Guidance Document

to

Reduce

Greenhouse Gas

Emissions

from

Landfills

!!!!!!FINAL REPORT DRAFT!!!!!!!!

Florida State University

Hinkley Center for Solid and Hazardous Waste Management

CHAPER I

INTRODUCTION AND BACKGROUND

1.1 BASISCS OF LANDFILL GAS

In municipal solid waste landfills, gas production is caused by the breaking down of organic matter. Microorganisms eat away at organic matter and this decomposition creates gas. Gas production in landfills occurs in five different phases. The first four phases are bacterial decomposition phases and the fifth phase is the stabilization period.

Stage One begins as soon as the garbage is placed in the landfill. The garbage is decomposed by bacteria that use oxygen and produce large amounts of carbon dioxide. No methane is produced in this stage. Stage One will continue until the bacteria can no longer receive oxygen. After the landfill is closed and no more oxygen is inside the landfill, Stage Two begins.

In Stage Two, different bacteria break down the garbage. Instead of consuming oxygen like the bacteria in Stage One, these bacteria consume the organics in the landfill and generate acid. They decompose and ferment organics in the landfill.

Since all the oxygen has been depleted in the landfill, the bacteria in Stage Three use sources other than oxygen and produce methane gas and carbon dioxide.

Once the methane and carbon dioxide levels have leveled out, the landfill is said to be in Stage Four. During the beginning of this stage, methane and carbon dioxide are at their highest levels. Gas composition in Stage Four is about 50/50 methane/carbon dioxide with small amounts of trace gases. After all of the carbon in the landfill has been decomposed, Stage Five will commence. As time progresses, bacteria will cease to break down the garbage and gas production will be minimal.

1.1.1 Landfill Gas Production and Composition:

In general, when there is no oxygen in the landfill, the composition of gas produced is approximately 50 percent methane and 50 percent carbon with trace amounts (<1 percent) of nitrogen, oxygen, hydrogen sulfide, hydrogen and non-methane organic compounds (NMOCs). The more organic waste and moisture are present in a landfill, the more landfill gas is produced by the bacteria during decomposition. The more chemicals disposed in a landfill, the more likely there will be volatile organic compounds (compounds that are easily evaporated) and other gases produced.

1.1.2 The Four Phases of Bacterial Decomposition:

Bacteria break down landfill waste in four phases. The types of gases produced changes with each of the four stages of decomposition. Since landfills often accept waste over a 20- to 30-year period, waste in different parts of the landfill may be experiencing different stages of decomposition at any given time. Waste in one area may be in a different phase of decomposition than more recently buried waste in another area.

Phase I bacteria consume oxygen while breaking down the long molecular chains of complex carbohydrates, proteins and fats that make up organic waste. This breakdown of organic waste produces carbon dioxide. Nitrogen levels are high at the beginning of this phase but decline as the landfill moves through the four phases. Phase I will continue until all of the oxygen in the landfill is depleted. This phase can last for days or months, depending on the amount of the available oxygen. The more oxygen present, the longer Phase I will last.

Phase II starts after the oxygen in the landfill has been used up. Phase II bacteria use the compounds the Phase I bacteria created. They convert these compounds into acetic, lactic and formic acids and they also form alcohols such as methanol and ethanol. During Phase II of Microbial Decomposition, the landfill becomes very acidic. As the acids mix with the moisture present in the landfill, they cause certain nutrients to dissolve, making nitrogen and phosphorus available to the increasingly diverse species of bacteria in the landfill. These processes release gases such as carbon dioxide and hydrogen. If the landfill is disturbed, or if oxygen is somehow introduced into the landfill, microbial processes will return to Phase I.

Phase III decomposition starts when a different kind of bacteria begins eating the organic acids produced by the Phase II bacteria. They consume it and produce acetate, another kind of organic acid. This process causes the landfill to become a more neutral environment in which methane-producing bacteria begin to establish themselves. Methane- and acid-producing bacteria have a symbiotic, or mutually beneficial, relationship. Acid-producing bacteria create compounds for the methane-producing bacteria to consume. Methanogenic, or methane-producing bacteria, consume carbon

dioxide and acetate. This benefits the acid-producing bacteria because large amounts of carbon dioxide and acetate kill acid-producing bacteria.

Phase IV decomposition is the last phase. It begins when both the composition and production rates of landfill gas remain relatively constant. The gas produced during Phase IV usually contains approximately 45 percent to 60 percent methane by volume, 40 percent to 60 percent carbon dioxide and 2 percent to 9 percent other gases, such as sulfide. During Phase IV, gas is produced at a steady rate for about 20 years, but that gas will continue being emitted for 50 or more years after the waste is placed in the landfill (Crawford and Smith 1985). Gas production might last longer, for example, if greater amounts of organics are present in the waste, such as at a landfill receiving higher than average amounts of domestic animal waste (ATSDR, 2001).

Because refuse is placed in the landfill at different times and consists of different types of solid waste at different moisture content, all four bacterial decomposition phases of degradation may be occurring simultaneously within the landfill.

1.1.3 Factors Affecting Landfill Gas Production

Gas production rates depend on the rate of decomposition, which is in turn affected by moisture content of the waste, temperature, and soil cover permeability, amount of precipitation, composition of the waste, refuse particle size, compaction and landfilling practices. Moisture is essential for bacterial survival. Ideal moisture for decomposition is one that approaches saturation. If the moisture content falls below 40 percent, biological reactions can be retarded. If the moisture content falls below 20 percent, biological reactions essentially stop. Therefore, in dry climates, methane-producing decomposition is highly unlikely to occur. Methane will only be produced when oxygen is no longer available inside the landfill, so it is essential to cap the landfill. However, by capping it, this reduces the availability of moisture because rain and humidity cannot enter the landfill. A balance needs to be sought to optimize both of these conditions. In addition, small particles decompose more quickly due to the high surface area to mass ratio. Therefore, shredded waste produces gas more quickly than non-shredded waste.

Prediction or measurement of these factors is impractical on the scale of a large landfill. Municipal Solid Waste is extremely variable in organic content and particle size. It is more useful to construct a model in terms of general trends observed in the profession over several years and in varying climates. This is the method adopted by the EPA, and is the only method currently accepted in modeling gas generation rates for the sizing of gas collection systems (USEPA, 1997). The Landfill Gas Emissions Model (LandGEM) currently used is based on a first order decomposition equation (USEPA 2005):

$$Q = \sum_{i=1}^{n} 2 \cdot k \cdot L_o \cdot M_i \cdot e^{-k \cdot t_i}$$

where n is the number of years of waste placement, Q is the volume of gas production per year (m3/year), k is the landfill gas generation rate constant in reciprocal years, Lo is the methane generation potential in cubic meters per megagram, Mi is the mass of the solid waste section placed in year i in megagrams, and ti is the age of the waste section placed in year i in years. To use the model, the annual acceptance rate (Mi) and age of each year's waste mass (ti) must be known, as well as the duration of the active period of the landfill. Factors that must be assumed using guidelines provided in the model are the methane generation potential of the waste (Lo, ft3/yr or m3/Mg), as well as the methane generation rate constant (k, yr-1). The New Source Performance Standards guidelines provide regulatory defaults to yield conservative estimates of landfill gas production in the event that actual coefficients are unknown (USEPA 2005). As the landfill grows, site specific values can be determined through testing (USEPA 2005). The software released by the EPA provides two sets of default values to determine the applicability of Federal regulatory requirements as well as provide for emission inventory needs.

The need for inventory of emissions is based on both economic concerns as well as environmental concerns. Methane, which accounts for nearly half of all landfill emissions, is an important resource for landfill operators looking to convert landfill gas to energy. Reasonable estimates of methane production rates are necessary in determining sizing of gas collection systems, energy conversion systems, and potential uses and customers for the energy produced. Duration of methane production is also an important estimate to have to plan for monitoring periods and to project future costs and earnings in

developing and operating a landfill gas to energy conversion system, or a gas scrubbing and flaring system. As for the environmental concerns, methane emissions must be kept under control due to their potential as a greenhouse gas.

1.1.4 Methane and Carbon Dioxide

Landfill gas is composed primarily of 50 percent methane and 50 percent carbon dioxide with small amounts of trace gases. Methane and carbon dioxide are generated through the biological decomposition of waste. Methane is a naturally occurring flammable, colorless and odorless gas and is the principal explosive component of concern in landfill gas. Carbon dioxide is colorless, odorless and slightly acidic.

Methane is one of the contributing gases to what is called the greenhouse effect. Methane levels in the atmosphere have been steadily increasing for centuries, but even more so since the beginning of the industrial revolution. According to the IPCC (2001), methane is augmenting global warming because of its capability to retain heat at a rate of 21 times that of carbon dioxide within a 100 year time span. Hummer and Lechner (1999) reported that methane concentrations within the atmosphere have increased at a rate of about 1% per year since 1978. They also reported that the amount of methane in the atmosphere has increased from 0.8 ppm to 1.7 ppm since the year 1800. Because of its ability to retain infrared radiation, methane, on a molar basis, is 20 to 30 times more potent than carbon dioxide as a greenhouse gas (Rodhe, 1990, Blake and Rowland, 1988). Methane has become a target for emissions reduction due to its relatively short decay time in the atmosphere, 9 to 10 years, and its higher effectiveness as a greenhouse gas (Rodhe, 1990; WMO, 1998; IPCC, 2001). Stern and Kaufmann (1996) stated that around 12% of worldwide methane emissions are caused by the decomposition of waste within landfills. A large number of these landfills are designed with passive venting systems which allow for the free movement of methane from within the landfill envelope out to the atmosphere. In recent years, research has been conducted to reduce methane atmospheric emissions from landfills by converting it into carbon dioxide through biofiltration.

1.1.5 Hydrogen Sulfide

The rotten egg smell that is sometimes emitted from landfills is caused by naturally occurring gases called sulfides. Even at low concentrations, sulfides can cause unpleasant odors. Hydrogen sulfide, a colorless, flammable gas, is one of the most common sulfides responsible for landfill odors. The concentration at which hydrogen sulfide is odorously detectable is different for every person. Also, "*at concentrations around 100 ppm,"* (parts per million) *"no odor is detected due to a loss of olfactory sensation, resulting in loss of warning properties at lethal levels.*" (Integrated Risk Information System (IRIS)). Therefore, smell alone is not a reliable sign to provide warning of elevated concentrations. Hydrogen sulfide is likely to pool in areas of low elevation because it is denser than air. Hydrogen sulfide is a natural gas, as mentioned before, but it can also be produced from manmade processes, like landfilling.

Although municipal solid waste landfills produce hydrogen sulfide, landfills that receive construction and demolition (C&D) waste usually produce much greater quantities than at landfills that do not accept C&D. The gypsum board component (e.g. wallboard) is believed to be the cause of the higher concentrations of hydrogen sulfide. The combination of gypsum, organic material, moisture and anaerobic conditions present in C&D landfills is believed to provide a favorable mixture and environment for bacteria to produce hydrogen sulfide gas. Concentrations of hydrogen sulfide levels detected in raw landfill gas samples have ranged from 50,000 ppb to 15,000,000 ppb (50 ppm - 15,000 ppm) for those landfills that accepted C&D solid waste. These samples were collected from within a landfill waste mass. Landfills which do not accept C&D generally have much lower concentrations of hydrogen sulfide in the raw gas, usually less than 100,000 ppb (100 ppm).

The gypsum contained in the construction and demolition landfills leaks out of the waste and contaminates the landfill. Sulfate is leached from this gypsum and a series of biological reactions reduce the sulfate to hydrogen sulfide. In general, wallboard consists of a facing and backing made of paper with a gypsum core ($CaSO_4 \times 2H_2O$) in between. The microorganisms responsible for generating hydrogen sulfide include sulfate-reducing bacteria (bacteria that react with and convert sulfate) and sulfur-reducing bacteria (bacteria that react with and convert sulfur) (Florida, 2002). Sulfate reducing bacteria require a sulfate source (gypsum), a carbon source (organic material), anaerobic

conditions (the presence of oxygen), and moisture to produce hydrogen sulfide. Studies in the lab have suggested that under anaerobic conditions (conditions where oxygen is present) "*...decaying drywall, even alone, leached enough sulfide ions and organic matter for sulfate reducing bacteria to generate large hydrogen sulfide concentrations*" (Florida, 2002). "*Factors contributing to hydrogen sulfide production are anaerobic conditions, pH, moisture, organic matter content, in the presence of sulfate*" (Florida, 2002). Additional information on factors affecting hydrogen sulfide production can be accessed through the State University System of Florida.

Hydrogen sulfide production from municipal solid waste ("MSW") and C&D waste has been measured under laboratory conditions. Lab experiments have shown that C&D waste landfills produce more sulfide than non-C&D landfills but C&D debris landfills do not produce the large volumes of methane gas that MSW landfills generate**.** The lower methane production can actually be a negative feature of C&D landfills because one of the most effective technologies for treating landfill gas (including hydrogen sulfide) is combustion using methane as the fuel source. The lower methane production in C&D landfills is probably due to the failure of the landfill to reach an anaerobic state (no oxygen present.) Anaerobic conditions are necessary for methane production. Furthermore, hydrogen sulfide is toxic to methane producing bacteria; therefore, methane gas production is reduced even under anaerobic conditions.

1.1.6 Landfill Gas Migration

There are several types of physical processes that cause gas migration in a landfill environment. These processes include advection and Fickian transport. Advection is the bulk movement of fluid with a specific direction and length. Fickian transport includes diffusion and dispersion. Diffusion is the process where there is movement of a substance from higher concentration to lower concentration. Molecular diffusion in a landfill can occur even without fluid flow. Dispersion is the process of a substance being scattered over a range, area or volume. Gas molecules can be dispersed through porous medias in the landfill such as soil layers or garbage. In landfills, advective flow is the major transport process for gas migration. This advective flow is caused by the difference in pressure within and outside of the landfill. The decomposition of the refuse produces gas,

causing pressure to build up in the landfill. As a result, the gas is going to migrate from an area of high pressure within the landfill to an area with lower pressure outside of the landfill.

There are several factors that influence the direction of gas migration within a landfill but none are as important as landfill design. The design of the landfill will determine if the gas in the landfill migrates vertically or horizontally. Horizontal migrations of gases out of the landfill boundary are unwanted. Gases that migrate horizontally out of a landfill and into the surrounding community can cause health and public relations problems. To control this potential problem, and to avoid pressure buildup, it is important to provide a venting system for the gases to escape.

1.2. SUMMARY OF PAST BMPs

Previous efforts, that looked at technologies and management practices for reducing greenhouse gas emissions from landfills, **have focused on optimizing the collection and use of landfill gas**. These BMPs addressed operational, management, and design associated with the gas collection system. SCS Engineers, along with the state of California has issued a set of BMPs to address these issues. The following are few examples:

- **Use Horizontal collectors or Surface Collectors**

  Horizontal and surface collectors are used to collect LFG before vertical wells can be installed. These collectors may not be feasible in wet conditions. Horizontal collectors can be installed during the filling process, however, to avoid damage, this must be coordinated with fill planning. A vacuum should not be applied until the collectors is effective for gas collection.

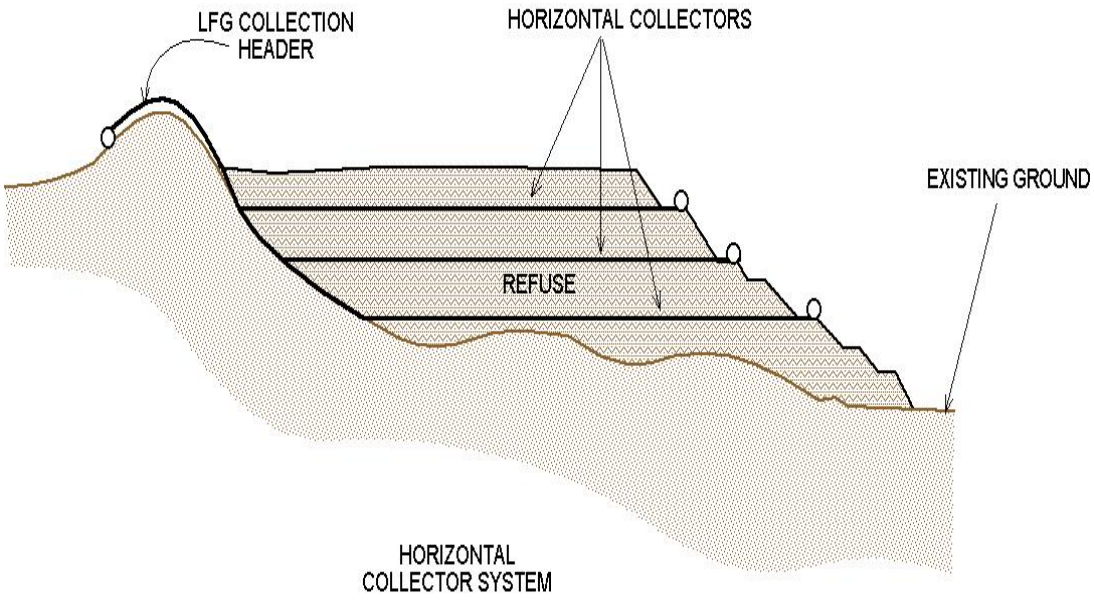


Fig. 1.1. Sketch of Horizontal Collectors (After……)

- **Tighter spacing of LFG wells**

If monitoring data shows that there are potential surface leaks from surface collectors, vertical wells may be closely spaced to increase overlap of the radius of influence (ROI). The surface monitoring data should be the basis for the exact spacing requirements.

- **Mixed horizontal/vertical well systems**

  Vertical wells should be placed in areas where they are not at risk of damage, while horizontal wells are typically placed in active areas. This must be coordinated with the landfill operations and it may be costly, but this BMP is feasible for most landfills. This method is especially useful for deep landfills that take years to fill each section.

- **Connection of leachate collection and recirculation (LCRS) layer to gas collection and capture systems (GCCS)**

  The LCRS and the GCCS can be connected to collect LFG along the bottom of the landfill. This is feasible for landfills where the LCRS contains LFG. The LCRS should be monitored to determine when there is enough gas to apply vacuum. To prevent blockage of gas flow, the upslope side of the LCRS should be connected to the GCCS.

- **Deep multi-depth vertical wells**

  Wells operating near a landfill slope and in deep unlined or clay lined landfills should be designed to extract gas from different depths. Wells are to placed in the same borehole but at multiple depths. Wells can also alternate between deep and shallow with closer spacing.

- **Maximize borehole well diameters**

  This BMP can be used for all vertical well systems. Pipe diameters of 4” to 6” or greater are used to collect LFG. When considering this BMP, it is best to select conservative values and select the largest diameter.

- **Enhance seals on LFG wells/boreholes**

  A maximum of 3 types of seals can be placed on wells to allow more vacuum to be applied to LFG wells. A least two seals should be used and alternative seals should be used in arid regions where bentonite seals crack.

- **Dewater gas wells**

This BMP is used to prevent the blocking of the flow of gas to the LFG wells by water. Pumping can be used to remove the water from the wells, but pumping can be complicated. Installation is less difficult when large diameter pipes are used.

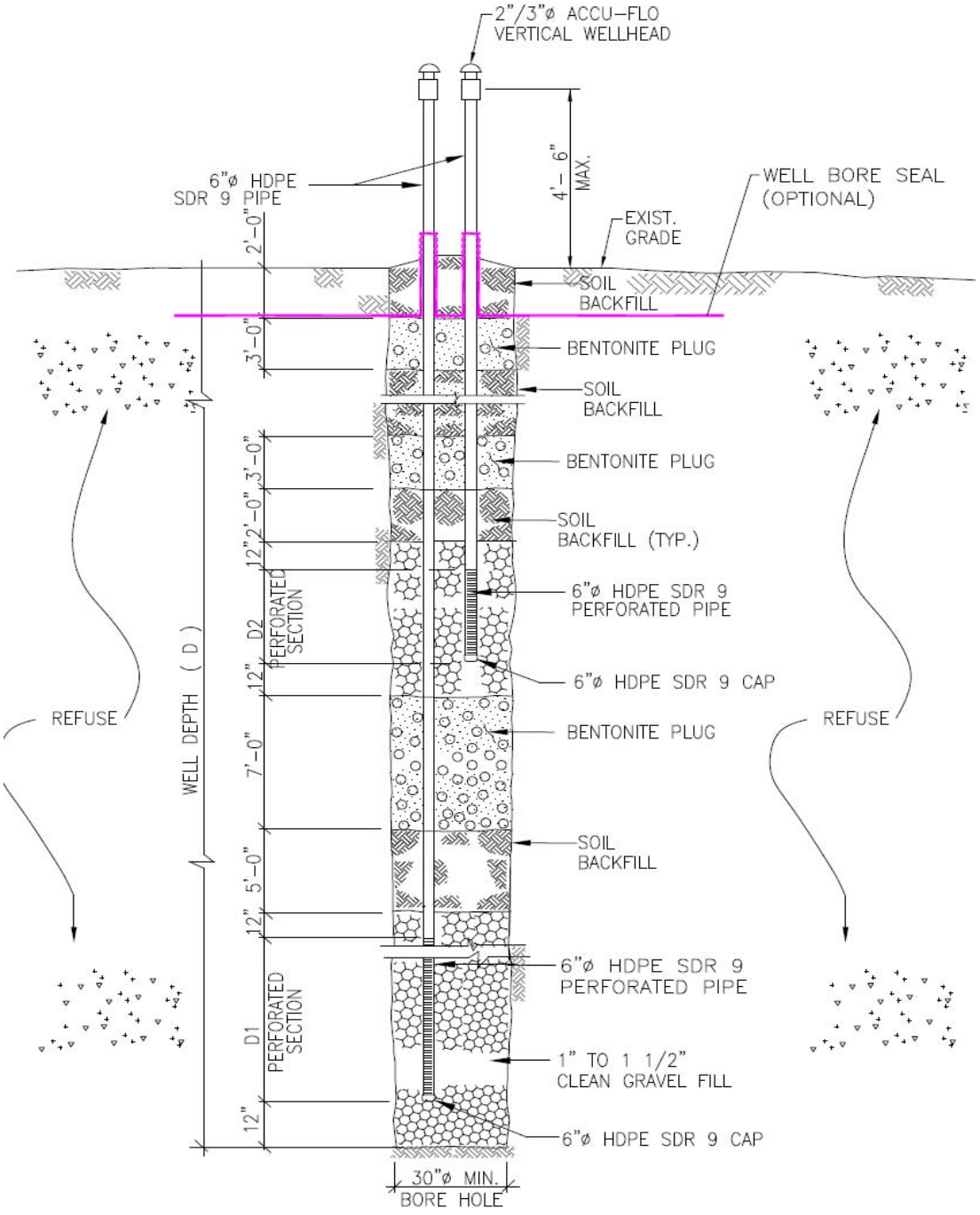


Fig. 1.2. Sketch of Multi-depth Gas Collection Well (After…….)

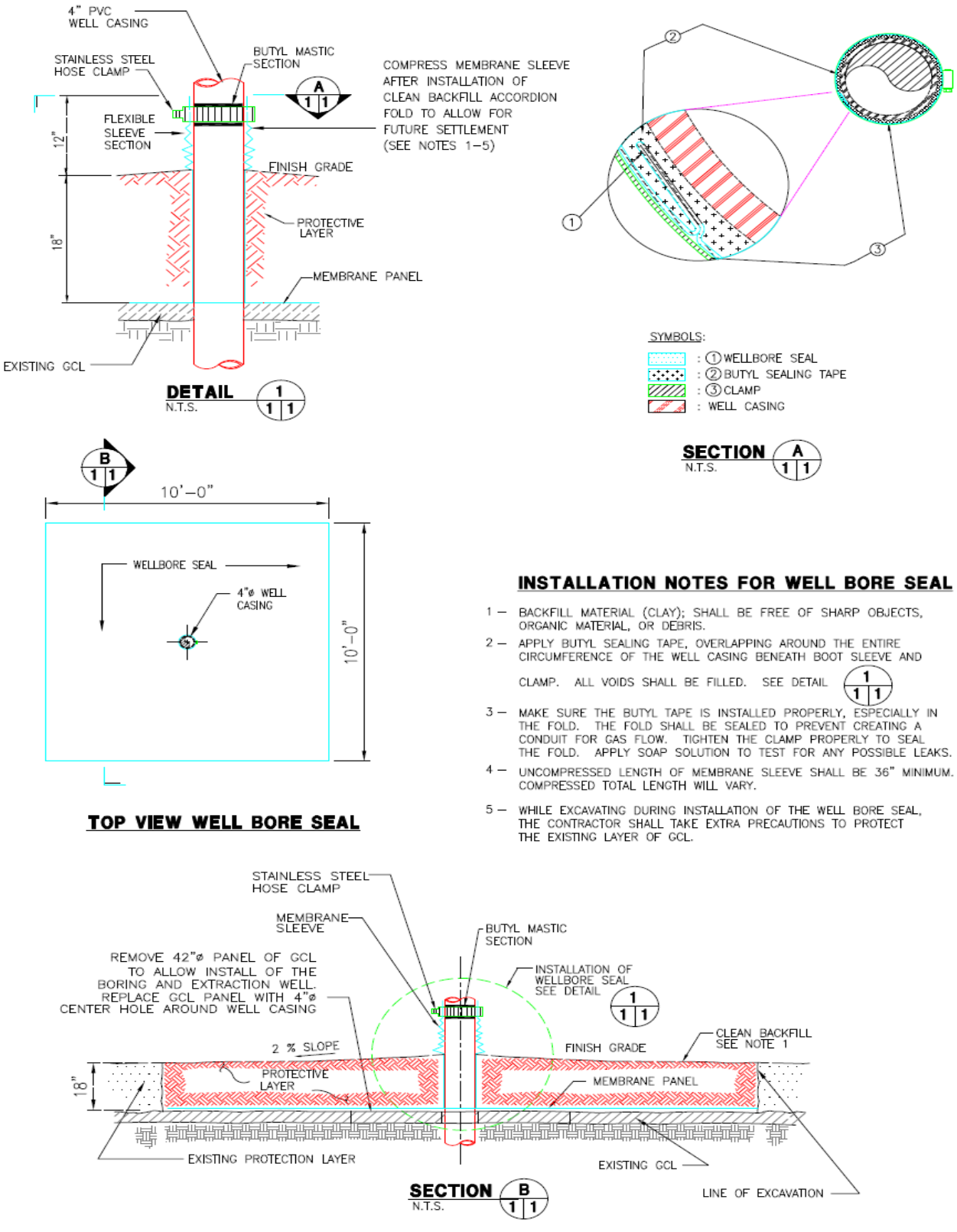


Fig. 1.3. Typical Gas Well Seal (After…….).

Collection systems are never 100% efficient. Emissions can escape the landfills from around wells and along routes of installed landfill equipments. Some report that gas utilization and/or flaring systems associated with active gas collection begins to falter and fail periodically leading to high emissions. *Historical practice suggests that collection systems may operate less than half the time that landfill gas is produced because these systems are economically and operationally feasible only when methane concentrations are high*. As low level methane production may continue for a long time, the net accumulation of methane during the non-collection period is not negligible. One methane concentrations fall below 35 to 40% and the total production rates are below 30 to 50 m3 per hour, the treatment of landfill gas in combined heat and power plants becomes technically and economically not feasible (Haubrichs and Widmann 2006). When methane concentrations reach 20 to 25%, and landfill gas flow rates dip below 10 to 15 m3 per hour, the most suitable treatment methods become high temperature flaring. Below these values, the treatment of poor landfill gas becomes more expensive and complex (Huber-Humer et al 2008). Fluidized combustion or catalytic oxidation are two possible options for such quality of landfill gas**.** One of the most promising options under these conditions is the bio-oxidation of LFG, as will be described in the next section.

1.3 BIO-OXIDATION OF "NOT COLLECTED" LANDFILL GAS

One of the most promising and cost-effective options being employed for control of low-level methane emissions is the use of biological oxidation-based systems.

1.3.1 Methane Oxidation

Methane migrates through zones of low pressure in cover soils, and eventually reaches the surface. The cover soil will oxidize about ten percent of the generated methane unless it is captured by a recovery system and used as an energy source (Liptay et al. 1998). Methane oxidation occurs in many natural systems and soils without human interference and serves as an important sink for natural methane emissions. The process of methane oxidation consists of the conversion of methane into carbon dioxide, water, and biomass by means of microbial activity. Biological oxidation is greater in magnitude than that of chemical oxidation when the total methane budget is considered and not only the initial transport to the atmosphere. Microbial methane oxidation reduces methane released in certain methanogenic areas. The following are determining factors for methane oxidation:

- Existence of methanotrophic microorganisms
- Supply of oxygen
- Suitable carrier that offers adequate nutrient supply and facilitates colonial formation
- Adequate moisture content and ambient conditions in the medium.

Environmental factors have a decisive impact on the activity of methanotrophic bacteria. Among these factors are pH, temperature, water content, oxygen supply, inhibitors, soil condition and nutrient supply, barometric pressure, and retention time.

1.3.2 Factors Affecting Methane Oxidation

Water Content Profile

Water content is a very important factor affecting CH_4 oxidation in landfill cover soils. Water content is defined as the mass of water lost from the soil by oven-drying at

105°*C* for 24 hours divided by mass of dry soil. In geotechnical engineering, the water content is measured in accordance with ASTM D2216 (ASTM, 1998). Boeckx et al. (1996) used a multiple linear regression analysis under different incubation conditions and concluded that water content has more influence on CH_4 oxidation than temperature. Christophersen et al. (2000) used statistical methods to analyze the effect of soil water content on CH_4 oxidation. They also concluded that water content can explain most of the variation observed in CH_4 emission data.

Water plays three important roles. First, the optimum environment for CH_4 oxidizing bacteria (methanotrophic) is obtained at certain water content. Second, water content affects the penetration of oxygen (O_2) into the soils, which is the main reactor for CH_4 oxidation. As the water content increases in the soil, the O_2 diffusion into the soil is hindered. Thirdly, water content affects the air filled porosity of the soil and influences gas transport through the soil. As water fill up the pores in the soil, it blocks the flow of gas upward. At the same time, the blocking of flow might lead to CH_4 emission due to the excess pressure built-up in the landfill (Boeckx et al., 1996).

When these factors come to such a balance, the soil will have its optimum water content for maximum CH_4 oxidation. Below this water content, the oxidation rate will increase as the water content increases. Above this water content, the oxidation rate will decrease as the water content increase (Fig. 2). At this optimum water content, there is both rapid gas phase molecular diffusion and a sufficient microbial activity to oxidize the delivered CH_4. The reduced CH_4-oxidizing capacity at higher water contents is caused by a shift of gas-phase molecular diffusion to aqueous-phase molecular diffusion, which is $\sim 10^4$ fold less rapid (Boeckx et al., 1996). The low CH_4 oxidation at the low range of soil water content may be caused by the less methanotrophy activity. Oxidation peaks when balance is achieved (Czepiel et al., 1996). The optimum oxidation soil water content will be different for different soil types and depends on temperature and other environmental factors. The optimal oxidation water content ranged between 15.6 and 18.8% for soils tested by Boeckx et al. (1996) and Christophersen et al. (2000). Czepiel et al. (1996) measured an optimal oxidation water content of 15.7%. Whalen et al. (1990) reported a value of 11%. Visvanathan et al. (1999) also reported values ranging from 15 to 20%.

Christophersen et al. (2000) point out that soil environment needs some time

before the growth of methanotrophic bacteria is established. The longest lag phase occurs in the lowest water content range. Methanotrophic microorganisms tend to become inactive under ambient conditions when the water content falls below 13% of the maximum water capacity (Bender and Conrad, 1992). Visvanathan et al. (1999) reported that at water content lower than 6%, oxidation became zero. When the water content increases to saturation, the oxidation rate decreases by ~56% (Nesbit, 1992), because water fills all voids of soil and inhibit O_2 diffusion into the soil.

Organic Matter Content

Generally, the oxidation rate increase with increasing organic matter content in soils. Organic matter content is defined by the loss mass on ignition at $550°C$ for 1 hour of oven dried($105°C$ for 24 hours) soil divided by the mass of oven dried soil. The organic matter content is typically determined using procedures described in ASTM D2974 (ASTM, 2000). Using a higher organic matter content soil as a cover would be an efficient way to mitigate CH_4 emission (Borjession and Svensson, 1997; Christophersen et al., 2000). Soil from old landfill, which have been exposed to CH_4 emission for a long time, have higher oxidation rate than fresh soils (Nozhevnikova et al., 1993; Visvanathan et al., 1999). Christophersen et al. (2000) reported that CH_4 oxidation rate increased with increasing organic matter content using incubation test. They also pointed out that there was a relationship between optimal water content and organic matter content. The optimal oxidation water content increases with increasing organic matter content. Visvanathan et al. (1999) reported that higher organic matter content was associated with higher CH_4 oxidation rate in their column test. High organic material has proved to be very efficient CH_4 oxidizer, such as compost (Humer and Lechner, 2001). They reported that compost covers enriched with organic matter were able to entirely oxidize all CH_4 emitted from their landfill. Organic matter provides nutrient for methanotrophic bacteria and have high porosity allowing more O_2 penetration.

Porosity

The porosity of soil directly influences the penetration of O_2 into the soil. O_2 is the main reactor of the oxidation process. Porosity can provide the channel for O_2 penetration and the contact surface area with methanotrophic bacteria. Borjesson et al. (2004) reported a significant relationship between CH_4 oxidation and particle size

distribution. Soils with high porosity retain CH_4 and O_2 longer in the pores leading to higher oxidation rate (Humer and Lechner, 1999).

Climatic and Ambient Conditions

Soil temperature

Generally, CH_4 oxidation rate increases with increasing temperature (De Visscher et al., 2001). Low temperatures inhibit CH_4 oxidation (Whalen et al. 1990; Nozhevnikova et al., 1993; Borjesson and Svensson, 1997; Visvanathan et al., 1999). Borjesson and Svensson (1997) even reported that soil temperature is the controlling factor of CH_4 oxidation, and can explain 85% of the variation in measured CH_4 oxidation. Methanotrophic bacteria favor a certain range of temperatures. Czepiel et al. (1996) reported that oxidation rate increased as temperature increased to 36°*C*. They also reported that CH_4 oxidation essentially stopped when temperature reached 45°*C*.

Humer and Lechner (2001) reported that CH_4 oxidation rate was 70-80% at 18°*C*. At a lower temperature of 4°*C*, little oxidation was observed. Borjesson and Svensson (1997) reported that the optimum temperatures for CH_4 oxidation were ~25 to 35°*C*. Boeckx et al. (1996) reported that the optimal incubation temperature for CH_4 oxidation is ~20 to 30°*C* and decreases with increasing water content. Dunfield et al. (1993) measured an optimum temperature of ~20 to 25°*C*.

In tropical landfills, temperature is the dominant factor of CH_4 oxidation and is ~30 to 36°*C* (Visvanathan et al., 1999). Whalen et al. (1990) reported an optimum temperature of 31°*C*. Boeckx et al. (1996) reported optimal temperatures between 25 and 30°*C*. Nesbit (1992) reported the optimal temperatures between 20 and 30°*C*.

To quantify the influence of temperature, we usually use Q_{10} value, which is the value for how many times the oxidation rate increases when temperature is increased by 10°*C*. With this value, we can easily evaluate the effect of temperature on CH_4 oxidation. Higher Q_{10} indicates that temperature dominates CH_4 oxidation more than other factors. Crill et al. (1994) concluded that low Q_{10} values (<2) indicate that temperature has less influence on oxidation than water content. Boeckx et al. (1996) measured an average Q_{10} value of 1.88 ± 0.14 between 10 and 20°*C*. Dunfield et al. (1993) reported values between 1.4 and 2.1 in peat soils. Christophersen et al. (2000) measured Q_{10} values between 4.13 and 5.60. A Q_{10} of ~2.8 was reported by De Visscher et al. (2001), 2.4 by

Czepiel et al. (1996), and 1.9 by Whalen et al. (1990). Moreover, Christophersen et al. (2000) have reported that Q_{10} values for CH_4 oxidation in landfill cover soil can be as high as 4.10 to 7.26. Borjesson and Svensson (1997) explained their high Q_{10} values (3.4 to 7.3) by the negative relationship between temperature and water content.

Barometric pressure

The pressure difference is believed to be one factor that controls the advection flow through soils. A decline in atmospheric air pressure can "pump" gas out of the landfill body (Borjesson and Svensson, 1997). Some gas-flow models use air pressure as the main factor of gas transmission in soils (Lu and Kunz, 1981; Young, 1990). Czepiel et al. (2003) reported a very strong negative relationship between measured CH_4 emissions and atmospheric air pressure. However, Borjesson and Svensson (1997) reported that there is no relationship between CH_4 emission and air pressure or change in air pressure over the seasons. This contradiction indicates that when the flow is governed by advection, pressure difference plays a role. However, when the flow is mainly a diffusive flux, it does not depend on pressure difference.

Vegetation

Vegetation influences the properties of soil, such as its pH , water content, and gas transport. De Visscher et al. (1999) points out that plant in landfill soil cover may inhibit CH_4 oxidation by N uptake. However plants also provide channels for O_2 penetration into the soil and therefore enhance CH_4 oxidation. The root system of vegetation can also induce a more suitable microbiological environmental for CH_4 oxidation. In general vegetation can be used to enhance CH_4 oxidation (Maurice et al., 1999).

1.3.3 Inhibiting Factors

Chemical Inhibiters

Studies on sieved cover soil from a municipal solid waste landfill showed that several compounds completely inhibited CH_4 oxidation. These compounds include but are not limited to C_2H_2 (0.01 mL L^{-1}) or C_2H_4 (1 mL L^{-1}), as reported by Chan and Parkin, (2000). Studies on forest sieved soils and soil cores also showed that HNO_3 strongly inhibited net CH_4 oxidation in both of these soils. Bradford et al. (2001) reported that

even when the concentration of $(NH_4)_2SO_4$ is high, its negative effect on the CH_4 oxidation is not as great as HNO_3.

Research on three adjacent areas in eastern Scotland, with contrasting land use, showed that CH_4 oxidation rate negatively correlated with soil water content in woodland soil, positively correlated with soil temperature in set aside soils (not farmed soils), and positively correlated with soil temperature in dry summer conditions in arable soil (Dobbie and Smith, 1996). Research on peat bog hummocks and hummocks soil in New Galloway (Scotland) showed that penetration of O_2 into the peat increased under illumination when photosynthesis was active, but decreased in the dark (Nedwell and Watson, 1995). Four different soils (meadow cambisol, forest luvisol, cultivated cambisol, paddy soil) were studied and incubated under different CH_4 mixing ratios and showed that the CH_4 incubation activity was strictly O_2 dependent and was inhibited by acetylene or autoclaving, demonstrating that CH_4 oxidation in the soil was due to the methanotrophic bacteria. The induction process was influenced by soil moisture, pH, temperature, NH_4 concentration, Cu concentration and aggregate size (Bender and Conrad, 1995).

A negative correlation was found between NH_4 concentration and CH_4 oxidation rate in woodland soil (Debbie and Smith, 1996). The addition of supplemental NH_4 or NO_3 ions in excess of 30 $\mu g\ g^{-1}$ dry soil yielded lower CH_4 oxidation rates (Chiemchaisri et al., 2001). In contrast, according to Kammann et al. (2001), there was no direct correlation between the amount of the N fertilizer applied and average CH_4 oxidation rates.

Physical Inhibiters

In landfill cover soils, there is an optimum zone for CH_4 oxidation, where optimum condition for methanotrophs growth, O_2:CH_4 ratio, retaining time and suitable environmental condition exist. In landfill cover soil, the gas profile is very complicated. The distribution of gas concentration across the soil profile depends on diffusion, reaction, and gas flow (De Visscher et al., 1999). High CH_4 concentration will increase CH_4 oxidation (De Visscher et al., 2001); however the high flow rate of CH_4 from the underlying waste hinders the diffusion of O_2 into soil, which will inhibit CH_4 oxidation. Czepiel et al. (1996) measured oxidation rates at different soil depth using incubation

tests. They reported that maximum oxidation occurred in the top 5 cm to 10 cm of the soil profile. Visvanathan et al. (1999) reported that maximum oxidation occurs at a depth between 15 and 40 cm. Several researchers reported different maximum CH_4 oxidation zone of different depths, between 40 and 60 cm by Nozhevnikova et al. (1993) and Borjesson and Svensson (1997), 15 and 60 cm by Barratt (1995), 3 to 12 cm by Whalen et al. (1990), and 20 to 30 cm by Kightley et al. (1995). At the top of the soil profile, soils are drier and inhibit lower CH_4 oxidation and below a certain depth the soil will become in anaerobic condition. Humer and Lechner (2001) found that in their field scale test the maximum zone to CH_4 oxidation is between 40 cm and 90 cm in sewage sludge compost and municipal solid waste compost. Table 3 shows a summary of reported maximum oxidation zones in several soil profiles.

Czepiel et al. (1996) and Bender and Conrad (1995) reported that 30 mL L^{-1} of O_2 concentration is a threshold to CH_4 oxidation to occur, which means above 30 mL L^{-1} the concentration has very little influence on oxidation but will decrease dramatically when the concentration is <30 mL L^{-1}. That can be the reason of why there is a sharp slope in CH_4 concentration with depth profiles.

Chanton et al. (2009) summarizes 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^{-2} d^{-1} (52 to 102 g $m^{-2}d^{-1}$) across the different soils. The overall mean % oxidation across all studies was 35% with a standard error of 4% (se = standard deviation/($n^{1/2}$)). The overall mean oxidation rate across all studies was 4.5 mol m^{-2} d^{-1} with a standard error of 1.0 (72 ± 16 g $m^{-2}d^{-1}$).

1.3.4 Engineering Methane Oxidation (Biocovers, Biocells, Biofilters, Biowindows,..)

Biocovers and Biocells:

The preceding review indicates that CH_4 oxidation can be achieved in landfills. In Florida and other states, several old landfills have been closed or are in need for closure. Because of their small size, they have no gas management plans. However, these

landfills can be significant source of greenhouse gases. Gas extraction is not required for such landfills and tends to be expensive: out of the reach of most small communities managing their solid waste facilities. An attractive alternative is to incorporate biocovers or biocells on existing landfill covers. Biocovers can also be designed as part of a daily or an interim cover to reduce CH_4 emissions. Biocells can be used in areas with significant release of gas into the atmosphere (typically referred to as hot spots). Biocells are an excellent option for landfills with limited access to inexpensive compost.

Biofilters, Biowindows,….

Many landfills also passively vent CH_4 to prevent its horizontal migration. These passive vent pipes serve as direct conduits for CH_4 to pass from deep within the landfill to the atmosphere. Passive biofilters that uses CH_4-oxidizing bacteria to reduce passive vent CH_4 release can be incorporated into LFG management plans. Such biofilters can be contained in cylinders open at the top to allow air to diffuse into it from above. The cylinder can contain a porous substrate for the bacteria to grow upon. As LFG passes though this scrubber, entering at the base, CH_4 and non-CH_4 organic compounds would be oxidized.

A large scale biofilter system could also be used in later stages on larger landfills when active recovery is no longer economically feasible. As landfills age, they produce less CH_4 and at some point it will no longer be economical to power the pumps that extract the CH_4 and direct it to engines and/or flares. The engineering challenge for the design of biofilters is to keep the gas permeability of the filter media high enough so the presence of the biofilter does not hinder flow. This can be performed by using coarse grained compost and by keeping the compost at target water content.

Biofilters can also be designed as biowindows. Biowindows are used when a full a biocover is too expensive or unneeded, and when there is no gas collection system to direct the gas to a biofilter. The window is typically composed of compost and it is integrated into the landfill cover. The gas flows beneath the window and then progresses up through the window. The gas does not need to be directed because it will naturally migrate to the window because it is the path of least resistance due to its low permeability.

CHAPTER II
BIOCOVERS, BIOCELLS

2.1 HISTORICAL PRESPECTIVE

One of the first attempts to quantify methane oxidation in landfill cover soils was made by Whalen *et al.* (1990), who documented about 45 g CH4 m–2 day–1 (= 63 L CH4m–2 day–1 at normal conditions: 1013 mbar; 0 °C) uptake rates in laboratory tests of soil cores taken from a closed municipal solid waste (MSW) landfill. Since then, various landfill sites and types of cover soils have been tested for their methane oxidation capacity in laboratory and field trials, the latter mostly using stable isotope techniques (Liptay *et al*. 1998, Chanton *et al.* 1999, Chanton & Liptay 2000, Borjesson *et al.* 2001). The findings generally converge to suggest that high oxidation capacity is associated with coarse, porous and well-structured substrates that are often rich in organic matter.

Some of the first field trials to investigate compost covers were carried out on two different Austrian MSW landfills between spring 1999 and winter 2002. The purpose was to design a cover to enhance biological methane removal as well as to minimize leachate generation under mid-European seasonal conditions. After testing various designs over several years, a simple but efficient two-part cover system proved most effective. It consisted of a layer of up to 1.2 m of mature, well structured compost underlain by a 0.3–0.5 m coarse gravel layer to provide high gas permeability. While the function of the sub-layer was to homogenize gas fluxes, the porous upper layer served to support good methane oxidation activity by providing, a balanced oxygen and methane supply, proper moisture content and viable temperature regimes. Although a strong decline in methane oxidation was evident in conventional or shallow cover soils during the winter (Liptay *et al.* 1998, Chanton & Liptay 2000, Börjesson *et al*. 2001), there was no decrease in methane emission mitigation at the Austrian study site, which was probably due to the good insulation properties afforded by the biocover design.

In optimally designed compost covers the year-round methane removal rate (relative to an adjacent open landfill reference cell) was 95–99%, depending on the kind of compost applied (Humer & Lechner 2001a, Huber-Humer 2004b). The high removal rate was mostly linked to the installation of a coarse gas distribution layer for balanced methane fluxes and the good insulation effect due to sufficient cover dimension and the

use of proper substrates. In contrast, variants in the same trials that had shallower compost layers (about 30– 40 cm) and no gas-balancing layer removed only 68–74% of the emitted methane. Within the constraints of meeting certain permeability and stabilization requirements, biocover modifications can be made to adapt designs to meet local site-specific conditions and performance objectives. Thus, the dimensions and thickness of a biocover may vary depending on the nature of available materials, likely settlement behavior (particularly when the covers are placed without any artificial compaction), climate conditions (precipitation, temperature, frost penetration depth), expected gas fluxes, the purpose of the cover (final or temporary), and the intended after-use of the site (vegetation, land use).

Currently, Austria has at least five closed MSW landfills or sections of landfills covered with systems designed according to Humer & Lechner (2001a) (i.e. 0.5 m gravel gas distribution layer overlain by up to 1.2 m of mature, well-structured compost or waste substrates). These biocovers are serving either as the sole means to mitigate methane emissions on smaller, older sites or in combination with an operating gas extraction system as an additional measure to capture emissions that escape gas collection. Presently, these sites are fitted with a quasi state-of-the-art biocover design, the construction of which has been officially approved in Austria as an acceptable interim MSW landfill cover for a period of about 20 years. During this period, the biocover performance must be thoroughly monitored and documented. To date, the longest practical operating biocover is in Austria and has been monitored for 6 years. The data show that flat, undisturbed biocover areas have been consuming nearly 100% of the potentially emitted methane over the entire investigation period. Only in border areas and zones around physical installations such as drainage or gas wells are sporadically high methane emissions detected, particularly when the gas extraction system is turned off. Thus, site-specific trouble spots such as this will require particular attention during the life of a biocover. The quality of a biocover can be checked with a flame-ionization detector (FID) mapping unit to detect surface methaneconcentrations that may indicate leaks., but for determining the overall effectiveness of such a system, the methane influx (reference flux or emission) into a biocover must be known, which is typically a more complex measurement. These reference values can be defined by flux measurements on

the site prior to biocover application, on adjacent control cells. In some instances, landfill gas production data for the specific site can be surveyed, calculated or modeled. Due to temporal and/or spatial variability, however, noticeable discrepancy can occur between reference influxes and day-to-day values.

2.2 FLORIDA PRESPECTIVES

Mulch Biocovers Overlaying a Daily Cover

A mulch biocover was placed on 2+ year old waste that had a daily cover of about 15 cm of compacted sandy clay. Crushed fluorescent tube glass was placed 10 to 15 cm deep on the two 18 m by 32 m areas to be used for the mulch treatment. Three treatment areas were used to test the effect of mulch on methane emissions reduction from an area covered with only daily cover. The addition of mulch to the daily covers constituted an interim cover that can stay for a longer period of time. One biocover area had deep mulch (60 cm), another had shallow mulch (30 cm) and the last area used no mulch. Each treatment area measured 18 m by 32 m (Fig. 2.2.1). A static chamber (Fig. 2.2.2) consisting of a 0.4 m2 aluminum collar and lid was used to measure the methane emissions rates. The chamber had two outlets. One of the outlets was plugged with a stopper and gas samples were taken from the other outlet using a syringe. Samples of methane were collected at time intervals of 0, 2, 5, 10, 15 and 30 minutes after the chamber was placed on the site. Methane oxidation was estimated using the stable isotope technique. Stable isotope method has been employed to determine the oxidation of CH_4 in landfill cover soils (Liptay et al., 1998; Chanton and Liptay, 2000, Abichou et al. 2006a and b; Stern et al., 2006). There are two stable isotopes of carbon, ^{13}C and ^{12}C, which comprise 1% and 99% of carbon atoms, respectively. Microbiology studies have shown that the methanotrophic bacterial prefer to consume $^{12}CH_4$ rather than $^{13}CH_4$, leaving residual CH4 enriched in $^{13}CH_4$. Methane oxidation rates can be calculated with this ratio of $^{13}CH_4/^{12}CH_4$ changing (Chanton and Liptay, 2000; Abichou et al., 2006a and b). Stable carbon isotopes were determined by using a Hewlett Packard Gas Chromatograph coupled via a combustion interface to a Finnegan Mat Delta S Isotope Ratio Mass Spectrometer (GCC-IRMS) (Merrit et al., 1995). Replications of measurement were performed for most samples with the standard deviation of about 0.15‰. Stern et al. (2006) provided more detailed methods of methane emission and oxidation measurement.

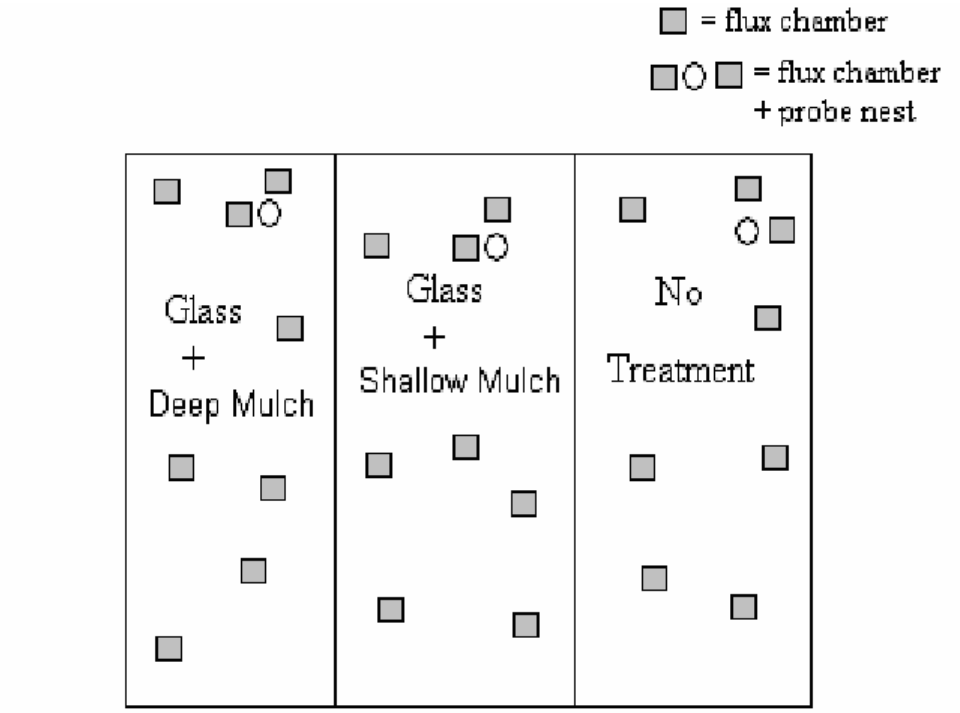


Fig. 2.2.1. Experimental Layout: Eight chambers and one probe nest per treatment.

Figure 2.2.2. Static flux chamber attached to collar with gas collection tube.

Fig. 2.2.3 shows the surface emissions from the daily cover before the installation of the biocovers. Pre treatment results for the area of the deep mulch treatment consisted of an average flux of 72 with a standard error of ± 41g $CH_4m^{-2}d^{-1}$, the shallow treatment area had an average flux of 23 with a standard error of ± 6g $CH_4m^{-2}d^{-1}$, and the no mulch area had an average flux of 64 with a standard error of ± 29g $CH_4m^{-2}d^{-1}$.

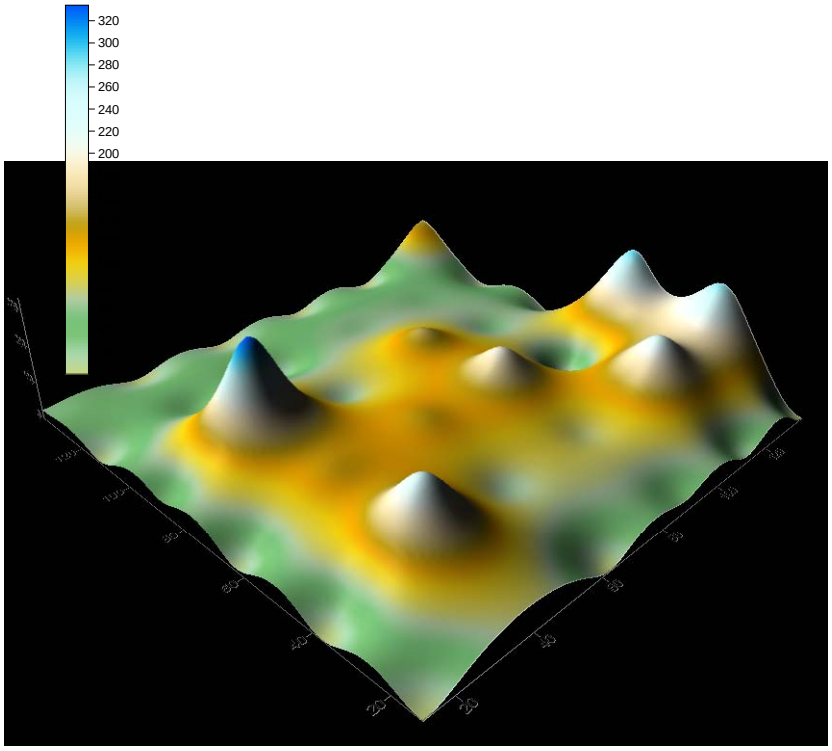


Fig. 2.2.3. Contour maps of surface emissions before installation of the biocovers.

Fig. 2.2.4 shows the emissions from the deep, shallow and no mulch areas after the biocovers were installed. Throughout the whole study, the deep mulch treatment average monthly values was 19 ± 4 g $CH_4m^{-2}d^{-1}$, and average monthly fluxes ranged from 7 to 51 g $CH_4m^{-2}d^{-1}$. The shallow mulch treatment had a monthly average flux of 121 ± 21 g $CH_4m^{-2}d^{-1}$, and monthly fluxes ranged from 49 to 208 g $CH_4m^{-2}d^{-1}$. The no mulch treatment (control) had an average monthly flux of 104± 12 g $CH_4m^{-2}d^{-1}$, and monthly fluxes ranged from 17 to 259 g $CH_4m^{-2}d^{-1}$.

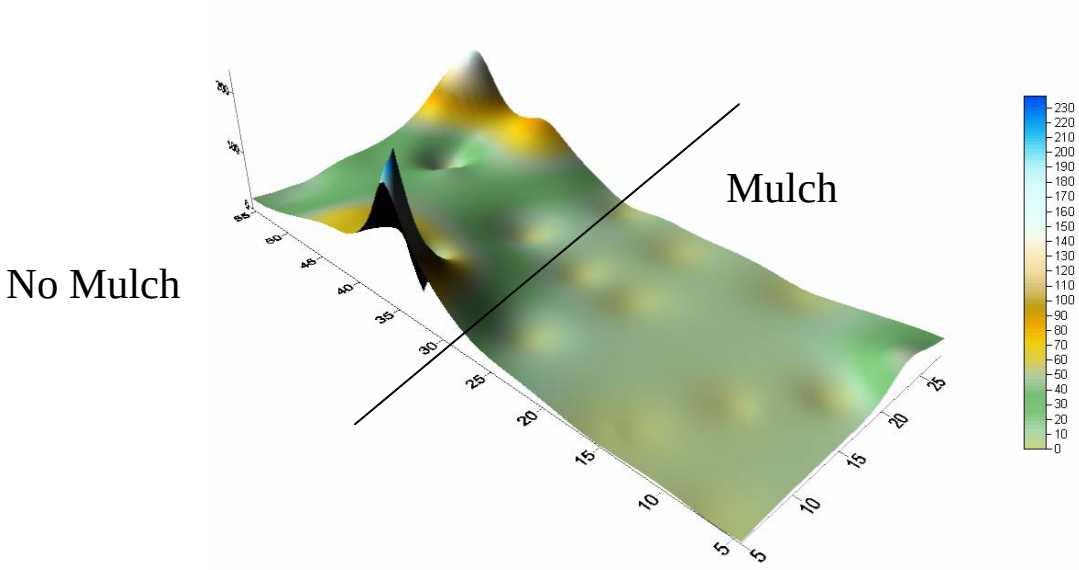


Fig. 2.2.4. Surface emissions after installation of biocovers.

The results show that the deep mulch treatment section had low fluxes and that the flux stayed relatively consistent over time. The shallow mulch treatment section showed a low flux at first, but the flux gradually increased. The results also show that there were a number of times that the deep mulch layer and the shallow mulch layer had methane gas fluxes of zero. A zero flux indicates that there was 100% methane oxidation below the surface of the landfill. This means that the mulch did not allow for any methane to flow out of the landfill into the atmosphere. The results also show that the mulch test sections actually drew methane gas into the landfill from the atmosphere. This is known as a negative flux. This shows that the mulch covers are effective at mitigating methane emissions.

2.3 FLORIDA PRESPECTIVES

Compost Biocells Overlaying an Interim Cover

This study was conducted at the Leon County Solid Waste landfill in Leon County, Florida. Following preliminary spatial assessment of methane emissions at the site (Abichou et al. 2006a), a portion was chosen based on the location of methane "hotspots" for the biocell experiment. Three biocells and three control cells, all 7.6 m x 7.6 m, were selected as shown. Fig. 2.3.1 shows a cross-section of each constructed biocell. In the biocells, a 100-mm-thick layer of glass was placed over the entire 7.6 x 7.6 m surface of the cell. The in place dry unit weight of the top of the compost was measured using the sand cone method (ASTM D1556-00). The compost extends 3.8 m beyond the edges of the glass edges. The compost was provided by the landfill, and consisted of chipped yard waste that had been windrowed and turned for three years. The pH of compost in distilled water was 7.5.

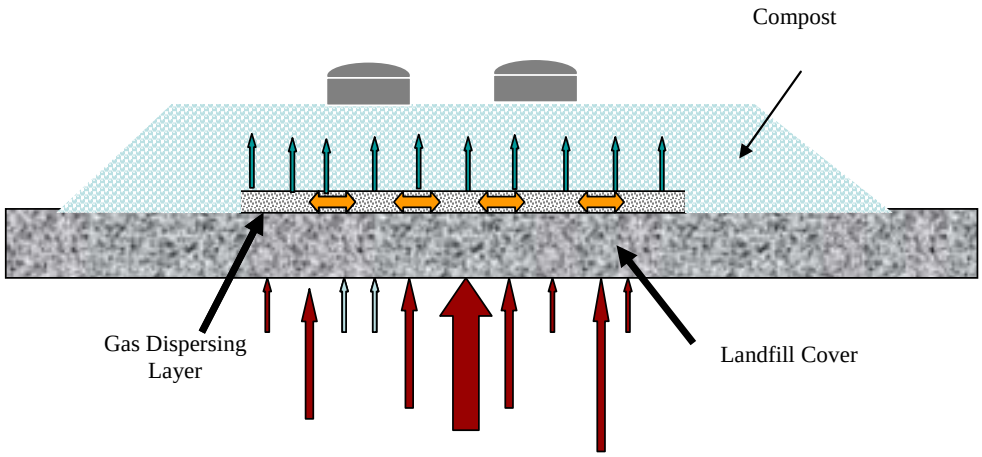


Fig. 2.3.1. Schematic of a Biocell cross-section.

Four flux collars were installed on each cell. A nest of gas sampling probes to various depths were installed in each hot spot location. Profiles of water content were monitored at two locations (control and biocell) using water content reflectrometers (Campbell Scientific CR615). An automated data acquisition system was used to collect

and store water content data (Campbell Scientific CR23X). Measurements from all sensors were collected at a 60-minute interval and periodically downloaded to a computer. Power for the system was supplied by lead-acid batteries and two solar panels.

Methane emission rates from the landfill surface were determined using a static chamber technique in this study (Abichou et al. 2006a, b; Stern et al. 2006). Landfill Static chamber method is the most common measuring method for gas fluxes from the surface of soil, has its advantage of low cost, simple operation and practical observations by discretizing space and time. The concept of static chamber is to monitor concentration changing in a certain volume above a gas-emitting or consuming surface. Gas samples for testing CH_4 concentration were collected sequentially within 15min using 60mL disposable plastic syringe with 3-way plastic stopcocks. Gas Samples were analyzed for CH_4 concentration with a Gas Chromatograph (GC) with a Flame Ionization Detector (FID) within 4hr of collection. Methane fluxes were determined from the slope of a linear regression curve of concentration data (C in ppmv) plotted versus elapsed time (t in minutes). This change in volumetric concentration was converted to a mass flux by using the ideal gas law, the CH_4 flux (g CH_4 m^{-2} d^{-1}).

Stable isotope method has been employed to determine the oxidation of CH_4 in landfill cover soils (Liptay et al., 1998; Chanton and Liptay, 2000, Abichou et al. 2006a and b; Stern et al., 2006). There are two stable isotopes of carbon, ^{13}C and ^{12}C, which comprise 1% and 99% of carbon atoms, respectively. Microbiology studies have shown that the methanotrophic bacterial prefer to consume $^{12}CH_4$ rather than $^{13}CH_4$, leaving residual CH4 enriched in $^{13}CH_4$. Methane oxidation rates can be calculated with this ratio of $^{13}CH_4/^{12}CH_4$ changing (Chanton and Liptay, 2000; Abichou et al., 2006a and b). Stable carbon isotopes were determined by using a Hewlett Packard Gas Chromatograph coupled via a combustion interface to a Finnegan Mat Delta S Isotope Ratio Mass Spectrometer (GCC-IRMS) (Merrit et al., 1995). Replications of measurement were performed for most samples with the standard deviation of about 0.15‰. Stern et al. (2006) provided more detailed methods of methane emission and oxidation measurement.

Performance of Gas Distribution Layer

The design of the biocells included a gas distribution layer constructed with crushed glass culets (Fig. 2.3.1). This layer was provided to equalize the inflow of gas into the compost. Fig. 2.3.2 shows a contour map of emissions from biocells (2D and 6D) before and after placement of compost. All contours were obtained using inverse distance weighting (IDW) with a power value of two. Abichou et al. (2006a) and Spokas et al. (2003) both reported that IDW is an acceptable method to map methane flux emissions from landfill surfaces. Prior to compost placement, an intensive flux survey was performed on all six cells to establish pre-compost conditions. The flux in each cell was measured 12 to 14 times during the month prior to compost placement. The pre-compost contours were obtained using the average of flux measurement during the period between January 14th and February 2nd 2004 for biocell 6D and from January 19th to February 2nd for biocell 2D. The post compost contours were obtained by averaging the emissions during the period between March 19th and June 29th, 2004.

Emissions from Cell 2D were very high and were characterized by a high peak in the Southeast quadrant. The flux contours after compost placement show significantly lower peak fluxes. They also show that the Southeast quadrant has remained the area of higher emissions as compared to the rest of biocell 2D. The glass layer placed below the compost was thought to act as a dispersing layer. The flux contours seems to indicate that the glass layer was not able to disperse the high flux measured before the glass was placed. This might be due to the limited thickness of the glass layer and to the lack of separation between the distribution layer the adjacent soils. Cover soil from below and compost from above might have migrated to the more porous distribution layer and might have caused clogging of this layer.

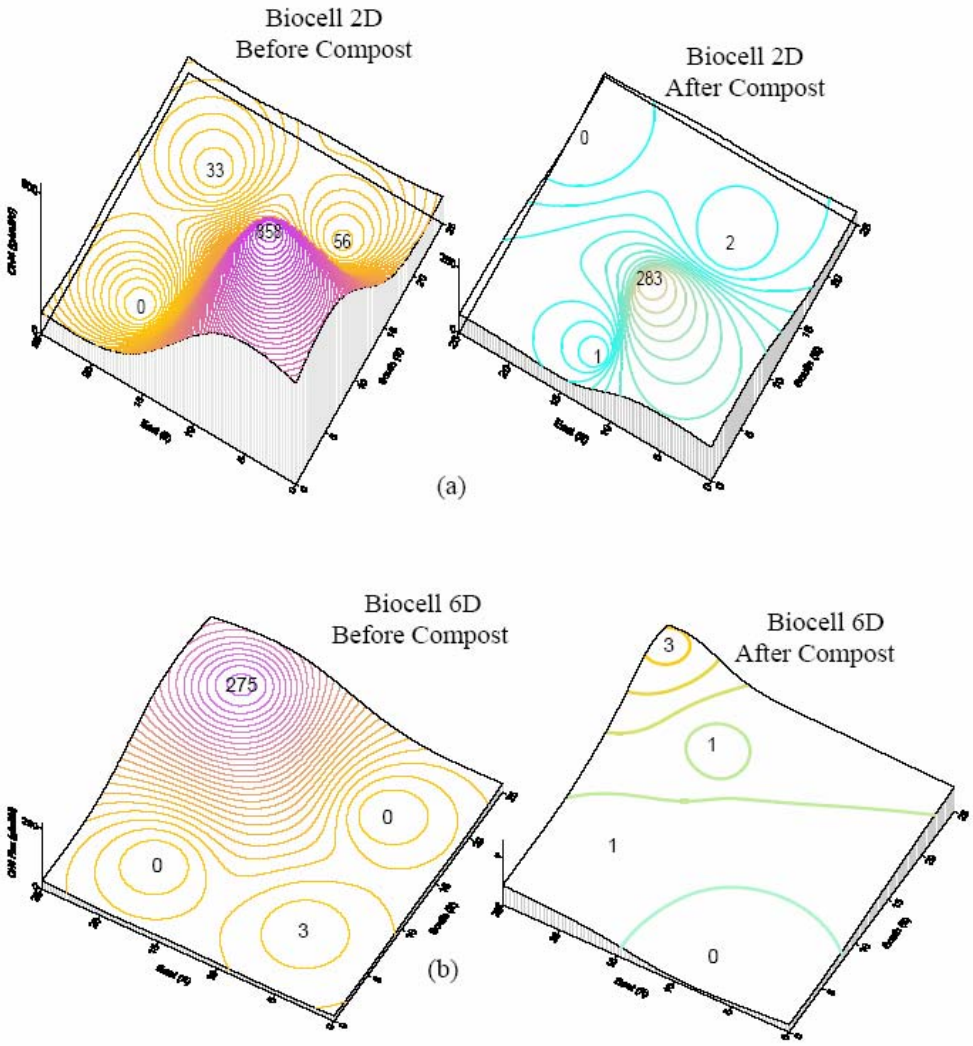


Fig. 2.3.2. Contours of Surface Methane Flux Pre and Post Placement of Compost for Biocell (a) 2D; (b) 6D.

Table 3.2.1. Summary of methane emissions from all cells

	Pre Biocells**		Post Placement of Biocells		
Cell	**Average Flux ($g/m^2/day$)**	**Peak Flux ($g/m^2/day$)**	**Average Flux ($g/m^2/day$)**	**Peak Flux ($g/m^2/day$)**	**Number of Negative Flux**
2B	37	247	**23**	740	**11**
4B	20	58	**13**	218	**21**
8B	2	8	**2**	49	**25**
2D	306	1743	**32**	1159	**25**
4D	11	77	**1**	22	**33**
6D	65	330	**0**	11	**30**

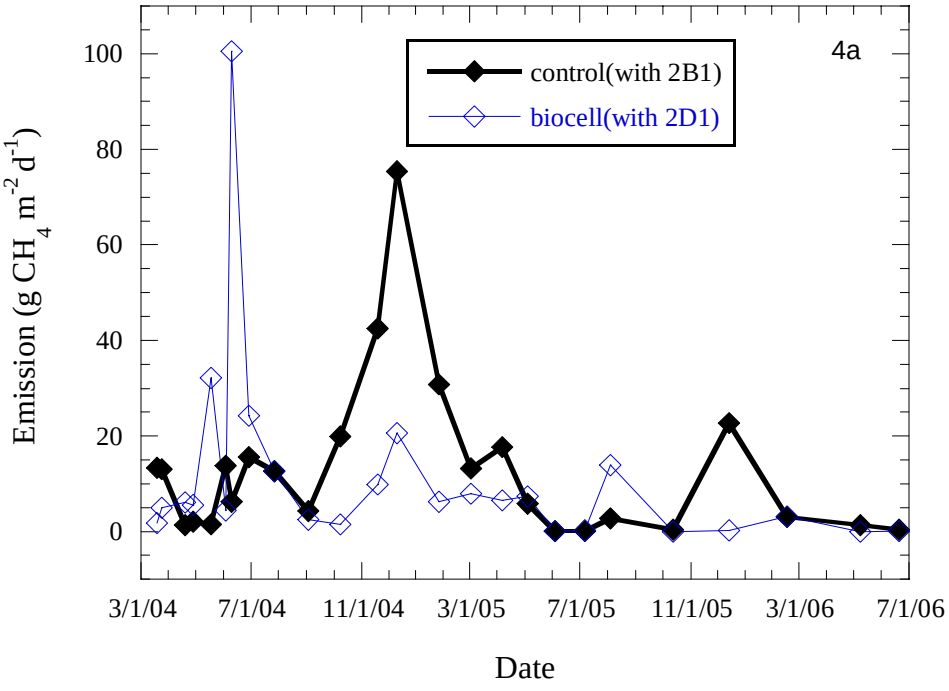
** Pre-biocells emissions were monitored for a period of 3 months

Comparison of Emissions from Treated and Non-treated Areas

Table 3.2.2 shows average and peak methane flux measured from each control and biocell before and after placement of biocells. The peak measured methane flux in the control cells before placement of the biocells were 247, 58, 8 g m^{-2} d^{-1} for Cell 2B, 4B, and 8B respectively. In contrast, peak emissions measured on Biocells 2D, 4D, and 6D were 1743, 77, and 330g m^{-2} d^{-1}, respectively, during the same period. The peak measured flux increased for all control cells during the post-biocell period (740, 218, 49 g m^{-2} d^{-1}). In contrast, the peak emissions for all biocells decreased during the post-biocell period (1159, 22, 11 g m^{-2} d^{-1} for Cells 2D, 4D, and 6D respectively). During the post biocell period, the average emission from the control cells was 13 g m^{-2} d^{-1}, which 65% from the average of fluxes measured during the pre biocell period. Emissions from the biocells (2D, 4D, and 6D) during the post biocells period averaged 11 g m^{-2} d^{-1}, which is only 8.7% of the average emission before the placement of the biocells. These observations are expected since the placement of compost on Cells 2D, 4D, and 6D should increase the resistance to gas flow from the inside the landfill to the surface. At the same time, compost has been reported to oxidize more methane. Additional analysis will be provided later to separate these two effects.

The distributions of flux measurements from the biocell and the control cells were both highly skewed. At the same time, one flux chamber in the biocells (2D1) and one flux chamber in the control cells (2B1) were orders of magnitude higher than all other flux values. As described in Stern et al. (2006), statistical analysis was performed to exclude flux values that did not fall within two standard deviations of the mean for each treatment on a given date and did not pass the Q-test. The Q-test is generally considered to be the most legitimate statistical test available for the rejection of deviant values from a small sample with a Gaussian distribution (Rorabacher, 1991). Values are placed in rank order and the difference between the outlier and the next closest value is divided by the range of values, i.e. the gap divided by the range. This quotient is compared to a table of critical Q values, and if larger than the critical Q value at a specified confidence level, the outlier can be rejected.

Fig. 2.3.3a shows the average emissions measured in the control cells and the biocells over the entire period of this study (March, 2004 to June, 2006), without excluding flux measurements 2D1 and 2B1. The average flux measured in the biocells was 10.87 $gm^{-2}d^{-1}$ as compared to 12.76 $gm^{-2}d^{-1}$ for control cells. ANOVA analysis showed that $p=0.72$, which means there was no significant difference between them.



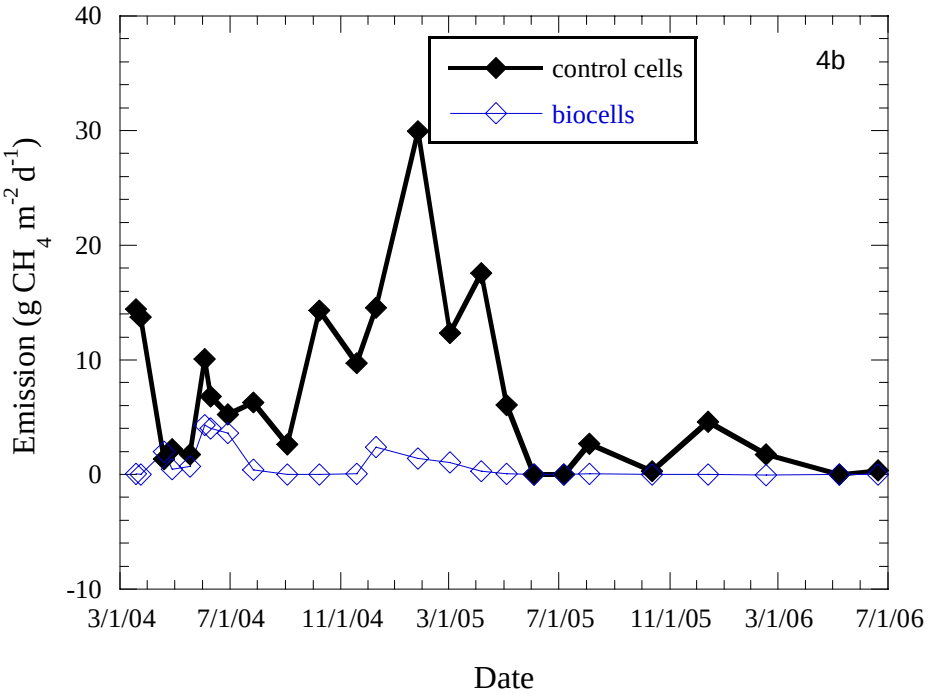


Fig. 2.3.3. Average Surface Flux Measured at the Control Cells and Biocells: Including all flux data (a), and excluding 2D1 and 2B1 (b).

When fluxes from 2D1 and 2B1 were excluded, the average flux in the control cells and biocells were significant different (one-way ANOVA, p=0.00103), with a mean flux 7.15 g m^{-2} d^{-1}, for the control site and 0.084 g m^{-2} d^{-1} for biocells. The first period corresponds to the time just after the compost was placed on the biocells. It is believed that during this period, the placement of compost on the biocells has led to disturbance in the flow patterns of landfill gas from the underlying waste. This period was considered to be a flow equilibration period. Outside the early equilibration period (From June 29th 2004 to the end of the study), emissions from the biocells seems to be always lower than those measured on the control cells.

Negative fluxes were measured on all control and biocells, which indicate areas where methane in the atmosphere was being consumed by the soil cover. The numbers of negative fluxes measured for the control and the biocell areas are also shown in Table 3.2.2. The biocells were a sink for atmospheric methane 88 times during the monitoring period as opposed to 57 times for the control cells. Negative fluxes might also an index of higher methane oxidation. Further analysis of oxidation results is provided later in this section.

Methane Oxidation Results

Table 3.2.2 shows methane oxidation measured during this study before and after the placement of compost biocells. Percent oxidation was calculated from stable isotope data as described earlier. Fig. 2.3.4 shows the percent oxidation measured during the entire study. Over the entire course of the experiment, oxidation in the control and biocell were significantly different (p=0.037), with a mean oxidation of 26.4% for the control and 39.7% for the biocell (Table 4.3). Negative flux values indicate uptake of methane from the atmosphere by the landfill (Bogner et al. 1997a). A negative flux indicates a downward flux of atmospheric CH_4 into the landfill, so it may be that all of the methane coming from below is oxidized before it reaches the soil surface. It is possible to assume that these fluxes represent 100% oxidation (Stern et al., 2006). Alternatively, negative fluxes could indicate blockage or failure of methane to be transmitted through a less permeable zone below the surface. Probably both explanations serve to describe different areas. When values for 0 and negative fluxes are included in

the averages as if they represent 100% oxidation, the mean oxidation for the biocells was 72.9%, and the mean oxidation for the control was 51.7%, with p = 0.014 for the entire monitoring period (Fig. 2.3.5).

Table 3.2.2. Summary of Methane Oxidation from all Biocells and Control Cells from March 2004 to June 2006.

	Pre Biocells* Average Oxidation	**Post Biocells Average Oxidation Excluding Zero and Negative Fluxes as 100% oxidation**	**Post Biocells Average Oxidation Zero and Negative Fluxes as 100% oxidation**
Row B (Control)	25.2%	26.4%	51.7%
Row D (Biocell)		39.7%	72.9%

* Pre-biocells oxidation were monitored for a period of 3 months in the area where this study was performed (Abichou and Chanton, 2004).

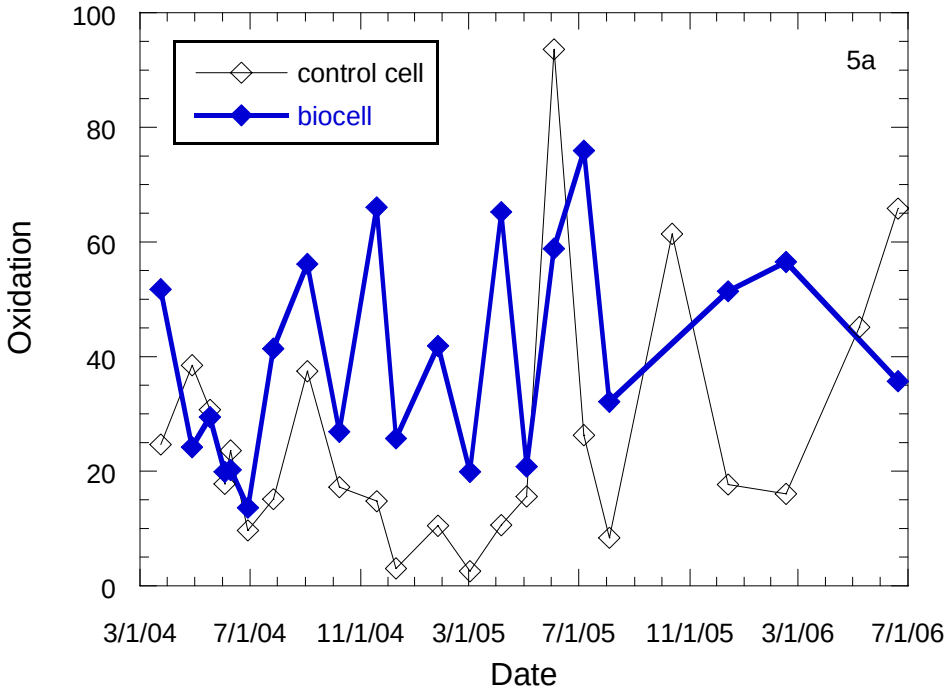


Fig. 2.3.4. Oxidation Results Measured in Biocells and Control Cells without assume zero and negative flux to be 100% oxidation.

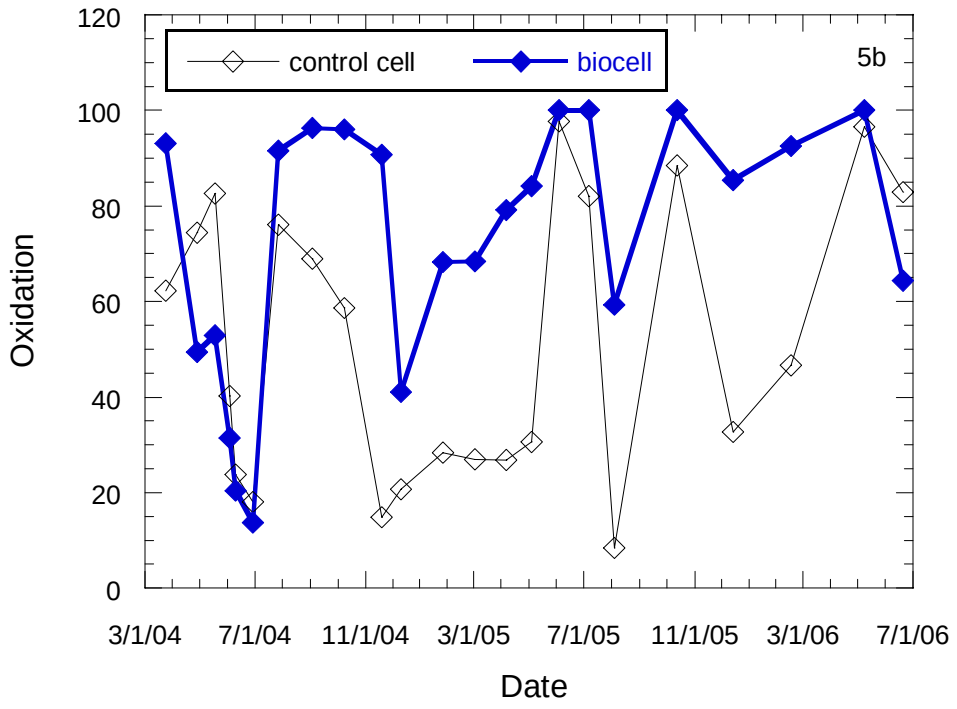


Fig. 2.3.5. Oxidation Results Measured in Biocells and Control with assume zero and negative flux to be 100% oxidation.

2.3. RECOMMENDATIONS

Daily Cover Applications: It was concluded from our study that the deep mulch treatment significantly increased methane oxidation. The shallow mulch treatment also increased methane oxidation in the landfill cover. On average, the biocover with the deep mulch was the most effective at oxidizing methane. The layer without mulch was the least effective at reducing methane emissions. The annual average methane oxidation rate of the deep mulch layer was 41%. The shallow mulch layer oxidized 22% of the methane and the no mulch layer only oxidized 18% of the methane. The deep mulch treatment oxidized over twice as much methane as the no mulch treatment.

We recommend adding mulch to any thin cover and especially daily cover. The addition of mulch will keep the soil cover moist and warm and therefore enhance methane oxidation. The mulch can be removed when filling is to continue in the ame location. The mulch can then be re-applied to the same area. A dispersion alyer for this application is not necessary.

Interim Covers Applications:

The glass dispersing layer in our study was not effective in equalizing the methane influx into the overlaying compost and was deemed to be too thin (10 cm) to achieve its functionality. Methane oxidation occurred in the biocells and the control cells. The biocells, were more effective in reducing methane emissions from the surface of the landfill via methane oxidation and blockage of landfill gas from underlying waste. Methane oxidation averaged 26.4% in the Control cells as compared to 39.7% in the biocells. These averages increase to 51.7% and 72.9% when the negative fluxes are considered as 100% oxidation. When outliers, were removed from the data, methane emission averaged 0.084 $gm^{-2}d^{-1}$ from the biocells and was significantly different from emissions from the control cells (7.15 $gm^{-2}d^{-1}$).

A numerical model was also used to separate blockage of landfill gas from the underlying waste and the actual biological oxidation of methane by methanotrophic

bacteria. Simulations were performed for 1 year (2005) on two soil columns depicting a biocell (soil column with a layer of compost) and a control cell (soil column only).

Simulations of methane transport through both columns indicated lower methane emissions from the biocell as compared to control. The simulated emissions were in the same range as emissions measured in the field and average 0.72 g m^{-2} d^{-1} in the biocell simulations and 9.18 g m^{-2} d^{-1} in the control simulations. In addition, influx of methane into the biocell (9.7 g m^{-2} d^{-1}) was lower than that into the soil cover alone (27.9 g m^{-2} d^{-1}), and thus indicating that part of the lower emissions from the biocell were caused by the presence of the compost layer (blockage). In addition to blockage, methane oxidation in the compost layer was also responsible for the lower emissions obtained from the biocell simulations. Methane oxidation was associated with lower water content in the top layer of the biocell. On the other hand, methane blockage was associated with higher water content in the lower layer of the biocell.

When there is a limited supply of mulch or compost biocells are a good option. The dispersion layer of these biocells is critical. It should be at least 30 cm thick. The compost should be at least 1 m thick. Surface drainage of the biocell should be investigated.

Application to gas well seals

Gas well seals can also be redesigned based on the results of our biocells and biocovers. For instance, Fig. 2.3.1 shows a survey of surface emissions from a landfill in Florida. The final cover of this landfill consist of a geomembrane-clay composite cover. Surface emissions were very low almost everywhere on the landfill surface. Surface emissions in the vicinity of gas wells and pipe intrusions were extremely high (up to 9500 g/m2/d). The well seals were constructed as recommended by membrane manufacturers and consisted of a boot with a bentonite seal. Fig. 2.3.2 shows a photograph of a typical gas well seal. The bentonite seams to be drying out and cracked. The fractures in the bentonite are responsible for the high gas leaks around these wells. Placing Compost around wells would have kept bentonite from cracking leading to less gas leaks. Also compost could have oxidized emissions from these areas. Fig. 2.3.3 shows our recommended design.

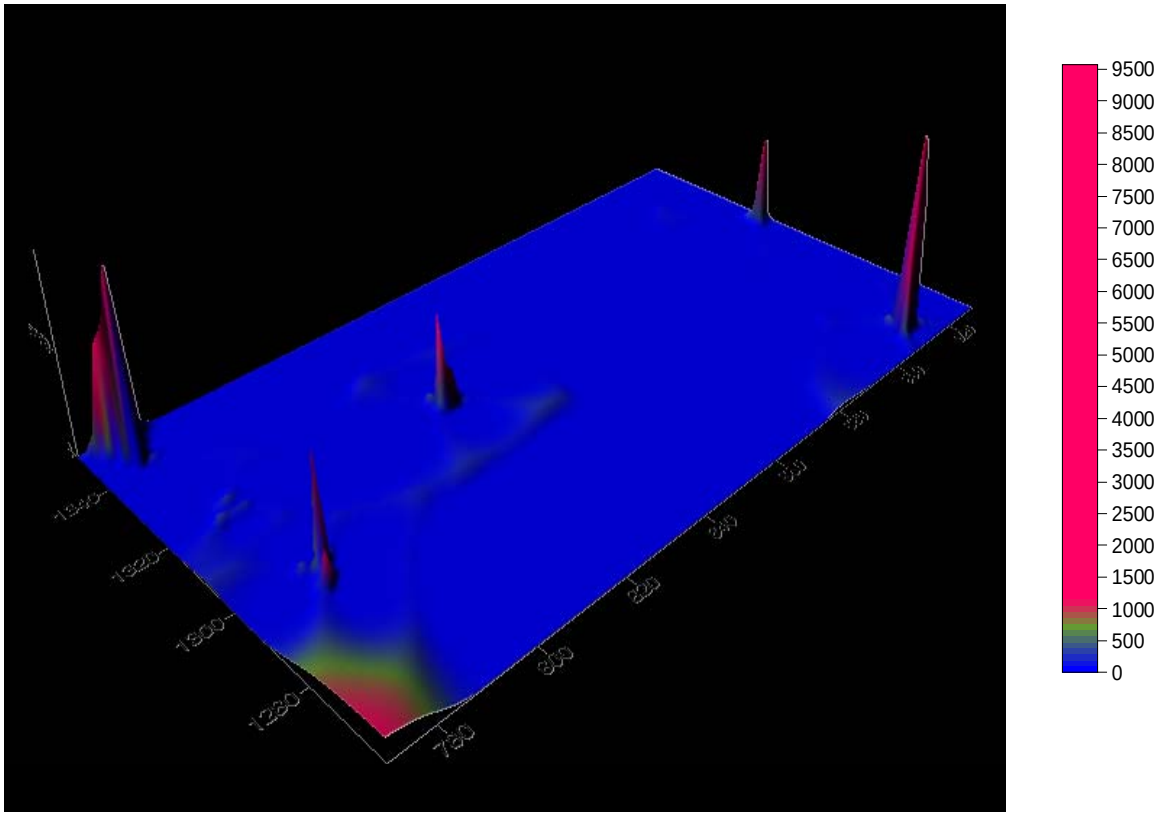


Fig. 2.3.1. Surface emissions from geomembrane-clay covered landfill.

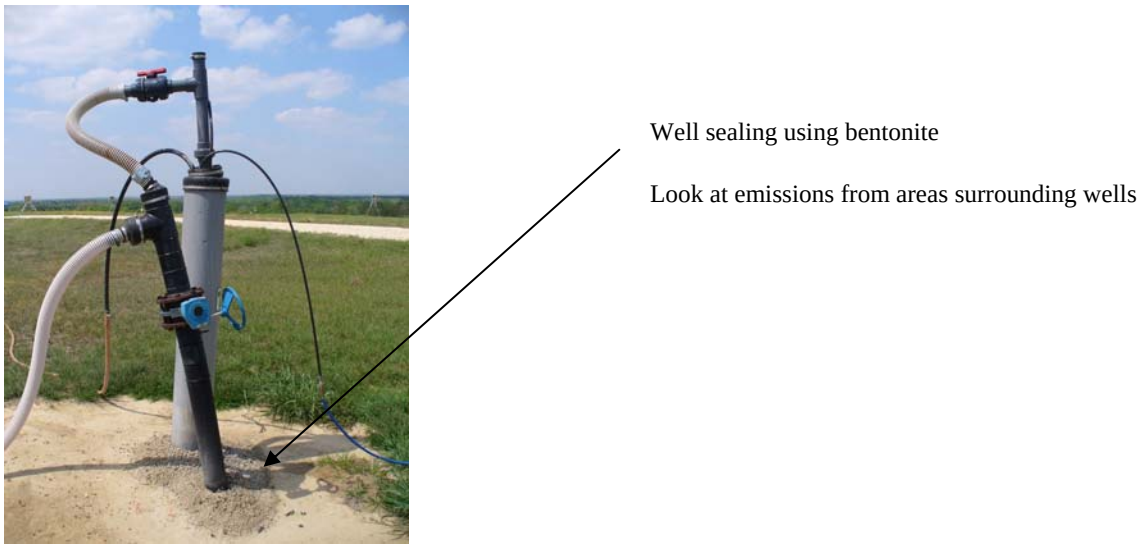


Fig. 2.3.2. Typical gas well seal design.

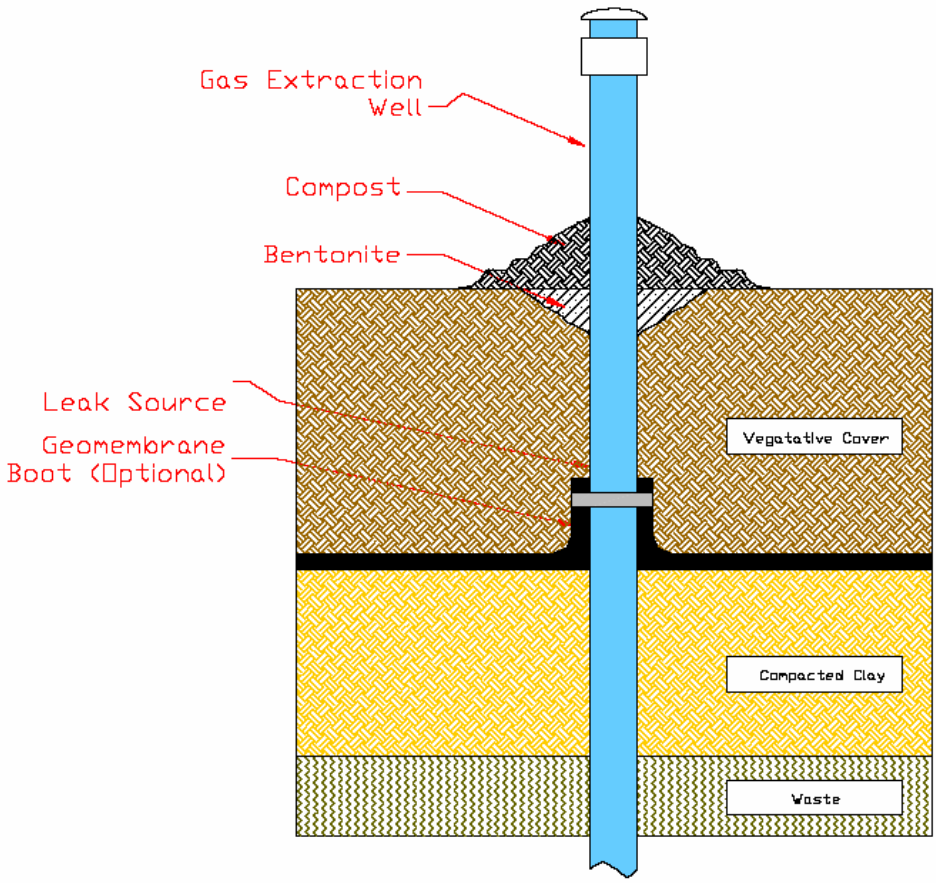


Fig. 2.3.3. Recommended gas well seal.

CHAPTER III
BIOFILTERS, BIOWINDOWS

3.1 HISTORICAL PERESPECTIVES

Biofilters are reactors that contain materials that support methane consuming bacteria. The media must sustain good environmental conditions for the bacteria and have gas permeable pore space. Biofilters are most useful when active gas extraction and energy recovery is no longer, or not yet, practical. The filters can either be operated with active gas feed or passive gas feed. Passive gas feed occurs because of the pressure gradient between the landfill and the atmosphere, but active gas feed can be controlled. There are two types of biofilters: closed bed and open bed. Open bed biofilters can be integrated into the cover system, allowing landfill gas to enter through the bottom of the filter and progress upward through the methane oxidizing material. They do not require supplementary heating or irrigation. These filters receive oxygen from diffusive ingress or from a supply line. Closed bed biofilters are completely enclosed and require that gas be fed in through a supply line. Closed bed biofilters are more easily regulated than open biofilters because the temperature, moisture conditions and gas flow can be controlled. Closed beds also have higher capital and operational costs. Open filters require less maintenance, but they are not as easily regulated because they are part of the cover and temperature, moisture content and gas flow cannot be controlled easily.

The biofiltration media should have a large pore area, a large surface area and good environmental conditions for the microbes, including water-holding capacity, appropriate pH, conductivity and nutrient availability. High permeability is also a necessity because it is important to reduce pressure loss. The optimal media is one that is homogenous so as to minimize segregation and aggregation. Several medias have been found to be effective biofilter materials. Gerbert and Grongroft (2006b) found that inorganic porous clay pellets topped with densely grassed top soil achieved methane removal rates of 80 g m^{-3} h^{-1}. Streese (2005) found that actively vented compost reached methane removal rates of 10 – 20 g m^{-3} h^{-1} when operated in a down-mode at 20 ^{o} C. Straka et al. (1990) found that mixtures of compost and bark or compost and wood chips

were effective at offering an environment that supports methanotropic bacteria and therefore produced methane removal rates of over 90%.

There are limitations to biofilters. When the landfill gas flow upward is so great that it reduces the downward flow of oxygen, the biofilter is not as effective at reducing methane emissions because methanotrophic bacteria need oxygen to consume the methane. Also, low ambient temperatures will slow the methane uptake process. One way to overcome this challenge was found by Powelson et al. (2006). They installed differently textured sands into an upward gradient, causing the gas-filled pore space to increase downward. Another problem that can occur in biofilters is the formation of exopolymeric substances (EPS). These substances cause the biofilter material to adhere to itself, limited the void space in the filter. This decreases the ability of gas to flow through the filter. EPS gas formation may be avoided by controlling the rate of inlet flux to the landfill biofilter.

Smaller, as well as older landfills, typically emit methane into the atmosphere through passive vents, vents that do not have vacuum applied to them. A passive vent is a vent that has no mechanical workings, such as a blower or a pump, to pull the landfill gas out. Instead of relying on a power-driven device, passive vents rely on advection and Fickian transport to expel gases out of the landfill envelop. It was the objective of this study to research biofilters as a way to reduce landfill methane emissions from passive venting into the atmosphere.

Biofilters can also be referred to as biowindows. Biowindows are used when a full biocover is too expensive or unneeded, and when there is no gas collection system to direct the gas to a biofilter. The window is typically composed of compost and it is integrated into the landfill cover. The gas flows beneath the window and then progresses up through the window. The gas does not need to be directed because it will naturally migrate to the window because it is the path of least resistance due to its low permeability.

A field-scale biowindow was also tested on an MSW landfill in Louisville, Kentucky that had an operating gas extraction system. Both a flat and a sloped biowindow section were tested, where each was made of a 0.15 m clay layer overlain with a 0.15 m layer of tire chips for gas distribution and a 1 m layer of yard waste

compost. The control plots contained a 1 m thick conventional clay cover (Barlaz *et al.* 2004). With the gas collection system off, methane emissions from the biowindow cells did not increase, whereas those from the conventional soil cover rose significantly. When the collection system was operational, the soil cover generally performed well, although it occasionally released large quantities of methane thought to be mainly associated with desiccation cracks. No such cracks were observed in the biowindow cells, leading the authors to conclude that compost-based biowindows not only reduce emissions through biotic mitigation, but also, through their propensity to resist erosion and cracking, can vent large gas flows.

3.2 FLORIDA PRESPECTIVES

Gas Vent Biofilter

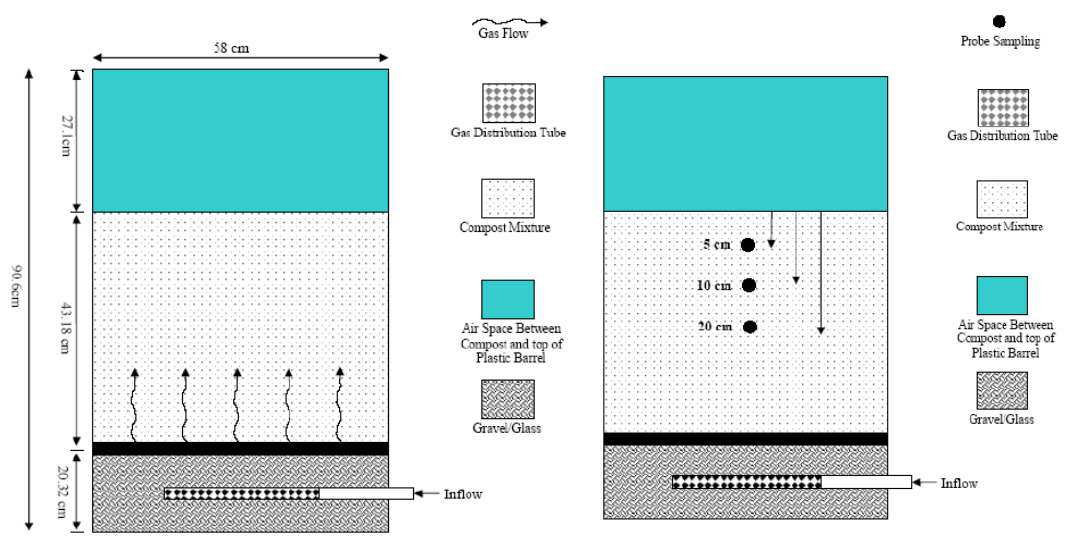
A biofilter study was performed on Phase One of the Leon County Landfill on Apalachee Parkway, approximately 8 miles west of Tallahassee, Florida. The objective of this study was to investigate the feasibility of using methane oxidation to reduce emissions from passive landfill vents. This low technology approach has the potential of reducing greenhouse gas emissions from landfills in developing countries and in landfill with low methane production. Two designs of biofilters that vary in construction (vertical and radial biofilters) were developed and assessed in the field.

3.2.1 Biofilter Designs

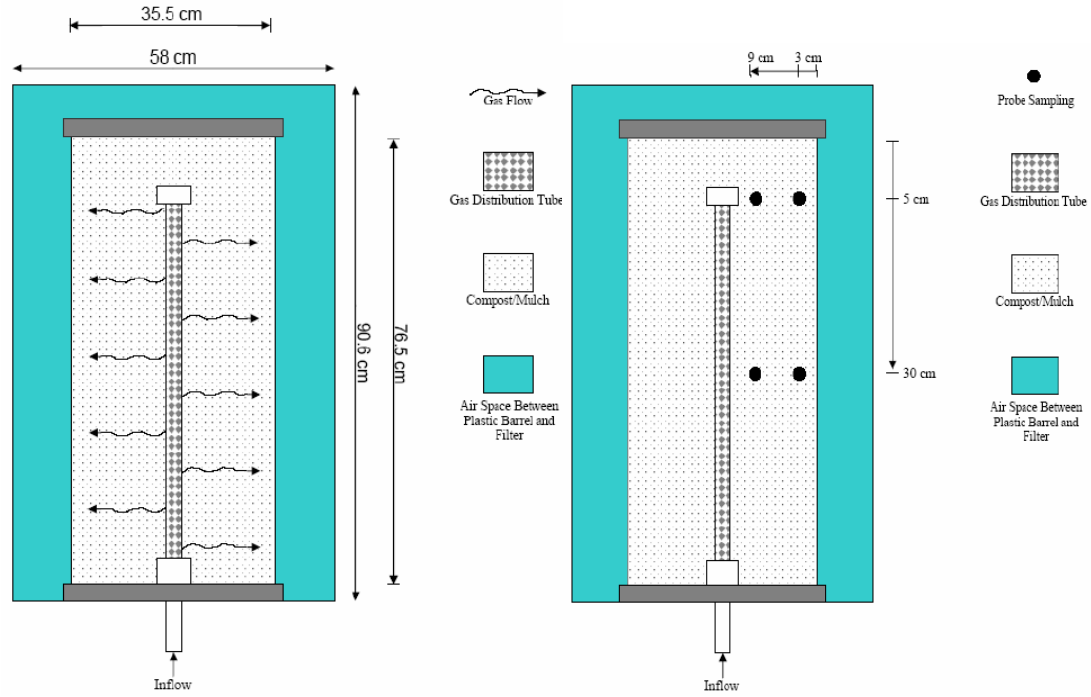
The vertical biofilter design is named after the direction of gas flow within the filter. As shown in Fig. 3.1a, landfill gas enters the bottom of the biofilter and migrates vertically up through the compost media mixture and out the top. This design was constructed using a 231 liter barrel in which it is housed. The filter has an inflow near the bottom which is embedded in a drainage layer of gravel and/or recycled glass. This drainage layer also acts as a gas distribution layer. A geotextile/geonet composite separates the drainage layer from the compost mixture and provides additional gas dispersion. The compost mixture is made up of a 50/50, by volume, mix of compost and peanut foam or compost and rubber tire chips. Peanut foam and the rubber tire chips were used to increase the air filled porosity of the compost and to increase air permeability of the media.

The concept of a “radial” biofilter was developed because of the need to increase oxidation of methane by biofilters. The radial design increases the surface area of flow. The surface area for the radial filters is 1.212 m^2, an increase of 459% of that of the vertical filters (0.264 m^2). Fig. 3.1b shows a diagram, with dimensions, of the radial biofilters. The radial biofilters are then housed in an identical plastic barrel as the vertical filters for protection. It is important to note that there is a gap between the sides of the

radial biofilters, and the side of the barrels to allow for diffusion of atmospheric oxygen into the filter.



(a)



(b)

Fig. 3.1. Diagram of Biofilters with flow patterns and location of gas sampling probes: Vertical Design (a) and Radial Design (b).

3.2.2 Measuring of Emissions and oxidation from Biofilters

Measurements of CH_4 emissions from each biofilter were performed using static chamber technique similar to methods described by Czepiel et al. 1996, Hutchinson and Livingston 2002, and Powelson et al. 2006. For the vertical biofilters, the "chamber" consisted of the empty head space between the top of the biofilter media and the top of the plastic barrel. For the radial biofilters, the "chamber" consisted of the empty space between the filter media and the top and sides of the plastic barrels. To enclose the headspace, the original plastic barrel lid along with an equipped rubber gasket were placed on top of the barrels and sealed with a clamp. A sampling time increment of 0, 1, 2, 3, and 5 minutes were chosen to minimize pressure build-up in the head space. Samples were taken at these time intervals using plastic 60mL syringes which were then used to inject into a Shimadzu 8A Gas Chromatograph with a flair ionization detector to establish CH_4 concentrations. The change in concentration, C (ppmv), with time, t (min) was then used to

To determine methane oxidation in the biofilters, a stable carbon isotope technique was used. This technique has been widely used for this purpose (Liptay et al. 1998, Chanton et al. 1999, Abichou et al. 2005, Abichou et al. 2006, and Powelson et al. 2006.

Gas probes constructed with stainless steel tubes were used to obtain gas samples within each biofilter. Using these samples, gas concentration profiles were constructed for methane, oxygen, carbon dioxide, and nitrogen. For the vertical biofilters, the sampling depths were at 5, 10, and 20 cm depths (Fig. 1a). For the radial biofilters, samples were taken at 3 cm and 9 cm from the side of the filter at a distance of 5 and 30 cm from the top (Fig. 1b). Gas probe samples were analyzed in the lab using a Shimadzu 8A gas chromatograph (GC) thermal conductivity detector (TCD).

3.2.3 Vent Methane Output Patterns

The following is a mass balance based formula to calculate the influx of methane into the biofilters. This quantity is also the methane output from the vent to which the biofilter is attached.

$$J_{ines} = J_{out} /(1 - f_{oxm})$$

where, J_{ines} is the estimated methane influx, J_{out} is the measured methane outflux, and f_{oxm} is the fraction oxidized. The use of this approach allowed us to estimate the methane output from each passive vent, except for fluxing events with 100% oxidation. Average emissions or methane output from the four vents were 3815, 3644, 3508, and 2372 gd^{-1} respectively. Vent 1 methane output varied from a high of 7845 gd^{-1} to a low of 296 gd^{-1}. Vent 2 methane output varied from a high of 8298 gd^{-1} to a low of 5 gd^{-1}. Vent 3 varied from 5911 gd^{-1} to 0 gd^{-1}. Vent 4 varied from a low methane emission of 0 gd^{-1} to a high of 13120 gd^{-1}. These findings show that vents are effective in limiting horizontal migration of landfill gas by providing a vertical path to the escaping. The high methane output also underscores the fact that landfill vents can be a major source of greenhouse emissions from landfills even in older closed landfills. Similar to emissions from landfill surfaces, emissions from vents vary tremendously. In fact emissions from the same vent can vary up to 4 orders of magnitude. Table 3.1 shows vent methane output per sampling event, as well as average vent outputs for the study period. Blank cells in the Table below indicate that the methane output was not calculated due to 100% oxidation in all or one of the biofilters connected to that vent.

Average vent methane output decreased during the study. During the study air temperature increased due to change of seasons, from winter to summer. However, the decrease in methane output cannot be correlated to an increase in temperature. The decrease in methane output from the vents might be caused by the reduction in gas production from the aging waste.

3.2.4 Performance of Vertical and Radial Biofilters

At the beginning of this study Vents 1, 2, and 3 all were attached to vertical biofilters. However, Vent 1 biofilters were changed to radial biofilters and collection of data for these biofilters started on February 28, 2006. The two designs were evaluated by comparing performance data from the same time period, April 7 to August 4, 2006. The performance in oxidizing methane by the two design was compared in terms of percent oxidation and in terms of methane oxidation rate, because methane input into each type of biofilters were not found to be significantly different (P-value = 0.23). This means that all biofilters were operating in similar conditions.

Table 3.1. Summary of Vent output.

Date	Vent 1	Vent 2	Vent 3	Vent 4	Average
10/07/2005	6786	6641	5911		6446
10/12/2005	1229	5880	176		2428
10/20/2005	5639	7341	3571		5517
11/9/2005	1748	5893	5008	1867	3629
12/1/2005	5601	8298	3725	3640	5316
1/24/2006	5898	3258	4592		4583
2/9/2006	2276	679		310	1088
2/28/2006	7845				7845
4/7/2006					
4/25/2006	6431	6231		13120	8594
5/3/2006	1811	706		7811	3443
5/8/2006					
5/25/2006	953	1120	342	1489	976
6/7/2006	296	5	0	0	75
6/29/2006	4322	1190	198	4967	2669
7/18/2006	1459			1868	1664
8/4/2006	2369	1696	195	763	1256
Average	**3644**	**3815**	**2372**	**3508**	

For the period of April 7 to August 4, 2006 the highest percent methane oxidation achieved for a vertical biofilter was 100%. The highest percent oxidation, other than 100%, reached by a vertical biofilter was 67% with a methane input of 4.6 gd^{-1}. A minimum methane oxidation value of 0% was observed on a number of occasions for the vertical biofilters. Not including zero methane inflow events, the lowest calculated methane inflow into the vertical biofilter that is associated with 0% oxidation was measured was 169 gd^{-1}. This inflow corresponds to a flux of 640 $gm^{-2}d^{-1}$. One might

deduce that for the vertical biofilters, this flux of methane coming from the bottom was able to keep oxygen from diffusing into the biofilters and therefore practically stopping methane oxidation.

The highest methane percent oxidation achieved by a radial biofilter for this same time period was 100%. The highest percent oxidation value achieved by a radial biofilter, not including 100% oxidation events, was 53%, with a methane input of 628.7 gd^{-1}. Not including zero methane inflow events, the lowest calculated methane inflow into the radial biofilter that is associated with 0% measured oxidation was 296 gd^{-1}. This inflow corresponds to a flux of 247 $gm^{-2}d^{-1}$. One might deduce that for the radial biofilters, this flux of methane coming from the bottom was able to keep oxygen from diffusing into the biofilters and therefore practically stopping methane oxidation.

Table 3.2 shows the average methane inflows, average methane percent oxidation, and averaged methane oxidation rate for the two biofilter designs by flux date events for the time period of 4/7/06 to 8/4/06. One hundred percent oxidation events were not included in the averages, because input may not be calculated, as well as zero methane input events (since oxidizing methane is not possible without methane). Fig. 3.2 shows methane percent oxidation over time (a), as well as methane oxidation rate over time (b) for both types of biofilters. During the study period, vertical biofilters averaged a percent oxidation of 12.2% (N = 9, stdev = 0.21) and a methane oxidation rate of 10.7 g/d (N = 9, stdev = 22.4) at an average methane input rate of 836.6 g/d (N = 9, stdev = 1086.1).

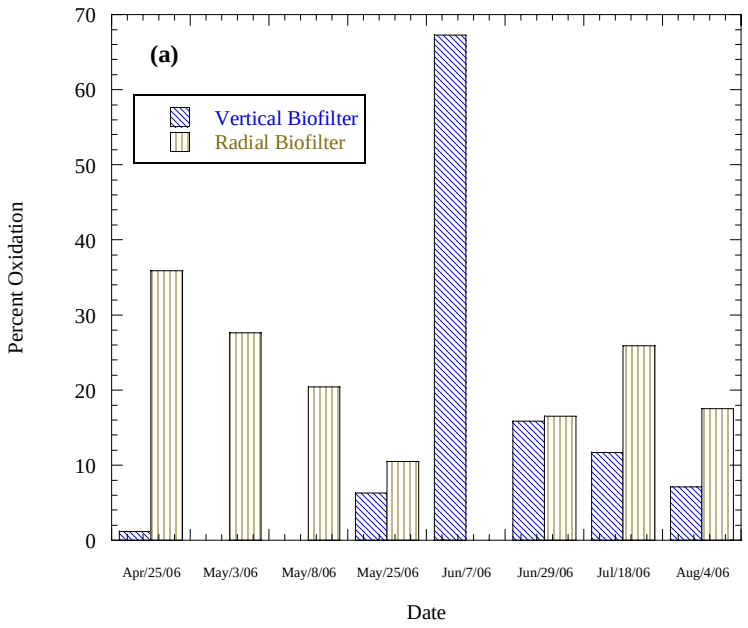
For the same time period, the radial biofilters averaged 19.3% (N = 8, stdev = 0.11) percent methane oxidation, 292.8 g/d (N = 8, stdev = 270.9) methane oxidation rate at an average methane input rate of 1580.1 g/d (N =8, stdev = 1420.6). Statistically, the two filter designs averaged a similar methane percent oxidation, 19.3% for the radial filters vs. 12.2% for the vertical filters with a P-value of 0.41. In terms of real performance, a better assessment of the two deign is the comparison the average methane oxidation rate (10.7 g/d for the vertical and 292.8 g/d for the radial). These two means were found to be significantly different (P-value = 0.0069). This analysis shows that a radial biofilter can oxidize as much as 30 times more methane as a vertical biofilter.

Vent 1 was the only passive vent that was attached to vertical biofilters, and later to radial biofilters. Comparison of the results for the two biofilter designs, vertical and

radial, from this same vent is of interest since the methane source is the same. Data from Vent 1 for vertical biofilters from 10/7/05 to 2/9/06 is compared to data from Vent 1 for radial biofilters from 2/28/06 to 8/4/06. The average calculated methane input for vertical biofilters on Vent 1 was 2084 gd^{-1} compared with 1855 gd^{-1} for the radial biofilters. Average methane removal rate for the vertical biofilters was 137.4 gd^{-1} compared to 300 gd^{-1} for the radial biofilters. The radial biofilters also outperformed the vertical biofilters in oxidizing methane (18.2% vs. 6.6%) Using this comparison, the radial biofilters achieved a higher methane removal efficiency at similar methane input.

Table 3.2: Summary of CH4 input, percent oxidation, and oxidation rate for vertical and radial biofilters.

	Vertical Biofilters			**Radial Biofilters**		
Date	**CH_4 Input (gd^{-1})**	**CH_4 % Oxidation**	**CH_4 Oxidation Rate ($gm^{-2}d^{-1}$)**	**CH_4 Input (gd^{-1})**	**CH_4 % Oxidation**	**CH_4 Oxidation Rate ($gm^{-2}d^{-1}$)**
4/25/2006	3116	1.2	69.7	3215	35.9	715.8
5/3/2006	353	0.0	0.0	906	27.6	188.7
5/8/2006	2235	0.0	0.0	1183	20.4	237.1
5/25/2006	366	6.3	7.2	953	10.5	99.8
6/7/2006	5	67.3	3.0	148	0.0	0.0
6/29/2006	347	15.9	2.6	4322	16.5	714.8
7/18/2006	165	11.7	4.9	730	25.9	185.5
8/4/2006	473	7.1	8.5	1185	17.5	201.0
Average	**837**	**12.2**	**10.7**	**1580**	**19.3**	**292.8**


(a)
Vertical Biofilter
Radial Biofilter
Percent Oxidation
0
10
20
30
40
50
60
70
Apr/25/06
May/3/06
May/8/06
May/25/06
Jun/7/06
Jun/29/06
Jul/18/06
Aug/4/06
Date

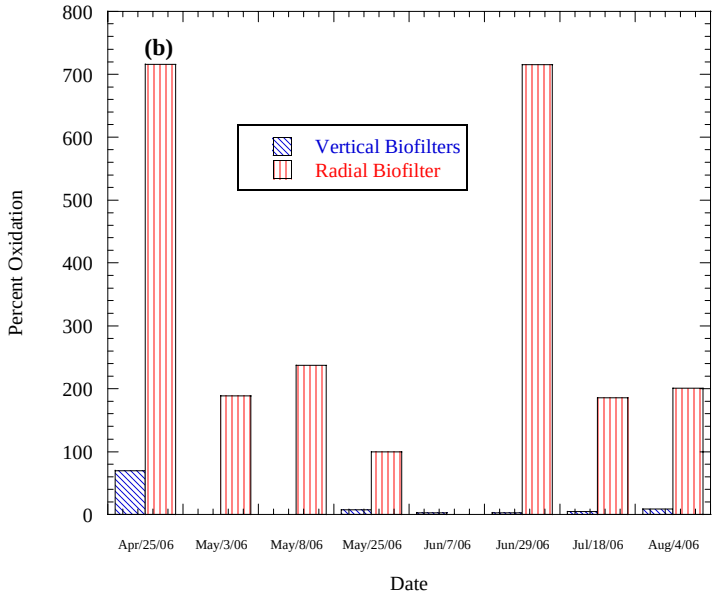

(b)
Vertical Biofilters
Radial Biofilter
Percent Oxidation
0
100
200
300
400
500
600
700
800
Apr/25/06
May/3/06
May/8/06
May/25/06
Jun/7/06
Jun/29/06
Jul/18/06
Aug/4/06
Date

Fig. 3.2. Percent Oxidation (a) and Rate of Oxidation (b) for Vertical and Radial Biofilter.

3.2.5 Gas Concentration in Biofilters

Fig. 3.3 shows oxygen and methane concentration inside the biofilter obtained on 4/25/2006. This date was chosen because both biofilters had similar methane inputs of about 5800 gd^{-1}. As seen Fig. 3.3, the radial biofilter attained higher oxygen content throughout its filter medium, with an oxygen content of 20% at 3 cm from the edge (12 cm from center where the input source is located), and 8% at 9 cm from the edge (3 cm from the input source where the input source is located). The vertical biofilter, on the other hand had an oxygen content of only 2% at 5 cm from the surface (20 cm from bottom where the input source is located) and 1% at a depth of 20 cm (10 cm from the bottom where input source is located). During this sampling event, the radial biofilter achieved a higher oxidation rate (18.9%) under the same methane input rate than the vertical biofilter (2.4% oxidation). This difference in performance is due directly to the increased surface area of the radial biofilter over the vertical biofilter. The increase in surface area allows for a lower methane flux (4787 $gm^{-2}d^{-1}$ for the radial biofilter vs. 22096 $gm^{-2}d^{-1}$ for the vertical) at the same input rate which results in an increase in oxygen penetration in the radial biofilter.

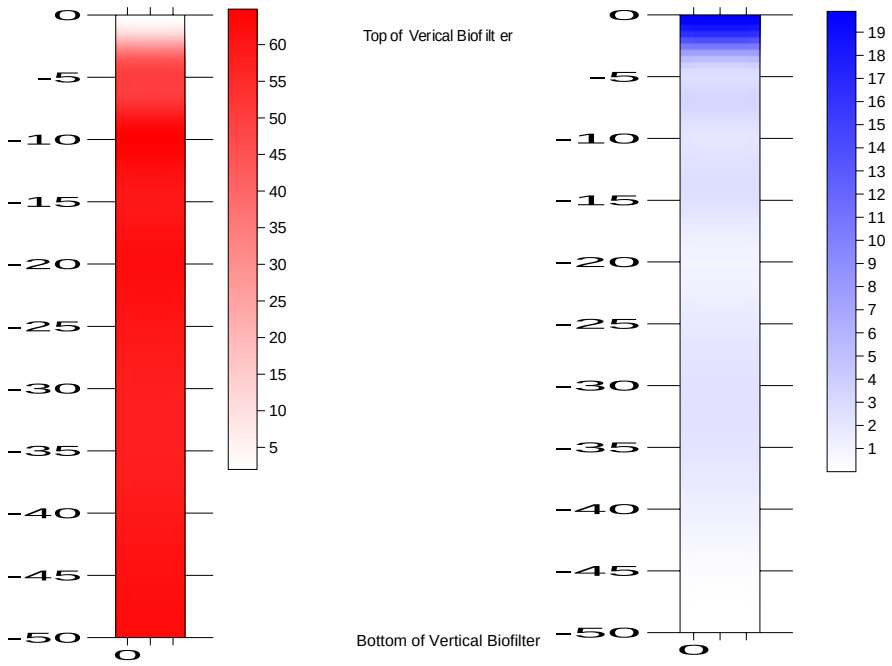
Conclusions and Practical Implications

The average methane output for the study period was 3644 g/d for Vent 1, 3815 g/d for Vent 2, 2372 g/d for Vent 3 and 3508 g/d for Vent 4. Emissions from these vents were high considering that the landfill cell where these vents were located is 20 to 30 years old. Although atmospheric pressure was recorded during testing events during the study, there was no correlation found between the atmospheric pressure and vent methane emissions.

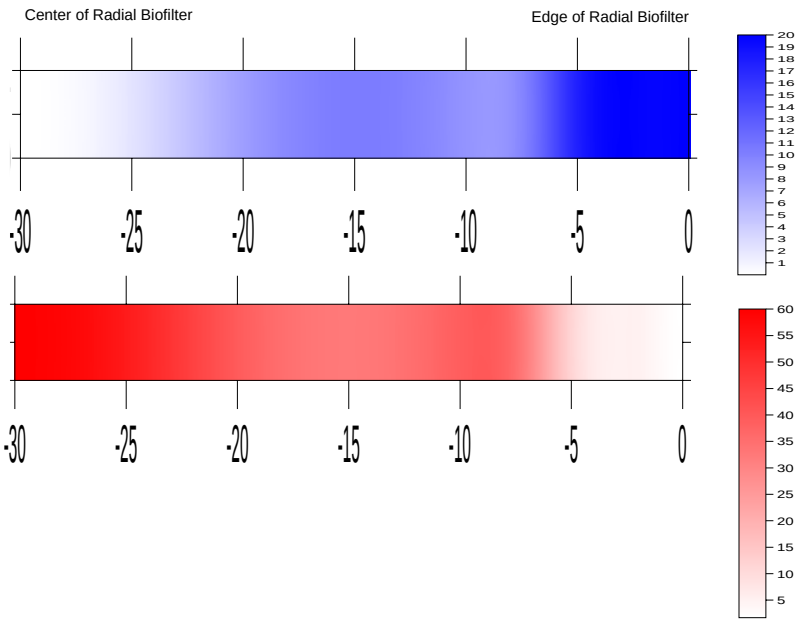
The highest percent oxidation achieved by a vertical biofilter was 100%, as was obtained with the radial biofilters. The lowest methane percent oxidation achieved was 0% by both designs. From April 7th 2006 to August 4th 2006 the radial biofilters outperformed the vertical biofilters. During this time period the radial biofilters attached

to Vent 1 averaged a 19.3% methane oxidation at an average methane input of 1580.1 g/d and an average methane removal rate of 292.8 g/d. The vertical biofilters on Vent 2 and 3 averaged a similar oxidation of 12.2 %, but at a lower methane input of 836.6 g/d, and a much lower methane removal rate of 10.7 g/d.

The overall better performance of the radial design is attributed to its larger surface area (1.212 m^2) than that of the vertical design (0.264 m^2). The increased surface area results in a smaller influx, increased detention time, and better oxygen penetration. All these factors combined make the radial design a much better design for oxidizing passive methane emissions from landfills. The finding of this study also showed that availability of oxygen to the methanotrophic bacteria, filter influx, and filter surface area are key factors for optimizing methane oxidation in biofilters. It might be concluded that passive oxidation for methane will be a more viable option if vent emissions were lower, i.e., at older landfills. The study also suggests that introducing air into biofilters with higher flows might improve their performance and should be investigated.



(a) Methane Left, Oxygen Right



(b) Oxygen Top, Methane Bottom

Fig. 3.3. Methane and Oxygen Concentrations within Biofilters.

3.3 FLORIDA PERESEPECTIVES

Large Scale Biofilter

3.3.1 Experiemental Set-up

This study expands upon the previous research on gas vent biofilters conducted at the Leon County landfill, and also examines the possibility of using a low cost, passive horizontal gas collection system to redirect landfill gas to the biofilters. This study was conducted at the Leon County landfill from December of 2005 to March of 2007.

In November of 2005, we selected a site for the implementation of a large scale biofilter project at the Leon County Landfill. This site presently has a temporary cover and has been closed for approximately two years. This site was found to be suitable due to significant flux of methane coming through the cover as determined by use of a portable gas chromatograph (GC) and subsequently by several tests done using the static chamber flux method. The study area was divided into a grid of 36 squares (cells) in a 6 by 6 pattern. The total site was 46 meters by 46 meters square. Each node was 7.6 meters by 7.6 meters, giving a total of 36 cells for data collection. Surface mapping of methane emissions began as soon as the site was selected. The emissions from the site were measured a total of eight times between January 2006 and May 2006 using the static chamber method. Each time emissions were taken, flux from every cell was measured the same day for consistency.

Flux data for each of the 36 nodes was plotted using Surfer 8, mapping software that is capable of using various weighting functions to determine the distribution of flux rates throughout the site, presented as a topographic map. Variations of flux between data points can be determined by either the Kriging method (default) or by means of the Inverse Power Weighting (IDW) method, whereby the effect of a data point decreases as a function of the distance raised to some user defined power. The software calculates total cut and fill volumes, which in this scenario, represent the mean flux rate for the day for the entire site in terms of $g/m^2/day$. The IDW method was the chosen method for initial data evaluations since prior investigations have shown that using this method with a power of 2 is adequate for soil covers (Spokas et al., 2003; Abichou et al., 2004).

The more accurate and much more frequently performed static chamber tests have shown that the site has a geospatial mean flux ranging from 60 to 120 g/m^2/day (Fig. 3.4). This is significantly higher than many of the tests performed on other sites at the same landfill, sometimes a whole magnitude higher. The site cover consists of a slightly sandy fat clay (CH with sand) varying from 2.5 to 3 feet in thickness. An exception is the southwest corner, where red sandy lean clay (SC-CL) predominant.

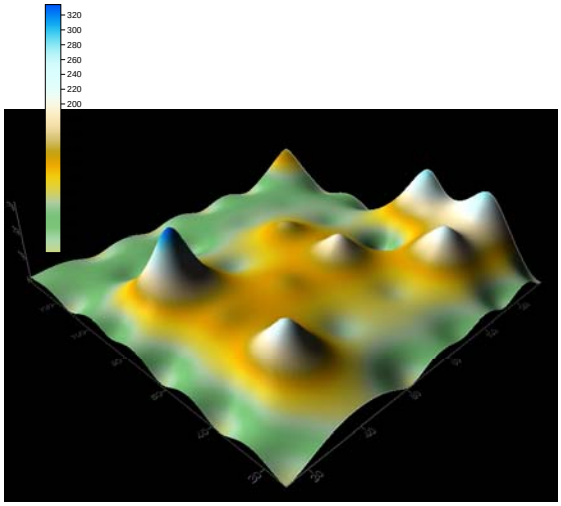
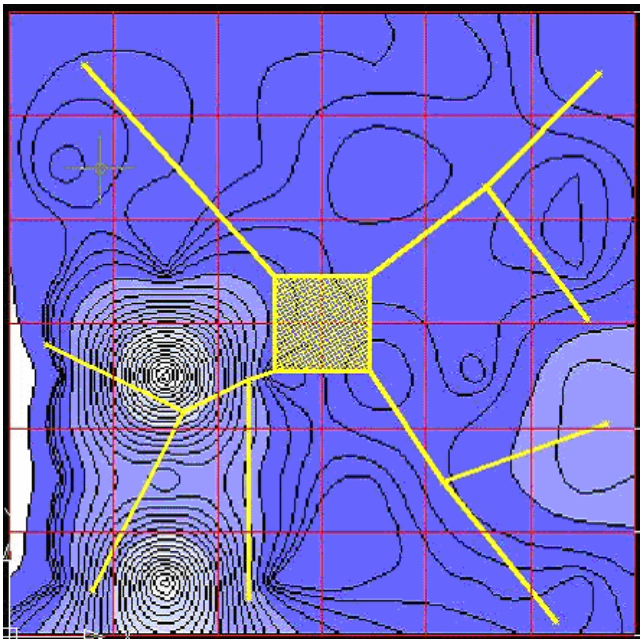


Fig. 3.4. Contour Map of Methane Emissions from the Biofilter Area Prior to Trenching.

Following several fluxing sessions, a consistent pattern was found where the hot spots remained in the southwest corner and on the center of the west side as can be seen Fig. 3.4. Contour maps of flux emissions from four dates between January 20th and February 23rd were used to determine suitable locations for the horizontal, passive gas collection trenches. A total of 140 meters of trench were installed, the positions and proportions of which were intended to coincide with locations of high methane output (Figure 3.5).

The trenches were constructed during the week of March 6-11, 2006. The trenches were approximately 0.6 meters wide and 1.2 meters deep (Figure 3.6). They were first lined with geotextile, and then filled with 0.5 meters of rubber tire chips. A 10.2 centimeter diameter, perforated PVC pipe was embedded in the tire chip layer to direct the landfill gas toward the proposed biofilters (Figure 3.7). The geotextile was

then wrapped around the top of the tire chip layer to form a sort of ‘burrito’, and then covered with a geomembrane and 0.7 m of soil cover (Figure 3.8).



(a)

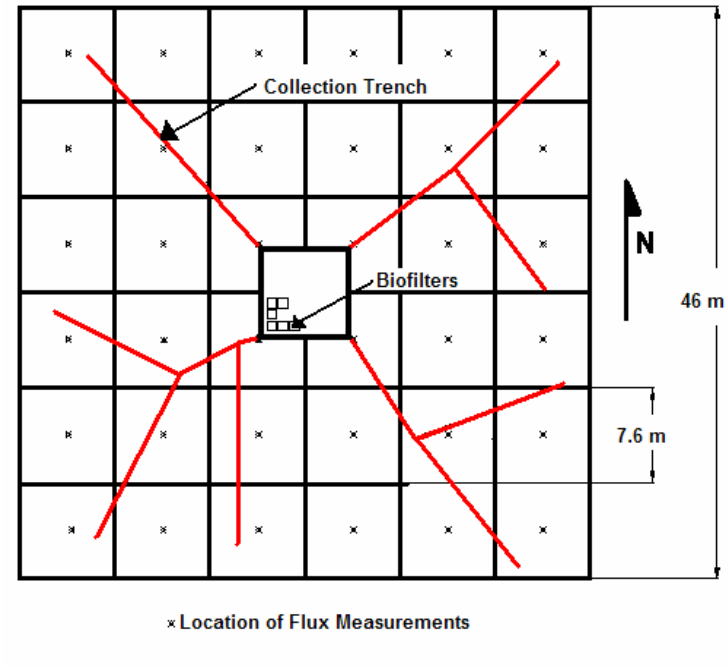


Fig. 3.5. (a) Location of trenches and example of hot spots on flux map, and (b) location of measurement points.

Fig. 3.6. Installation of trenches using a backhoe.

Fig. 3.7. (a) Placement of Tire Chips into geotextile wrap. (b) Perforated pipe laid into tire chips before being covered by more tire chips and wrapped in geotextile.

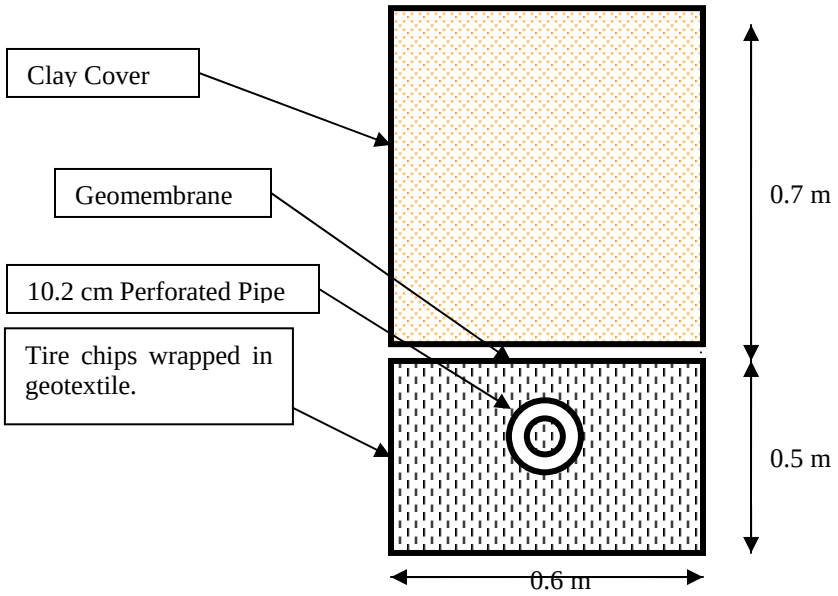


Fig. 3.8. Illustration of horizontal passive gas collection trench (Lengthwise Profile).

(a)

(b)

Fig. 3.9. (a) Biofilters with screen mesh walls to allow for oxygen penetration. (b) Biofilter with vertical vent pipe.

Fig. 3.10. Manifold for distributing landfill gas from the two south wells to each biofilter.

3.3.2 Trench Performance

Two months after installation of the trenches, the geometric mean was measured on two consecutive days, with an average of 163 g/m2/day. This was a return to pre-trench means. Comparisons of pre- and post- trench means (excluding March 23rd data) showed that the means were not significantly different. This return to pre-trench surface emissions can be explained by the fact that the trenches had become clogged with precipitation that had seeped through the soil cover, as was later discovered on June 6th, 2006. This diverted attention away from the surface emissions study and focused the efforts on getting gas to the biofilters in time for the completion of the study. Leachate was evacuated from the trenches by means of a bilge pump lowered into the vent pipes. The leachate was pumped to an area away from the trenches, but within the capture zone of the leachate collection system. It was observed that each time it rained the trenches would become completely clogged, with the exception of the southwest trench, which would sometimes retain partial flow after light rains.

Two attempts were made to improve subsurface drainage of the trenches by increasing holding capacity of the trenches and increasing surface area for the precipitation to percolate through. This was done by adding sumps, roughly 1.2 m wide by 1.8 m long and 1.2 m deep on

each attempt, for a total of two sumps per trench (Fig. 3.11. This increased the volume of the trenches by approximately 5.2 m^3. The sumps were filled with tire chips and then covered with a geotextile before replacement of the clay cover. Clogging continued to occur after each rainfall, indicating that the volume of water draining into the site was substantial and that the hydraulic conductivity of the compacted waste was much lower than expected; possibly due in part to plastic garbage bags. Due to the amount of time involved with pumping the trenches; it was decided that the study would proceed using only the two south trenches, leaving the two north trenches full of water.

Fig. 3.11 (a) Construction of sump where water filled trench has just been uncovered, and (b) sump filled with tire chips prior to being covered with geotextile and clay cover.

Practical Implications

Performances of the biofilters under the conditions encountered at this site are not encouraging; nor are the results of the trench performance. A truly passive horizontal gas collection system is not possible under the climatic conditions encountered in Florida. The extremely low capacity for drainage encountered with the compacted waste requires that an automated sump pump be installed if the biofilters are to receive uninterrupted flow of gas and thus achieve optimal oxidation rates. This defeats the purpose of the passive design however, since automated pumping would require more expense, a constant power source and of course, more monitoring. On site personnel would have to frequently inspect the site to ensure that the leachate is not creating channels that guide it away from the leachate collection area.

Methane emissions from municipal solid waste landfills make up a significant portion of anthropogenic greenhouse gas production. Present methods of mitigating methane emissions are effective on large scale applications but prove to be economically impractical on small scale landfills and old landfills where gas production is too low to create adequate energy for financial returns or to justify the expense of installing an active collection and flaring system. For this reason, alternative mitigation strategies like the one presented here should be investigated in order to provide inexpensive and effective means for mitigating methane emissions.

The site chosen at the Leon County Landfill provided gas flows that proved to be too much for the evaluation of this design, considering its intended application. Estimated production rates for the cover area as determined by the static chamber method were as high as 260 kg/day, or an average flux rate of 1240 kg/hectare/day. This is a very large flux rate compared to other emission surveys at the same landfill, indicating that the waste in this cell was near peak production at the time of this investigation. Though the percentage of surface emissions captured by the trenches ranged from 26% to 49%, it was far more than intended for these biofilters.

The trenches did redirect significant amounts of landfill gas as indicated by the change in surface emissions witnessed shortly after installation as well as by the summation of inflows calculated for the individual biofilters. This performance was undermined by the extremely low conductivity of the compacted waste which soon

caused precipitation that had seeped through the cover to accumulate and clog the trenches. The trenches then had to be pumped following each rain event in order to provide the biofilters with methane.

The changes in flow rates caused by clogging and then clearing of the trenches may have caused the most detriment to the biofilters' performance. During the times that the trenches were clogged with water, the biofilters could not 'feed' on methane and methanotrophic bacteria populations likely decreased. When the trenches were cleared, flow rates were so high that oxygen may have had difficulty penetrating the compost layer to nourish the bacteria. This phenomenon is reflected in the trend towards higher oxidation rates with decreasing flux found in previous studies at the same landfill. Only when the flux is low enough to allow for the presence of both oxygen and methane in the right amounts is oxidation optimal.

The biofilters performance varied in terms of flow performance more than in terms of oxidation rates. The M1 steel biofilters received the highest flow rates throughout the study, followed by the tire chip mixtures, and finally the compost only biofilters. The compost only biofilters ceased to provide fluxes within two months of starting the study. This may have been due to clogging of the material or complete oxidation. Perhaps partial clogging of the material lowered the flux enough to provide for complete oxidation, but there is no way to verify this with the methods chosen for this study. In general, average percent oxidation for the biofilters ranged from 3.4% to 10.6%. The mass oxidation rates were highest in the M1 and CT biofilters with rates ranging from 12.9 to 49.4 g/m2/d. The compost only biofilters averaged 8.2 g/m2/d.

The lowest flux encountered during this study yielded a percent oxidation of 64.3% in the M1-A biofilter, indicating that the trend towards higher oxidation rates with lower influxes applies to this application. If this is so, future testing could benefit from controlling flow rates with a control valve or by means of designing shorter trenches to decrease the zone of influence. In any case, these biofilters should only be applied to mitigating much lower methane production rates than those found at this site.

3.4 FLORIDA PRESPECTIVES

Bifilters to Remove Hydrogen Sulfide

3.4.1 Study Description

Hydrogen Sulfide, H2S, is the source of most odors in landfill gas, LFG. The typical concentration of H2S in LFG from municipal solid waste is 35 ppm. It smells offensive at low concentrations of 0.1 - 1 ppm. Odor is one reason for landfills to manage H2S concentrations of their LFG. (SWANA, 1997). LFG from construction and demolition landfills can have H2S concentrations as great as 12,000 ppm (Lee, et al. 2006). Landfill cover materials have shown potential to attenuate high concentrations of H2S, greater than 50,000 ppm, in laboratory scale simulated columns. (Plaza et al., 2006). This study characterized the capacity of different material to attenuate H2S from LFG generated from a municipal solid waste landfill. The reduction of H2S was measured in bucket filters attached to a vertical passive LFG vent. The vent emitted a steady 40 liters per minute flow of LFG. The concentration of H2S ranged from 20 to 40 ppm.

The fieldwork was conducted at U.S. South 27 Landfill, 8 miles east of Tallahassee, Florida. The landfill opened in 1977 and contains 1.68 million tons of municipal solid waste, MSW, and construction and demolition, C&D waste. Disposal of MSW ended in 2006, disposal C&D waste is planned until 2015. At the time of this study, landfill gas was vented through a passive gas collection system; LFG generation estimates were over 20,000 liters per minute. A vacuumed controlled LFG collection system was installed in 2007.

To quantify the reduction of H2S, the filter would need to receive a constant flow of LFG. One passive vertical vent was selected, as it maintained a constant flow of 40 liters per minute. The landfill vent was fitted with couplers and reducers and connected to a manifold where the LFG flow rate could be divided between the different filters. Although the passive landfill vent was subject to flow variations from atmospheric and internal landfill conditions, the LFG flowrate was continuously controlled into the filter with Dwyer Visi-Float Flowmeter. The LFG had H2S concentration of 20-40 ppm.

Filters were constructed from 5-gallon buckets, Figure 3.11. Gas entered the bucket through a grommet inlet drilled into the lower sidewall of the bucket. Gas exited

the filter through a similar grommet inlet drilled through the lid of the bucket. Filter seals were verified by flow balance in and out of the bucket.

Fig. 3.11. Photo of bucket filter.

Four different media were used in the bucket filter. The first was M1 steel, a waste by-product from tire shredding recycling operations. The shredded steel was rusted with an acid solution to form a Ferrous Oxide (rust). The second media was 'red' soil found at the landfill. The soil is classified as Orangeburg Series, a fine-loamy, kaolin tic, themic Typic Kandiudulat by the National Resources Conservation Service of the US Department of Agriculture. The third media was wood mulch; a material produced from a grinding operation at the Leon County Landfill. The forth media was soil compost, a material that was produced from the composting of shredded mulch from the composting facility from the site.

Sampling occurred at the filters attached to a LFG well, Figure 2. Filter media was exposed to LFG for up to 120 days. Sample frequency was 3 to 4 days per week. LFG was collected in Tedlar gas sample bags. Two samples of unfiltered LFG were collected for each sampling event. The flow rate to each filter was measured and controlled with Dwyer Visi-Float Flowmeter. The filter either was closed or open. For sampling, opened filters were closed, sample bags were filled directly from filter lid outlet. Flow rates were recorded at the time of each sample.

Samples were labeled and carried to laboratory in an ice chest. Laboratory work was conducted within 2 hours of data collection. Samples were injected into a Shimadzu, GC-8A with an Alltech Carbograph 1 SC Column and a flame photometric detector to determine sulfur content. A deccident water filter was used to prevent moisture from the LFG to be injected into the GC. Gas standards of H2S, methylmercaptan, and Carbonyl were used. Of the three sulfur compounds analyzed, only H2S was found.

Fig. 3.12. Photo of Filters at LFG well.

Two applications for the filters were considered. The first, a filter for a LFG well. Shredded steel from tire recycling process was used in the filter to reduce sulfur. H2S reacts with iron oxide (Fe2O3) to form iron sulfide and water. Landfill gas can easily flow through the wool like texture of the shredded steel with formed iron oxide.

$2Fe_2O_3 + 6H_2S \rightarrow 2Fe_2S_3 + 6H_2O$

The filters were connected to the LFG vent with a closed lid. The amount of sulfur deposited in the filter was calculated until the filters no longer reduced H2S by 100 percent. After which, the regenative ability of the media was simulated by opening the bucket lids. The presence of oxygen has potential to effect the removal of H2S from LFG in two ways; the (1) Iron Sulfide coating of the steel media can be regenerated back to iron oxide and (2) the presence of oxygen itself can oxidize H2S to elemental sulfur or sulfur dioxide. It is assumed the sulfur delivered to the filter would remain in the filter.

Disposal of the sulfur collected from filters on a larger scale should consider the effect of further H2S generation in the landfill.

$$2Fe2S3 + 3O2 \rightarrow 2Fe2O3 + 6S$$

$$2H2S + 02 \rightarrow 2Fe2O2 + 6S$$

The second filter application, a H2S reducing layer of a landfill cover; a cover that could reduce H2S through a thin layer of iron rich soil. Two inches of red soil were placed in the filters and the filter lids remained closed. The amount sulfur that remained in the bucket filter could be calculated. Additional material was also tested, compost and wood mulch.

3.4.2 Results

The capacity of the media to reduce H2S was established by determining the total amount of sulfur retained in each filter at 100 percent sulfur reduction. Flow rates, and sulfur concentrations were used to quantify the amount of sulfur delivered to the filter, retained in the filter, and released from the filter. In the discussion, the amount of sulfur attenuated and the mass of filter media are used to project media life and establish a range of performance possibilities. Two applications are considered; vent filters that emit less than 6 liters per minute and landfill covers that emit less than 100 liters per day per square meter.

The total amount of sulfur retained in the filter was calculated given the LFG flow rate and the H2S concentration of the LFG before and after the filter. The H2S reducing capacity of the filter is the amount of sulfur retained in the closed filter until the filter fails to perform at 100 percent reduction, as summarized in Table 3.3.

Table 3.3. Cumulative Sulfur retained in M1 Steel Closed Filter at 100 percent H2S Reduction

LFG Flow into filter and H2S concentration	Mass of Media, Kilograms	Total Sulfur retained in filter at 100% H2S reduction	Time in Filter at 100% H2S reduction, Days
1 liter per minute and 20 to 40 ppm of H2S	3.7	5.85	112
3 to 6 liters per minute and 20 to 40 ppm of H2S	3.65	2.90	15
3 to 6 liters per minute and 20 to 40 ppm of H2S	3.60	4.48	35
1 to 6 liters per minute and 20 to 40 ppm of H2S	1.85	0.0	0

The closed M1 filters were opened after they performed below 50 percent H2S reduction. Once the filters were opened, 100 percent H2S reduction occurred for the remainder of the sample period. The total amount of sulfur delivered and retained in the filter for the duration of the closed and opened filter is summarized in Table 3.4.

Table 3.4. Cumulative Sulfur Retained in M1 Steel Filter for Both Opened and Closed Conditions

LFG Flow into filter and H2S concentration	Mass of Media, Kilograms	Total Sulfur delivered to filter, Grams	Total Sulfur retained in filter, Grams	TotalTime in Filter, Days
3 to 6 liters per minute and 20 to 40 ppm of H2S	3.60	21.66	17.41	112
1 to 6 liters per minute and 20 to 40 ppm of H2S	1.85	21.47	17.98	108

3.4.3 Vent Filters, Discussion and Projections

For the M1 Closed filters, the life expectancy performance is the total amount of sulfur that is delivered to the filter at 100 percent H2S reduction. To provide a sense of design possibilities, the life of the filter was projected by dividing the total sulfur attenuated by a mass of M1 Steel at 100 percent H2S reduction by the rate of sulfur delivered to the filter from a range LFG flow rates and H2S concentrations. The Closed M1 Filter performed best at low, 1 liter per minute or less LFG flowrate, in 112 days, 3.7 kilograms of shredded M1 Steel attenuated 5.8 grams of sulfur at 100 percent sulfur reduction. The life expectancy of a filter filled with 3.7 kilograms of M1 filter at a range of 0.1 to 1 liter per minute LFG and H2S concentrations from 20 to 40 ppm is shown in Table 3.5. The performance life ranges from 24 months for a LFG flow rate of 0.1 liter per minute with an H2S concentration of 20 ppm to 2 months for a LFG flow rate of 1 liter per minute with an H2S concentration of 40 ppm.

Table 3.5: Life Expectancy of 3.7 kilograms of M1 Steel in Closed Filter for a range of LFG Flow Rates and H2S Concentrations

LFG Flow rate, Liters per Minute	**Concentration of H2S in LFG, ppm**	**Mass of Sulfur delivered to Filter, Grams per Day**	**Life of Filter to Perform at 100 percent Reduction of H2S, Months**
0.2	20	0.01	24.4
0.4	20	0.02	12.2
0.6	20	0.02	8.1
1.0	20	0.04	4.9
0.2	40	0.03	12.2
0.4	40	0.05	6.1
0.6	40	0.05	4.1
1.0	40	0.08	2.4

The performance life of the Opened Filters was calculated similarly to the Closed Filters, Table 3.6. Since the Opened bucket performed at 100 reduction once the lid was opened, until the end of the sample period, the total amount of sulfur delivered to the filter includes both open and closed conditions. Results indicate that an opened filter containing 3.7 kilograms of rusted M1 Steel could attenuate 43 grams of sulfur. This is based on the doubling performance of the filter containing 1.85 kilogram or rusted M1 steel; it attenuated 21.5 grams of sulfur in 58 days. The performance life ranges from 36 months for a LFG flow rate of 1 liter per minute with an H2S concentration of 20 ppm to 3 months for a LFG flow rate of 6 liters per minute with an H2S concentration of 40 ppm.

Table 3.6. Life Expectancy of 3.7 kilograms of M1 Steel to Attenuate 43 grams of Sulfur in Opened Filter for a range of LFG Flow Rates and H2S Concentrations

LFG Flow rate, Liters per Minute	**Concentration of H2S in LFG, ppm**	**Mass of Sulfur delivered to Filter, Grams per Day**	**Life of Filter to Perform at 100 percent Reduction of H2S, months**
1	20	0.04	35.9
2	20	0.06	17.9
4	20	0.16	9.0
6	20	0.24	6.0
1	40	0.08	17.9
2	40	0.16	9.0
4	40	0.33	4.5
6	40	0.49	3.0

3.4.4 CONCLUSION

The purpose of this field study was to quantify the sulfur reducing capacity of low-cost the filters made from recycled material. The performance of the filters indicates

a potential to eliminate H2S from municipal solid waste from low flow LFG vents and LFG emissions through landfill covers.

CHAPTER IV

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