	October	0.8-9.8	2.8-11.8	29-83
								February	0.7-2.7	2.7-4.8	25-57
								June	3.8-23	8.2-27.3	46-84
Philopoulos et al. 2008	9.3 m^2	Canada	Yard waste compost	1.5	0.69	0.8	Carbon mass balance	Oct-May	37	28	76
	9.3 m^2					0.8 With gas well			54	36	68
	20.9 m^2					0.8			1.2	0.4	35
Dever, 2009	3 x 3 m	Australia Temperate Climate	Yard waste compost and 10 % wood chips	1.2	0.64	0.5		Seasonal	168	101	60
	3 x 3 m		MSW compost and 10 % wood chips	1.2	0.71	0.5	Carbon mass balance		288	94	32
	3 x 3 m		Yard waste compost	1.2	0.72	0.5			542	62	12
	3 x 3 m		MSW compost and 20 % wood chips	1.2	0.73	0.5			160	108	67

[a] The result is an average between the results in the flux chamber and in the shallow subsurface as this was the method recommended by Chanton et al. (2008a)

5 CH_4 oxidation and temperature in compost biocovers

5.1 CH_4 oxidation at high temperatures

CH_4 oxidation is temperature dependent and optimum temperature is 25-35 °C (Scheutz *et al.,* 2009a). At 10 to 30°C the temperature response is approximately exponential with Q_{10} values from 1.7 to 4.1 (Scheutz *et al.,* 2009a). Results indicate that in landfill settings type I methanotrophs tend to have lower temperature optimum than type II methanotrophs (Scheutz et al., 2009a). These results were based on investigations done on mineral soils, but Mor *et al.* (2006) investigated the temperature response of CH_4 oxidation in garden waste compost. They found that if the methanotrophic bacterial community was allowed to adapt to the specific temperature during a period of days, similar results were reached from 15 to 30 °C. For another compost with a lower water content a different picture was seen, as the highest CH_4 oxidation rate was found at 40°C for five out of seven measurement of CH_4 oxidation rate over time (Mor *et al.,* 2006). For studies on landfill cover soil a significant decrease in CH_4 oxidation rate is reported above 30-35°C degrees (Stein and Hettiaratchi, 2001; Scheutz and Kjeldsen, 2004) and in Scheutz & Kjeldsen (2004) the CH_4 oxidation rate was zero at 50°C and very reduced at 45 °C (approximately 10 $\mu g\ CH_4\ g^{-1}\ DM\ h^{-1}$ compared to 120 $\mu g\ CH_4\ g^{-1}\ DM\ h^{-1}$ at optimum temperature).

Due to the results found by Mor *et al.* (2006) and the high temperatures found in the biocover at Fakse Landfill (Pedersen *et al.,* III) it could be hypothesised that there exist methanotrophic bacteria, which is adapted to higher temperature than the optimum reported by Scheutz et al. (2009a). In this case such methanotrophs should exist in tropical landfill cover soils. Visvanathan *et al.* (1999) investigated how the methanotrophic activity of tropical landfill soils was affected by temperature. From batch incubations they found that the optimum temperature was 30-40 °C and the CH_4 oxidation rate was close to zero at 45 °C. In a column study the dependency on temperature was assessed by determining CH_4 oxidation rates diurnally with temperatures, which ranged from 30 °C (at night) and 37-40 °C (during the day). The authors found the highest CH_4 oxidation rate during the night and this was explained by CH_4 oxidation being limited by the high temperatures during the day (Visvanathan *et al.*, 1999). An indirect affect of the high temperatures were that the upper layers in the soil column dried out, which resulted in almost zero CH_4 oxidation capacity in this zone (Visvanathan *et al.*, 1999). This phenomenon is well known in the literature on biofilter performance and Mysliwiec *et al.* (2001) found that it was one of the factors having the greatest detrimental effect on biofilter performance. For biofilters, which are run under very intensive conditions this is modeled in order to be able to add the right amount of water to achieve optimum moisture conditions for CH_4 oxidation (Chandrakanthi *et al.*, 2005).

Park *et al.* (2009) also investigated landfill cover soil collected from a tropical landfill (Korea). They found the highest CH_4 oxidation rates at 35°C, which was the highest temperature investigated in batch incubations (Park *et al.*, 2009).

From the above it is clear that the CH_4 oxidation rates at high temperatures should be investigated further. The investigation should be done with compost samples taken in the autumn where high temperatures are measured in the compost (Fig. 14) and if it can be concluded that inhibition of the CH_4 oxidation rates exist above 35 to 40 °C, this has implications for the suitability of composts, as biocover material. Indeed the maturity of the compost materials and their thermal conductivity should be assessed thoroughly as these two parameters together are believed to cause the high temperatures (together with CH_4 oxidation). In Pedersen *et al.* (I) it was found that the main part of the respiratory activity originated from the branches in the raw compost, therefore a sieved compost would produce less heat due to the lower microbial activity. A sieved compost could also be expected to have higher thermal conductivity due to the lower air filled pore space, which will allow the heat to escape to the atmosphere to a greater extend. Thermal conductivity will be assessed in the next chapter.

5.2 Thermal conductivity and heat capacity

The thermal conductivity describes a mediums ability to conduct heat and it describes the power loss (Watt) through a medium.

$$\alpha = \frac{K}{\rho \cdot C_p}$$ (Chandrakanthi *et al.*, 2005)

Where α is the thermal diffusivity ($m^2\ s^{-2}$), K is the thermal conductivity ($W\ m^{-1}\ K^{-1}$), ρ is the density ($kg\ m^{-3}$) and C_p is the specific heat capacity ($J\ kg^{-1}\ K^{-1}$). The specific heat capacity is the thermal energy required (J) to increase 1 kg the compost 1 ^{o}C.

The thermal conductivity of compost depends on several factors, such as texture, organic content, water content and bulk density (Chandrakanthi *et al.,* 2005). Hettiarachchi (2005) showed that the thermal conductivity of compost was significantly lower than a range of mineral soils. This means that the composts ability to conduct heat is lower and therefore heat will be stored in the compost compared to more mineral biocover material (Hettiarachchi, 2005).

Chandrakanthi *et al.* (2005) investigated the thermal conductivity of leaf compost, which showed high CH_4 oxidation rates in a column study (Wilshusen *et al.,* 2004). They found that compost thermal conductivity increased linearly with the degree of saturation. This phenomenon is caused by the fact that the thermal conductivity of water (0.6 W m^{-1} K^{-1}) is higher than that of air (0.025 W m^{-1} K^{-1}) (Chandrakanthi *et al.,* 2005). Ahn *et al.* (2009) investigated the thermal properties of a range of composting bulking materials. The highest thermal conductivities were found for saw dust, soil compost blend, beef manure and turkey litter. Silage showed medium values and low values of thermal conductivity was found for wheat straw, oat straw, soy bean straw, corn stalks, alfalfa hay and wood shavings (Ahn *et al.*, 2009). The study was performed in order to determine, which bulk materials could be used to maximise temperature in the composting process for successful inactivation and killing of pathogens (Ahn *et al.*, 2009). Thus it can also be used in the evaluation of bulk materials suitable for biocovers if too high temperatures should be avoided. Generally very coarse materials such as hay and shredded wood had low thermal conductivities and these materials should be avoided if it is believed that there is a risk of high temperatures limiting CH_4 oxidation. In a temperate climate the biggest concerns will be the performance of the biocover system in the winter, where atmospheric temperature is low and the water content is high (Christophersen *et al.*, 2001), which could promote the choice of very coarse and structured materials.

Huber-Humer *et al.* (2008) also found higher temperatures in coarse sewage sludge compost, than in finer municipal solid waste compost. This difference they attributed to the structure of the materials, as the respiratory activity of the two materials was similar (1.7 and 3.1 mg O_2 g DM^{-1}). They found that the coarse structure of the compost ensured a nearly constant temperature throughout the year in 130 to 80 cm depth (Huber-Humer *et al.,* 2008). This was explained by the lower thermal conductivity of coarse substrates with a high amount of air filled pore space and a lower compaction (Huber-Humer *et al.,* 2008).

5.3 Temperatures in a compost based biocover system

Pedersen et al. (III) measured temperature profiles during 1 year in a compost based biocover window from November 2008 to October 2009. The temperatures were averaged over depth and month and the results can be seen in Fig. 13 together with the atmospheric temperature. The temperatures in the compost biocover were generally higher than the atmospheric temperature. This was mainly attributed to the heat produced by aerobic degradation of the compost and the low thermal conductivity of the compost compared to mineral materials.

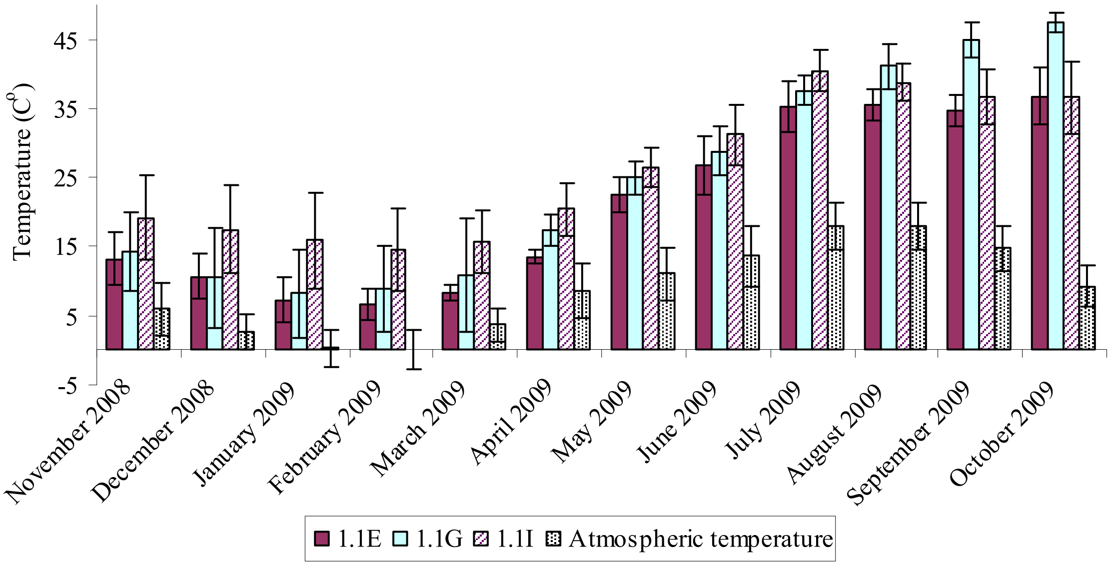

Fig. 14: Seasonal change in temperature for the 3 transect point E, G and I. Average over depth and time, n≥7195=5(depth)*1439 (time). Compared with atmospheric temperature from DMI (Danish Meteorological Institute) 14.5 km SW of the landfill (Pedersen *et al.,* III).

The low thermal conductivity of composts insulates well and ensures an environment with stable temperatures where methanotrophic bacteria are allowed to adapt to lower temperatures (Mor *et al.,* 2006) and CH_4 oxidation should be able to proceed right through the winter. This is especially the case if a low CH_4 load is ensured and the CH_4 oxidation zone is located in the deeper layers in the biocover. In Fig. 15 the gas concentration profiles were depicted together with the temperature profile from Pedersen et al. (IV). In A) December 1st 2008 the landfill gas load is high and the CH_4 oxidation zone is pushed upwards where it is subject to low temperatures (5 °C), whereas the temperature would have been

15 to 20 °C if the CH_4 oxidation zone was located in 50-80 cm. In B) the landfill gas load is lower and the CH_4 oxidation zone is situated between 20-80 cm based on the CH_4 and CO_2 profile. In this depth the temperature is around 15 °C in April and generally the temperature profile in April is straighter than in December. The temperature is actually lower in the bottom of the biocover in the beginning of April than in the beginning of December. Generally the temperature in the bottom of the filter (80 cm) was never below 15 °C and the temperature in 60 cm was never below 10 °C for the point G. Therefore it could be concluded that a low landfill gas load would result in temperatures (>15 °C), around the year, which were close to optimum for CH_4 oxidation (Mor *et al.,* 2006). Thus high temperatures in late summer and early autumn could diminish CH_4 oxidation as discussed above (Fig. 14).

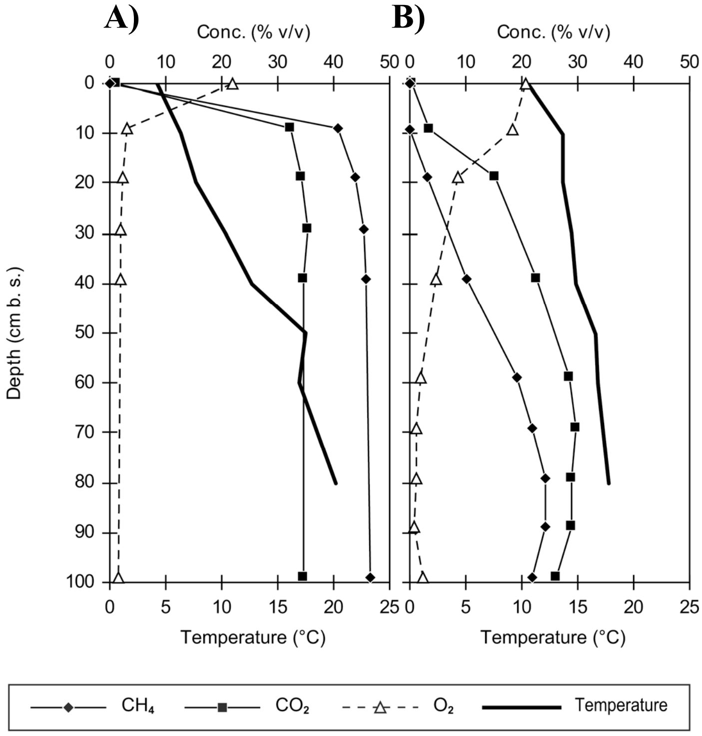

Fig. 15. A) Temperature and gas concentration in the profile G on Dec 1st 2008, B) Temperature and gas concentration in the profile G on April 9th 2009 (Pedersen *et al.,* IV).

6 Conclusions

Composts are suitable materials for biocovers as high CH_4 oxidation rates can be obtained. If suited mineral materials are locally available they should be considered as their stability will be better over time. At Fakse Landfill no suitable mineral materials were available and the transport of mineral materials to the site was found both economically and environmentally unsound. The compost chosen for Fakse Landfill was 4 year old raw compost as it achieved an average CH_4 oxidation rate of 108 g CH_4 m^{-2} d^{-1} in a column incubation and was cheap and readily available at the landfill. Thus the CH_4 oxidation rates were declining over time and the CH_4 oxidation rate on the last day (Day 111) of measuring was 76 g CH_4 m^{-2} d^{-1}. Based on these two rates the size of the biocover was calculated and this resulted in a size of 6,000 m^2 and 9,700 m^2, which corresponded to 7 % and 9 % of the total area of the landfill. The five materials tested in a column incubation were ranked from the results of batch incubations and column incubations and similar results were achieved. Therefore it was concluded that batch incubations were sufficient when wanting to rank locally available materials for full-scale biocover application. It was attempted to calculate CH_4 oxidation rates per surface area from the batch incubations, but the achieved results were different from the CH_4 oxidation rates obtained in the column incubation. Therefore CH_4 oxidation rates for design purposes cannot be determined solely from batch incubations. Thus it was advised that the CH_4 load to full scale biocovers was kept low, despite the CH_4 oxidation rates achieved in the laboratory, as this could diminish the risk of EPS formation and other negative impact from environmental factors in the field.

The performance of the full scale biocover at Fakse landfill was diminished by inhomogeneous gas distribution giving rise to low loads and hotspot formation. Even that a range of initiatives to improve the load to the biocover windows were performed FID screenings of CH_4 concentrations and flux measurements of CH_4 and CO_2 revealed that a big part of the CH_4 was emitted around the leachate collection wells. Hotspots were observed on some biocover windows and they were located around the edges or were trenches were dug in order to improve the inflow of landfill gas to the windows. The uneven gas distribution in the windows showed that the depth of the gas distribution layer (15 cm) was not sufficient. Therefore it was concluded that future biocover systems installed at landfills with a leachate collection system should attempt to use this installation for distributing the load of landfill gas to the biocover system. Additionally gas distribution layers should be thicker.

It was found that the performance of a selected biocover window was diminished by both spatial and temporal differences in the load. Under the gas distribution layer 4 m of clay mixed with waste was identified and despite this, high emissions were seen close to the edges of the biocover window. It was therefore speculated that the load of landfill gas to the biocover window originated from other locations in the landfill, as the gas distribution layer was covering the entire cell and therefore landfill gas could flow from other locations to the biocover window. Additionally it was suggested that the gas transport in the window was not only vertical but also horizontal, as both gas concentration profiles and flux measurements indicated this. However the phenomenon was found hard to prove. The emission from the biocover window was very dynamic and depended strongly on the development in the atmospheric pressure. The CH_4 oxidation zone moved upwards during pressure decreases where it was subject to diurnal temperature variations. Advective flux of landfill gas into the biocover was not diminished by the measured water content, but diffusion of O_2 was limited during winter. Generally it was found that the physical gas transport was very important, both for the full scale system (securing the landfill gas load to the biocover windows) and also within the individual windows (distribution of the load of landfill gas in time and space). Different factors affect these issues; landfill properties, dimensioning of the biocover, gas distribution layer and the gas filled porosity, which depends on the selected material and the water content.

CH_4 oxidation rates were determined in 13 campaigns over the year by the carbon mass balance approach and were calculated by two different methods 1) assuming that compost respiration was zero and 2) accounting for temperature dependent compost respiration. For 1) the average CH_4 oxidation rate for the 13 campaigns was 109 g CH_4 m^{-2} d^{-1} corresponding to 32 % of the calculated CH_4 load and for 2) the average CH_4 oxidation rate for the 13 campaigns was 70 g CH_4 m^{-2} d^{-1} corresponding to 23 % of the calculated load.

In general it was found that easily applicable, cheap and accurate methods measurement of CH_4 oxidation rates in passive systems is scarce. However the carbon mass balance was identified as a sufficient tool to assess performance of biocover windows in development. It is recommended that attention is drawn to the determination of the raw gas composition as it affects the results. For each measurement campaign raw gas composition both locally and deeper in the landfill should be determined. The use of local raw gas composition measured in the bottom of the biocover window is the most appropriate method, but it will underestimate CH_4 oxidation rates if CH_4 oxidation takes place in the gas distribution system and in the waste. Additionally this approach is not appropriate if the gas transport is not vertical and in this study it is plausible that a horizontal CH_4 oxidation zone exists. In these cases it will be more appropriate to adapt the raw gas composition found in deep probes in the landfill. The carbon mass balance was concluded to be an adequate method because it is cheap and therefore high frequency measurement in both time and space can secure good estimates and taking into account the magnitude and number of uncertainties when measuring emissions the uncertainty on the carbon mass balance was not found to be mayor.

For the stable isotopic approach the results were very sensitive to the sample location (surface chamber or subsurface sample) and especially at high CH_4 load the result were very different. The results were less sensitive to the fractionation factor, but sampling for the determination of the fractionation factor was difficult especially in biocover systems were CH_4 load varied both spatially and over time. Thus this variation will most likely be seen in most biocover systems. It was concluded that it would not cause very different results if a fractionation factor from the literature was adopted.

Very high temperatures (up to 50°C) were measured in the biocover window in October and this could diminish CH_4 oxidation. The high temperatures were attributed to the heat produced from aerobic degradation of mainly the branches in the compost and the low thermal conductivity of the compost. It was found that materials for biocovers could be engineered depending on whether higher or lower insulation properties were desirable. Generally very coarse and structured materials would have better insulating effect.

Future biocovers should have a gas distribution layer of at least 0.5 m and a compost layer of at least 1.2 m. Due to the high cost associated with buying and transporting mineral materials to the site, local material availability should preferably be a part of the very initial assessment, when identifying landfills suited for implementation of biocovers.

To secure higher efficiency during the entire year in northern European conditions load should be lower than in the inherent study. A load of 50 g CH_4 m^{-2} d^{-1} to 100 g CH_4 m^{-2} d^{-1} is suggested. An even lower load will increase the efficiency especially during situations of atmospheric pressure decrease combined with low temperature (autumn and winter situation). In general a low CH_4 load will diminish many of the negative effects on CH_4 oxidation efficiency identified in the inherent study, for example; low temperature in the upper layers, atmospheric pressure changes causing overload, formation of EPS and drying of the upper layer.

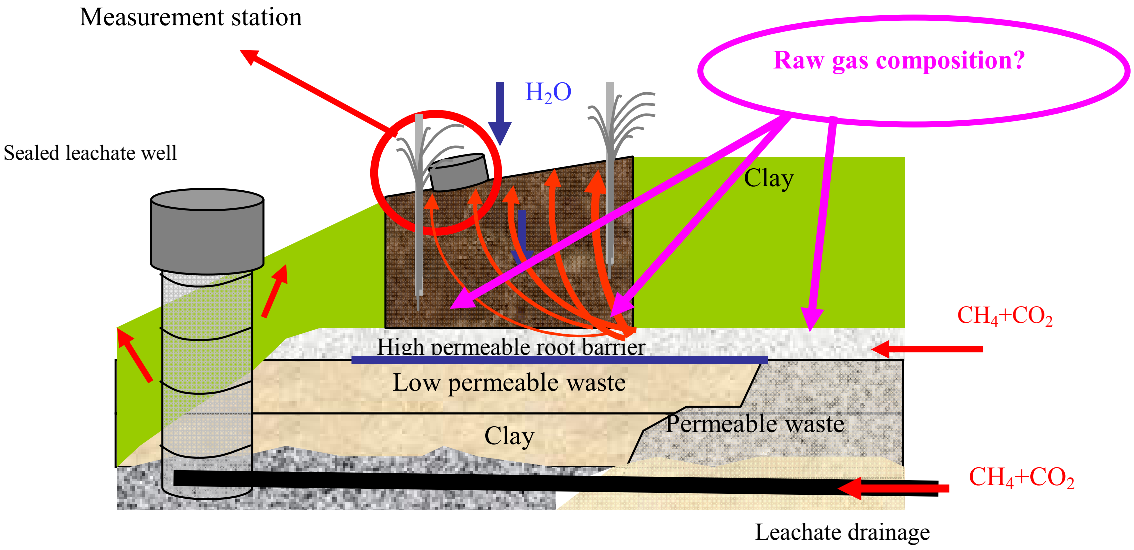

Fig. 16. Conceptual model of the gas transport at the Fakse Landfill Biocover system

7 Further research

The research done during my PhD and reported in this thesis has identified some areas where further investigations are needed

- The implementation and research on different designs of full scale biocovers should aim at reaching higher efficiencies, but the focus should be kept on low tech solutions.

- The necessary depth of the gas distribution layer should be investigated as it is a very cost sensitive parameter.

- The above should be done in combination with testing less permeable biocover materials, as this is believed to reduce the dynamics of the system and improve the gas distribution, thus the diffusion of O_2 should be ensured. It is hypothesized that lower permeability of the biocover material compared to the gas distribution layer will increase gas distribution

- High temperature batch incubations should be performed with compost samples taken from the biocover in autumn.

- Seasonal studies of potential CH_4 oxidation at different temperature and moisture regimes should be done to investigate the seasonal changes in the methanotrophic activity.

- Diffusivity of the compost should be measured at different water contents, which, together with the temperature and water content dependency studies, would allow modelling of CH_4 oxidation rates from gas concentration profiles.

- Generally the track of interdisciplinary studies with both physical gas transport studies and methanotrophic activity should be continued. Batch incubations of collected compost samples from the field biocover during the year, is a cheap and efficient way of determining potential CH_4 oxidation and evaluate, which parameters limit CH_4 oxidation.

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9 Appendices

I Pedersen, G.B., Scheutz, C., Kjeldsen, P (2010a). Mitigation of methane emissions from Fakse Landfill using a biocover system II: Material availability and properties. Manuscript submitted to Waste Management. 2010a.

II Fredenslund, A.M., Pedersen, G.B., Scheutz, C., Kjeldsen, P. (2010). Mitigation of methane emissions from Fakse Landfill using a biocover system III: Design and performance. Manuscript for submission to Waste Management. 2010.

III Pedersen, G.B., Scheutz, C., Kjeldsen, P. (2010b) Field study on a biocover window at Fakse Landfill, Denmark I: Short term dynamics and seasonal changes in methane emission. Manuscript for submission to Science of the Total Environment. 2010b.

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The papers are not included in this www-version, but can be obtained from the Library at DTU Environment. Contact library@env.dtu.dk or Department of Environmental Engineering, Technical University of Denmark, Miljoevej, Building 113, DK-2000 Kgs. Lyngby, Denmark.

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