



## **Current MSW Industry Position and State-of-the-Practice on LFG Collection Efficiency, Methane Oxidation, and Carbon Sequestration in Landfills**

Prepared For:

**Solid Waste Industry for Climate Solutions  
(SWICS)**

Presented by:

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Version 2.2, Revised January, 2009

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## EXECUTIVE SUMMARY

In July 2007, the Solid Waste Industry for Climate Solutions (SWICS) released its first white paper titled *Current MSW Industry Position and State-of-the-Practice on LFG Collection Efficiency, Methane Oxidation, and Carbon Sequestration in Landfills*. In the 2007 paper, SWICS proposed a revision to the existing California Air Resources Board methodology for calculating methane emissions from landfills. SWICS intent was to create a methodology that would result in more accurate inventories of methane emissions from landfills.

This document is a revision and update to the July 2007 SWICS white paper based on the critical review of the previous white paper by several academic experts in the field of waste management. The expert review re-evaluated the existing peer-reviewed sources, added several additional sources, and arrived at more supportable conclusions about greenhouse gas (GHG) emissions from landfills.

Methane is a very important contributor to global warming, with a 100 year global warming potential 21 to 25 times that of carbon dioxide (IPCC 2007). Municipal solid waste (MSW) landfills are recognized sources of methane through emissions of landfill gas (LFG).

The objective of this paper is to present the MSW industry position and state-of-the-practice on LFG collection efficiency, methane oxidation, and carbon sequestration in landfills based upon reviewed literature of the industry. Literature was reviewed to:

- Compile and critically analyze published information on LFG collection efficiencies;
- Compile and critically analyze published information on methane oxidation in different types of landfill cover soils;
- Evaluate carbon storage factors (CSFs) calculated for different types of MSW;
- Evaluate the impact of landfill carbon sequestration on GHG emissions accounting and development of reduction strategies and policies.

LFG collection efficiency is the percentage of generated LFG a system collects. Current inventory methodologies are very sensitive to changes in collection efficiency. Even small changes in efficiencies can result in large changes in calculated emissions. LFG system owners and operators believe that collection efficiencies greater than 75 percent, the default value now in common use, are commonly achieved at sites with well designed and operated LFG collection and control systems. LFG system owners and operators believe the use of default values prevents individual landfill sites from demonstrating higher collection efficiency using available site-specific information. The exclusive use of the default value also creates disincentives for those owner/operators to put in the extra effort to achieve a greater collection efficiency because those efforts would not be recognized by the default value and may in fact result in higher calculated emissions. Based on a literature review, LFG collection efficiency values ranging from 50 percent to 99 percent are proposed.

LFG that is not collected passes through the landfill cover where a fraction of the methane is oxidized into carbon dioxide. It is clear that the fraction of methane oxidized in landfill cover soils is much greater than the default value of 10 percent. Of the 47 determinations of methane oxidation reviewed in this document, only four oxidation values were less than 10 percent. The default value of 10 percent should be updated, based upon technological advancements in measurement approaches and the current

research on the topic. Based on a literature review, the values for the percent of methane oxidized in a landfill covers ranges from 22 percent to 55 percent.

SWICS reviewed and has herein summarized the positions of the United States Environmental Protection Agency (USEPA), Intergovernmental Panel on Climate Change (IPCC), California Energy Commission (CEC), and the California Air Resources Board (CARB) on carbon sequestration in landfills and other industries. An exhaustive review of the available technical literature was also conducted. Based on this review, SWICS is proposing that the research by Dr. Morton Barlaz of North Carolina State University and the USEPA be used to develop carbon storage values for organic wastes contained in the MSW stream.

This paper also describes how these proposed values for collection efficiency, methane oxidation, and carbon sequestration could be used to replace the current CARB values for collection efficiency (75%), methane oxidation (10%), and carbon sequestration (informational only).

In conclusion, if the proposed new values for collection system efficiencies, methane oxidation in cover soils, and carbon storage factors for carbon sequestration are used in GHG emissions inventories, GHG emissions from landfills will be significantly lower and the reduction potential will be higher than the current estimates. The differences in the previously used values and the proposed values need to be taken into account in order to more accurately define the state-of-the-practice technology used in the engineered landfills of today and allow site-specific estimates of landfill methane emissions.

# 1 INTRODUCTION AND OBJECTIVES

Methane is a very important contributor to global warming, with a global warming potential 21 to 25 times that of carbon dioxide (IPCC 2007). Municipal solid waste (MSW) landfills are recognized sources of methane through emissions of landfill gas (LFG).

Current methods used for calculating methane and other pollutant emissions from landfills have been the subject of intense scrutiny. One concern about the current methods is they use a generic default data that do not consider local conditions that can influence emissions. A number of scientific advances have been published in the last 10 years that require the updating of these default values. Issues that need to be addressed include collection efficiencies for LFG systems, methane oxidation in cover soils, and the importance of carbon sequestration in landfills.

Currently, the solid waste industry is evaluating various methods to more accurately determine methane emissions from landfills. Due to the high spatial variability of methane emissions, this determination has proven to be difficult, although there are a number of promising approaches on the immediate horizon. This white paper identifies the need for updates to the methane emissions protocol in this interim period where statewide inventories are being established and accurate measurement techniques are being validated. The following sections summarize a proposed strategy to update the current inventory protocol.

## 1.1 STATE OF CRITICAL NEED

This white paper details the current default methods for determining LFG collection efficiency and methane oxidation. With an understanding of the current status of these concepts, the critical need for the update of the default values associated with these methods becomes obvious. With respect to carbon sequestration, Section 5 identifies the importance of landfill carbon sequestration and that landfill greenhouse gas (GHG) inventories should include a sequestration offset value.

### 1.1.1 LFG Collection Efficiency

According to the United States Environmental Protection Agency's (USEPA's) "Compilation of Air Pollutant Emission Factors" (AP-42) (USEPA, 1997), researchers and practitioners estimated collection efficiencies to typically range from 60 to 85% at landfills with LFG collection installed. The most commonly assumed default efficiency has been 75% although higher efficiencies have been demonstrated at some sites, particularly those engineered to control emissions.

A review of available data and industry information regarding LFG collection efficiency was conducted by the USEPA in 2002. Eastern Research Group, Inc. (ERG), a USEPA contractor, conducted this review. In an internal memorandum dated October 24, 2002, the USEPA recommended a 75 percent default LFG collection efficiency (Leatherwood, 2002). Other key points from USEPA's review are included below.

- Overall, minimal data on LFG collection efficiency exist. Industry contacts cited the difficulty in documenting uncontrolled LFG emissions as the primary reason.
- Documenting uncontrolled LFG emissions is problematic because:
  - The high spatial variability of LFG emissions makes it very difficult to accurately quantify LFG emissions.
  - LFG generation rates are highly variable due to the heterogeneity of MSW and variations in rainfall and landfill temperature.
- LFG emission levels are site-specific and vary over time and spatially; therefore, representative collection of samples is difficult.
- LFG systems historically have been satisfied with capturing a majority of the LFG generated to achieve compliance or some other goal. As a result, LFG system owners and/or operators have not been particularly interested in expending additional efforts in trying to determine actual LFG emissions.

Most of the published sources cited by the memorandum are from 1993 or earlier, at least 15 years old. Consequently, these sources do not reflect LFG system operational experience after implementation of USEPA's New Source Performance Standards (NSPS) 40 Code of Federal Regulations (CFR) Part 60, Subpart WWW. By December 1998, higher efficiencies were necessary for NSPS Subpart WWW compliance. Most of the collection efficiency estimates in the Leatherwood memorandum were based on speculation, not on field studies. A current review of published information on LFG collection efficiency is presented in Chapter 3.

Furthermore, the default 75% collection efficiency does not take into account the different LFG collection systems that are utilized at landfills. For example, a LFG collection system designed for NSPS compliance is far more capable of higher collection efficiencies than a LFG migration control system. Using a default value of 75% for both of these systems is invalid.

### **1.1.2 Methane Oxidation in Landfill Cover Soils**

The USEPA AP-42 document categorizes emission sources at landfills as uncontrolled emissions or controlled emissions. Controlled emissions are defined as emissions that are typically controlled by collection of gas through a gas collection system and destruction of the gas through combustion, most typically a flare. As stated in the previous section, uncontrolled emissions from landfills are difficult to estimate. USEPA's AP-42 document uses a theoretical, first-order methane production model as the basis of the uncontrolled emissions estimate. Equation 5 of Section 2.4 of AP-42, below, shows the formula used to calculate the uncontrolled emissions rate:

$$CM_p = [UM_p * (1 - \eta_{col}/100)] + [UM_p * \eta_{col}/100 * (1 - \eta_{cnt}/100)]$$

Where:

$CM_p$  = Controlled mass emissions of pollutant P, kg/yr,



$UM_p$  = Uncontrolled mass emissions of P, kg/yr (from equation 4, section 2.4-4, AP-42 using first-order model)  
 $\eta_{col}$  = Collection efficiency of the LFG collection system, percent; and  
 $\eta_{ent}$  = Methane destruction efficiency of the LFG control or utilization device, percent.

This equation assumes all of the gas that is not collected by the LFG control system is emitted into the atmosphere. This equation does not take into account methane oxidation in the landfill cover soils that can dramatically reduce methane release.

Furthermore, the AP-42 states that “average oxidation of methane (on a volumetric basis) in some laboratory and case studies on landfill covers have indicated ranges from 10 percent to over 25 percent with the lower portion of the range being found in clay soils and higher in topsoils.” Due to the uncertainty involved and the lack of a standard method to determine oxidation rate, the USEPA recommends the default factor of 10% by volume methane oxidation for landfills with low permeability cover systems. This is termed a “conservative approach” by the USEPA. This default methane oxidation rate is dated and needs to be updated based on current engineering technologies of landfill cover soils and more recent research on this topic as detailed in this paper.

### **1.1.3 Carbon Sequestration in Landfills**

Carbon storage in landfills can significantly offset GHG emissions from landfills. The decision to include these factors and how they are utilized in a statewide inventory will depend on the accounting protocol employed.

A number of international and domestic protocols including the Intergovernmental Panel on Climate Change (IPCC), the USEPA, the Oregon Climate Trust, and the California Air Resources Board (CARB) recognize carbon storage in landfilled material as a sink in calculating carbon emissions inventories. These protocols recognize that when wastes of a biogenic origin are deposited in landfills and do not completely decompose, the carbon that remains is effectively removed from the global carbon cycle, or sequestered.

For example, the USEPA has published reports that evaluate carbon flows through landfills to estimate their net GHG emissions. The methodology the USEPA employed recognizes carbon storage in landfills. In these studies of MSW landfilling, the USEPA summed the GHG emissions from methane generation and transportation-related carbon dioxide emissions, and then subtracted carbon sequestration (treated as negative emissions) (USEPA, 2006).

Furthermore, the 2006 GHG emissions inventory published by the California Energy Commission (CEC) indicated that landfill disposal of urban wood waste and yard trimmings is a GHG sink. The report included only the categories of yard trimming and wood waste, and neglected sequestration from paper, boxes, yard waste, lumber, textiles, diapers, demolition, medical waste, sludge, and manure. In California, urban wood waste and yard trimmings represent only 16.4% of the total California waste stream and only 46% of sequestered carbon within landfills; therefore, restricting estimates of carbon storage to only these waste types

produces an extremely low value of overall carbon storage for the total amount of waste disposed.

The 2007 CARB landfill sequestration estimate includes sequestration from paper, boxes, yard waste, lumber, textiles, diapers, demolition, medical waste, sludge, and manure. CARB estimates the total carbon sequestration in landfill to be 4.94 million MTCE in 2005, which is 17.2 million metric tonnes carbon dioxide equivalent (MMTCO<sub>2</sub>E). CARB estimates that GHG emissions from landfills were 5.62 MMTCO<sub>2</sub>E in 2004, much less than the value of the carbon stored in the landfill.

In order to adequately calculate the net emissions from a landfill, all aspects of the carbon cycle as they relate to sequestration and emissions must be addressed. Therefore, the acceleration of carbon storage in all carbon sinks should be a part of any integrated GHG emissions plan to create an accurate GHG emissions inventory for landfills.

## 1.2 OBJECTIVES

Currently, CARB uses the following factors in its statewide inventory for landfills:

- 75% default value for collection efficiency where site-specific data are not provided;
- 10% default value for methane oxidation as a percentage of the gas not collected by a collection system;
- Carbon sequestration included as an informational item only for the landfill category.

The objective of this paper is to present the MSW industry position and state-of-the-practice on LFG collection efficiency, methane oxidation, and carbon sequestration in landfills based upon reviewed literature of the industry. Literature was reviewed to:

- Compile and critically analyze published information on LFG collection efficiencies;
- Compile and critically analyze published information on methane oxidation in different types of landfill cover soils;
- Evaluate carbon storage factors (CSFs) calculated for different types of MSW;
- Evaluate the impact of landfill carbon sequestration on GHG emissions accounting and development of reduction strategies and policies.

This paper will also provide interim techniques for inventorying baseline and annual GHG emissions for landfills until the CEC Study on landfill emissions is completed, and a new emission estimating method is available.

## 1.3 REVISIONS TO JULY 2007 SWICS METHODOLOGY

SWICS released an earlier version of this paper titled *Current MSW Industry Position and State-of-the-Practice on LFG Collection Efficiency, Methane Oxidation, and Carbon Sequestration in Landfills* in July 2007. In that paper, SWICS proposed revisions to the CARB landfill GHG methodology. This revision to the July 2007 SWICS methodology is based additional literature review, which includes the IPCC Fourth Assessment Report and revisions to the 2007 CARB GHG inventory.

In addition, this January 2009 revision is based on a critical review of the previous version by academic experts in the field. These experts re-reviewed literature sources, added literature sources, and completed a thorough evaluation of the research data to arrive at the most scientific supportable conclusions regarding appropriate default values.

The major changes to the previous SWICS methodology for this January 2009 revision are:

- The presence of a liner is no longer quantitatively considered when determining collection efficiency based on literature review;
- The collection efficiency ranges for each cover type have been changed based on a more detailed literature review and expert review;
- The methane oxidation percentage or rate is based on the cover material instead of the cover type based on a more detailed literature review and expert review;
- The methane oxidation percentage and rates have been updated based on a more detailed literature review and expert review;
- The carbon sequestration defaults have been updated to reflect a correction in the source material.

SWICS is comprised of public and private solid waste and recycling service providers throughout North America dedicated to advancing strategies and technologies to address the challenge of climate change and to introduce strategies to reduce and mitigate GHG emissions. We look forward to working on effective strategies to implement effective programs in California, the United States, and internationally to identify, evaluate, and implement ways to reduce GHG emissions from landfills.

The group responsible for the 2009 revisions to the methodology consists of:

- Morton Barlaz, Ph.D., Professor, Department of Civil, Construction, and Environmental Engineering, North Carolina State University;
- Jeff Chanton, Ph.D., Professor, Chemical Oceanography, Florida State University;
- Roger Green, Senior Scientist, Waste Management, Inc.;
- David K. Powelson, Ph.D., Associate in Research, Department of Oceanography, Florida State University;
- Patrick S. Sullivan, Senior Vice President, SCS Engineers.

## REFERENCES (SECTION 1)

Leatherwood, C. 2002. *Review of Available Data and Industry Contacts Regarding Landfill Gas Collection Efficiency*, Draft Memorandum to Brian Guzzone, Meg Victor, U.S. EPA, October 24, 2002.

USEPA, 1997. Compilation of Air Pollution Emission Factors, Report Number AP-42, 5<sup>th</sup> Ed. Supplement C, Office of Air Quality Planning and Statistics, U.S. Environmental Protection Agency, Washington, D.C.

USEPA, 1998. Greenhouse Gas Emissions from Management of Selected Materials in Municipal Solid Waste, U.S. Environmental Protection Agency, Washington, D.C. (EPA 530-R-98-013).

## 2 General Background

In the United States, as well as in California, municipal solid waste (MSW) is composed of approximately 30 to 50 percent cellulose, 7 to 12 percent hemicellulose, and 15 to 28 percent lignin on a dry weight basis, with cellulose and hemicellulose representing about 90 percent of the biodegradable portion of the MSW (Hilger and Barlaz, 2001). When MSW is buried in a landfill, a complex series of reactions occur in which anaerobic microorganisms decompose a portion of the organic fraction of the waste to carbon dioxide and methane.

The methane produced may be collected and flared or converted to energy, which oxidizes the methane to carbon dioxide. The methane can also be oxidized to carbon dioxide by methanotrophic bacteria in the landfill cover soil. Therefore, the ultimate fate of carbon placed in the landfill is sequestered or emitted as methane or carbon dioxide (Barlaz, 2007). In terms of atmospheric input, methane from landfills is considered an anthropogenic source of carbon while the carbon dioxide is considered biogenic in origin and not an anthropogenic source. Figure 1 depicts a simplified representation of the methane mass balance in landfills.

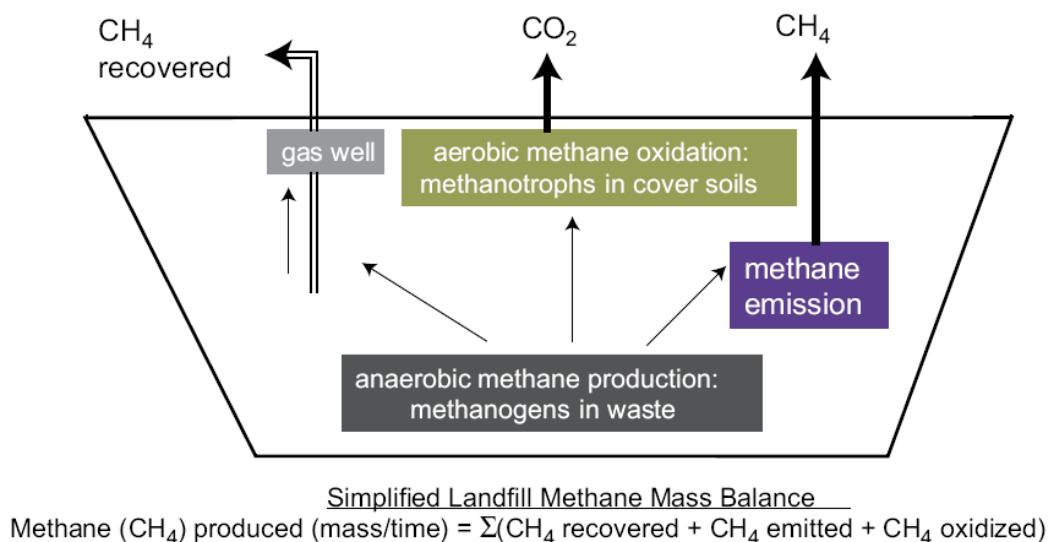


Figure 1. Simplified Landfill Methane Mass Balance (from Bogner et al, 2007)

Waste management affects greenhouse gas (GHG) emissions in several ways. The first is landfill methane emissions. The United States Environmental Protection Agency (USEPA) (1999) suggests that landfill methane accounts for about 4% of all GHG emissions, measured in terms of global warming potential (GWP). According to the Intergovernmental Panel on Climate Change (IPCC), the waste sector, including solid waste and wastewater, account for less than five percent of global GHG emissions. In the Fourth Assessment Report, the IPCC estimated that mitigation of landfill methane could reduce methane emissions from landfills by 70 percent at negative to low costs by 2030.

The other impacts are less visibly part of the waste management process. Both recycling and waste reduction often lead to decreased energy use and process emissions in the industry. Energy recovery from waste displaces fossil fuel consumption. Controlling landfill gas (LFG) has two benefits: landfill methane can be substituted for natural gas, a fossil fuel; and combustion converts methane to carbon dioxide, reducing its GHG impact.

## REFERENCES (SECTION 2)

Barlaz, M.A., 2007. *Recommended Protocol for Estimating Greenhouse Reductions Due To Landfill Carbon Storage and Sequestration*. Report submitted to Solid Waste Industry Climate Solutions Group (SWICS), 2007.

Higler, H., and Barlaz, M.A. 2001 “Anaerobic Decomposition of Refuse in Landfills and Methane Oxidation in Landfill Cover Soils”, Manual of Environmental Microbiology, 2<sup>nd</sup> Ed., Am. Soc. Microbiol., Washington D.C., pp. 696-718.

Intergovernmental Panel on Climate Change, 2001. Summary for Policy Makers of the IPCC WG III Third Assessment Report.

Okereke, V. 2006 *Analysis of Carbon Sequestration at the Cedar Hills Regional Landfill*, Presented at SWANA's 21st Annual Pacific Northwest Regional & Canadian Symposium, April 5-7, Richmond, B.C., Canada

USEPA, 1998. Greenhouse Gas Emissions from Management of Selected Materials in Municipal Solid Waste, U.S. Environmental Protection Agency, Washington, D.C. (EPA 530-R-98-013).

USEPA, 1999. Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-97, U.S. Environmental Protection Agency, Washington, D.C. (EPA 236-R-99-03).

Bogner, J., M. Abdelrafie Ahmed, C. Diaz, A. Faaij, Q. Gao, S. Hashimoto, K. Mareckova, R. Pipatti, T. Zhang. 2007. Waste Management, In Climate Change 2007: Mitigation. Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change [B. Metz, O.R. Davidson, P.R. Bosch, R. Dave, L.A. Meyer (eds)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

## 3 LANDFILL GAS COLLECTION EFFICIENCY

### 3.1 EXECUTIVE SUMMARY

To avoid the use of default values that have no bearing on site-specific conditions, it is critical to develop a methodology where new values can be calculated for landfill gas (LFG) system collection efficiencies in order to more accurately determine emissions from landfills with different types of cover and LFG collection systems. To this end, a detailed review of recent publications and available literature regarding LFG collection efficiency was conducted to ascertain the collection efficiency values from actual field tests where LFG emissions were measured and numeric collection efficiencies were calculated. These data were used to establish collection efficiency percentages relative to landfill cover types. These new values are presented in the bullets below:

- 50-70% (mid-range default = 60%) for a landfill or portions of a landfill that are under daily soil cover with an active LFG collection system installed (note that due to limited test data on daily soil covers, the selected range is based on the opinion of experts involved with the creation and review of this document);
- 54-95% (mid-range default = 75%) for a landfill or portions of a landfill that contain an intermediate soil cover with an active LFG collection system;
- 90-99% (mid-range default = 95%) for landfills that contain a final soil and/or geomembrane cover systems with an active LFG collection system.

Background information on LFG collection efficiency is summarized in Section 3.2 below. The literature review conducted for this document is detailed in Section 3.3, along with a summary of the research data. Section 3.4 describes landfill cover types, LFG collection system types, and LFG monitoring data, which can be used to make engineering judgments regarding collection efficiency values for a particular site. The proposed new collection efficiency values are restated in Section 3.5, including a methodology for site-specific application.

### 3.2 BACKGROUND ON COLLECTION EFFICIENCY

LFG collection efficiency is the amount of LFG that is collected relative to the amount generated by the landfill. The United States Environmental Protection Agency (USEPA), state, and local regulators often use assumed gas collection efficiencies to calculate landfill emissions for regulatory purposes. An efficiency of 75% (as a default value from a range of 60 to 85%) is frequently assumed as this value as set forth in USEPA's AP-42 document. The collection efficiency presented in AP-42 is based on engineering judgment, which was compiled by USEPA from various practitioners in the LFG industry, as opposed to test data.

In many cases, landfill owners/operators and regulatory agencies calculate the collection system efficiency from the actual volume of gas collected relative to the volume projected by LFG models for generation, for example using USEPA's LFG emissions model (LANDGEM). We believe that this approach is flawed. LANDGEM and other LFG models predict LFG generation using a variety of inputs to the model, which are difficult to determine on a site-specific basis. LANDGEM commonly



over predicts LFG generation in California where drier climates are commonplace, resulting in lower than actual collection efficiencies. Overall, the use of a LFG generation model introduces another source of error and uncertainty into the collection efficiency calculation and is not recommended at this time. Instead, it is preferred to estimate site-specific collection efficiency using certain landfill criteria combined with the measured amount of recovered LFG to estimate fugitive LFG emissions.

LFG system owners and operators believe that collection efficiencies greater than 75% are commonly achieved, and the use of default values prevents the sites from demonstrating this using available site-specific information. It also creates disincentives for those owner/operators to put in the extra effort to achieve a greater collection efficiency because those efforts would not be recognized by the default value.

The objective of this section is to present information on the factors involved in an evaluation of LFG collection efficiency and to summarize available literature, which can be used to develop site-specific values. In evaluating collection efficiency, it is important to recognize that the efficiency of a gas extraction system will vary continuously while a landfill is in operation. While waste is received daily, gas extraction systems are extended to new sections of a landfill less frequently (Emcon, 1997) such as on an annual basis. Thus, at any time in the life of a landfill, there may be sections with only daily cover and no gas extraction system, sections with intermediate cover and limited to near complete gas extraction, and areas with final cover and optimal placement of gas extraction wells. In addition, there are landfills that use horizontal trenches and other means of gas extraction from active areas and this may allow for a reduced period of time between refuse placement and gas recovery.

### 3.3 LITERATURE REVIEW

The objective of this section is to review literature reports on methane collection efficiency. This section is restricted to studies in which an attempt was made to quantify collection efficiency in some manner. It is recognized that there are other reports in which landfill experts have expressed judgment. For example, Leatherwood (2002) summarized a review of the literature and consultation with a number of experts, which later became the basis for the current AP-42 values. However, no quantitative estimates of collection efficiency were generated. While this judgment may in fact be quite reasonable, it is difficult to defend.

The only way to rigorously quantify the methane collection efficiency is to have a measure of collected methane and fugitive methane emissions from the same area at the same time. While measures of collected methane are readily available, measures of fugitive emissions are considerably more difficult to obtain and have only been reported for a few landfills. Spokas et al. (2006) summarized intensive field studies of the methane mass balance for nine individual landfill cells at three French landfills with well-defined waste inputs. The collection efficiency was calculated as the ratio of recovered gas to empirically modeled gas generation. Specifically, Spokas et al. used the following equation:

$$\text{CH}_4 \text{ generated} = \text{CH}_4 \text{ emitted} + \text{CH}_4 \text{ oxidized} + \text{CH}_4 \text{ recovered} + \text{CH}_4 \text{ migrated} + \Delta \text{CH}_4 \text{ storage}$$

Methane generation was estimated from a gas production model. Emitted methane was measured by using either static chambers or an atmospheric tracer technique. Methane oxidation was measured by using a stable isotope technique that provides a conservative estimate of oxidation. Recovered methane was based on direct measurements at each landfill, and methane migration was based on calculations of methane diffusion through liners. Maximum potential methane storage was calculated from an estimate of waste porosity and changes in methane concentration and used as an upper limit of the value required to close a mass balance. For this report, the data were recalculated where collection efficiency was defined as:

$$\frac{\text{methane collected}}{\text{methane collected} + \text{emissions} + \text{oxidation} + \text{migration}} \quad (1)$$

The results are summarized in Table 3.1. As presented, collection efficiencies for final clay covers were uniformly above 90% while the collection efficiency for the temporary cover was slightly above 50% in the summer but over 90% in the winter. Collection efficiencies were then recalculated to be consistent with other literature, which exclude the oxidation and migration terms that can introduce more uncertainty. The difference between equations 1 and 2 is minor in consideration of the uncertainty of these types of studies.

$$\frac{\text{methane collected}}{\text{methane collected} + \text{emissions}} \quad (2)$$

**Table 3.1. Collection Efficiency for Various Covers Reported in Spokas et al. (2006)**

<b>Cover Type</b>	<b>Collection Efficiency (%)<sup>a</sup></b>	<b>Collection Efficiency (%)<sup>b</sup></b>
Final clay cover (1 meter) with LFG recovery	91.5	92.6
Final geosynthetic clay with LFG recovery	51.5	53.0
Final clay cover (1 meter) with LFG recovery - summer	90.7	92.9
Final clay cover (1 meter) with LFG recovery - winter	97.8	98.6
Thin clay temporary cover (30 cm) with LFG recovery - summer	53.9	54.7
Thin clay temporary cover (30 cm) with LFG recovery - winter	93.2	95.1
Final clay cover (1 meter) with LFG recovery	99.2	100
Final geomembrane with horizontal gas collection	98.1	99.2

a. Calculated as methane collected/(methane collected + emissions + oxidation + migration)

b. Calculated as methane collected/(methane collected + emissions)

Borjesson et al. (2007) reported on methane oxidation and gas collection at six Swedish landfills using Fourier-transformed infrared (FTIR) in combination with a tracer. While the emphasis of their study was on methane oxidation, sufficient data were published to calculate collection efficiency as in Equation 2. Data from two of the landfills (Hagby and Visby) were excluded from this review because it was reported that the gas collection system was not working well during the test period. The results for each landfill test are presented in Table 3.2. All landfills reported were active and only minimal information was reported on the cover type. These data are likely applicable for daily covers in the context of U.S. landfills, although U.S. landfills do not typically use sludge as a cover material.

**Table 3.2. Collection Efficiency for Various Covers Reported in Borjesson et al. (2007)**

<b>Cover Type</b>	<b>Collection Efficiency (%)<sup>a</sup></b>
Fiborna (wood chips and sludge)	68.4
Fiborna (wood chips and sludge)	65.0
Fiborna (wood chips and sludge)	70.0
Heljestorp (sewage sludge and soil)	57.8
Hogbytorp (sewage sludge and soil)	33.9
Hogbytorp (sewage sludge and soil)	43.2
Sundsvall (sewage sludge and soil)	63.3

a. Calculated as methane collected/(methane collected + emissions)

Mosher et al., (1999) reported a summary of methane emissions from nine landfills in the northeastern US. Emissions were measured by both static chambers and a tracer flux technique. Two of the landfills collected gas, making it possible to compare emissions to collected gas. One of the two landfills was closed and had a geomembrane plus soil cover. A collection efficiency of 90.5% was calculated as in Equation 2. The authors indicate that the gas collected was not measured accurately which casts some doubt on this value. This collection efficiency is nonetheless likely to be reasonable from two perspectives. First, this landfill had the lowest emissions of the sites studied and second, the collection efficiency is consistent with other values in this review. A collection efficiency of 70% was calculated for an active landfill in which part of the landfill was covered with a geomembrane but other parts had daily cover only.

Huitric and Kong (2006) reported collection efficiencies for the Palos Verdes Landfill (PVLf) in Los Angeles County. The PVLf was closed in 1980, has a 5 foot thick clay cap and an active gas collection system. Gas emissions were measured using an SEM-500 flame ionization detector (FID) monitor. An “integrated surface methane” (ISM) concentration was measured by surface scans at 3 inches above the surface after dividing the landfill into approximately one acre grids. The measured ISM was compared to the ISM that was calculated using the US EPA’s Industrial Source Complex (ISC) model. The calculated ISM assumes no gas collection, and the source term was based on the volume of collected gas at the PVLf. Collection efficiency was calculated as follows:

$$\frac{ISM_r}{(ISM_r + ISM_e)} \quad (3)$$

where  $ISM_r$  is the modeled reduction in surface emission due to collection and  $ISM_e$  is the measured surface methane concentration due to emissions. In this method, emissions due to methane oxidation are incorporated into the measured ISM. This calculation assumes that methane concentration is correlated to flux. The efficiency of the LFG collection system at the PVLf was calculated to be 94 to 96%. While these results are consistent with Spokas et al. (2006), methane generation at the PVLf is relatively low given the refuse age. Total gas collection was equivalent to 36.3 gm CH<sub>4</sub> (m<sup>-2</sup> d<sup>-1</sup>). In Huitric et al (2007), the collection efficiency determined using Equation 3 was supported by the results of a static flux chamber study completed at the PVLf. In fact, even higher collection efficiencies were reported for the PVLf when using flux chamber results.

Lohila et al. (2007) reported methane fluxes for a section of a Finnish landfill that included an active disposal area and a sloped area. The active area was covered daily with soil and construction and demolition waste rejects, and the sloped area had a cover that included 0.2 to 0.5 meters of compost over 0.5 to 2 meters of diamicton<sup>1</sup> and clay. Three estimates of collection efficiency were reported. First, it was reported that the mean methane flux over seven days was reduced by 79% when the gas collection system was turned on. This measurement was made by using methane concentration data coupled to an eddy covariance method. A second measure of methane flux with the gas system activated suggested a reduction of only 39%. However, this second estimate was based on 7 to 10 static chamber measurements and is not likely representative of the entire landfill. The third estimate was made by comparing the mean methane emission to the volume of gas collected and assuming that methane production was the sum of emissions plus collection. This resulted in an estimate of 69% collection efficiency.

### **3.4 SUB-CATEGORIZATIONS FOR COLLECTION EFFICIENCY NUMBERS**

The following sections describe how the engineered controls of the landfill affect the overall collection efficiencies of a LFG extraction system and can be used in making engineering judgments in the application of the collection efficiency values. In addition, the importance of reviewing surface emissions monitoring (SEM) and LFG migration data is discussed as an additional means of making site-specific judgments.

#### **3.4.1 Type of Cover**

The cover is part of the gas control system (in fact, it may be the only gas control system in old, abandoned landfills with no gas extraction wells), so the potential diffusion or leakage of gas through the cover needs to be quantified. Gas flows through refuse or soils either by advection or by diffusion. Advection occurs when total gas pressure is not uniform throughout the system. Advective flow is in the direction in which total pressure decreases. Diffusive flow of a gas is in the direction in which its concentration (partial pressure) decreases.

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<sup>1</sup> unsorted and unstratified sedimentary deposit of any origin, for example: till, landslide debris, mudflows, turbidites, *etc.* (Aber, 2008)

Three types of cover are generally used at landfills; final, intermediate, and daily cover. Daily cover is a layer (usually about six inches) that is placed on top of the active fill area of a landfill at the end of each day. Soil is usually used as a daily cover, but other types of materials may be used as well:

- Textile cover (man-made material rolled over the top of the landfill)
- Chemical foams
- Tire chips (tires that have been shredded into small pieces)
- Wood chips or shredded green waste
- Tarps
- Compost

Intermediate cover is used on the areas of a landfill that will not be used for a long period of time (for example, greater than 12 months) but are not yet ready to be closed. Covering the parts of a landfill that are not used will reduce the amount of liquid that will infiltrate into the landfill. Intermediate covers usually consist of whatever type of soil is available at the landfill site. The thickness of the intermediate cover is usually one foot or more, twice as much as is used for the daily cover. Vegetation is usually allowed to grow on top of the cover as protection against erosion.

The final cover on a landfill is meant to seal the landfill and reduce the amount of water that will enter the landfill after it is closed. It usually consists of the following five layers of material, starting at the top of the cover:

- Top soil - This is a layer of nutrient-rich soil. Vegetation is planted here (grass, flowers, etc.). The layer of topsoil is usually about six inches thick.
- Protective layer - The protective layer will protect the barrier layer underneath from cracking, freezing, and thawing during cold months. It will also allow roots from vegetation to grow. This layer usually consists of soil and is about two feet thick.
- Drainage layer - The drainage layer is used to provide drainage in order to keep as much moisture away from the barrier layer below as possible. This layer usually contains coarse sand and it is usually about eight inches thick.
- Barrier layer - The barrier layer is put in place to keep as much water out of the landfill as possible. It usually contains clay and it is usually about two feet thick. A geomembrane may be used in place of or in addition to the clay layer.
- Grading layer - This material is usually very coarse soil or sand. It is put on top of the waste to make the surface uniform and level. This layer is usually six inches to two feet thick.

The type of cover is directly related to the efficiency of the LFG collection system. Thick clay covers have a lower conductivity and are more resistant to diffusion of gas (or infusion of air) through the cover than a thinner daily cover. The increasing pressure drop through a cover with decreasing hydraulic conductivity also decreases the effectiveness of a gas collection system. As such, low permeability daily or intermediate covers, which are ultimately buried and become part of the waste, can actually impede gas collection when they are buried in the waste. Geomembrane liners reduce both diffusion and advective flow of gas from the landfill even further.

### **3.4.2 Type of LFG Collection Systems**

The effectiveness of a LFG collection system is dependent upon its design and operation. In general, two types of gas collection systems are in use at landfills: active gas collection systems and passive gas collection systems. A passive system is a system that relies on pressure or concentration gradients to function (i.e. vertical vents, gravel trenches, etc.). An active system is a system that includes a prime mover that creates a vacuum on a landfill applied at collection points (i.e. vertical gas wells, horizontal collectors, etc.). For the purpose of this paper, only active systems are discussed in further detail. The type of active LFG collection system is directly related to the gas collection efficiency in a landfill. For example, a LFG collection system designed for New Source Performance Standards (NSPS) (or similar air quality) compliance is far more capable of greater collection efficiencies than a LFG migration control system. Furthermore, a site with a collection system that is used solely for energy recovery may not be capable of achieving as high a collection efficiency as compared to one that is compliant with NSPS regulations because it is difficult to maximize gas quality when trying to achieve the highest level of gas collection. In summary, the scale or manner of operation of the collection system has a direct effect upon the determination of collection efficiencies.

### **3.4.3 LFG Monitoring Data**

There are two type of monitoring data, which can assist in determining the effectiveness of a LFG collection system. Most importantly, SEM data, whenever available, is important to consider when assessing the effectiveness of a LFG system. This would include the frequency and magnitude of surface emissions exceedances above the NSPS 500 parts per million by volume (ppmv) methane standard as well as the average level of detected concentrations across the site. SEM is not a direct measurement of LFG flux, but it can provide semi-quantitative information on the effectiveness of a LFG system. Additionally, although LFG migration data cannot be directly related to LFG emissions, it can be used to assess whether an existing LFG system is achieving a high level of collection efficiency by providing an indirect measure of LFG, which is escaping collection and migrating away from the refuse mass.

## **3.5 PROPOSED NEW VALUES FOR COLLECTION EFFICIENCY**

Based upon review of recent publications regarding this issue, it is of utmost importance that new values can be calculated for LFG collection efficiencies in order to adequately determine site-specific emissions from landfills with different types of cover and LFG collection systems. These new values are presented in the bullets below:

- 50-70% (mid-range default = 60%) for a landfill or portions of a landfill that are under daily soil cover with an active LFG collection system installed (note that due to limited test data on daily soil covers, the selected range is based on the opinion of experts involved with the creation and review of this document);
- 54-95% (mid-range default = 75%) for a landfill or portions of a landfill that contain an intermediate soil cover with an active LFG collection system;
- 90-99% (mid-range default = 95%) for landfills that contain a final soil and/or geomembrane cover systems with an active LFG collection system.

The high ends of the range of these values are proposed for sites with NSPS or similar quality LFG collection systems, which are designed for and achieve compliance with air quality regulations and surface emissions standards. Before making a selection at the high end of the range, a detailed review of the LFG system design, LFG system coverage, age of waste, and SEM and LFG migration monitoring data should be conducted. The reviewer should be sufficiently satisfied that the particular landfill or landfill area meets all of the criteria for the highest level of LFG collection.

The low end of the range would be for full LFG systems that are installed and operated for other purposes, such as energy recovery, migration control, or odor management; those landfills with multiple exceedances of the 500 ppmv SEM standard and/or significant LFG migration; more limited coverage for the LFG system design; and/or operational issues that have limited the effectiveness to the LFG system. A mid-range default value is provided for those landfills for which detailed information about the landfill or landfill area is not known.

A composite collection efficiency should be determined for each site by evaluating the amount of landfill area under each cover type; selecting the appropriate low, medium, or high value for collection efficiency; and calculating a weighted-average based on cover area. This composite value can then be applied on a site-wide basis for the inventory year for which it was determined.

For landfill areas that are not under any form of LFG collection and are not influenced by LFG collection systems from neighboring areas, a collection efficiency of zero (0) should be used and included in the site-wide weighted average. In evaluating this condition, it should be noted that many active or recent disposal areas may still be under LFG control due to vertical wells installed in deeper portions of the waste in the same area, horizontal collectors, LFG connections to the leachate system, and/or vacuum influence from neighboring areas. In these cases, the daily cover values should be used, exercising the necessary engineering judgment based on site-specific factors to select the low, middle, or high value.

These values do not address LFG migration control systems that do not provide complete coverage of the landfill (e.g., perimeter system) since the collection efficiencies of such systems can vary over a wide range. For those, an average value of 50% may be appropriate to capture the possible range of efficiencies. Also, biologically active materials used as alternative daily cover (ADC) will be assumed to have similar efficiencies as daily soil covers. This assumption is based upon work performed by Barlaz et al. (2004) and other research summarized in Section 4.0, where biocovers were compared to soil covers in their ability to oxidize methane as it passes through the covers. It was shown that biocovers oxidize methane just as well as, and sometimes better, than soil covers and do not create impedances to gas flow once buried, like soil cover can. Collection efficiencies for other ADC types, besides the biologically active covers, have not been studied and are suggested to be subject to a 50% default as well.

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## 4 METHANE OXIDATION

### 4.1 EXECUTIVE SUMMARY

In developing our approach to methane oxidation, we summarized literature from 47 determinations of the fraction of methane oxidized and 30 determinations of methane oxidation rate in a variety of soil types and landfill covers. Both column measurements and in situ field measurements are included. For differing soil covers, the mean values for percent (%) oxidation ranged from 22% to 55% from clay to sand. Mean values for oxidation rate ranged from 3.72 to 6.43 mol m<sup>-2</sup> d<sup>-1</sup> (52 to 102 g m<sup>-2</sup>d<sup>-1</sup>) across the different soils. The overall mean % oxidation across all studies was 35% with a standard error of 4% (se = standard deviation/(n<sup>1/2</sup>)). The overall mean oxidation rate across all studies was 4.5 mol m<sup>-2</sup> d<sup>-1</sup> with a standard error of 1.0 (72 ± 16 g m<sup>-2</sup>d<sup>-1</sup>). As will be discussed within, many of the in situ determinations employed the stable isotope approach which is known to yield conservative lower limit values. Therefore the values listed below are conservative.

It is clear that the fraction of methane oxidized in landfill cover soils is much greater than the default value of 10% that is in common use. Of the 47 determinations of methane oxidation reported in this chapter, only 4 report values less than 10%. The default value of 10% should be updated, based upon technological advancements in measurement approaches, soil engineering and state-of-the-practice applications in cover design.

The mean methane oxidation rate and percent oxidized values proposed for use in lieu of current default values are summarized in Table 4.1.

**Table 4.1 - Summary of Methane Oxidation Rates**

	Oxidation Rate (standard error) mol m <sup>-2</sup> d <sup>-1</sup>	Oxidation Rate (standard error) g m <sup>-2</sup> d <sup>-1</sup>	Percent oxidized (standard error)
Organic Covers	3.96 (2.33)	63.6	38 (7)
Clay Cover	3.88 (2.17)	52.1	22 (5)
Sand Cover	6.43 (2.77)	102	55 (9)
Other Mixtures	3.72 (1.27)	59.5	30 (6)
Overall	4.51 (1.0)	72.0 (16)	35 (4)

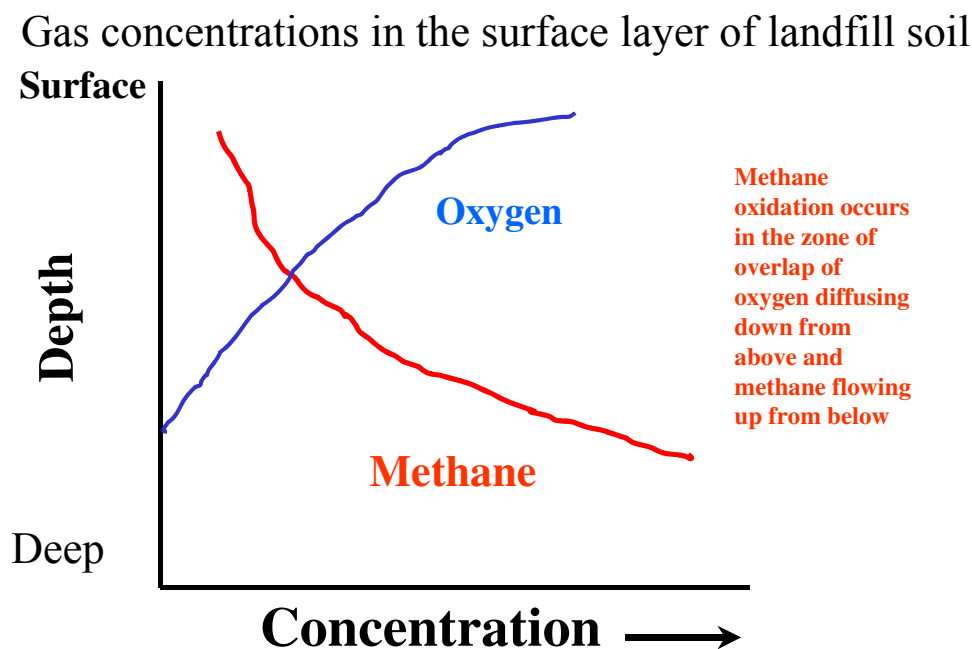
n = 47 for % oxidation and 30 for oxidation rate.

Sandy soils exhibited the greatest rates of oxidation. Sand is quite porous, allowing more methane and oxygen exchange and greater rates of oxidation. Oxygen is a factor which can limit methane oxidation, and it can diffuse into sand more readily than into clay soils due to the higher permeability of sand. However, sand is also not effective in blocking methane flow towards the surface and its use as a landfill cover soil would considerably reduce landfill gas capture efficiency.

## 4.2 BACKGROUND ON METHANE OXIDATION

The process of methane oxidation reduces the emissions of methane and other volatile hydrocarbons from the surface of landfills (Bogner et al., 1995, 1997a,b; Scheutz et al. 2003). The quantification of methane oxidation is one of the major uncertainties in estimating national or global methane emissions from landfills. Landfill gas (LFG) passes through landfill cover soils prior to being released to the environment. Bacteria consume methane and other volatile hydrocarbons that are produced by decomposition in the underlying waste by reacting it with oxygen near the landfill surface. These bacteria harness the energy from this chemical reaction to fuel their respiration. A portion of the methane is also incorporated into the biomass of the microbial cells.

The source of oxygen is downward diffusion from the air above the soil surface, while methane is transported up towards the surface from the waste by two processes, diffusion and advection. Gas can migrate against a total pressure differential via diffusion. The net direction of diffusion is always from higher concentration to lower concentration. Advection is the flow of gas driven by a pressure differential.



**Figure 4.1. Gas Concentrations in The Surface Layer of Landfill Soil.**

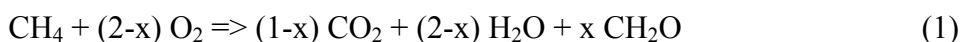
A report conducted by the United States Environmental Protection Agency (USEPA) in 2004 stated that “average oxidation of methane (on a volumetric basis) in some laboratory and case studies on landfill covers have indicated ranges from 10 percent to over 25 percent with the lower portion of the range being found in clay soils and higher in topsoils,” (USEPA, 2004a; see also USEPA, 1998). Due to the uncertainty involved and the lack of a standard method to determine oxidation rate, the USEPA recommended the default factor of 10% by volume

methane oxidation for landfill cover soils. A value of 10% oxidation is also recommended by the Intergovernmental Panel on Climate Change (IPCC) (2006) guidelines for national greenhouse gas inventories. The value of 10% came from Czepiel et al., (1996a). This study was the first to report the fraction of methane oxidized in landfills, and because of this, it has received undue weight. It should be noted that the 10% was an estimate of an annual average based on microbial kinetics and the effect of temperature on these kinetics. The work was done in a northern climate (New Hampshire, USA) and activity was very low in the winter.

Table 4.2 lists literature values for landfill cover methane oxidation rates (moles CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) and the fraction of methane oxidized. Fraction oxidized is that portion of methane that is oxidized in transported through the soil. Two approaches, column experiments and in situ field studies are reported. We also, for comparison, compile the results of closed-flask laboratory incubations of cover soils (Table 4.3). Laboratory and field oxidation measurement methods and their strengths and weaknesses are reviewed. Finally, our best estimates of methane oxidation rate and fraction oxidized are presented and discussed.

#### 4.2.1 Microbiological Background

Most biological methane oxidation is carried out by a group of aerobic bacteria called methanotrophs. They are nearly ubiquitous, and found where methane concentrations are at atmospheric levels (1.7 ppmv) or greater, oxygen is available, and other environmental conditions exist for typical soil bacteria. Although found everywhere in soil, methanotrophs are concentrated above anaerobic regions of wetlands where methane is produced. They are also found at high concentrations in the rhizosphere of wetland plants (Chanton et al., 1992). They can even be found in forest soils where they consume methane directly from the atmosphere. Biological methane oxidation is similar to chemical methane oxidation except that some energy is used to produce biomass (represented as CH<sub>2</sub>O):



Note that about 3 volumes of gas react to produce 1 volume of gas, resulting in a decrease in total pressure where methane oxidation occurs.

Methane oxidation is optimal when oxygen is greater than 5 % by volume, but some methane oxidation occurs at even at oxygen concentrations as low as 0.3 % (Czepiel et al., 1996a). Oxygen in the air is about 21%. Gebert et al. (2003) found that oxidation was maximum when methane is greater than 6 % and oxygen is greater than 12 %. The optimum temperature is in the range of 30 to 40 °C (Gebert et al., 2003). Optimal water content depends on soil texture and bulk density—more water is better as long as the remaining gas-filled porosity is adequate so that methane and oxygen diffusion is not attenuated. Some methanotrophs are able to fix nitrogen, so available nitrogen is not strictly needed for methane oxidation (Hanson and Hanson, 1996), but a small amount of added nitrogen (25 mg nitrogen / kg soil dry weight) was found to increase methane oxidation (De Visscher et al., 1999). Large amounts of nitrogen can inhibit methane oxidation, however, because the methane oxidation enzyme also reacts with ammonia (Hanson and Hanson, 1996). Methanotrophs also require a small amount of copper (0.02 to 4.3 mM) to synthesize one of the methane oxidizing enzymes (Hanson and Hanson, 1996).

One common approach towards obtaining in situ estimates of methane oxidation in cover soils is by use of subtle shifts in the stable carbon isotope ratio of methane as it passes from anaerobic zones through zones of oxidation. There are two stable isotopes of carbon, carbon-13 ( $^{13}\text{C}$ ) which is about 1% abundant and carbon-12 ( $^{12}\text{C}$ ) which comprises 99% of carbon atoms. Stable isotopes are useful for determining methane oxidation because as it occurs, the remaining methane becomes  $^{13}\text{C}$  enriched due to preferential utilization of the lighter  $^{12}\text{C}$  isotope by bacteria (Coleman et al, 1983). Carbon isotopic composition is expressed in the  $\delta$  notation, which is defined as follows:

$$\delta^{13}\text{C}_{\text{‰}} = ((R_{\text{sample}}/R_{\text{standard}})-1)*1000 \quad (2)$$

where  $R_{\text{sample}}$  is the  $^{13}\text{C}/^{12}\text{C}$  ratio of the sample and  $R_{\text{standard}}$  is the  $^{13}\text{C}/^{12}\text{C}$  ratio of the marine carbonate standard (Peedee belemnite standard (PDB), 0.01124). Typical biogenic landfill methane is produced at values of around -55 to -58‰. (Chanton et al., 1999). Following oxidation, methane may exhibit  $^{13}\text{C}$  enriched values of -30 to -50‰. Typical organic matter is  $^{13}\text{C}$  enriched relative to methane with a  $\delta^{13}\text{C}$  value of -26‰. The negative  $\delta$  value indicates that the sample is  $^{13}\text{C}$  depleted relative to the carbonate standard, which is by definition equal to 0 ‰. The more negative the value, the more  $^{13}\text{C}$  depletion is indicated. More positive values indicate  $\delta^{13}\text{C}$  enrichment.

Methanotrophic bacteria consume methane with  $^{12}\text{C}$  ( $^{12}\text{CH}_4$ ) at a slightly faster rate than methane with  $^{13}\text{C}$  ( $^{13}\text{CH}_4$ ) (Silverman and Oyama, 1960). This results in a shift in the isotopic ratio, or fractionation, as oxidation progresses. It is possible to calculate how much methane oxidation has occurred from the change in isotopic composition of methane before and following methane oxidation and the degree of fractionation by methanotrophs. The rate of oxidation is dependent on the volumetric methane concentration ( $\phi$ ) and a first-order rate constant ( $k$ ) for each isotope:

$$\frac{d\phi_{12}}{dt} = -k_{12}\phi_{12} \quad (3)$$

$$\frac{d\phi_{13}}{dt} = -k_{13}\phi_{13} \quad (4)$$

where  $k$  for  $^{12}\text{CH}_4$  is greater than that for  $^{13}\text{CH}_4$ . The ratio  $k_{12}/k_{13}$  is the kinetic isotope effect or fractionation factor  $\alpha_{\text{ox}}$ .  $\alpha_{\text{ox}}$  may be obtained by finding the slope of the regression for  $\ln \phi$  on the y-axis and  $\ln (\delta + 1000)$  on the x-axis (DeVisscher et al., 2004):

$$\alpha_{\text{ox}} = \frac{\text{slope}}{1 + \text{slope}} \quad (5)$$

The fraction of methane oxidized in a closed system ( $f_{\text{oxc}}$ ) may then be calculated from initial ( $\delta_0$ ) and final ( $\delta$ ) isotope ratios using the simplified Rayleigh equation (Mahieu et al., 2006):

$$f_{oxc} = \left( \frac{\delta + 1000}{\delta_o + 1000} \right)^{\alpha_{ox} / (1 - \alpha_{ox})} \quad (6)$$

## 4.3 LITERATURE REVIEW

### 4.3.1 Laboratory Methods

#### 4.3.1.1 Column Measurements (reported in Table 4.2) ---

The most realistic laboratory microcosm is a large undisturbed column containing soil where CH<sub>4</sub> and CO<sub>2</sub> are applied at a known rate to the bottom and the top is open to air. CH<sub>4</sub> emission from the top is monitored by periodically by enclosing the headspace and measuring the increase in CH<sub>4</sub> concentration within the enclosure. From the CH<sub>4</sub> influx (J<sub>in</sub>, mol m<sup>-2</sup> d<sup>-1</sup>) and outflux (J<sub>out</sub>, mol m<sup>-2</sup> d<sup>-1</sup>) the flux-based or mass balance determined oxidation rate (J<sub>oxf</sub>) and fraction oxidized (f<sub>ox</sub>) can be directly calculated:

$$J_{oxf} = J_{in} - J_{out} \quad (7)$$

$$f_{ox} = J_{oxf} J_{in}^{-1} \quad (8)$$

It is sometimes difficult to obtain an undisturbed column due to gravel, wood, etc. in the cover material, and there is likely to be considerable variability between undisturbed columns. For these reasons some research is conducted with repacked columns where soil is dug out and sieved before being packed in a column. This method allows better control of soil composition, but the disruption of soil structure makes repacked columns not very representative of field conditions.

It is also possible to measure methane oxidation by measuring the loss of CH<sub>4</sub> from the headspace as it and O<sub>2</sub> diffuse into a soil core. This simple method makes it easier to determine oxidation rates at various CH<sub>4</sub> concentrations and to have many replicates. It is likely that oxidation rates by this method are too high, however, because CH<sub>4</sub> and O<sub>2</sub> diffuse together and a large volume of soil will have the right proportions of these gases for oxidation. In a landfill CH<sub>4</sub> and O<sub>2</sub> diffuse from opposite directions, resulting in a narrower depth where both are present in adequate amounts.

#### 4.3.1.2 Flask Incubations (reported in Table 4.3) ---

In vitro incubations are used to determine the Michaelis-Menton kinetic parameters (V<sub>max</sub> and K<sub>m</sub>):

$$V = V_{max} S / (K_m + S) \quad (9)$$

where V is the CH<sub>4</sub> loss rate and S is the aqueous CH<sub>4</sub> concentration (usually determined from φ<sub>CH<sub>4</sub></sub> and Henry's law). Sieved and homogenized soil is typically used to minimize variability. A small amount of soil is placed in a flask along with CH<sub>4</sub> in air so that gas diffusion is rapid throughout the soil, and measurements of V are taken as quickly as possible to minimize growth of methanotrophs. The Michaelis-Menton equation was originally intended for use with a

constant amount of an enzyme, so enzyme production or loss through changes in the bacterial condition or numbers are confounding factors. It is theoretically possible to calculate an oxidation rate ( $J_{oxv}$ ) from  $V_{max}$  ( $\text{mol g}^{-1} \text{d}^{-1}$ ), field bulk density ( $\rho_b$ ,  $\text{g m}^{-3}$ ), and oxidation depth ( $z$ , m):

$$J_{oxv} = V_{max} \rho_b z \quad (10)$$

If  $V_{max}$  is not determined the maximum measure  $V$  can be used in approximation. Table 4.3 lists four studies where oxidation rate determined from  $V_{max}$  is compared to flux-based oxidation rate with the same soil material. Two of the studies show similar results, but two studies have 22.2 and 33.6 times greater oxidation rates using the  $V_{max}$  method. This might be expected because  $V_{max}$  is a theoretical maximum oxidation rate where  $\text{CH}_4$  and  $\text{O}_2$  are optimal in depth  $z$ , which is not likely to be the case for  $J_{oxf}$ . Furthermore, the incubated soil is broken up, which allows gases better access to methanotrophs, and nutrients are mixed and more available. Because there is considerable uncertainty in applying  $V_{max}$  to field conditions and in determining  $z$ , this method of estimating oxidation rate is likely to be less reliable than other methods. These results are reported in Table 4.2 for the purpose of comparison, but they are not included in any reported averages or recommendations.

#### 4.3.2 Field Methods (reported in Table 4.2) ---

Calculation of methane oxidation rate is straightforward if the influx to the bottom of the oxidation region is known ( $J_{in}$ , eq. 7). Christophersen et al. (2001) used the mass balance of methane and carbon dioxide to estimate  $J_{in}$  in steady-state conditions. Knowing the total outflux of methane and carbon dioxide and the methane and carbon dioxide concentrations below the zone of oxidation,  $J_{in}$  can be calculated:

$$J_{\text{CH}_4+\text{CO}_2} = J_{\text{CH}_4\text{out}} + J_{\text{CO}_2\text{out}} = J_{\text{CH}_4\text{in}} + J_{\text{CO}_2\text{in}} \quad (11)$$

$$J_{\text{CH}_4\text{in}} = (J_{\text{CH}_4\text{out}} + J_{\text{CO}_2\text{out}}) \frac{\phi_{\text{CH}_4\text{in}}}{\phi_{\text{CH}_4\text{in}} + \phi_{\text{CO}_2\text{in}}} \quad (12)$$

where  $\phi$  is volumetric gas concentration. The main assumptions in this technique are: there is no net change in carbon dioxide due to its dissolution in water entering or leaving the oxidation zone, and production of these gases in the soil is negligible. The last assumption for carbon dioxide is the most uncertain due to oxidation of soil organic compounds other than methane.

As a result of the difficulty in estimating  $J_{in}$  in the field, carbon isotope fractionation has been widely used. This technique compares the  $^{13}\text{C}/^{12}\text{C}$  ratio before and after exposure to methanotrophic bacteria (Section 4.1). The advantage of this method is that only gas samples from the anaerobic zone and the surface or shallow sub-surface are required, along with the fractionation factor (eq. 5).

There are several important issues that must be addressed in the application of this method, however. As will be discussed below, these issues result in the approach yielding a lower limit appraisal of methane oxidation.

First, most researchers do not calculate  $f_{ox}$  by the Rayleigh equation (eq. 6) because the *closed-system* assumption that a sample of gas moves through soil without mixing with other methane may not be realistic. Instead, they use an open-system equation:

$$f_{oxo} = \frac{\delta - \delta_o}{1000(\alpha_{ox} - \alpha_{trans})} \quad (13)$$

where  $\alpha_{trans}$  is the transport fractionation factor. The open-system equation was adapted from an equation used by Monson and Hayes (1980) to study fatty acid synthesis (Liptay et al., 1998).

The term  $\alpha_{trans}$  is difficult to access, because it depends on the relative importance of diffusion relative to advection in the transport of methane. If methane is transported by advection, then  $\alpha_{trans}$  is equal to one and the approach is accurate. However, if diffusion is important, then  $\alpha_{trans}$  is greater than one. It is currently not possible to easily assess the relative importance of diffusion versus advection in transporting methane through the soil covered surface of a landfill. Certainly advection is a dominant process, as methane production produces gas volume and this excess pressure is relieved by flow outwards towards the surface. It has been shown that landfill gas emission varies as a function of variations in atmospheric pressure implying that advection is an important process (Czepiel et al., 2003; Bergamaschi, et al., 1998). However landfill covers are quite variable in terms of their gas permeability so advection/diffusion may vary in their relative importance from location to location. Additionally, landfill gas recovery systems reduce pressure within the landfill, reducing outward advection and increasing the importance of diffusion in gas transport towards the surface. Generally, in applying Eq 13,  $\alpha_{trans}$  is assumed to be equal to one. The effect of this assumption is that the isotopic approach results in lower-limit conservative values for methane oxidation.

The lower limit effect of diffusion in applying the isotope approach results because diffusion, as well as oxidation, fractionates isotopes.  $^{12}\text{CH}_4$  diffuses to the atmosphere faster than  $^{13}\text{CH}_4$ , thereby impacting measured isotope fractionation of surface samples and isotope-derived  $f_{ox}$  (De Visscher et al., 2004). De Visscher et al. (2004) clearly demonstrated that isotopic based measurements, particularly those applied on emitted methane captured in chambers, are always lower-limit conservative estimates. In controlled lab experiments when isotopic and mass balance approaches have been directly compared, the isotope approach consistently underestimates methane oxidation (Powelson et al., 2007; De Visscher et al., 2004). The diffusion effect likely explains the observation that  $\delta^{13}\text{C}$  values from 5 to 10 cm probe samples were less negative than surface samples, which meant that subsurface samples had greater calculated oxidation (Chanton et al., 2008).

A second weakness of the method is that literature values for the fractionation factor  $\alpha_{ox}$  range considerably (Templeton et al., 2006; Chanton and Liptay, 2000), depending on temperature and other factors. Small differences in  $\alpha_{ox}$  have a large impact on  $f_{ox}$  (eq. 6 or 13). Generally  $\alpha_{ox}$  is determined for each soil type where oxidation is quantified by the isotopic approach to reduce this source of uncertainty (e.g. Borgesson, et al., 2007).

Third, the heterogeneity of soil pore sizes results in a range of methane transport rates. Methane following the slowest routes may be completely oxidized before reaching the surface, and the

loss of this isotope signature is an additional factor which results in the underestimation of  $f_{ox}$  by the isotope approach (Powelson et al., 2007; Chanton et al., 2008).

**Table 4.2. Literature values for methane oxidation rate and fraction oxidized (mean and standard error, SE), sorted by cover material and method (column or field)**

Cover material	Method	Oxidation rate	SE rate	Fraction oxidized	SE fraction	Gas Collection	Cover Depth	Soil temperature	Reference
<b>ORGANIC</b>		----- mol m <sup>-2</sup> d <sup>-1</sup> -----					m	°C or month and state	
<b>ORGANIC</b>	<b>COLUMN</b>								
Compost	Repacked column with CH <sub>4</sub> advection	9.423 <sup>†</sup>	na <sup>†</sup>	1.00 <sup>†</sup>	na <sup>†</sup>	na <sup>†</sup>	0.6	18	Humer and Lechner, 1999
Compost	Repacked column with CH <sub>4</sub> advection	6.25	0.6250 <sup>†</sup>	0.19	0.019 <sup>†</sup>	na <sup>†</sup>	0.5	22	Wilshusen et al., 2004
		7.836		0.6					AVERAGE
<b>ORGANIC</b>	<b>FIELD</b>								
Compost landfill cover	Isotope fractionation	0.041 <sup>†</sup>	0.0331 <sup>†</sup>	0.55	0.136	yes	1.15	Apr,Jun,Sep in KY	Barlaz et al., 2004
Wood chips and sludge	Plume tracer and isotope fractionation	nr <sup>†</sup>	nr <sup>†</sup>	0.16	0.031	yes	nr <sup>†</sup>	7	Borjesson et al., 2007
Wood chips and sludge	Plume tracer and isotope fractionation	nr <sup>†</sup>	nr <sup>†</sup>	0.25	0.003	yes	nr <sup>†</sup>	17	Borjesson et al., 2007
Compost	Isotope fractionation	nr <sup>†</sup>	nr <sup>†</sup>	0.29	0.065	no	0.47	all year, FL	Chanton et al., 2008
Compost	Isotope fractionation (subsurface)	nr <sup>†</sup>	nr <sup>†</sup>	0.44	0.048	no	0.47	all year, FL	Chanton et al., 2008
Mulch	Isotope fractionation	nr <sup>†</sup>	nr <sup>†</sup>	-0.01	0.045	no	0.3	all year, FL	Chanton et al., 2008
Mulch	Isotope fractionation (subsurface)	nr <sup>†</sup>	nr <sup>†</sup>	0.37	0.031	no	0.3	all year, FL	Chanton et al., 2008
Mulch	Isotope fractionation	nr <sup>†</sup>	nr <sup>†</sup>	0.4	0.033	no	0.6	all year, FL	Chanton et al., 2008
Mulch	Isotope fractionation (subsurface)	nr <sup>†</sup>	nr <sup>†</sup>	0.59	0.046	no	0.6	all year, FL	Chanton et al., 2008
Yard-waste compost	Isotope fractionation	0.108	0.0338	0.38	0.031	no	0.5	all year, FL	Stern et al., 2007
		0.074		0.34					AVERAGE
<b>ORGANIC</b>	<b>ALL</b>	3.955 ± 2.335		0.38 ± 0.07					AVERAGE



Cover material	Method	Oxidation rate	SE rate	Fraction oxidized	SE fraction	Gas Collection	Cover Depth	Soil temperature	Reference
<b>CLAY</b>		----- mol m <sup>-2</sup> d <sup>-1</sup> -----					m	°C or month and state	
CLAY	COLUMN								
Silty clay landfill cover	Undisturbed column with 135 to 145 ppmv CH <sub>4</sub> diffusion	0.004 <sup>†</sup>	0.0005 <sup>†</sup>	nr <sup>‡</sup>	nr <sup>‡</sup>	na <sup>‡</sup>	nr <sup>‡</sup>	Jun, Sep, Nov in IL	Bogner et al., 1997
Silty clay landfill cover	Undisturbed column with 1 to 8 % CH <sub>4</sub> diffusion	1.156 <sup>†</sup>	0.2927 <sup>†</sup>	nr <sup>‡</sup>	nr <sup>‡</sup>	na <sup>‡</sup>	nr <sup>‡</sup>	Jun, Sep, Nov in IL	Bogner et al., 1997
Clay landfill cover	Repacked column with CH <sub>4</sub> advection	6.8	0.3	0.4	0.018	na <sup>‡</sup>	1	19	Kightley et al., 1995
		2.653		0.4					AVERAGE
CLAY	FIELD								
Sandy clay landfill cover	Isotope fractionation	0.564 <sup>†</sup>	0.3420 <sup>†</sup>	0.14	0.039	no	0.15	Feb to May in FL	Abichou et al., 2006
Clay landfill cover	Isotope fractionation	1.216 <sup>†</sup>	0.8693 <sup>†</sup>	0.21	0.056	yes	1	Apr, Jun, Sep in KY	Barlaz et al., 2004
Clay landfill cover	Isotope fractionation	13.517 <sup>†</sup>	1.8114 <sup>†</sup>	0.14	0.02	no	0.15	6 to 32	Chanton and Liptay, 2000
Clayey landfill cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.08	0.03	no	0.15	all year, FL	Chanton et al., 2008
Clayey landfill cover	Isotope fractionation (subsurface)	nr <sup>‡</sup>	nr <sup>‡</sup>	0.34	0.054	no	0.15	all year, FL	Chanton et al., 2008
		5.099		0.18					AVERAGE
CLAY	ALL	3.876 ± 2.175		0.22 ± 0.05					AVERAGE

Cover material	Method	Oxidation rate	SE rate	Fraction oxidized	SE fraction	Gas Collection	Cover Depth	Soil temperature	Reference
<b>SAND</b>		----- mol m <sup>-2</sup> d <sup>-1</sup> -----					m	°C or month and state	
<b>SAND</b>	<b>COLUMN</b>	-							
Coarse sand landfill cover	Repacked column with CH <sub>4</sub> advection	10.4	0.1	0.61	0.006	na <sup>‡</sup>	1	19	Kightley et al., 1995
Fine sand landfill cover	Repacked column with CH <sub>4</sub> advection	6.9	0.3	0.41	0.018	na <sup>‡</sup>	1	19	Kightley et al., 1995
Loamy sand landfill cover	Repacked column with CH <sub>4</sub> advection of 4.8 mol m <sup>-2</sup> d <sup>-1</sup>	4.608 <sup>†</sup>	0.0842 <sup>†</sup>	0.96 <sup>†</sup>	0.017 <sup>†</sup>	na <sup>‡</sup>	1	22	Scheutz and Kjeldsen, 2003
Loamy sand landfill cover	Repacked column with CH <sub>4</sub> advection of 65 mol m <sup>-2</sup> d <sup>-1</sup>	23.725 <sup>†</sup>	1.7299 <sup>†</sup>	0.37 <sup>†</sup>	0.027 <sup>†</sup>	na <sup>‡</sup>	1	22	Scheutz and Kjeldsen, 2003
Sandy soil	Repacked column with CH <sub>4</sub> advection	4.366 <sup>†</sup>	0.2634 <sup>†</sup>	0.44 <sup>†</sup>	0.033 <sup>†</sup>	na <sup>‡</sup>	1.2	30 to 40	Visvanathan et al., 1999
		10		0.56					AVERAGE
<b>SAND</b>	<b>FIELD</b>								
Sand landfill cover	Isotope fractionation	0.172 <sup>†</sup>	0.2771 <sup>†</sup>	0.42	0.095 <sup>†</sup>	no	0.3 to 0.8	0.4 to 24.8	Borjesson et al., 2001
Sand adjacent to landfill	CH <sub>4</sub> and CO <sub>2</sub> mass balance	1.097 <sup>†</sup>	0.4969 <sup>†</sup>	0.89	nr <sup>‡</sup>	nr <sup>‡</sup>	na <sup>‡</sup>	2 to 25	Christophersen et al., 2001
Sand adjacent to landfill	Isotope fractionation	0.149 <sup>†</sup>	0.1372 <sup>†</sup>	0.28	0.152	nr <sup>‡</sup>	na <sup>‡</sup>	2 to 25	Christophersen et al., 2001
		0.473		0.53					AVERAGE
<b>SAND</b>	<b>ALL</b>	6.427 ± 2.770		0.55 ± 0.09					AVERAGE
<b>OTHER MATERIAL</b>	<b>COLUMN</b>								
Sandy-clay loam landfill cover	Incubation and model	1.830 <sup>†</sup>	0.2642 <sup>†</sup>	0.1	nr <sup>‡</sup>	no	1.0 to 2.0	19	Czepiel et al., 1996a
Loamy agricultural soil	Repacked column with CH <sub>4</sub> advection	8.236 <sup>†</sup>	0.2725 <sup>†</sup>	0.61 <sup>†</sup>	0.020 <sup>†</sup>	na <sup>‡</sup>	0.6	18 to 26	De Visscher et al., 1999
Sandy loam landfill cover	Repacked column with CH <sub>4</sub> advection	15	0.2725 <sup>†</sup>	0.65	0.020 <sup>†</sup>	na <sup>‡</sup>	0.6	18 to 26	De Visscher et al., 1999
Sandy loam landfill cover	Repacked column with CH <sub>4</sub> advection	3.535	0.0757	0.21	0.005	na <sup>‡</sup>	0.31	22	Hilger et al., 2000
Sand with clay landfill cover	Undisturbed column with CH <sub>4</sub> diffusion	3.813	0.2914	nr <sup>‡</sup>	nr <sup>‡</sup>	na <sup>‡</sup>	0.1	25	Whalen et al., 1990

Cover material	Method	Oxidation rate	SE rate	Fraction oxidized	SE fraction	Gas Collection	Cover Depth	Soil temperature	Reference
<b>OTHER MATERIAL</b>		----- mol m <sup>-2</sup> d <sup>-1</sup> -----					m	°C or month and state	
OTHER MATERIAL	<b>FIELD</b>								
Sandy loam landfill cover	Isotope fractionation	0.456 <sup>†</sup>	0.2158 <sup>†</sup>	0.25	0.028	no	0.45	Sept to Feb in FL	Abichou et al., 2006
European landfill covers	Isotope fractionation	4.239 <sup>†</sup>	1.5460 <sup>†</sup>	0.84	0.023 <sup>†</sup>	yes	1	5.8 to 26.9	Bergamaschi et al., 1998
Sandy loam landfill cover	Isotope fractionation	3.795 <sup>†</sup>	2.6892 <sup>†</sup>	0.26	0.048 <sup>†</sup>	yes	0.4 to 1.0	-0.3 to 26.8	Borjesson et al., 2001
Sewage sludge and mineral soils	Plume tracer and isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.07	0.023	yes	nr <sup>‡</sup>	6	Borjesson et al., 2007
Sewage sludge and mineral soils	Plume tracer and isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.15	0.011	yes	nr <sup>‡</sup>	-2	Borjesson et al., 2007
Mineral soil	Plume tracer and isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.4	0.026	yes	nr <sup>‡</sup>	9	Borjesson et al., 2007
Mineral soil	Plume tracer and isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.38	0.032	yes	nr <sup>‡</sup>	5	Borjesson et al., 2007
Mulch and topsoil landfill cover	Isotope fractionation	1.678 <sup>†</sup>	0.2114 <sup>†</sup>	0.26	0.04	no	1.09	8 to 35	Chanton and Liptay, 2000
Loamy landfill cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.12	0.025 <sup>†</sup>	no	0.35	all year, FL	Chanton et al., 2008
Loamy landfill cover	Isotope fractionation (subsurface)	nr <sup>‡</sup>	nr <sup>‡</sup>	0.31	0.045	no	0.35	all year, FL	Chanton et al., 2008
Landfill "A" cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.28	0.108	nr <sup>‡</sup>	nr <sup>‡</sup>	21 to 30	Liptay et al., 1998
Landfill "B" cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.19	0.014	nr <sup>‡</sup>	nr <sup>‡</sup>	20 to 28	Liptay et al., 1998
Sandy-clay loam landfill cover	Chambers and estimated fraction oxidized	0.906 <sup>†</sup>	0.2506 <sup>†</sup>	nr <sup>‡</sup>	nr <sup>‡</sup>	no	1 to 2	Sept to Oct in NH	Czepiel et al., 1996b
Sandy-clay loam landfill cover	Plume tracer and estimated fraction oxidized	1.031 <sup>†</sup>	0.0586 <sup>†</sup>	nr <sup>‡</sup>	nr <sup>‡</sup>	no	1 to 2	Sept to Oct in NH	Czepiel et al., 1996b
Sandy-clay loam landfill cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.68	0.093	nr <sup>‡</sup>	nr <sup>‡</sup>	22 to 29	Liptay et al., 1998
Rochester landfill cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	-0.03	0.035	nr <sup>‡</sup>	nr <sup>‡</sup>	Aug in NH	Liptay et al., 1998
Springfield landfill cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.01	0.042	nr <sup>‡</sup>	nr <sup>‡</sup>	14 to 18	Liptay et al., 1998
Wayland landfill cover	Isotope fractionation	nr <sup>‡</sup>	nr <sup>‡</sup>	0.32	0.053	nr <sup>‡</sup>	nr <sup>‡</sup>	18 to 23	Liptay et al., 1998

Cover material	Method	Oxidation rate	SE rate	Fraction oxidized	SE fraction	Gas Collection	Cover Depth	Soil temperature	Reference
Fine sandy loam landfill cover	Isotope fractionation	0.141	0.0281	0.19	0.027	no	0.35	all year, FL	Stern et al., 2007
		1.749		0.27					AVERAGE
OTHER MATERIAL	ALL	3.722 ± 1.266		0.3 ± 0.06					AVERAGE
OVERALL AVERAGE		4.51		35±4					
All studies		± 0.48							
<sup>†</sup> Estimation methods listed by reference below. <sup>‡</sup> na = not applicable; nr = not reported. ± = standard error = (standard deviation)/(n <sup>1/2</sup> ) Note, fraction oxidized = % oxidized /100									

### Table 4.2 footnotes:

Abichou et al., 2006. Oxidation rates ( $J_{ox}$ ) calculated from emission fluxes ( $J_{out}$ , their Table 1) and oxidation fractions ( $f_{ox}$ , their Table 2):

$$J_{ox} = \frac{J_{out}}{f_{ox}^{-1} - 1} \quad (f1)$$

Barlaz et al., 2004. Oxidation rates calculated using eq. f1 from Supplemental Information using the average emission fluxes in Tables S3 and S4 and the oxidation fractions in Table S6. Bergamaschi et al., 1998. The oxidation rate and SE had to be approximated by averaging the emission fluxes, and the SE approximated by using the average, minimum, and maximum fluxes listed for covered areas in their Tables 7 and 8. The oxidation rate SE accounted for propagation of error:

$$s_{rel,a} = \sqrt{s_{rel,b}^2 + s_{rel,c}^2} \quad (f2)$$

where  $s_{rel}$  is the relative standard deviation ( $s \text{ mean}^{-1}$ ) and in this case a, b, and c refer to oxidation rate, emission flux rate, and fraction oxidized, respectively (Christian, 1986). The fraction oxidized SE was estimated from the relative SD of emitted  $\delta^{13}\text{C}$ .

Bogner et al., 1997. Oxidation estimated from their Fig. 3 for two ranges of initial  $\text{CH}_4$  concentration: 135 to 145 ppm<sub>v</sub> and 1 to 8 percent by volume.

Borjesson et al., 2001. The oxidation rates and standard errors were calculated by averaging data in their Table 3 and 5 and the propagation of error for the averages was accounted for by:

$$s_{avg} = \sqrt{s_1^2 + s_2^2 + \dots} \quad (f3)$$

where  $s_{avg}$  is the standard deviation of the average and  $s_1$ ,  $s_2$ , etc. are standard deviations of the values contributing to the average (Christian, 1986).

Chanton and Liptay, 2000. The reported average oxidation rates were used with each average  $\text{CH}_4$  emission flux over the course of a year (estimated from their Fig. 1) to calculate oxidation rate.

Chanton et al., 2008. Emitted fraction oxidized from closed-system equation (their Table 1). SEs estimated from  $\delta^{13}\text{C}$  SEs. Subsurface samples taken from 5 to 10 cm depth with probes.

Christophersen et al., 2001. For the mass-balance method, the reported average oxidation rate for the Field transect, 89%, was used with each Field average  $\text{CH}_4$  emission flux (their Table

1) to calculate oxidation rate. For the isotope fractionation method, the fraction oxidized for three locations (their Table 3) was used with the corresponding emission flux to calculate oxidation rate.

Czepiel et al., 1996a. Jar incubations of homogenized soil. Oxidation rate estimated from their Fig. 1 (5 - 15 cm depth) and assuming bulk density of  $1.7 \text{ g cm}^3$ . Fraction oxidized was modeled over a year using oxidation rates “in soil columns” adjusted for field temperature and moisture content and field-measured outflux. This approach is similar to our eq. 7, 8, and 10. Standard error or other measure of confidence was not given.

Czepiel et al., 1996b. Both chamber and plume emissions converted to oxidation rates (eq. f1) using their estimated oxidation fraction of 0.20.

De Visscher et al., 1999. There are no true replicates; mean and standard error for the agricultural soil were estimated from repeated sampling from one column shown in their Fig. 1. Data for the landfill cover column were not shown; the landfill standard error was assumed to equal that for the agricultural column.

Hilger et al., 2000. Oxidation values are for steady state (their Table 1, Exp. 1, Live + LFG). Oxidation rate calculated from column parameters.

Humer and Lechner, 1999. Oxidation values are for steady state in a single column where there was complete oxidation.

Scheutz and Kjeldsen, 2003. Oxidation estimated from their Fig. 5 at inlet flow rates of  $0.24$  and  $3.18 \text{ m}^3 \text{ m}^{-2} \text{ d}^{-1}$ .

Visvanathan et al., 1999. Oxidation rates estimated from their Fig. 6.

Whalen et al., 1990. Oxidation rate is Michaelis-Menton  $V_{\text{max}}$  reported on an area basis (their Table 1).

Wilshusen et al., 2004. Oxidation values are for steady state. SE was estimated from final values of three columns in their Fig. 2 (the low, non-steady state column was excluded).

#### 4.4 CALCULATING RATES OF OXIDATION (IN MOLES $\text{M}^{-2}\text{D}^{-1}$ ) FROM PERCENT OXIDATION VALUES AND EMISSION RATES

If the outflux or emission rate of methane ( $J_{\text{out}}$ ) and the fraction of methane oxidized in the cover ( $f_{\text{ox}}$ ) are known it is possible to calculate the rate of methane oxidation as shown below:

$$J_{\text{ox}} = \frac{J_{\text{out}}}{f_{\text{ox}}^{-1} - 1} \quad (14)$$

where  $f_{\text{ox}}$  is usually determined by the isotope method. Outflux may be found by the chamber method, where the increase in methane concentrations in a closed chamber covering the soil is determined. Outflux from a large area such as a landfill may be estimated by averaging outflux from numerous locations. It has been found that there is large spatial variability in methane outflux, which makes it difficult to determine the true mean flux (Czepiel et al., 1996b). Atmospheric tracers may also be used to find the methane emission ( $Q_m$ ,  $\text{mol d}^{-1}$ ) from large areas. This involves releasing a tracer gas like sulfur hexafluoride from evenly spaced locations at a known rate ( $Q_t$ ) and measuring the relative concentrations of the tracer ( $\phi_t$ ) and methane ( $\phi_m$ )

downwind (Czepiel et al., 1996b). If the released tracer is well mixed with the methane plume,  $Q_m$  may be calculated:

$$Q_m = Q_t \frac{\phi_m}{\phi_t} \quad (15)$$

The average outflux is  $Q_m$  divided by the source area. The main disadvantage of this technique is the expense of the gas and the collection of many plume samples downwind over a large area in a short time. It should also be mentioned that sulfur hexafluoride is the most potent greenhouse gas that the IPCC has evaluated, with a global warming potential 22,200 times that of carbon dioxide when compared over a 100 year period (Ramaswamy et al., 2001).

A new promising approach involves the use of optical remote sensing to determine methane emissions from large areas of landfills. The method uses a path-integrated optical remote sensing system in multiple beam configurations to locate “hot spots” and determine emissions (US-EPA, 2006, Hater et al., 2007; Chanton et al., 2007; Modrak et al., 2007). A tunable diode laser (TDL) is used as the optical sensor since methane absorbs strongly in the infrared portion of the electromagnetic spectrum. The approach is to turn the landfill into a giant open path spectrophotometer. A series of mirrors is placed across and above the landfill surface to form a 3-dimensional set of reflectors. The computer-controlled laser rotates automatically from mirror to mirror, focusing its beam at each one in turn. The laser signal returns from the mirror to a receiver that measures its strength. The attenuation of the laser beam in relation to the locations of the reflectors in the field are input to the field computer which sums the pathways and methane concentrations and produces horizontal and vertical maps of the methane plume above the landfill. Simultaneously a weather station determines wind velocity and speed and a model is used to calculate the mass of methane emitted from a selected area. Eddy correlation approaches have also been used (Lohila et al., 2007).

#### 4.5 PROPOSED NEW VALUES FOR METHANE OXIDATION

The results in Table 4.2 were organized by soil type and method. Oxidation rate is a better way to quantify methane oxidation than fraction oxidized because  $f_{ox}$  is a function of the methane loading rate as will be discussed below. Organic covers had an average percent oxidation of 38% and an average oxidation rate of 3.96 moles  $m^{-2} d^{-1}$ . Field oxidation rates were lower than column studies as the field compost and mulch covers were placed over clay soils. The compost covers retained moisture and thus prevented desiccation and cracking of the clay causing them to act as an effective barrier, thus effectively blocking upward methane flux to the compost (Stern et al., 2007). Clay soils had an average oxidation rate of 3.88 moles  $m^{-2} d^{-1}$  with an average % oxidation of 22%. Sandy soils had the greatest rate of oxidation at 6.43 moles  $m^{-2} d^{-1}$  with percent oxidation of 55%. Composite soils (other material) had an oxidation rate of 3.72 moles  $m^{-2} d^{-1}$  and 30% oxidation. Generally the column studies reported greater rates of oxidation and percent oxidation than the in situ field studies. If field studies are considered alone, the percent oxidation values are compost, 34%, clay, 18%, sand 53% and other 27%. However, the in situ field studies are based mostly on the isotope approach and thus are lower limit estimates.

Column studies are based on the mass balance approach which is more reliable, but impossible to apply in field settings.

Sand had the highest oxidation rate and the highest fraction oxidized (Table 4.2), probably due to better gas permeability. Oxygen is a factor which can limit methane oxidation and it can diffuse into sand more readily than into clay soils due to the higher permeability of sand. Sand is not effective in blocking methane flow towards the surface and would considerably reduce landfill gas capture efficiency.

Surprisingly, given the interest in using compost as biocovers, organic materials in the field had the lowest oxidation rate ( $0.07 \text{ mol m}^{-2} \text{ d}^{-1}$ ) in the field studies. This is because when compost covers are applied in the field, they often limit the gas transmissivity of the underlying clay cover by preserving the hydration of the clay. This effectively reduces methane emissions by a process termed blockage (Stern et al., 2007; Abichou et 2007). This value is the average of only two studies however, and in laboratory conditions organic materials had the second highest oxidation rate ( $7.84 \text{ mol m}^{-2} \text{ d}^{-1}$ ).

We suggest that the best approach to estimating the methane oxidation capacity of landfill covers is to use the average appropriate to each soil type (Table 4.2). We have included laboratory column studies and field methods in the average because each has strengths and weaknesses as discussed above. The column results were determined by mass balance, which produces a more exact estimate while the field studies employed the isotope approach for the most part, which yields a lower limit value for methane oxidation.

Thus, the best methane oxidation rate and percent oxidized estimates are (standard error):

**Table 4.1 - Summary of Methane Oxidation Rates**

	Oxidation Rate $\text{mol m}^{-2} \text{ d}^{-1}$	Oxidation Rate $\text{g m}^{-2} \text{ d}^{-1}$	Percent oxidized
Organic Covers	3.96 (2.33)	63.6	38 (7)
Clay Cover	3.88 (2.17)	52.1	22 (5)
Sand Cover	6.43 (2.77)	102	55 (9)
Other Mixtures	7.32 (1.27)	59.5	30 (6)
Overall	4.51 (1.0)	72.0 (16)	35 (4)

n = 47 for % oxidation and 30 for oxidation rate.

For differing soil covers, the percent oxidation means ranged from 22 to 55% from clay to sand. Mean values for oxidation rate ranged from  $3.72$  to  $6.43 \text{ mol m}^{-2} \text{ d}^{-1}$  ( $52$  to  $102 \text{ g m}^{-2} \text{ d}^{-1}$ ) across these different covers. The overall mean percent oxidation across all studies was 35% with a standard error of 4%. The overall mean oxidation rate across all studies was  $4.5 \text{ mol m}^{-2} \text{ d}^{-1}$  with a standard error of  $1.0$  ( $72 \pm 16 \text{ g m}^{-2} \text{ d}^{-1}$ ).

Based upon the literature summarized in Table 4.2 it is clear that the fraction of methane oxidized in all cover soils is much greater than the default value of 10%. Of the 47 determinations of methane oxidation reported in Table 4.2, only 4 report values less than 10%.

One reports a value of 10%, the aforementioned Czepiel et al., study (1996a). This study was the first to report the fraction of methane oxidized in landfills, and because of this, it has received undue weight. It should be noted that the 10% was an estimate of an annual average based on microbial kinetics and the effect of temperature on these kinetics. The work was done in a northern climate (New Hampshire, USA) and activity was very low in the winter.

#### **4.6 COVER THICKNESS, TEMPERATURE AND MOISTURE**

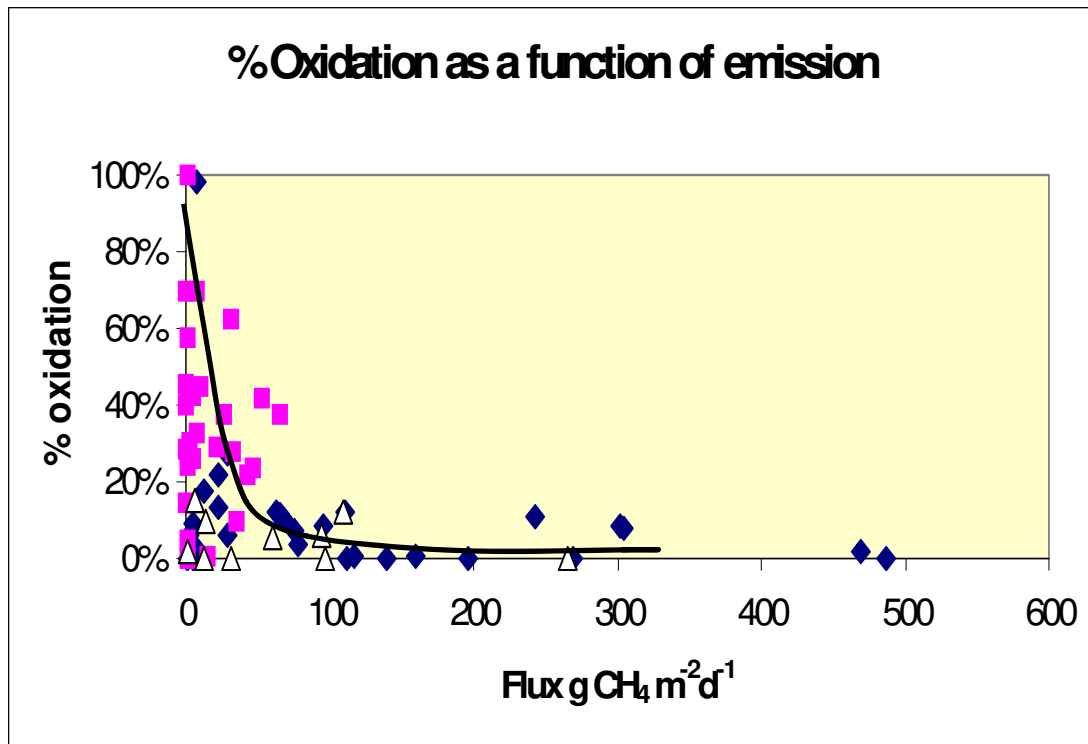
We cannot offer oxidation guidelines as a function of cover thickness as there was no trend in the data with oxidation as a function of cover thickness in the studies compiled. There are several reasons why we did not observe this trend. First, oxidation is confined to the surface-most zone of a cover where oxygen penetrates via diffusion from above. Typically this area will be restricted to the upper portion of the soil column so cover thickness overall does not affect oxygen penetration except in that a thicker cover may attenuate methane upward advection and not push oxygen out of the soil. Secondly, the data were compiled from a number of soils covers and include a variety of factors so any trend might have been obscured by other sources of variability. In general, however, it is known that a thicker cover will result in increased retention times for transported methane. This increased retention of methane in the landfill soil covers results in lower emission rates and in a higher fraction of methane being oxidized (Stern et al., 2007).

Temperature and moisture are also known to affect the efficiency of microbial methane oxidation. Methanotrophs have an optimum temperature range of 30 to 40 °C. Optimal water content depends on soil texture and bulk density—more water is better as long as the remaining gas-filled porosity is adequate so that methane and oxygen diffusion is not attenuated. Factoring in these parameters will require the development of models (Abichou et al., 2007).

#### **4.7 METHANE OXIDATION, ABSOLUTE RATES OR PERCENTAGES?**

It is also important to consider how methane oxidation rates are expressed. Currently oxidation is generally expressed as a percent of the transported methane that is oxidized, that is, a constant fraction of landfill emissions, increasing linearly as emissions increase. Recent studies show that the percent oxidation and emission have the opposite trend. Methane oxidation percentage is an inverse function of the rate of emission (Stern et al., 2007; Chanton et al., unpublished data, Figure 4.2). At lower rates, the methanotrophs in the soil cover can consume a larger portion of the methane delivered to them, oxidizing up to 95 to 100% (Humer and Lechner, 1999, 2001a, Huber-Humer 2008; Powelson et al., 2006, 2007). As flux rates increase, their percent oxidation decreases and they can become overwhelmed with methane. As methane emission increases, percent oxidation decreases (Powelson et al., 2006, 2007).





**Figure 4.2. Data Collected from a Mid-Continent Landfill in the USA Located in a Temperate Climatic Region.** *The percent oxidation was determined with the isotope technique on emitted methane samples, while the emission rate was determined with chambers (Chanton, Abichou, Hater, Green, and Goldsmith, unpublished data, 2007). These results show that at low methane emission rates, the landfill cover has a greater capacity to oxidize methane, up to 100% of methane emissions. As methane emissions increase, the percent oxidation decreases. The most widely accepted current model of methane oxidation, which it is 10% of methane emissions is not supported by these data.*

From these data, it is clear that predictions of soil oxidation cannot be accurately made independent of an estimate of flux. If flux through the cover is very high, oxidation will be limited by a lack of oxygen in the pore space. If the flux is zero, methane is not available to oxidize. When the flux is low but above zero, the soil has the capacity to oxidize 100% of the methane delivered (Humer and Lechner, 1999, 2001a, Huber-Humer 2008; Powelson et al., 2006, 2007). The correct model for evaluating cover soil oxidation is similar to that of a biofilter. The cover soil has a certain oxidation capacity, and as long the flux is controlled within the limits of that capacity, 100% oxidation can be expected. Excessive flux is analogous to overloading a biofilter and negates the oxidation capacity of the cover soil.

In a design scenario, a landfill designer could eliminate the methane flux by constructing a gas collection system and soil barrier that limits the upward flux to a range less than or equal to the oxidation capacity of the cover system. A reasonable goal would be a functionally layered cover system in which the bottom layer regulates LFG flux as a barrier (typically clay soil). The upper

layer would function as an oxidation medium and work in tandem with the underlying barrier layer (eg Stern et al., 2007).

The current accounting system, which unequivocally states that oxidation is 10% of emissions, discourages innovation to reduce methane emissions from landfills. Particularly discouraging are regulations that assume capture efficiency is 75% of generated methane and that fugitive emissions and oxidation make up the 25% balance. Under this scenario, a landfill that improves its gas collection system to achieve results better than 75% is “rewarded” by having their emissions estimate increased. In other words, the more methane a landfill captures by improving its gas collection system, the more methane they are charged with emitting. Alternatively, if the landfill turns down its gas collection system, they would be charged with emitting less methane. It’s a lose-lose situation. This use of default values also affects the methane oxidation because as the landfill increases methane collection efficiency, methane oxidation rates increase too; however, the use of a single default value does not recognize this increased oxidation

The average value for methane oxidation compiled from the Vmax flask incubation results (Table 4.3) is  $28.0 \text{ mol m}^{-2} \text{ d}^{-1}$ , roughly a factor of 6 times greater than the results compiled here. These flask incubations represent the maximum possible oxidation rate because they occur in unlimiting conditions for methane and oxygen, and the soil is broken up and fully exposed to the gas in the flask. In similar incubations of wetland rhizosphere material the rate of methane uptake is considerably greater than rates of methane emission, even on occasion outstripping rates of in vitro methane production (Gerard and Chanton, 1993). Nonetheless, this value is useful, as it may represent the maximum value that can be achieved by soil methane oxidizing bacteria. The value represents a goal for the design of landfill covers to achieve maximum rates of methane oxidation.

**Table 4.3. Comparison of Oxidation Rates Calculated from Vmax or Maximum V and Flux.**

Material	$V_{\max}$	Bulk Density	Oxidation rate from $V_{\max}$ ( $J_{\text{oxv}}^{\dagger}$ )	Oxidation Rate from flux ( $J_{\text{oxf}}$ )	$J_{\text{oxv}} / J_{\text{oxf}}$	Reference
	$\text{mol Mg}^{-1} \text{ d}^{-1}$		$\text{mol m}^{-2} \text{ d}^{-1}$	$\text{mol m}^{-2} \text{ d}^{-1}$		
Coarse sand	24	1.65 <sup>‡</sup>	11.86	10.4	1.14	Kightley et al., 1995
Sandy-clay loam	62.3	1.63 <sup>‡</sup>	30.44	0.91	33.59	Czepiel et al., 1996a&b
Loamy agricultural soil	22.5	1	6.72	8.24	0.82	De Visscher et al., 1999
Compost	720	0.64	138.46	6.25	22.15	Wilshusen et al., 2004
	Maximum $V^{\S}$					
	$\text{mol Mg}^{-1} \text{ d}^{-1}$					
Loam	24	1.59 <sup>‡</sup>	11.47	nr <sup>¶</sup>	na <sup>¶</sup>	Stein and Hettiaratchi, 2001
Sandy loam	3.6	1.62 <sup>‡</sup>	1.75	nr <sup>¶</sup>	na <sup>¶</sup>	Hilger et al., 2000a
Sandy loam	70.8	1.62 <sup>‡</sup>	34.5	nr <sup>¶</sup>	na <sup>¶</sup>	De Visscher et al., 2001
Loamy sand	177	1.62 <sup>‡</sup>	85.84	nr <sup>¶</sup>	na <sup>¶</sup>	Scheutz and Kjeldsen, 2004
Sandy silt	2.3	1.36 <sup>‡</sup>	0.92	nr <sup>¶</sup>	na <sup>¶</sup>	Scheutz et al., 2003
Sandy soil	37.5	1.62 <sup>‡</sup>	18.28	nr <sup>¶</sup>	na <sup>¶</sup>	Park et al., 2005
Humic soil	129.6	1.59 <sup>‡</sup>	61.92	nr <sup>¶</sup>	na <sup>¶</sup>	Figuerola, 1993
Till	60	1.59 <sup>‡</sup>	28.67	nr <sup>¶</sup>	na <sup>¶</sup>	Figuerola, 1993
Biowaste compost	192	0.64 <sup>‡</sup>	36.92	nr <sup>¶</sup>	na <sup>¶</sup>	Figuerola, 1993
Sandy loam	0.3	1.62 <sup>‡</sup>	0.15	nr <sup>¶</sup>	na <sup>¶</sup>	Jones and Nedwell, 1993
Sand mixed with clay	4.1	1.62 <sup>‡</sup>	1.96	nr <sup>¶</sup>	na <sup>¶</sup>	Whalen et al., 1990
Sandy loam	0.004	1.62 <sup>‡</sup>	0.002	nr <sup>¶</sup>	na <sup>¶</sup>	Boeckx and Van Cleemput, 1996
Sandy loam	0.015	1.62 <sup>‡</sup>	0.007	nr <sup>¶</sup>	na <sup>¶</sup>	Boeckx et al., 1996
Sandy loam	28.5	1.62 <sup>‡</sup>	13.89	nr <sup>¶</sup>	na <sup>¶</sup>	Christophersen et al., 2000
Loamy clay	0.014	1.43 <sup>‡</sup>	0.006	nr <sup>¶</sup>	na <sup>¶</sup>	Bender and Conrad, 1994
Sandy loam	28.2	1.62 <sup>‡</sup>	13.74	nr <sup>¶</sup>	na <sup>¶</sup>	Borjesson et al., 2004

Material	Maximum $V^{\S}$	Bulk Density	Oxidation rate from $V_{\max}$ ( $J_{\text{oxv}}^{\dagger}$ )	Oxidation Rate from flux ( $J_{\text{oxf}}$ )	$J_{\text{oxv}} /$ $J_{\text{oxf}}$	Reference
	$\text{mol Mg}^{-1} \text{ d}^{-1}$					
Sandy loam	37.8	1.62 <sup>*</sup>	18.42	nr <sup>¶</sup>	na <sup>¶</sup>	Borjesson et al., 2004
Silty loam	259.5	1.49 <sup>*</sup>	115.74	nr <sup>¶</sup>	na <sup>¶</sup>	Borjesson et al., 1997
Sandy loam	72	1.62 <sup>*</sup>	35.09	nr <sup>¶</sup>	na <sup>¶</sup>	Borjesson, 1997
Sand	24	1.66 <sup>*</sup>	11.93	nr <sup>¶</sup>	na <sup>¶</sup>	Borjesson and Svensson, 1997

<sup>†</sup>  $J_{\text{oxv}} = (V_{\max}) (\text{Bulk Density}) (\text{Depth})$ . An oxidation depth of 0.3 m was assumed.

<sup>\*</sup> Bulk density estimated from saturated water content of the soil texture (Leij et al., 2003) and a mineral density of  $2.65 \text{ g cm}^{-3}$ , except for biowaste compost which was estimated from leaf compost (Wilshusen et al., 2004).

<sup>§</sup> Maximum oxidation rates taken from Scheutz et al. (in press, Table 2).

<sup>¶</sup> nr = not reported in Scheutz et al. (in press); na = not applicable.

Specific estimation methods listed by reference below.

**Table 4.3 Footnotes:**

Czepiel et al., 1996a&b.  $J_{\text{oxf}}$  from average of chamber outflux and assumed  $f_{\text{ox}}$  of 0.2.

De Visscher et al., 1999.  $V_{\max}$  averaged from most active depths of agricultural soil.

Wilshusen et al., 2004.  $J_{\text{oxf}}$  from steady-state rate in repacked compost column.

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## 5 CARBON SEQUESTRATION

### 5.1 EXECUTIVE SUMMARY

In developing our approach to carbon sequestration, Solid Waste Industry for Climate Solutions (SWICS) reviewed and has herein summarized the positions of the United States Environmental Protection Agency (USEPA), the Intergovernmental Panel on Climate Change (IPCC), the California Energy Commission (CEC), and the California Air Resources Board (CARB) on carbon sequestration in landfills and other industries. An exhaustive review of the available technical literature was also conducted. Based on this review, SWICS is proposing that the research by Dr. Morton Barlaz of North Carolina State University and the USEPA be used to develop carbon storage values for organic wastes contained in the MSW stream.

It is clear that carbon is being stored in landfills and removed from the carbon cycle, and inventory methods must account for this carbon sink. SWICS proposes the following carbon storage values for refuse placed in landfills:

<b>Table 5.1 - Summary of Carbon Storage Values</b>	Grass	Leaves	Branches and Wood	Food	Coated Paper, Magazines and 3 <sup>rd</sup> Class Mail	Old Newsprint	Old Corrugated Containers	Office Paper	Municipal Solid Waste (15% moisture)	Municipal Solid Waste (20% moisture)	Municipal Solid Waste (25% moisture)
USEPA Carbon Storage Factor (MTCE/Wet Short Ton Refuse)	0.07	0.25	0.31	0.02	0.23	0.36	0.22	0.04	0.09	0.09	0.08

The weighted average carbon storage factor (CSF) for all California solid waste disposed in 2003 is about 0.082 megagrams (Mg) carbon equivalent (MTCE)/wet ton of refuse disposed. Using this CSF, one ton of waste disposed would result in storage of 0.30 Mg carbon dioxide equivalent (MTCO<sub>2</sub>E).

Use of this factor would result in substantially more carbon storage than is currently estimated by the CEC and CARB GHG inventory. If applied to the 40.2 million tons of MSW disposed in California in 2003, a CSF of 0.082 MTCE/wet ton of refuse would result in an estimated sequestration of 12.1 MMTCO<sub>2</sub>E. This is almost two times the carbon storage of 6.42 MMTCO<sub>2</sub>E calculated by the CEC for 2003 and more than the estimated greenhouse gas (GHG) emissions from landfills that were calculated to be 5.67 MMTCO<sub>2</sub>E by the CEC for 2003. CARB estimates that 4.62 MMTC were stored in California landfills, which equates to 17.0 MMTCO<sub>2</sub>E. However, the carbon sequestration value calculated by CARB is not included in the GHG inventory totals and is only presented as an informational item.

### 5.2 BACKGROUND ON CARBON SEQUESTRATION

In the United States, MSW is composed of 30 to 50 percent cellulose, 7 to 12 percent hemicellulose, and 15 to 28 percent lignin on a dry weight basis, with cellulose and hemicellulose representing about 90 percent of the biodegradable portion of MSW (Hilger and

Barlaz, 2001). Because not all of the cellulose and hemicellulose decompose, and none of the lignin decomposes, significant carbon sequestration occurs in landfills.

Increases in atmospheric levels of carbon dioxide are attributed mainly to fossil fuel burning and deforestation. While efforts to reduce emissions of carbon dioxide continue, increases in carbon dioxide emissions can also be offset, to a degree, by accumulation in carbon sinks such as plant biomass and oceans. Landfills are a carbon sink and should be included in carbon inventories of landfills.

### **5.2.1 Summary of How Sequestration has been Allowed for Other Industries**

The inclusion of carbon sequestration in GHG emissions inventory and accounting efforts is important because carbon sequestration is a mechanism that reduces the rate of accumulation of carbon dioxide in the atmosphere. Carbon sequestration concepts in greenhouse gas (GHG) accounting and related guidelines in the Kyoto Protocol (Protocol) have been evaluated by numerous researchers. Marland and Schlamadinger (1999) noted that although the Protocol would sometimes allow emissions credits for sequestered carbon, it would always give credits when fossil-fuel carbon dioxide is displaced. Gillespie (1999) added that the limitation by the Protocol of allowable activities to land-based activities, specifically, reforestation and deforestation, combined with the largely unspecified nature of the inclusion of sequestration and uncertainties in the estimating methodology, contributed to the U.S. decision in 2001 to not participate in the Kyoto Treaty.

The revised guidelines for national emissions inventories published in 2006 by the IPCC stipulate the inclusion of carbon sequestration through land use and forestry in national GHG inventories as an offset to gross GHG emissions from other sources (IPCC, 2006). Table 5.2 shows the forestry practices that sequester or preserve carbon.

**Table 5.2 – Forestry Practices Recognized as Carbon Sinks**

<b>Key Forestry Practices</b>	<b>Typical definition and some examples</b>	<b>Effect on greenhouse gases</b>
Afforestation	Tree planting on lands previously not in forestry (e.g., conversion of marginal cropland to trees).	Increases carbon storage through sequestration.
Reforestation	Tree planting on lands that in the more recent past were in forestry, excluding the planting of trees immediately after harvest (e.g., restoring trees on severely burned lands that will demonstrably not regenerate without intervention).	Increases carbon storage through sequestration.
Forest preservation or avoided deforestation	Protection of forests that are threatened by logging or clearing.	Avoids CO <sub>2</sub> emissions via conservation of existing carbon stocks.
Forest management	Modification to forestry practices that produce wood products to enhance sequestration over time (e.g., lengthening the harvest-regeneration cycle, adopting low-impact logging).	Increases carbon storage by sequestration and may also avoid CO <sub>2</sub> emissions by altering management. May generate some N <sub>2</sub> O emissions due to fertilization practices.

Instead of being accounted for in landfills, carbon storage that results from forest products and yard trimmings disposed in landfills is accounted for in the Land-Use Change and Forestry chapter, as proposed in the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006).

Practices such as revegetation, forest management, crop management, and grazing land management have also been allowed as sequestration by the United Nations Framework Convention on Climate Change (UNFCCC) conference of parties. However, inclusion of carbon sequestration by landfilling is not currently included in emissions accounting.

### **5.3 SUMMARY OF LITERATURE ON CARBON STORAGE FROM MUNICIPAL SOLID WASTE**

Most of the literature reviewed to support the inclusion of carbon sequestration of landfills in GHG emission inventories is based on work performed by Barlaz (1998) and the USEPA (2006).

Barlaz conducted laboratory-scale biodegradation studies to quantify carbon sequestration for the major biodegradable components of MSW including grass, leaves, branches, food, coated paper, old newsprint, old corrugated containers, and office paper. A CSF was calculated that represented the mass of carbon stored (not degraded) per initial mass of the component. The CSF for the MSW components ranged from 0.05 to 0.47<sup>2</sup> kg of carbon sequestered per dry kg of the waste component.

USEPA has used these same factors, with some slight modification, in their report: “Solid Waste Management and Greenhouse Gases: A Life-Cycle Assessment of Emissions and Sinks” (USEPA, 2006). The differences between Barlaz’s study and the USEPA’s study are based on the following updates:

1. The CSF for leaves was corrected after an error was found in the original calculation. (Barlaz, 2008a).
2. USEPA equated Magazines and 3rd class mail with coated paper and uses the same CSF. Barlaz suggests using the average CSF for newspaper, office paper, and coated paper, which results in a CSF of 0.25. (Barlaz, 2008a).
3. The CSF for MSW was corrected after an error was found in the original calculation. (Barlaz, 2008b)

The wet CSF was calculated from the dry CSF and the moisture content. Table 5.3 shows the dry storage factor, as well as the typical moisture content. Because the composition of MSW can vary by region, the CSFs at 15%, 20%, and 25% moisture are shown in Table 5.3.

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<sup>2</sup> The original range as published was 0.05 to 0.54. However, the upper end of this range was the result of a calculation error that was later corrected.

**Table 5.3 – Carbon Storage Factors (Barlaz 1998, 2008a, 2008b)**

Component	CSF (dry basis) (kg C/ kg waste)	Moisture Content (mass fraction)	CSF (wet basis) (kg C/ kg wet waste)	CSF (Metric Tons C/wet short ton)
Grass	0.24	0.7	0.072	0.065
Leaves	0.385	0.3	0.27	0.25
Branches	0.38	0.1	0.34	0.31
Wood	0.38	0.1	0.34	0.31
Food Waste	0.08	0.7	0.02	0.02
Coated Paper	0.27	0.05	0.26	0.23
Magazines/3rd Class Mail	0.25	0.05	0.24	0.22
Newspaper	0.42	0.05	0.40	0.36
Old Corrugated Containers	0.26	0.05	0.25	0.22
Office Paper	0.05	0.05	0.05	0.04
Municipal Solid Waste	0.12	0.15	0.10	0.09
Municipal Solid Waste	0.12	0.2	0.10	0.09
Municipal Solid Waste	0.12	0.25	0.09	0.08

These studies, as well as the literature, indicate that significant carbon sequestration occurs in landfills because of incomplete biodegradation of lignocellulosic components of MSW. These findings are important because they are used in the development of national GHG emission inventories. In addition, carbon-trading protocols require defensible data on which to base trades and purchases.

#### **5.4 INDUSTRY POSITION ON CARBON SEQUESTRATION OF LANDFILLS**

An accurate GHG inventory for a landfill would be derived from a carbon mass balance for the landfill and would reflect the net carbon emissions from the landfill. This mass balance approach would account for the amount and types of carbon containing wastes placed in the landfill and their fates over time.

Carbon sequestration estimates by the USEPA only account for wood waste and yard trimmings within a landfill (USEPA 2008) and conflicts with a 2006 US EPA study. Restricting carbon sequestration estimates to only these categories produces an estimate that is biased low as it neglects paper and food waste. Based upon the literature summarized in Section 5.3, the industry focus on estimating carbon sequestration in landfills is geared towards accounting for all types of biogenic carbon containing wastes.

#### **5.5 PROPOSED NUMERIC VALUES FOR CARBON SEQUESTRATION FOR LANDFILL DISPOSAL**

The 2006 inventory of GHG emissions published by the CEC indicates that landfill disposal of urban wood waste and yard trimmings is a GHG sink. The report calculated the GHG sink for yard trimming and wood waste, and assumed that these were the only waste categories that

contributed to storage in a landfill. The report used a CSF of 0.26 MTCE/short wet ton of woody waste deposited, and calculated that the GHG sink for lumber and yard trimming disposal was 6.42 million MTCO<sub>2</sub>E for 2003. However, in California, these waste categories (yard trimmings and lumber) represent only 16.4% of the total California waste stream; therefore, restricting estimates of carbon storage to only these waste types produces an estimate of carbon storage for total waste disposed of about 0.043 MTCO<sub>2</sub>E/short wet ton of total MSW disposed which is biased low. This carbon storage is only about 20% of the amount of carbon storage attributed to landfill disposal by USEPA (2004). If paper, cardboard, and other MSW components are included, the overall carbon storage for total waste disposed in California is 0.12 MTCO<sub>2</sub>E/ short wet ton as derived in the following section. Thus, the total amount of carbon storage in California landfills may be up to three times greater than the CEC inventory estimated.

In the 2007 CARB inventory of California's GHG emissions from 1990 to 2004, CARB estimated that 4.62 MMTC were stored in California landfills in 2003. The carbon storage value calculate by CARB includes the woody wastes that were included in the CEC inventory, plus paper, cardboard, textiles, diapers, construction and demolition waste, medical waste, sludge and manure. This carbon storage is equal to 17.0 MMTCO<sub>2</sub>E, but it is included only as an informational item, not in the inventory totals.

It is apparent that landfills sequester more carbon than is currently credited in California. The following section presents proposed carbon storage factors for estimation of carbon sequestration in landfills.

## 5.6 SUGGESTIONS FOR HOW TO INCORPORATE AND USE CARBON STORAGE FACTORS IN STATEWIDE GHG INVENTORIES

For purposes of computing the carbon footprint of a landfill, it is proposed that the CSF values presented in Table 5.3 be used. The CSF should be applied to the tons of waste placed into each state's landfills by refuse type for each inventory year. If a site specific waste characterization is available, then waste composition data should be used to calculate carbon storage. If no site specific data are available, or the data are insufficient to calculate the carbon storage, regional, statewide, or national data can be used instead. Tons disposed are typically on a wet-weight basis; therefore, the most appropriate CSF value should be selected based on the known or estimated moisture content. The final step is to convert tons of sequestered carbon equivalents to sequestered tons of carbon dioxide equivalents. This is done by multiplying by the molecular weight ratio of carbon dioxide to carbon ( $44/12 = 3.67 \text{ MTCO}_2\text{E/MTCE}$ ). Thus to convert one short ton of material disposed to the relative GHG reduction factor, the following formula should be used:

$$\text{Short Wet Tons of Material} \times \text{CSF (MTCE/short wet ton)} \times (3.67 \text{ MTCO}_2\text{E/MTCE}) = \text{Sequestered Carbon in MTCO}_2\text{E}.$$

To calculate a composite CSF for a mixed waste stream, the carbon storage for each component should be calculated and summed. Table 5.4 shows an example of this summation using the composition of the California waste stream, which was characterized by the California Integrated

Waste Management Board in 2004. The resultant CSF for California is 0.082 MTCE/wet ton of refuse. The CIWMB study did not distinguish between leaves and grass, which comprised 4.2% of the waste stream. It was assumed that leaves and grass were present in equal proportions.

**Table 5.4 – Calculation of the California Waste Stream CSF**

Major Waste Category	Waste Sub-category	Wet Mass <sup>a</sup> thousand Mg wet/yr	% of MSW (wet basis) <sup>b</sup>	Estimated % Moisture Content	Dry Mass thousand Mg-dry/yr	Carbon Storage Factor (kg C dry/kg or ton C/dry ton)	Carbon Sequestered for MSW (thousand Mg C/yr)
Paper		7,662	27.4%	5.7%	7,227	0.25	1,811
Newspapers		805	2.9%	6.0%	756	0.42	318
High-Grade/Office Paper		712	2.5%	6.0%	670	0.05	33
Mixed Paper		1,352	4.8% <sup>c</sup>	6.0%	1,271	0.24	299
Magazines/Catalogs		282	1.0%	6.0%	265	0.27	72
Corrug. Cardboard/Kraft Bags		2,448	8.8%	5.0%	2,325	0.26	605
Remainder/Comp Paper		2,063	7.4% <sup>d</sup>	6.0%	1,940	0.25	485
Organics		10,358	37.0%	50.7%	4,311	0.16	704
Tires		115	0.4%	2.0%	113	0	0
Textiles		860	3.1%	10.0%	774	0.01 <sup>e</sup>	9
Wood (non-C & D)		109	0.4%	20.0%	87	0.38	33
Food Waste		5,311	19.0%	70.0%	1,593	0.08	127
Yard Trimmings <sup>f</sup>		2,374	8.5%	60.0%	949	0.34 <sup>g</sup>	322
Misc. organics		1,590	5.7% <sup>h</sup>	50.0%	795	0.27 <sup>i</sup>	212
Other categories		9,947	35.6%			0	0
	TOTAL =	27,967	100.0%		21,231		2,515
				% Moisture Content of MSW =	24.1%		
						MSW Carbon Sequestration Factor (tons C/dry ton) =	0.118
						MSW Carbon Sequestration Factor (MTCE/wet ton) =	0.082

a. California Integrated Waste Management Board. (2004). Statewide Waste Characterization Study. CIWMB Pub. No. 340-04-005. [www.ciwmb.ca.gov/WasteChar/DBMain.htm](http://www.ciwmb.ca.gov/WasteChar/DBMain.htm). Accessed 2/1/2008

b. Moisture contents from Tchobanoglous et al., 1993.

c. Mixed paper moisture content and CSF are an average of newspaper and office paper.

d. Composite paper moisture content and CSF are an average of newspaper, office paper, glossy paper and OCC/Kraft bags.

e. CSF values assumed the cotton portion of textiles were similar to office paper. Thus, the CSF values were obtained by multiplying the cotton fraction of textiles by the CSF for office paper. Roughly ~23.7% of textiles consumed in the U.S. from 2001-05 were made of cotton (Fiber Economics Bureau, 2006).

f. Yard trimming moisture content and CSF are a weighted average based on relative contribution of grass (30.3%), leaves (40.1%) and brush (29.6%) (Oshins and Block, 2000). CSF values of 0.24, 0.385 and 0.38 kg C dry kg refuse<sup>-1</sup> were used for grass, leaves and brush, respectively, are corrected values initially published by Barlaz (1998).

g. CSF values for branches were used for wood (non-C&D) and lumber.

h. Misc organics moisture content and CSF are an average of wood (non-C&D), food waste and yard trimmings.



The resulting CSF for California is 0.082 MTCE/wet ton of refuse. Using this CSF, one ton of waste disposed would yield the following amount of sequestered C in MTCO<sub>2</sub>E:

$$1 \text{ Short Wet Ton of MSW} \times (0.082 \text{ MTCE/short wet ton}) \times (3.67 \text{ MTCO}_2\text{E/MTCE}) = 0.30 \text{ MTCO}_2\text{E}.$$

Use of this factor would result in substantially more carbon storage than is currently estimated by the CEC GHG inventory. If applied to the 40.2 million tons of MSW disposed in California in 2003, a carbon storage factor of 0.082 MTCE/wet ton of refuse would result in an estimated sequestration of 12.1 MMTCO<sub>2</sub>E which is nearly identical to the value suggested in the 2007 CARB study.

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## 6 PROPOSED METHODOLOGY FOR USE OF PROPOSED FACTORS

This section describes how these proposed values for collection efficiency, methane oxidation, and carbon sequestration could be used to replace the current California Air Resources Board (CARB) values for collection efficiency (75%), methane oxidation (10%), and carbon sequestration (informational only).

Collection Efficiency – For each year:

- Obtain actual landfill gas (LFG) recovery data for the LFG collection system;
- Determine and evaluate the design, operation, performance, and the type of LFG collection system (New Source Performance Standards (NSPS)/air quality compliance or other);
- Determine the percent of landfill surface under each cover type (daily, intermediate, final, or geomembrane) during the year;
- Calculate a weighted average collection efficiency based on cover type and other factors;
- Divide the actual recovery data by a calculated average collection efficiency value based on a weighted average of land surface area by cover type.

Methane Oxidation – For calculating the methane oxidation potential for each year:

- Determine the percent of landfill surface area under each cover type (organic, clay, sand, or other) during the year;
- If oxidation rates are used, apply the methane oxidation rate factor for each cover type to calculate the additional methane emission reduction; or
- If percent oxidation is used, calculate an average oxidation percentage based on a weighted average of land surface area by cover type. Apply this factor to the amount of methane not collected by the LFG system.

Carbon Sequestration – for calculating amount of carbon sequestered:

- Determine the weighted average computed carbon storage factor (CSF) for the MSW in the landfill;
- Determine the amount of MSW landfilled for the year (in short wet tons);
- Multiply the weighted average computed CSF for the MSW in the landfill by the amount of MSW landfilled for the year (in short tons) to calculate the sequestered amount in metric tonnes carbon dioxide equivalent (MTCO<sub>2</sub>E);
- Include as a separate line item in landfill inventory for avoided emissions by carbon sequestration.

## 7 CONCLUSIONS

In conclusion, if the proposed new values for collection system efficiencies, methane oxidation in cover soils, and carbon storage factors for carbon sequestration are used in the state's GHG emissions inventory, GHG emissions from landfills will be significantly lower and the reduction potential will be higher than the current estimates. The differences in the previously used values and the proposed values need to be taken into account in order to more accurately define the state-of-the-practice technology used in the engineered landfills and the type of MSW landfilled.