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Atmospheric Dispersion Modeling to Inform A Landfill Methane Emissions Measurement Method

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Atmospheric Dispersion Modeling to Inform A Landfill Methane Emissions Measurement Method

by

Diane Margaret Taylor

A dissertation submitted in partial satisfaction of the requirements for the degree of Doctor of Philosophy in Engineering - Civil and Environmental Engineering in the Graduate Division of the University of California, Berkeley

Committee in charge:
Professor Fotini Chow, Chair
Professor Dennis Baldocchi
Professor Evan Variano

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Atmospheric Dispersion Modeling to Inform A Landfill Methane Emissions Measurement Method

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Diane Margaret Taylor
Abstract

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Landfills are known to be a significant contributor to atmospheric methane, yet emissions are difficult to quantify because they are heterogeneous over a large area (up to 1 km$^2$) as well as unsteady in time. Many different measurement methods have been developed, each with limitations and errors due to various factors. The most important difference among different measurement methods is the size of the measurement footprint. Flux chambers have the smallest footprint, typically 1 m$^2$, radial plume mapping mass balance and eddy covariance can have footprints between 100 and 10,000 m$^2$, and aircraft-based mass balance and the tracer dilution method can measure whole landfill emissions. Whole landfill measurement techniques are considered the best because they account for spatial heterogeneity, and the tracer dilution method (TDM) in particular has gained popularity because it is relatively noninvasive and cost-effective.

The TDM works by comparing ratios of methane and tracer gas plumes downwind of the landfill. A tracer gas such as acetylene is emitted from the landfill at a known, steady rate. Downwind plume transects are collected using a gas analyzer on a moving vehicle to obtain both methane and tracer gas concentrations. The idea behind the method is that at the transect measurement location, the gas plumes are well mixed enough that the ratio between the methane and tracer gas concentrations is approximately equal to the ratio between the methane and tracer gas emissions rates. Methane emissions are calculated by equating the ratio of concentrations to the ratio of emissions rates and solving for methane emissions. Field studies of the TDM with controlled methane releases over a flat field have quantified TDM-related measurement errors, but these errors are impossible to assess in real landfill measurements because the true landfill emissions are unknown.

Numerical modeling of the TDM is an advantageous way to study the errors in TDM-measured emissions as well as how the error changes under a variety of different conditions. With a numerical model, the true methane emissions are prescribed, so they can be compared to the TDM-measured emissions to evaluate the method’s accuracy. The TDM is examined in this dissertation using numerical simulations with the Weather Research and Forecasting
model (WRF). WRF is a mesoscale numerical weather prediction model with large-eddy simulation (LES) capabilities, allowing for high-resolution simulations with resolved large-scale turbulent motions. To examine the TDM, two real landfills in the U.S. are selected, with high resolution topography data and real atmospheric data informing the initial and boundary conditions of the model. The simulations are run with a nested grid configuration, starting from 2.25 km resolution and nesting to 150 m resolution and then 30 m horizontal grid resolution over the landfill area, with the lowest vertical level \( \sim 15 \) m. To simulate the TDM, three components are needed: tracer emissions with a specified configuration and emissions rate, landfill emissions specified at every grid point on the surface of the landfill, and simulated transect measurements with a specified transect path and transect collection speed. In this dissertation, tracer emissions are prescribed as steady values at grid points on the surface of the landfill constituting various tracer configurations to be examined, and methane emissions are either prescribed as steady values or calculated using prescribed soil methane concentrations and a surface flux parameterization.

To our knowledge, the work presented in this dissertation constitutes the first time WRF simulations have been used to examine the TDM. In the first part of this dissertation, steady landfill methane emissions are prescribed to study the effects of various aspects of the TDM setup and various external factors on the accuracy of the TDM-measured emissions. Factors tested include tracer location relative to the methane emissions hot spot of the landfill, distance from the landfill to the transect path, transect angle relative to the wind direction, and transect speed. Tracer location relative to the emissions hot spot is found to have the most significant effect on TDM accuracy, while transect angle relative to wind direction and transect vehicle speed are found to have negligible effects. The roles of wind direction and topography are also examined and found to have significant effects of the TDM's accuracy.

In the second part of the dissertation, the same landfill area is simulated, and a surface flux parameterization is added to WRF to introduce wind-dependent variability to the landfill emissions. Significant standard deviations were seen in the TDM-measured emissions in the previous chapter despite the prescribed landfill emissions remaining constant, and when TDM-measured emissions for steady and unsteady emissions simulations are compared, the variability in the TDM-measured emissions is found to be essentially the same even though the variability in true emissions is significantly different, pointing to possible errors inherent in the TDM's ability to capture true emissions short-term variability. TDM-measured emissions standard deviation and TDM error are compared for eight different time periods over two different days to see whether different times of day and different atmospheric conditions affect the TDM. The smallest measured standard deviations and smallest errors are seen at night on both days, and measured standard deviation increases over the course of the day for both days, with the largest standard deviations seen in late afternoon shortly before sunset. TDM percent error does not exhibit a noticeable diurnal trend. Two different tracer configurations are used for the TDM simulations to obtain a range of standard deviations and percent errors for an optimal and less ideal tracer placement.

In the last part of the dissertation, a different landfill area is simulated and emphasis is placed on extensive comparison to field measurements. Four different days from different
seasons are simulated to examine the seasonal and diurnal effects of wind-dependent variability on emissions. These simulations aim to help inform how limited measurement data can be used to extrapolate annual landfill methane emissions.
To my parents
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Chapter 1

Introduction

1.1 Motivation and background

Quantifying landfill methane emissions and understanding the errors in estimated emissions are both complex problems. Modeling and field measurements are both commonly used to estimate annual landfill methane emissions. Most modeling approaches for annual landfill emissions are overly simplified and rely on tunable parameters that vary between landfills. Field measurement methods have their own limitations due to the heterogeneity and unsteadiness of landfill methane emissions, cost and labor required for the field work, the need for ideal wind conditions. Error estimates for the predicted emissions are also difficult to obtain. This work aims to address some of these issues by using three-dimensional, high-resolution atmospheric dispersion model to simulate a particular landfill methane emissions measurement technique, the tracer dilution method (TDM). This chapter presents motivation for this work, followed by an overview of the results of this dissertation research.

Landfills

Landfills are estimated to be the third largest contributor to anthropogenic methane emissions in the U.S., after natural gas leaks and enteric fermentation (U.S. EPA, 2015). Methane is considered one of the most important greenhouse gases, with a 100-year Global Warming Potential of 25: over a 100-year period, the amount of heat trapped by a mass of methane is 25 times the amount of heat trapped by the same mass of carbon dioxide (U.S. EPA, 2015). Since landfills are such an important contributor to emissions of this potent greenhouse gas, it is essential to understand the basics of landfill design, landfill methane production and emissions, and what is currently happening at landfills to reduce emissions.

Landfills are divided into sections which are categorized by three different cover types: daily, intermediate, and final. Daily cover is the portion of the landfill that has new waste added daily. At the end of each day, the waste is compacted with heavy machinery and then covered with 15 to 30 cm of soil. This daily cover soil reduces the odor of the landfill and prevents waste debris from being scattered by wind or animals. Sometimes at the
beginning of the next day, this cover soil is scraped off to save space before new waste is added. Intermediate cover is any portion of the landfill which has received waste in the past, and has not yet reached full capacity, but will not be receiving any waste for at least 180 days. This cover soil is typically at least 30 cm thick. Final cover is an area that has permanently stopped receiving new waste because it has reached full capacity. A final cover generally has several layers, including a plastic cap to help prevent methane emissions and a surface layer of top soil and vegetation to prevent erosion. The amount of methane produced by a landfill steadily increases over the life of the landfill. It begins to decrease once the landfill is no longer operational, though the amount of methane emitted continues to be significant for decades after landfill closure. Because of the continued emissions, landfill aftercare procedures must be maintained for at least 30 years after closure (Huber-Humer et al., 2008). Figure 1.1 illustrates the differences between the different cover types.

\begin{figure}[h]
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\includegraphics[width=\textwidth]{landfill_cover_types.png}
\caption{Sketches of the three different landfill cover types.}
\end{figure}

Understanding the mechanisms governing methane production in landfills is necessary to mitigate their environmental impacts. Methane is created inside landfills by methanogenic archaea. There are several important factors that affect landfill methane production: waste composition, oxygen concentration, moisture content, temperature, and age of waste (Williams et al., 2001). Waste composition is important because the more organic waste present in the landfill, the more methane is produced by microbial decomposition. Some types of organic
waste contain certain nutrients such as calcium and potassium which help microbes thrive, increasing methane production. Oxygen concentration is important because methane can only be produced once oxygen has been depleted. If waste is loosely buried or frequently disturbed, more oxygen is available, so aerobic microbes persist for longer producing carbon dioxide and water. If the waste is highly compacted, methane production will begin earlier. Moisture inside the landfill can increase methane production because moisture encourages microbial growth and transports nutrients and microbes to other areas within the landfill. Warm temperatures also increase microbial activity, thus increasing the rate of landfill gas production. Weather changes can affect gas production in shallow landfills, but deep landfills usually maintain a fairly stable interior temperature because microbial activity releases heat. Lastly, the age of the waste is important because younger waste produces more methane than older waste. Methane production usually begins within 1 to 3 years of the waste being added, with peak production at 5 to 7 years after the waste has been added. Almost all gas production occurs within 20 years of the waste being added, but small quantities may continue to leak out of the landfill for 50 years or more.

Landfill gas undergoes four phases of development once waste is added. During the first phase of decomposition, aerobic bacteria deplete the oxygen inside the landfill on the order of a few days. In the second phase, anaerobic microbes create acids that cause compounds inside the landfill to dissolve. This phase takes several months, and the importance of dissolving these compounds is to make more nitrogen and phosphorous available for bacteria to consume, therefore allowing a more diverse population of bacteria to thrive. During the third phase, the acids are consumed by certain anaerobic microbes, creating an environment inside the landfill that is more hospitable to methanogenic archaea. This phase takes between half a year and three years. The final stage begins when the composition of landfill gas becomes constant with time, approximately 50% methane and 50% carbon dioxide. This mixture, referred to as landfill gas, is what landfills emit into the atmosphere.

When landfill gas is produced inside the landfill, it expands to fill the available space, so the gas moves through the pore spaces within the waste and soil covering the landfill. The natural tendency of methane, which is lighter than air, is to move upward, through the landfill surface and into the atmosphere. Upward movement of methane can be inhibited by densely compacted waste or landfill cover material (e.g. plastic caps), so the gas follows the path of least resistance. The three main factors that influence gas transport within a landfill are diffusion, pressure, and permeability (Williams et al., 2001). Diffusion is the natural tendency of gas to achieve uniform concentration within a given space, so the methane flows from regions of higher concentration to regions of lower concentration. Since the concentration of methane within the landfill is usually much higher than the concentration of methane in the atmosphere, methane diffuses out of the landfill through the soil pores.

Pressure can also affect methane transport within the landfill. Gas accumulated in a landfill can create areas of high pressure in which gas movement is restricted by compacted waste or impermeable cover material (Williams et al., 2001). Variations in pressure throughout the landfill result in gas moving from areas of high pressure to areas of low pressure. As more gas is generated, the pressure within the landfill increases, causing subsurface pressure
Landfill emissions measurements

Landfill emissions are difficult to measure because there are large uncertainties in several aspects, from estimates of the type and amount of waste material to the role of methane oxidizing microbes (Hovde et al., 1995). Even when the landfill is a strong source of methane emissions, flux measurements can be difficult because the concentration in the atmosphere may not be much higher than the background concentration of methane (~1.8 ppm). Several strategies are used to measure or estimate landfill methane emissions, using both field instrumentation and models. The most common methods, discussed in this section, are flux chambers, eddy covariance, the tracer dilution method (TDM), and the Intergovernmental Panel on Climate Change (IPCC) first order decay model.

Flux chambers are one of the oldest techniques used to measure methane emission rates (Hovde et al., 1995). A flux chamber is a small box placed on the ground surface that can measure methane concentrations within the chamber. Flux chambers can be static or dynamic. With a static chamber, the top of the chamber is closed, the chamber fills with gas being emitted from the ground, and the amount of gas accumulated over a certain time period is used to calculate the ground flux. With a dynamic chamber, clean air is added to the chamber through an inlet at a fixed rate, there is an outlet in the chamber for the gas of interest to exit, after some time the concentration within the chamber reaches a steady state, and the ground flux can be calculated using the flow rate of clean air and the concentration of the gas of interest at the outlet during steady-state. Flux chambers have advantages which include low cost, high sensitivity, and easy implementation (Hovde et al.,
1995). Their size is an extremely limiting factor in measuring landfill emissions because a single flux chamber has a footprint of \( \leq 1 \text{ m}^2 \) while a landfill is often on the order of \( \sim 1 \text{ km}^2 \). Landfill emissions are known to have significant heterogeneity, which cannot be accurately accounted for by limited measurements from flux chambers (Abichou et al., 2006; Hovde et al., 1995). Additionally, flux chambers may introduce measurement bias because it is a relatively invasive measurement technique (Moore and Roulet, 1991). Börjesson and Svensson (1997) used static chambers to measure landfill methane emissions at a Swedish landfill and found both seasonal and diurnal patterns in the emissions, which they found to have a negative correlation with soil temperature, a regulating factor of methane oxidation.

Eddy covariance is a micrometeorological method which provides continuous, direct measurements of unperturbed methane fluxes. Eddy covariance rapidly measures both instantaneous methane concentration and vertical wind velocity, using fluctuations from the mean to determine the vertical turbulent flux. Eddy covariance measures the flux from a footprint which will change in size depending on the meteorological conditions, with wind direction and atmospheric stability playing critical roles in the footprint size and location. Hovde et al. (1995) used eddy covariance to measure methane emissions from a landfill near Oak Ridge, Tennessee, USA, and found that over seven 30-minute measurement periods over one day, they found fluxes that were 20 to 200 times greater than fluxes observed from natural ecosystems such as northern peatland and subarctic tundra. They hypothesized that their measurements might underestimate the emissions by up to 15% due to the measurements being taken at 2.5 m above the ground.

The tracer dilution method (TDM) uses a tracer gas emitting at a known rate collocated with the emissions source of interest. The concept of using a tracer gas to estimate emissions from an inaccessible ground level source was first presented by Ludwig et al., who used Gaussian plume theory to develop rules of thumb for where to take measurements when using the method (1983). Use of the TDM to quantify landfill methane emissions has become increasingly popular in the past decade as it replaces more expensive and invasive methods such as flux towers and chamber measurements (Babilotte et al., 2010). For the TDM, one or more cylinders containing a tracer gas are placed on the surface of the landfill emitting the gas at a known rate. A gas analyzer located downwind of the landfill records concentrations of methane and tracer gas. A schematic of the TDM setup is shown in Figure 1.2. This figure refers specifically to the mobile TDM, in which a gas analyzer is placed in a vehicle and driven back and forth across the width of the plume along a road downwind of the landfill. The mobile TDM is the focus of the work in this dissertation, and further details about how it is used in this work are found in Section 1.2.

The IPCC first order decay model comes from the differential equation

\[
\frac{d}{dt}(DOC) = -k(DOC) \tag{1.1}
\]

where \( DOC \) is the mass of degradable organic carbon in the landfill at time \( t \) (which is an integer year since the landfill’s inception) and \( k \) is the decay rate constant (yr\(^{-1}\)) (Pipatti...
This differential equation yields the solution

$$(DOC) = (DOC)_0 e^{-kt}$$

where $(DOC)_0$ is the mass of DOC in the landfill at $t = 0$. The expression in Equation 1.2 describes how much degradable organic carbon is remaining in the landfill at time $t$. The quantity of interest, however, is the amount of DOC that has decomposed over one year, because $k$ has units of yr$^{-1}$. This quantity is described by

$$(DOC)_d = (DOC)_0 [e^{-k(t-1)} - e^{-kt}]$$

The amount of DOC that has decomposed in year $t$ can be related to the amount of methane generated by that decomposed matter:

$$G_{CH4} = (DOC)_d F(16/12)$$

where $F$ is the fraction of methane in the generated landfill gas (default value is 0.5) and (16/12) is the molecular weight ratio of methane to carbon. Finally, the amount of generated methane can be used to calculate the landfill methane emissions.

$$Q_{CH4} = [G_{CH4} - R_t][1 - (OX)_t]$$

In this equation, $Q_{CH4}$ is the amount of methane emitted in year $t$ over the whole year, $R_t$ is the amount of methane recovered over the whole year $t$, and $(OX)_t$ is the oxidation
fraction during year $t$. This model gives a crude back-of-the-envelope calculation for annual landfill methane emissions that depends heavily on factors that are known to be complex and variable such as methane oxidation rate. This IPCC first order decay model is more accurate than its predecessor, which did not take age of waste into account, and it is easy to use because it can be calculated quickly with a spreadsheet. This model, however, does not take into account many factors that are known to control emissions such as the thickness and soil type of landfill cover soils, and the effects that local climate has on methane oxidation due to variations in soil moisture and soil temperature.

**Issues with current measurement methods**

The impetus for this work comes from the need for a reliably accurate method to estimate total annual landfill methane emissions. To address this, we must first understand how annual landfill emissions are currently estimated and why the need remains for a more accurate way to estimate annual emissions. Currently, total annual emissions are usually estimated with a simple model rather than field measurements due to technical and economic limitations (Spokas et al., 2015; Figueroa, et al., 2009; De La Cruz, et al., 2016). The Intergovernmental Panel on Climate Change (IPCC) uses a first order decay (FOD) model, which estimates methane generation based on a few parameters such as the age of waste, quantity of waste, degradable organic carbon, and a decay rate constant with default values set by the IPCC. Once the methane generation is estimated, it is assumed that 10% of the methane is oxidized, so methane emissions is calculated as simply (methane generation) × 0.9 (IPCC, 2006). This 10% oxidation figure has been recently contested, however, and in reality has been found to fluctuate based on soil temperature, soil moisture, and soil oxygen concentration (Chanton et al., 2011, Spokas et al., 2011).

De La Cruz et al. (2016) compared IPCC modeled annual emissions to TDM-estimated annual emissions for a young landfill over the course of 4 years and found that the IPCC model’s predictions exceeded the TDM’s predictions by up to 31 times. This is clearly a large discrepancy worth addressing, and De La Cruz et al. evaluated both the possibilities of errors in the TDM and the model parameterization. The TDM measurements were compared to eddy covariance data of the same landfill emissions, which showed an average methane flux within 2% of the TDM-measured emissions. The IPCC model was run with the parameters changed from average values reported in the literature to extreme values, and the model emissions then matched the TDM-measured emissions much better. Both pieces of evidence point to the model parameters being significantly incorrect for this landfill, as De La Cruz et al. (2016) concluded.

The established modeling method for annual landfill methane emissions is very simple and lacks site specificity as well as important factors such as heterogeneity and meteorological effects, so a TDM estimation method seems like a promising replacement (Foster-Wittig et al., 2015; Mønster et al., 2015). Many uncertainties shroud the feasibility of such a method, however, due to limited field testing and the inability to truly quantify the method’s error.
in the field. This work aims to address these uncertainties and shed light on how the TDM can be used to estimate annual landfill emissions.

**Atmospheric dispersion modeling with WRF**

The Weather Research and Forecasting Model (WRF) is chosen for the work in this dissertation because it is a commonly used, state-of-the-art model that includes sophisticated turbulence models and is coupled to a land surface model (LSM) which can incorporate a methane emissions parameterization. WRF is a mesoscale numerical weather prediction model used for atmospheric research and operational forecasts. WRF can be used for a very wide range of grid resolutions, and this work uses a resolution as fine as 30 m with the large-eddy simulation (LES) approach. This approach to atmospheric turbulence modeling explicitly resolves larger turbulent eddies, and the subfilter-scale eddies are parameterized with a turbulence closure scheme. The simulations presented in this work use LES with the turbulent kinetic energy nonlinear backscatter and anisotropy (TKE-NBA) model for the subfilter-scale stress (Mirocha et al. 2010). WRF provides many options for parameterizing turbulence, and LES with TKE-NBA was chosen for this work because it provided wind speed and wind direction output that best agreed with observation data.

The focus of the work contained in this dissertation is the atmospheric dispersion modeling, which allows us to simulate the TDM. The process for simulating the TDM is outlined in Section 1.2. The novelty of this work lies in the fact that the landfill emissions are known in the model and can therefore be compared to the TDM-measured emissions with the simulated TDM using the WRF output. Details about simulating the TDM with WRF output are provided in Section 1.2.

**Landfill emissions modeling with CALMIM**

While the focus of this work is on the atmospheric dispersion modeling to understand the tracer dilution method, this work also contains a landfill methane emissions model, used in Chapters 4 and 5. The purpose of this landfill emissions model is to simulate wind-dependent emissions with significant short-term variability to understand how wind-dependent emissions affect the tracer dilution method. The variable emissions will affect the gas plumes that are sampled downwind. This work does not focus on the accuracy of modeling the landfill methane emissions. In Chapter 3, the simulations use a prescribed, steady emissions rate, and no landfill methane emissions model is used in this chapter. In Chapters 4 and 5, landfill methane emissions are calculated at each time step with a surface flux parameterization based on how WRF calculates heat and moisture surface fluxes. This landfill methane emissions parameterization uses Equation 1.6.

\[
Q_{CH4} = C_d u^* (c_{soil} - c_{atm})
\]

where \( C_d \) is a time-dependent drag coefficient, \( u^* \) is the shear velocity, \( c_{soil} \) is the soil methane concentration, and \( c_{atm} \) is the atmospheric methane concentration. This equation is discussed
extensively in Chapter 2. In the WRF model, $c_{atm}$ is in the middle of the lowest grid cell, between 7 and 10 m above the ground in our cases. Additionally, $c_{soil}$ is prescribed as a time-dependent model input, with the values applied at 5 cm depth, the same depth WRF uses for heat and moisture fluxes.

The values for $c_{soil}$ come from the California Methane Inventory Model (CALMIM). CALMIM was developed for use by California landfills because at the time the model was developed, California had special regulations on landfills that other states did not have. The model, however, can take into account meteorological data for any latitude and longitude on the planet, so it is used in this work for simulations of Sandtown Landfill in Delaware, USA, and a landfill in the Southeastern US.

CALMIM is a 1-D column model which calculates whole landfill methane emissions by accounting for transport within the soil of heat, moisture, oxygen, and methane. CALMIM is site-specific, and the user-specified inputs include: latitude and longitude, acreage of the landfill, cover type (multiple covers can be selected for the same landfill), percent of the landfill occupied by that specific cover, cover soil type, cover soil thickness, percent of organic matter in the waste, percent of the cover soil occupied by vegetation, and percent of gas recovered if gas collection system is present. CALMIM uses historical meteorological data averaged over several decades for the specified latitude and longitude to predict parameters such as solar radiation, air temperature, and precipitation events.

When the depth of the cover soil is specified, CALMIM divides the soil column into a model domain with 1-inch resolution. The model time step is 10 minutes, and the model writes output to an output file every hour. Methane transport is modeled according to the 1-D soil gas transport algorithm outlined in Campbell (1985). Methane concentration at the bottom soil node, which is defined to be at the soil-waste interface, is prescribed as a constant boundary condition. The methane concentration prescribed at this boundary condition has different default values that depend on the cover type, and the default values can be changed by the user if field data values are known. Methane diffusivity is calculated as an empirical function of soil moisture and soil temperature, and methane oxidation is a sink for the methane, based on the oxygen concentration. Surface methane emissions in CALMIM are calculated using the difference $(c_{soil} - c_{atm})$ times a static boundary layer conductance (where $c_{soil}$ is located at the top soil node, 1 inch below the surface). The boundary layer conductance has a default value of 0.01 m/s.

CALMIM is used in this dissertation for one main purpose: to supply realistic soil methane concentration values to the unsteady emissions parameterization, which is used for the landfill methane emissions in Chapters 4 and 5. The CALMIM soil methane concentration values used in Equation 1.1 are taken from the second soil node, at 2 inches deep, because 2 inches $\approx$ 5 cm. 5 cm is the depth in WRF where the soil values are defined for WRF to calculate the surface fluxes of soil moisture and heat.

The processes accounted for in the unsteady emissions parameterization used in Chapters 4 and 5 are summarized in the schematic in Figure 1.3.
Figure 1.3: Cartoon of the processes accounted for in the unsteady emissions parameterization used in Chapters 4 and 5.

1.2 Overview

Simulating the tracer dilution method

Chapters 3, 4, and 5 all use WRF to simulate the TDM. The process of simulating the TDM is outlined here for clarity. Simulating the TDM is in essence a replication of the field measurement technique, which relies on Equation 1.7 to calculate the landfill emissions based on the downwind concentration measurements of methane and tracer gas.

\[ Q_{CH4} = Q_{tr} \int \frac{c_{CH4}(x)dx}{c_{tr}(x)dx} \]

In this equation, \( Q_{tr} \) is the known tracer emissions rate, \( c_{CH4} \) is the measured methane concentration, \( c_{tr} \) is the measured tracer concentration, and \( x \) is the location in space along the transect path where the concentration is recorded.

1. Landfill emissions are prescribed (or calculated) at each grid cell on the surface of the landfill, and these prescribed emissions rates feed a certain amount of mass into the
atmospheric grid cells above the surface, and this mass is then transported by the advection-diffusion equation.

2. Tracer emissions rates are prescribed for point sources, which have various placements on the surface of the landfill for different simulations, and these tracer emissions rates match the tracer emissions rates used in the field, which were steady in time. These emissions rates also inject mass into the atmospheric grid cells above the surface, and this mass is transported by the advection-diffusion equation as a separate plume from the methane plume.

3. With these two separate gas plumes resulting from the emissions of landfill methane and tracer gas, downwind concentrations are collected as if there were a vehicle with a gas analyzer moving along a path downwind of the landfill roughly perpendicular to the gas plumes: after one concentration point is collected from the model output, the simulated vehicle moves along the transect path in both space (16 m) and time (2 seconds) so that the simulated vehicle is traveling at 8 m/s. In this way, transect measurements are collected that span the width of the plume in 2 min, and then the vehicle "turns around and collects a new transect along the same path, and so on until transect measurements have been collected over the entire simulation period.

4. Once these transect measurements are collected (all numerical simulations, no field data involved in this process), Equation 1.2 is used to integrate the concentrations along the transect path to calculate a modeled TDM-measured emissions rate.

These are the steps to obtain the modeled TDM-measured emissions to compare to the modeled true landfill emissions. Comparing the modeled TDM-measured emissions to the modeled true landfill emissions allows us to estimate the error of the TDM-measured emissions under various circumstances. Error of the TDM measurements is assessed in each chapter with Equation 1.8.

\[ P_{\text{error}} = \frac{Q_{\text{TDM}} - Q_{\text{model}}}{Q_{\text{model}}} \times 100 \] (1.8)

In Equation 1.3, \( Q_{\text{TDM}} \) is the average of the emissions values measured by the tracer dilution method and \( Q_{\text{model}} \) is the average of the emissions values calculated in WRF. Being able to quantify the errors incurred by this measurement method under different circumstances due to the use of known model landfill emissions is a major contribution of this work, the aim of which is to help improve the accuracy of the TDM in the field.

Summaries of each chapter

We begin in Chapter 2 with an in-depth discussion of the current state of research regarding atmospheric dispersion and scalar fluxes to thoroughly understand how we expect
CHAPTER 1. INTRODUCTION

the methane to behave as it is emitted from the landfill soil and as it travels through the atmospheric boundary layer to the gas analyzer downwind.

Chapter 3 describes the research conducted to simulate the TDM using a steady, prescribed landfill methane emissions rate. In this study, numerical modeling is used as an alternative to field measurements to study the sensitivity of the TDM and provide estimates of measurement accuracy. WRF is used with topography and wind conditions for a real landfill, Sandtown Landfill in Delaware, USA. In WRF, a landfill emissions rate is prescribed to emit methane from the surface at a steady rate at each time step, and this prescribed emissions is compared against the simulated TDM-measured emissions rate to evaluate the error of the TDM measurements. The prescribed methane emissions are heterogeneously distributed with a hot spot containing 80% of the total emissions where the daily cover area is located, as was measured to be the case in the field at this particular landfill. Factors tested include tracer setup, distance from landfill to transect path, speed of the transect measurement vehicle, and wind direction. Results show that location of the tracers relative to the hot spot of highest landfill emissions makes the largest difference in accuracy of the TDM-measured emissions. Wind direction and topography also play a significant role in the accuracy of the TDM measurements. These findings will inform the use of the TDM in the field, for example to help ensure that more care is taken in the placement of the tracer release locations so that the tracer gas plume is representative of the methane plume.

In Chapter 4, a methane surface flux parameterization is added to the WRF model to simulate time-varying landfill methane emissions. In field campaigns, large short-term variability has been measured, and short-term correlations between landfill emissions and surface wind speed have been observed, supporting the use of such a drag law parameterization. The surface flux parameterization used in these simulations has been adapted from the parameterizations already used in WRF for heat and moisture fluxes. TDM-measured emissions and their associated percent error are compared between steady emissions simulations and unsteady (drag law parameterized) emissions simulations, and the variability of the TDM-measured emissions is found to be approximately the same for both simulations. This indicates that the TDM introduces large variability into the measured emissions due to errors inherent in the method, and that fluctuations are not all due to wind dependence. Ideal and non-ideal tracer configurations are both simulated to determine a range of the percent error we could expect in TDM-measured emissions in the field. Lastly, several different time periods across two different days with different wind directions and different atmospheric conditions are simulated. Results show some correlation between standard deviation of TDM-measured emissions and time of day. This study again informs field implementation of the TDM and may guide understanding of measurements taken under different atmospheric conditions.

In Chapter 5, the TDM is simulated over 4 different days at a Southeastern US landfill with extensive field data measurements available for comparison. These simulations serve to help create a robust modeling framework for exploring the TDM and how it can be used to estimate annual landfill emissions. Results of this work show excellent agreement between model output and field data for both local wind conditions and tracer gas concentration.
at the locations of the real roads where field measurements were taken on all four days simulated. Results also show that longer measurement periods result in more accurate mean TDM-measured emissions, and that TDM-measured emissions are more accurate when wind speed is $\geq 5$ m/s. Methodologies for using limited measurements to estimate annual methane emissions are explored along with their challenges and limitations.

### 1.3 Summary of Contributions

In summary, the main contributions of this research are:

- development of a framework for numerical simulations of landfill methane emissions and the tracer dilution method for estimating landfill methane emissions

- recommendations for TDM setup (tracer placement and transect path location) for increasing accuracy of TDM-measured landfill methane emissions

- recommendations for when to perform TDM measurements regarding the relationship between the wind direction and topography of the landfill

- development of a drag law parameterization for unsteady landfill methane emissions, with rationale and supporting field data

- evidence that the magnitude of variability in TDM-measured emissions over a short period may be largely due to errors in the TDM and may not be representative of the true short-term variability in emissions

- recommendations for how accurate the TDM-measured emissions may be under different atmospheric conditions

- recommendations for how to better estimate annual landfill methane emissions based on limited TDM measurements
Chapter 2

Methane transport: atmospheric dispersion and soil-atmosphere fluxes

We begin with a description of turbulent dispersion of plumes in the atmospheric boundary layer before applying this background knowledge to investigate the tracer dilution method (TDM) in Chapter 3. The middle portion of this chapter addresses previous TDM studies and how this work builds on and complements those studies. The last part of this chapter is devoted to a discussion on the surface flux parameterization used to simulate unsteady landfill methane emissions in Chapters 4 and 5 of this work.

2.1 Plume dynamics

Understanding plume dynamics is a key component to understanding the principles behind the tracer dilution method and improving its accuracy. The accuracy of the method relies on the placement of the gas analyzer and tracer gas cylinders in relation to the landfill and the wind direction. A few rules of thumb have been developed for optimal setup of the tracer dilution method through Gaussian plume approximations as well as trial and error in the field. Studying the governing mechanisms and expected behavior of plumes of passive scalars in the atmosphere will lead to more informed decisions when performing the tracer dilution method as well as a better understanding to aid in interpreting the measurements.

2.1.1 The Advection-diffusion equation

The movement of mass constituting a plume is governed by the turbulent advection-diffusion equation, presented in equation 2.1:

$$\frac{\partial c}{\partial t} + \frac{\partial}{\partial x}(uc) + \frac{\partial}{\partial y}(vc) + \frac{\partial}{\partial z}(wc) = \frac{\partial}{\partial x} \left( D_x \frac{\partial c}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_y \frac{\partial c}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_z \frac{\partial c}{\partial z} \right) + S \tag{2.1}$$

In this equation, $\nabla \cdot (\bar{uc})$ is the advection term that describes mass moving with the air at mean wind speed $\bar{u} = (u, v, w)$, the partial derivatives on the right-hand side comprise the
diffusive term that describes down-gradient scalar flux, and $S$ represents one or more source terms. The result of the diffusive term is that the spatial gradients smooth out over time. In the atmosphere, diffusivity is dominated by turbulence, which is anisotropic, so $D_x$, $D_y$, and $D_z$ may be different. For the sections that follow, we are interested in mass transfer in all three dimensions where $x$ refers to the longitudinal, or downwind, axis; $y$ refers to the lateral, or crosswind, axis; and $z$ refers to the vertical axis.

### 2.1.2 Gaussian plume solution for point source

In this section we consider a Gaussian plume from a ground-level point source with advection acting in only the $x$-direction and anisotropic diffusion. This solution is a good approximation for a plume in the atmosphere averaged over a long enough time, or for a laminar flow, but cannot capture the instantaneous concentration distribution of a turbulent plume. This point will be discussed further in section 2.1.4. For a Gaussian plume from a ground-level point source with continuous release rate $Q$ and mean wind speed $\bar{U}$, concentration at any point in 3-D space can be found with the following equation:

$$c(x, y, z) = \frac{Q}{\bar{U}\sqrt{2\pi\sigma_y}} \exp\left(\frac{-y^2}{2\sigma_y^2}\right) \frac{1}{\sqrt{2\pi\sigma_z}} \exp\left(\frac{-z^2}{2\sigma_z^2}\right)$$  \hspace{1cm} (2.2)

In equation 2.2, $\sigma_y$ and $\sigma_z$ are the lateral and vertical standard deviations of the plume, which are a way to approximate the lateral and vertical extent of the plume. The $x$-dependence in equation 2.2 lies in $\sigma_y$ and $\sigma_z$, the general forms of which are presented in equations 2.3 and 2.4:

$$\sigma_y^2 = 2D_y \frac{x}{\bar{U}}$$  \hspace{1cm} (2.3)
$$\sigma_z^2 = 2D_z \frac{x}{\bar{U}}$$  \hspace{1cm} (2.4)

In the above equations, $D_y$ and $D_z$ are diffusivities with units m$^2$/s, and $\frac{x}{\bar{U}}$ can be replaced by $t$ to substitute longitudinal dependence for time dependence. These expressions for the width of the plume come from the cloud growth law.

If we wish to look more precisely at how a plume grows in the atmosphere from a point release of mass, it is necessary to integrate the full advection-diffusion equation, which reveals that the plume spreads like a Gaussian, but not with the variances presented in equations 2.3 and 2.4. Tadmor and Gur (1969) developed empirical formulas for $\sigma_y$ and $\sigma_z$ that depend on the atmospheric stability class:

$$\sigma_y \approx a_1 x^{0.9031}$$  \hspace{1cm} (2.5)
$$\sigma_z \approx a_2 x^b$$  \hspace{1cm} (2.6)

Equation 2.6 is only valid for 500 m $\leq x \leq$ 5000 m. Coefficients $a_1$, $a_2$, and $b$ take on different values for stability classes A through F.
If the source is elevated above ground level, then the solid boundary of the ground must be accounted for. To account for a solid boundary at the ground, the method of images is used, in which a new source is reflected about the ground with a source strength equal to the original source. This imposes a no-flux Neumann boundary condition at ground level \((z=0)\), and the solution to the model system is only considered for \(z \geq 0\). The equation for a plume from an elevated source located at \(z=H\) with the method of images to impose the solid bottom boundary is presented in equation 2.7:

\[
c(x, y, z) = \frac{Q}{U} \frac{1}{2\pi \sigma_y \sigma_z} \exp \left( -\frac{y^2}{2\sigma_y^2} \right) \left[ \exp \left( \frac{-\left( z + H \right)^2}{2\sigma_z^2} \right) + \exp \left( \frac{-\left( z - H \right)^2}{2\sigma_z^2} \right) \right] (2.7)
\]

### 2.1.3 Meandering and diffusion

The spreading of a plume can be separated into two types of motion: spreading due to turbulent eddies, and meandering of the plume center relative to a fixed longitudinal axis (Gifford, 1959). Meandering is due to turbulent motions larger than the width of the plume, and diffusion is due to turbulent motions smaller than the width of the plume. Meandering is more prominent closer to the source, where the lateral concentration gradients are sharper, and therefore produces larger fluctuations closer to the source. Fluctuations decrease with distance from the source as meandering becomes less prominent (Fackrell and Robins, 1982). Meandering affects fluctuations in the lateral direction the most, and for a ground level source has essentially no effect in the vertical direction because vertical motions are typically smaller than the vertical extent of the plume (Pasquill, 1975). This might not be true over complex terrain, however, because terrain can induce larger vertical motions.

### 2.1.4 Lagrangian and Eulerian frameworks

One consideration in studying plume dispersion is the distinction between using an Eulerian or Lagrangian framework for representing fluid flow. The Lagrangian framework tracks the locations of individual fluid parcels, while the Eulerian framework uses coordinates fixed in space (Bush, 2016). In the Lagrangian framework, each fluid parcel carries its own properties such as density and momentum, and these properties may change over time. The flow is described by recording the histories of each fluid parcel, including their individual trajectories. For the Eulerian description, on the other hand, the evolution of the flow properties can be recorded at every point in space as time advances. The Eulerian framework is used by WRF, which uses a fixed 3-D grid and updates the values of several properties such as velocity and density at each grid point.

The Lagrangian framework is well-suited to dispersion problems, especially when considering near-source dispersion, because Lagrangian models are independent of a numerical grid, so they will not instantaneously spread over an entire grid cell when a scalar is released from a point source. Additionally, Lagrangian models do not introduce numerical diffusion, like Eulerian models do (Stohl et al., 2005). Lagrangian models can give an idea of the travel
time of a particle from when it is emitted to a certain point, such as a measurement location. The Lagrangian framework can be worse for atmospheric modeling, however, because there are many ad hoc parameters that need to be tuned to get the particles to disperse properly under different atmospheric conditions.

### 2.1.5 Concentration fluctuations

Understanding concentration fluctuations has been a key component of environmental pollution problems for many decades. Although plumes are often thought of as smooth, steady Gaussians, in reality this model cannot be applied to instantaneous plumes which meander and fluctuate due to turbulent motions in the atmosphere. Gifford was one of the first researchers to develop a fluctuating plume model that extended the Gaussian model and allowed individual plume elements to shift randomly away from the longitudinal axis of the plume in both $y$ and $z$ (1959). Figure 2.1 shows Gifford’s sketches of the classical spreading disk plume model with no meandering, a real plume made of non-disk-shaped elements, and his meandering spreading disk model.

![Figure 2.1: Gifford's sketches of the classical spreading disk model (top), a real plume made of non-disk-shaped elements (middle), and Gifford's meandering spreading disk model (bottom). Source: Gifford, 1959.](image)
CHAPTER 2. METHANE TRANSPORT: ATMOSPHERIC DISPERSION AND SOIL-ATMOSPHERE FLUXES

One very visually informative study of concentration fluctuations in instantaneous plumes comes from Crimaldi et al. (2002). They released dye in a turbulent water flume and used a planar laser-induced fluorescence technique to record 2-D snapshots at various locations throughout the plume in the $x$-$z$, $y$-$z$, and $x$-$y$ planes, where $z$ is the vertical direction. Figure 2.2 shows images of the mean concentration field in all three planes.

![Mean concentration field in $x$-$y$ plane at two different vertical heights (top); mean concentration field in $x$-$z$ plane (middle); mean concentration field in $y$-$z$ plane (bottom). Source: Crimaldi et al., 2002.](image)

In these figures, mean concentration is normalized by the source concentration $C_0$. These mean plumes appear qualitatively to be Gaussian, and in fact 1-D transects in all directions fit a Gaussian curve very well. The instantaneous plumes, however, are made up of filaments of dye following the paths of turbulent eddies and creating irregularly distributed patches of high concentration, low concentration, and even intermittent patches with no concentration.
An example of an instantaneous plume in downstream sections of all three planes are shown in Figure 2.3.

![Instantaneous plume](image)

Figure 2.3: Instantaneous plume in a downstream subsection of the $x$-$z$ plane through the plume centerline (top), instantaneous plume in a downstream subsection of the $y$-$z$ plane (middle), instantaneous plume in a downstream subsection of the $x$-$y$ plane (bottom). Source: Crimaldi et al., 2002.

From figure 2.3, it becomes obvious that a Gaussian model could not adequately capture this structure, and the concept of predicting concentration at a particular point in space and time appears much more complicated. Crimaldi et al. were able to tease out a few general trends in the instantaneous concentration fields, however. Peak concentrations universally decreased as $x$-$z$ planes moved away from the centerline, with maximum peak concentration
occurring in the $x$-$z$ plane through the centerline, indicating the expected lateral diffusion. Comparing the heights of the mean and instantaneous plumes in the $x$-$z$ plane, we see that the instantaneous plume extends much higher ($\sim 3$ cm and $\sim 7$ cm respectively), indicating that the plume experiences a lot of intermittency in the vertical direction.

Looking at $y$-$z$ planes, the region of non-zero concentration grew wider and the peak concentration grew lower as the planes moved further downstream. In the $x$-$y$ planes, peak concentration decreases as the height of the plane moves away from the source (at ground level). These observed behaviors indicated that while an instantaneous plume appears very complex and difficult to predict, some expected behaviors can be observed.

The authors of the work found that looking only at the mean concentration field did not give an accurate picture of the instantaneous structure of the plume. For example, a low value of $C$ at a point in space almost never corresponded to a small local concentration peak. Therefore, they also looked at fields of standard deviation of concentration and intermittency (probability that the concentration is above a small threshold value) and concluded that a combination of mean concentration, standard deviation of concentration, and intermittency can provide a meaningful measure of the instantaneous structure. This work is particularly applicable to the landfill research in the coming chapters because the accuracy of the TDM depends on understanding how to best set up the tracer gas release points so that the tracer gas plume mimics the methane plume from a heterogeneous area source. The accuracy of the TDM is also affected by how to best take downwind measurements to capture the full plumes’ widths as they meander and change in time.

### 2.1.6 Gradient transfer theory

Gradient transfer theory is used to approximate turbulent scalar fluxes. The idea is that the flux of the scalar in a particular direction is proportional to the negative of the gradient of that scalar in that particular direction. In other words, the scalar will be transported from an area of higher concentration to an area of lower concentration. The conditions for gradient-transfer theory to apply are that length and time scales of the transporting motions need to be sufficiently small and sufficiently uniform compared to the length and time scales of variations in the mean field gradients of the scalar being transported. For example, in the atmosphere, gradient transfer theory is not sufficient to describe the motions of the cross-section of a plume because the lateral movement is affected by meandering, which is due to motions at the scale of or larger than the cross-section of the plume itself. For the case of concentration flux in the vertical direction, gradient transfer theory states that:

$$w'c' = -K \frac{\partial \bar{c}}{\partial z}$$  \hspace{1cm} (2.8)

In this equation, parameter $K$ is positive and has units $m^2/s$. Despite its limitations, gradient transfer theory is used in the majority of turbulence models and is how turbulence is parameterized in WRF with the large-eddy simulation (LES) turbulence closure used in this dissertation, the TKE-1.5 model with nonlinear backscatter and anisotropy (NBA). Note
that the NBA model allows for some counter-gradient transfer and therefore addresses some of the shortcomings of classic gradient transfer models.

2.1.7 Green’s function

The advection-diffusion equation can be thought of as a linear operator acting on a concentration field, and because of this, a Green’s function can be used to solve the advection-diffusion equation. This is useful because the consequence is that if we can solve an equation using a Green’s function, then if we solve the equation for a certain source strength and get a resulting concentration field, we can see how that concentration field would change if the source strength changed without having to solve the equation all over again. This idea is demonstrated in the following steps.

A Green’s function, \( G(t, \tau; x, \xi) \), of a linear differential operator \( \mathcal{L} \) at time \( \tau \) and location \( \xi \) is the solution to:

\[
\mathcal{L}G(t, \tau; x, \xi) = \delta(t - \tau, x - \xi) \tag{2.9}
\]

In equation 2.6, \( \delta(t - \tau, x - \xi) \) is the Dirac delta function. A Green’s function can be used to solve differential equations of the form:

\[
\mathcal{L}c(t, x) = S(t, x) \tag{2.10}
\]

We can use the Green’s function to solve for \( c(x) \) in the following manner:

\[
c(t, x) = \int_{\Omega} \int_{0}^{t} G(t, \tau; x, \xi)S(\tau, \xi)d\tau d\xi \tag{2.11}
\]

In equations 2.9 and 2.10, we use \( c(x) \) for our dependent variable because we are interested in concentration, and \( S(x) \) will be the source term for our advection-diffusion equation. \( \Omega \) is the spatial domain of interest. In the case of the advection-diffusion equation, our linear operator is:

\[
\mathcal{L} = \frac{\partial}{\partial t} + \nabla \cdot (\vec{u}) - \nabla \cdot (\vec{D}\nabla) \tag{2.12}
\]

We know that the advection-diffusion equation is linear because we are able to get it into the form of equation 2.9 by identifying the linear operator that acts on our dependent variable of interest, which means that the following is true for constant \( a \):

\[
\mathcal{L}(ac) = a\mathcal{L}c \tag{2.13}
\]

Looking back at equation 2.9, it follows that \( ac \) solves:

\[
\mathcal{L}(ac) = aS \tag{2.14}
\]

To simplify our Green’s function notation from equation 2.10, let us define function \( \mathcal{G} \) such that:

\[
c(t, x) = \mathcal{G}(S(t, x)) \tag{2.15}
\]
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Using the same linear principle from equations 2.12 and 2.13, we can apply our Green’s function to \((aS)\) instead of \(S\):

\[
\mathcal{G}(aS) = a\mathcal{G}(S) = ac
\]

(2.16)

The important implication of this result is that if we solve for the concentration field numerically and then want to look at the concentration field produced by a different source strength, we can simply multiply the current concentration field by the same constant we want to multiply the source strength by instead of performing a new simulation.

2.2 Previous tracer dilution method studies

The tracer dilution method was first developed to estimate emissions from inaccessible ground level sources such as manufacturing plants by Ludwig et al. (1983). This study presents the current general practice of the tracer dilution method that concentration measurements should be taken between 1 km and 3 km downwind. The derivation for the method begins by considering steady-state Gaussian plumes from an infinite line source for the tracer, which is near but not collocated with the area source, and an area source for the gas emissions to be quantified. Using the fact that ground-level concentration as a function of downwind distance from the source is dependent on emissions strength, the ratio of concentration of the gas of interest (e.g. landfill methane) to tracer gas concentration is rearranged to solve for emissions rate of the important gas. The resulting equation contains empirical constants that depended on atmospheric stability. The authors assert that measurements should be taken further than 1 km from the source to ensure that the vertical standard deviation of the Gaussian concentration distribution is at least 15 m. With a vertical spread of at least 15 m, the concentration distribution will be close to uniform when averaged over 1 h. Additionally, the measurements should be taken closer than 3 km from the source to ensure that the lateral standard deviation is less than 200 m. The method was tested at a halocarbon plant, where hexafluoride was released as the tracer gas from a cylinder in a moving vehicle located downwind of the plant as it drove along a line 800 m long perpendicular to the wind direction, and concentration measurements were taken with bag samplers placed 150 m apart in a line 1.6 km downwind from the hexafluoride release. This TDM setup is different from the way the TDM is performed in this dissertation work, in which the tracer gas is released from a few stationary points collocated with the area source of interest, rather than released from a moving vehicle near the area source, and the measurements are taken by a moving vehicle rather than stationary measurements collected at multiple points. The field test revealed several problems with the method including a large dependence on the assumptions made about the size of the emitting area and the fact that the emissions were highly temporally variable. The authors suggested that the method be improved by increasing the spatial and temporal resolution of downwind measurements.

Ludwig et al.’s tracer technique for estimating emissions was extended to measure methane emissions by Lamb et al. (1995), who performed a controlled methane release test to study the effect of location of the tracer relative to the methane as well as stationary measurements.
versus transect measurements. No apparent difference was found between the collocated and separated release tests, although the paper does not say whether the separated tracer was located upwind, downwind, or laterally displaced from the methane. The authors then conducted field tests at a natural gas facility. The results from the natural gas facility were successfully replicated on different days with different wind conditions at approximately 1.5 km downwind of the plant, leading the authors to conclude that a single tracer release provides a relatively accurate simulation of the methane emissions from a gas plant.

Tracer dispersion from landfills was modeled by Carpentieri et al. in a wind tunnel with the purpose of building an experimental data set to be used for validating numerical models (2008). Two different landfill scenarios were modeled: a taller landfill with flat downwind topography, and a shorter landfill both with and without a gently sloped hill placed downwind. Emissions were released through capillary tubes uniformly distributed over the top of the model landfill. Mean concentration profiles showed an exponential decay in the longitudinal direction, a fairly symmetrical Gaussian shape in the lateral direction, and a fairly linear vertical profile with the maximum at the ground level. The exception to the linear vertical profile occurred at the closest measurements to the landfill, where concentration was pooling at the surface to create a bulge at the surface of the vertical concentration profile. This bulge in the surface concentration directly downwind of the landfills was determined to be due to a combination of induced turbulence at the surface and decreased wind speed at the surface, due to the presence of the landfill.

The tracer dilution method was further developed for estimating landfill emissions by Mønster et al. (2014). A controlled release of methane was performed on a flat field with three different tracer placements: a single tracer collocated with the point source of methane, a single tracer upwind of the methane, and two tracers to the right and left of the methane forming a line perpendicular to wind direction. Mobile transect measurements were performed at three different distances from the methane. Methane emissions were then calculated for each tracer placement and each transect location with three different methods, including the plume integration method presented in equation 1.2 in Chapter 1. The plume integration method was found to yield the most accurate estimates of the different calculation methods. Placing the tracer upwind of the methane yielded the least accurate estimates of the different tracer placements. None of the transect distances was clearly the most accurate, which was surprising to the study because the objective was to determine the accuracy of the tracer dilution method as a function of distance from the methane source. This result could mean that all of the downwind transects were measured at distances where the plumes were well-mixed.

This last study by Mønster et al. (2014) inspired many aspects of the work presented in this dissertation, particularly Chapter 3. The authors set up many questions that could be answered more easily through numerical modeling than expensive and time-consuming field campaigns. Some of the questions explored in the next chapter that arose from the Mønster et al. study are: how do the distance from the landfill and angle from the wind direction of the transect path affect the accuracy of TDM-measured emissions, how does the interaction between topography and wind direction affect the accuracy of TDM-measured
emissions, and how does location of the tracers relative to the hot spot(s) of emissions affect the accuracy?

2.3 Methane surface flux parameterization

Understanding the processes that transport methane through the top soil layer and govern the emissions rate are crucial to accurately modeling the diurnal and seasonal variations in emissions. The numerical modeling in this dissertation is performed with the Weather Research and Forecasting model (WRF), which is a mesoscale atmospheric model with large-eddy simulation (LES) capabilities. WRF uses a fully compressible, Eulerian, non-hydrostatic equation set, and it is coupled to the Noah land surface model (LSM). The Noah LSM uses a radiation budget, water budget, and soil and vegetation properties to compute surface fluxes that drive atmospheric boundary layer growth. The heat and moisture surface flux parameterizations native to WRF are used as a template for the methane surface flux parameterization that we have added to WRF for this dissertation.

2.3.1 Flux parameterization background

The vertical flux \( Q_a \) of any variable \( a \) is assumed to be driven by the difference in \( a \) across the interface, demonstrated in Equation 2.17:

\[
Q_a = -U_T (a_{\text{above}} - a_{\text{below}})
\]  

(2.17)

where \( U_T \) is the transport velocity across the interface, and \( a_{\text{above}} \) and \( a_{\text{below}} \) are the values of \( a \) above and below the interface (Stull, 1988). Transport velocity is parameterized as a function of some mean horizontal wind speed at a specified height above the interface:

\[
U_T = C_D \overline{U}
\]  

(2.18)

In Equation 2.18, \( \overline{U} \) is the mean horizontal wind speed at a height \( z \) above the surface, and \( C_D \) is the bulk transfer coefficient for the same height. This parameterization comes from the assumption that wind speed right at the surface is zero while wind speed at some height above the surface will be greater than zero, and this gradient in wind speed will create shear at the surface, generating turbulence and supporting the transport of \( a \).

In atmospheric science, it is common practice to apply Equation 2.17 to heat and moisture fluxes from the soil surface, using the same bulk transfer coefficient for both processes (Stull, 1988; Bonan, 2002; Deardorff, 1972). These two fluxes can be parameterized by Equations 2.19 and 2.20 respectively:

\[
Q_\theta = -C_H \overline{U} (\theta_{\text{atm}} - \theta_{\text{soil}})
\]  

(2.19)

\[
Q_q = -C_H \overline{U} (q_{\text{atm}} - q_{\text{soil}})
\]  

(2.20)
where $\theta$ is potential temperature and $q$ is specific humidity, both with units of kg/m$^3$, and the bulk transfer coefficient is defined as:

$$C_H = \frac{k^2}{[\ln(\frac{z}{z_0}) - \Psi_M][\ln(\frac{z}{z_0}) - \Psi_H]}$$  \hspace{1cm} (2.21)

which is dimensionless (Stull, 1988). Note that $k$ is the von Karman constant, $z_0$ is the roughness length, and $\Psi_M$ and $\Psi_H$ are the stability functions for momentum and heat respectively. In the WRF code, moisture flux is parameterized as:

$$Q_q = \rho u^* q^* = \frac{\rho u^* k(q_{soil} - q_{atm})}{\ln(\frac{z}{z_0}) - \Psi_H}$$  \hspace{1cm} (2.22)

which is equivalent to equation 2.20 because $u^*$ is defined as:

$$u^* = \frac{\bar{U}k}{[\ln(\frac{z}{z_0}) - \Psi_M]^2}$$  \hspace{1cm} (2.23)

and $q^*$ is a simplified term representing $Q_q/\rho u^*$.

### 2.3.2 Relating diffusive properties of methane to moisture

Surface fluxes of other gases are analogous to moisture fluxes, and Equation 2.22 can be adapted to any gas by scaling by the ratio of molecular diffusivity of the gas to molecular diffusivity of water vapor (Pleim and Ran, 2011). In a large excess of air, molecular diffusivity of water vapor in air is $\sim 2.4 \times 10^{-5}$ m$^2$/s, while molecular diffusivity of methane in air is $\sim 1 \times 10^{-5}$ m$^2$/s (Marrero and Mason, 1972). Given this information, Equation 2.24 reflects the equation used in the WRF code to simulate unsteady landfill methane emissions:

$$Q_{CH_4} = \frac{D_{CH_4}}{D_{H_2O}} \rho u^* c^* = 0.42 \frac{\rho u^* k(c_{soil} - c_{atm})}{\ln(\frac{z}{z_0}) - \Psi_H}$$  \hspace{1cm} (2.24)

### 2.3.3 Relevant time scales

To the author’s knowledge, this surface flux modeling approach has not yet been used for landfill methane emissions. Landfill methane emissions are typically parameterized using a constant boundary layer conductance times ($c_{soil} - c_{atm}$), so the emphasis of the calculations is placed on determining the value of the soil methane concentration and how it changes in time in response to changes in soil temperature, soil moisture, and oxygen concentration, which causes methane oxidation.

The above soil-focused approach is used in the California Landfill Methane Inventory Model (CALMIM) to calculate average annual landfill methane emissions from a specific landfill given inputs about the landfill. We ran CALMIM over Sandtown Landfill in Delaware using input parameters specific to Sandtown such as the latitude and longitude, area of
the landfill, soil type, and soil thickness. One of the CALMIM output files contains soil methane concentration at every hour over a 365-day period, not corresponding to specific dates in a specific year, but representing average values for each day of the year given average atmospheric conditions and precipitation on each day. By running CALMIM over Sandtown Landfill, we hoped to gain an understanding of the type of variability we should expect in soil methane concentration over short time scales (1 - 2 hours) as well as seasonal time scales. Over 2-hour intervals, we saw an average of 1% change in the soil methane concentration value, with a maximum of 6% change. Over the entire 365-day period the values differed by as much as 96% from the mean value.

Given the large differences in soil methane concentration variability on different time scales, it makes sense that landfill methane emissions models whose goal is to predict annual methane emissions will focus on the seasonal variability in soil methane concentration, which is governed by complex empirical formulas that depend on soil temperature, soil moisture, and methane oxidation rate. For the purposes of our short-term simulations, however, updating the soil methane concentration at each time step when the value changes by only 1% over the course of the 1 - 2 hour measurement period produces a negligible effect on the landfill methane emissions and the accuracy of the tracer dilution method. In light of this CALMIM soil methane concentration investigation, we use constant soil methane concentration values in the calculation of surface fluxes. Thus the time variability in our unsteady emissions simulations comes from other aspects of the surface flux parameterization.

Significant variability (~20% variability from the mean) is seen in landfill methane emissions over short time scales, and this variability in landfill methane emissions cannot be replicated in the model solely through variability in \((c_{soil} - c_{atm})\) (Delkash et al., 2016; Foster-Wittig et al., 2015). This concentration gradient would have a small standard deviation over short time periods because of the small standard deviation of \(c_{soil}\) and the fact that \(c_{soil} \gg c_{atm}\), based on the \(c_{soil}\) values predicted by CALMIM and the \(c_{atm}\) values predicted by WRF with prescribed emissions. We therefore seek a model that will yield the expected short-term variability in landfill methane emissions through other factors than the soil methane concentration. In other words, if \((c_{soil} - c_{atm})\) is relatively constant over short time scales, but observed emissions are not, then there must be variability in the boundary layer conductance coefficient.

### 2.3.4 Breakdown of modeled emissions

One way to determine how realistic Equation 2.24 is for our landfill methane emissions on short time scales is to examine the magnitudes and variability of each component on the right hand side, which we will regroup into:

\[
Q_{CH_4} = C_d u^* (c_{soil} - c_{atm})
\]  

(2.25)

so that we can examine \(C_d\), \(u^*\), and \((c_{soil} - c_{atm})\) separately. We will examine these components of Equation 2.25 using WRF output from a Sandtown Landfill simulation, which
we know has realistic mean and standard deviation of wind speed as well as reasonable soil methane concentration values from the CALMIM simulations.

Figure 2.4: Methane emissions in kg/m$^2$/s (top row); coefficient $C_d$ in kg/m$^3$ (second row); $u^*$ in m/s (third row); ($c_{soil} - c_{atm}$) in kg/kg (bottom row). Each column is a different grid point on the landfill surface to examine heterogeneity.

In Figure 2.4 we see that the magnitude of emissions from each grid cell is $\sim 7 \times 10^{-7}$ kg/m$^2$/s. The magnitude of $C_d$ is $\sim 0.06$ kg/m$^3$, $u^*$ is $\sim 0.6$ m/s, and ($c_{soil} - c_{atm}$) is $\sim 2 \times 10^{-5}$ kg/kg. Figure 2.5 presents the same dataset, normalized by the means of each variable to show the influence of the variability of the three components on the variability of the emissions.

From Figure 2.5 it is clear that the variability in $Q_{CH_4}$ is larger than the variability in $C_d$ and smaller than the variability in $u^*$, and the variability in ($c_{soil} - c_{atm}$) is imperceptible in comparison. The peaks and troughs on $Q_{CH_4}$ match the peaks and troughs in $u^*$, while there appears to be an inverse relationship between $Q_{CH_4}$ and $C_d$. This inverse relationship makes sense because $C_d$ is inversely proportional to $\Psi_H$, the stability correction term for heat from similarity theory, and $\Psi_H$ is directly proportional to $u^*$. There also appears to be a direct relationship between $Q_{CH_4}$ and ($c_{soil} - c_{atm}$), which makes sense because a change in emissions will directly affect the atmospheric concentration in the grid cell right
above the landfill surface, and since soil methane concentration is kept constant, changes in atmospheric concentration are the only factors affecting this $\Delta C$ term.

Field data supports significant variability in landfill methane emissions on short time scales (Foster-Wittig et al, 2015; Delkash et al, 2016; Xu et al, 2014; Poulson and Moldrup, 2006). It is clear from Figure 2.5 that the variability in $(c_{\text{soil}} - c_{\text{atm}})$ over a short time period (90 minutes in this case) is too insignificant to produce the variability seen in emissions. Proportionally, the variability seen in emissions is better matched by the variability in $u^*$ (closely related to the variability in wind speed by Equation 2.23). It is clear from these figures that if emissions were formulated as $(c_{\text{soil}} - c_{\text{atm}})$ times a constant, the necessarily variability would not be produced in the emissions on short time scales. To induce the appropriate variability in landfill methane emissions on short time scales, the atmosphere should be considered the driver of emissions rather than the soil processes.
Chapter 3

Numerical simulations to assess tracer dilution method for measurement of landfill methane emissions

1 The work in this chapter uses numerical simulations to assess the accuracy of one method used to measure landfill emissions. Atmospheric dispersion simulations with the Weather Research and Forecast model (WRF) are run over Sandtown Landfill in Delaware, USA, using observation data to validate the meteorological model output. A steady landfill methane emissions rate is used in the model, and transects of methane and tracer gas concentration are collected along various lines downwind from the landfill to use the tracer dilution method to calculate methane emissions. The calculated methane emissions are compared to the methane emissions rate used in the model to find the percent error of the tracer dilution method for each simulation. The roles of different factors are examined: measurement distance from the landfill, transect angle relative to the wind direction, speed of the transect vehicle, tracer placement relative to the hot spot of methane emissions, complexity of topography, and wind direction. Results show that percent error generally decreases with distance from the landfill, tracer placement has the largest effect on percent error, and topography and wind direction both have significant effects. Transect angle and transect speed have small to negligible effects on the accuracy of the tracer dilution method. These tracer dilution method simulations help develop a framework for testing different scenarios that can build confidence in the use of the method for measuring landfill methane emissions.

1This chapter is a reproduction of the paper by the same title by Diane M. Taylor (first author), Fotini K. Chow, Madjid Delkash, and Paul T. Imhoff, published in Waste Management 56: 298 - 309 (Taylor et al., 2016), Copyright ©2016 Elsevier Ltd.
CHAPTER 3. NUMERICAL SIMULATIONS TO ASSESS TRACER DILUTION METHOD FOR MEASUREMENT OF LANDFILL METHANE EMISSIONS

3.1 Introduction

Landfills are one of the largest anthropogenic sources of atmospheric methane in the U.S. (U.S. Environmental Protection Agency, 2015), yet measuring these emissions is challenging. Many methods of estimating landfill methane emissions are expensive, labor intensive, and associated with high levels of uncertainty. The tracer dilution method is a cost effective and minimally-invasive method for estimating whole landfill emissions by comparing concentrations of a tracer gas and methane downwind of the landfill (Babilotte, 2010). The tracer gas is released at one or more point sources on the surface of the landfill at a known rate. Downwind measurements are obtained from either stationary or mobile sensors, and the ratios of the methane and tracer concentrations are used to estimate the methane source strength based on the known tracer source strength. The predicted methane emissions are sensitive to different factors of the method setup such as placement of the tracer release locations and distance from the landfill to the downwind measurement points, which have not been thoroughly studied (Mønster et al, 2014).

In this work, numerical modeling is used to study the sensitivity of the tracer dilution method to the tracer configuration and sampling strategy, as well as site specific factors such as topography and wind direction. The mobile sensor approach is the focus here; this method uses a gas analyzer mounted on a vehicle collecting transects of both the methane and tracer plumes as it traverses the plume roughly perpendicular to the wind direction (Czepiel et al., 2003; Foster-Wittig et al., 2015; Mønster et al., 2014). The methane emissions are estimated based on the downwind ratio of methane concentration to tracer concentration, as shown in Equation 3.1:

\[ Q_{CH_4} = Q_{tr} \frac{\int c_{CH_4}(x)dx}{\int c_{tr}(x)dx} \]  

(3.1)

where \( Q_{CH_4} \) is the methane emissions rate, \( Q_{tr} \) is the tracer emissions rate, and \( \int c_{CH_4}(x)dx \) and \( \int c_{tr}(x)dx \) are the integrals over a mobile transect measurement of methane and tracer gas respectively.

One of the main sources of error in the tracer dilution method arises from the different evolution of the two plumes. In reality, the methane is emitted from a large area source, on the order of hundreds of meters wide, with significant heterogeneity in the emissions (Abichou et al., 2006). The tracer gas is often emitted from a few different point sources often separated by a large distance to try to capture the variability from the larger methane area source. Far downwind from the landfill, the methane and tracer plumes become more similar, meaning that both plumes have experienced enough dispersion to eliminate evidence of the differences in source size. The method becomes more accurate when both plumes reach this well-mixed condition. In this context, accuracy of the method means how well the emissions measured by the method match the actual emissions from the landfill. With this definition of accuracy, it is clear the accuracy of the method is especially difficult to assess because in the field the true landfill emissions are not known. Tracer dilution method measurements are sometimes used to validate or improve IPCC models (Börjesson et al., 2009), but using tracer dilution
method results as the true landfill emissions may be inappropriate while the accuracy of the method remains uncertain (Mønster et al., 2014).

A recent study by Mønster et al., 2014 investigated the effect of measurement distance from the tracers on the accuracy of the tracer dilution method. Experiments were conducted at a field site with flat topography, a controlled methane point source and three different tracer configurations: 1) collocated with the methane source, 2) upwind of the methane source, and 3) forming a line perpendicular to the wind direction on either side of the methane source. Three distances were used to measure the concentrations: 370, 770 and 1200 m. For tracer setup 3, increasing the measurement distance from the emission source diminishes the uncertainty (12%). Tracer setup 2 showed more significant effects on emission estimation than tracer setup 3. Tracer setup 2 caused the highest error among all the experiments; further, it was concluded that increasing measurement distance led to increasing the accuracy of emission estimation (from 36±21% to 20±2%). The authors attributed this observation to more dilution of methane. The best approximation was found when the tracer gas releasing bottles were located right at the center of the methane gas emissions (6%). This study highlighted some important factors to keep in mind when performing the tracer dilution method. Whether similar results would be obtained over real landfills (where the terrain is not flat and the methane source more complex) is a question of interest for landfill researchers and landfill operators.

One other study has quantified measurement error of the tracer dilution method by comparing known emissions with measured emissions (Babilotte, 2010; Mønster et al., 2014). Both studies used a small number of gas cylinders releasing point sources of methane, and both studies were performed on flat fields. Therefore, neither took into account the possible effects of the methane being emitted from a large area source on the order of hundreds of km² or the complex topography often seen at landfill sites. The errors in these studies arose from measured gas concentrations, the gas flow releases, data treatment, and source and transect geometry (Mønster et al., 2014). Using Equation 1 to estimate emissions, measured percent errors ranged from 2±6% to 36±21% (Mønster et al., 2014) or 3.7% to 19.2% (Babilotte, 2010). Through numerical modeling, this study will address the question of whether this error range can be expected for real landfill conditions with large area sources and complex topography.

Numerical modeling of the tracer dilution method is a useful tool for evaluating the method without having the expense and labor commitment of multiple field campaigns. In this study, numerical modeling is used as an alternative to field measurements for studying the sensitivity of the methane emissions estimate to the tracer dilution method setup. A known landfill emissions rate is prescribed in the model and therefore can be compared against the emissions rates predicted by various configurations of the tracer dilution method. The code used for these numerical simulations is the Weather Research and Forecasting Model (WRF), which is a mesoscale and large-eddy simulation model used to study the atmospheric boundary layer (Skamarock et al., 2008). Large-eddy simulations of atmospheric dispersion have been used to study the spread of air pollutants (Michioka and Chow, 2008) but have not previously been applied to the specific study of the tracer dilution method.
CHAPTER 3. NUMERICAL SIMULATIONS TO ASSESS TRACER DILUTION METHOD FOR MEASUREMENT OF LANDFILL METHANE EMISSIONS

Given the current limitations of field studies and uncertainty in current applications of the tracer dilution method, assessment of the method through numerical studies is crucial to the improvement of the methodologies for better quantification of landfill methane emissions. Use of a sophisticated atmospheric dispersion model presents a unique opportunity to thoroughly explore the application of the tracer dilution method for the quantification of landfill methane emissions. The numerical model is largely able to account for complex but realistic external factors that may profoundly affect the robustness of the tracer dilution method. Furthermore, the model can overcome limitations in the field such as number of tests that can be done over the same time period and location of downwind concentration measurements. The analysis of the numerical simulations in this work helps build confidence in applying this method while understanding the limitations and interpreting results from the field.

3.2 Methods

3.2.1 Weather Research and Forecasting Model

The Weather Research and Forecasting Model (WRF) is a mesoscale numerical weather prediction model used for atmospheric research and operational forecasts (Skamarock et al., 2008). It is commonly used at a grid resolution that is much larger than the largest scales of turbulent motion in the atmosphere, which are limited by the height of the boundary layer (about 1 km). In this case, no turbulent eddies can be resolved explicitly, so all turbulence is parameterized by a planetary boundary layer (PBL) scheme. There are, however, many situations in which a much finer grid is needed to resolve more detail over complex terrain or to look at a small area of interest. When the grid spacing is fine enough, the model can explicitly resolve the turbulent eddies that are larger than a few grid cells across (Deardroff, 1970). This approach is called large-eddy simulation (LES), where the subgrid-scale eddies are parameterized with a turbulence closure scheme.

The boundary and initial conditions for this work come from the North American Mesoscale (NAM) model, which has a spatial resolution of 12 km. WRF is used with a standard grid nested approach, where several finer grids are forced with the NAM 12 km data to refine down to 30 m resolution. Three nested grids are used with grid resolutions of 2.25 km, 150 m, and 30 m, with horizontal domain sizes of 90, 300, and 300 grid cells respectively. This particular nesting configuration was chosen based on extensive prior experience with the WRF model as well as comparison with meteorological data. At the coarsest resolution, the YSU planetary boundary layer scheme is used, and at the 150 m and 30 m resolutions, LES is performed, using the TKE-1.5 nonlinear backscatter and anisotropy (NBA) model for the subgrid-scale stress (Mirocha et al. 2010). This turbulence closure is chosen because it produces model output that best matches meteorological observation data from this site.

With 30 m resolution on the smallest grid, WRF can represent some but not all the scales of turbulent motion affecting the plume dispersion. Because of this, the modeled concentra-
tion data is smoother than data collected in the field. This can affect the applicability of the model results to real landfills. Additionally, the model does not have varying background concentration of methane and tracer gas, which occurs in the field, so the influence of the background signal on measurement error that has been observed in the field is not reproduced in these simulations (Mønster et al., 2014; Foster-Wittig et al., 2015). The ability of WRF to represent turbulent motions using LES, however, makes these simulations a great improvement over Gaussian models and other simplified models that have been used in previous studies, which cannot account for the variability in the atmospheric flow field (Sykes and Henn, 1992).

3.2.2 Sandtown Landfill

The impetus for this numerical study came from a field study of the tracer dilution method at Sandtown Landfill in Delaware, USA. In the field study, concentration measurements of the methane and tracer gas were collected both at ground level and 85 m to determine if aerial measurements might be better for the tracer dilution method than ground measurements (Delkash et al., 2016). This work simulates the tracer dilution method at Sandtown Landfill using real meteorological data for the initial and boundary conditions of the atmospheric model, real topography data (which accounts for the landfill being at higher elevation than the surrounding area). Realistic heterogeneous landfill emissions are prescribed based on field surveys of emission source locations coupled with a geostatistical model for emission variability in the source area developed from previous landfill studies (Abichou et al., 2006).

Simulations of the tracer dilution method are run over Sandtown Landfill for March 6, 2010, when the tracer dilution method was performed in the field. Meteorological data and tracer concentration data from the site are used to confirm that model results represent observed conditions. Figure 3.1 shows the largest WRF domain with the middle and smallest grids (resolved at 150 m and 30 m respectively) delineated, as well as a Google Earth image of a subsection of the smallest domain to show the locations of the landfill and the measurement location used in the field work.

3.2.3 Model Validation

Comparisons of the model with observed wind data are shown in Figure 3.2. Both the observation data and model output are shown as one-minute averages. Table 3.1 shows that WRF captures the mean surface wind speed and its standard deviation quite well. The modeled wind direction also agrees very well with observations on average, though the observations show more deviations from the mean, indicating that the model does not perfectly capture the variability in the wind direction observed in the field. This variability may be due to external meteorological factors that are not capture by the model, but overall, the performance of the model compared to observations is quite good (compared to previous studies) and captures the general nature of the turbulent plume. The mean surface wind speed of about 5 m/s and the moderate insolation due to the measurement period being
Figure 3.1: WRF nested grids (left) and Google Earth image of subsection of smallest WRF domain (right), showing the landfill outlined in white, as well as the wind direction and measurement site for the real field campaign.

Sunny and midday in early spring indicate slightly unstable conditions for both observations and model output.

Table 3.1: Comparison of meteorological statistics for observation data and model output.

<table>
<thead>
<tr>
<th></th>
<th>Mean wind speed</th>
<th>Standard deviation of wind speed</th>
<th>Mean wind direction</th>
<th>Standard deviation of wind direction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Observation data</td>
<td>5.0 m/s</td>
<td>0.8 m/s</td>
<td>320°</td>
<td>54°</td>
</tr>
<tr>
<td>WRF output</td>
<td>4.9 m/s</td>
<td>1.0 m/s</td>
<td>324°</td>
<td>14°</td>
</tr>
</tbody>
</table>

In the field experiment, three acetylene gas cylinders were placed on the upwind edge of the active landfill region, identified as the main CH$_4$ source during preliminary ground-level atmospheric CH$_4$ measurements around the landfill in March 2010. The gas cylinders were arranged 70 m apart and approximately perpendicular to the mean wind direction (Delkash et al., 2016). Mass flow controllers were used to discharge C$_2$H$_2$ at a constant mass flow rate of 52.23 g C$_2$H$_2$/min. Methane and acetylene concentrations were measured with a cavity ring-down spectrometer (CRDS) (Picarro Inc., Santa Clara, CA, USA) 1.6 km downwind of the landfill at 2.5 m and 85 m above ground level (Delkash et al., 2016).

Figure 3.3 shows a comparison of modeled and observed tracer concentration at the field sampling location for two different heights. Concentrations are normalized by the mean ground concentrations, which are 0.47 ppb for the model output and 1.26 ppb for
Figure 3.2: Time series of meteorological observed data and model output. Data was measured on the surface of the landfill, and field data was collected at 1-minute resolution.

the observation data. Normalized concentration values are plotted because the range and peaks of the observations are not captured as well by the model due to the grid resolution limitations. Figure 3.3 also shows a sample of plan view plume snapshots, which demonstrate that the flow field is turbulent and highly variable. The model captures the overall variability of the tracer plume well. The observed data is discontinuous because sometimes the gas analyzer sampled gas at 2.5 m above the ground, and sometimes through tubing connected to a tethered balloon at approximately 85 m above the ground, with time gaps needed to purge sample tubing between measurements. Due to the turbulent flow field, the peaks in model concentration are not expected to align with observed concentration fluctuations; this is due to the complex nature of atmospheric turbulence, which cannot be predicted exactly with current models. With 30 m resolution, the LES cannot capture all of the small scale turbulent fluctuations seen in the field (Michioka and Chow, 2008; Lundquist et al., 2012), but the mean fields agree adequately to allow further model simulations, which are the target of this paper. Another reason for the differences between observation data and model output and the reason for normalizing the data by the mean ground concentrations is the difference between sample volume of the gas analyzer and size of the model grid cells. The gas analyzer used in the field has a sample volume of 10 mL, representing a point measurement, while the model concentrations shown in Figure 3.3 are calculated for grid cells which are 30 m × 30 m × 15 m. This grid discretization is relatively fine as far as atmospheric models are concerned, but nonetheless represents much larger volumes than actual field samples.
The model thus captures the overall plume behavior quite well, but yields lower peaks and smoother concentration data than are seen in the field data (Michioka and Chow, 2008). Statistical comparisons between model-predicted concentrations and field observations are not reported, since the field data are sufficiently sparse to preclude the value of such an analysis. The remainder of the paper will focus on model-to-model intercomparisons to examine sensitivity and accuracy of the tracer dilution method.

### 3.3 Base Case Simulation

A base case simulation over Sandtown Landfill is presented first to aid with interpretation of the model output. The simulation uses steady, heterogeneous landfill methane emissions and two point source tracers.
3.3.1 Methane and tracer emissions

Sandtown Landfill is composed of five different cells. Two cells have a final cover on them, and the emissions from these cells are negligible. Two cells are entirely intermediate cover, and the remaining cell has some intermediate cover and a portion that is daily cover (Delkash et al., 2016). Measurements of methane concentration on the day of the field campaign indicated that the majority of the Sandtown Landfill emissions originate from the daily cover area and adjacent intermediate cover regions where landfill gas extraction was minimal. In the model, this hot spot of emissions is set to emit 80% of the total mass of methane from the surface of the landfill and covers 17% of the surface area. The emissions in the hot spot are log-normally distributed with a geometric mean of 24.9 g/m²/day (16.0 g/grid cell/min) and a geometric standard deviation of 6.1 g/m²/day (3.81 g/grid cell/min). These statistics describe emissions data from newly placed waste with a 15-30 cm daily cover reported in a recent study (Abichou et al., 2006). Spatial correlation of emissions is neglected at the 30 m grid scale in the numerical simulations, since spatial correlation at this scale was small in other investigations (Spokas et al., 2003; Abichou et al., 2006). The emissions on the remainder of the landfill, which is intermediate cover, are log-normally distributed with a geometric mean of 3.4 g/m²/day (2.13 g/grid cell/min) and a geometric standard deviation of 2.8 g/m²/day (1.75 g/grid cell/min). These statistics describe emissions data at a site containing 14-year old waste with a 45 cm intermediate cover (Abichou et al. 2006). Log-normal distributions with the aforementioned statistics are randomly generated with Matlab for 51 grid points for the daily cover and 249 grid points for the intermediate cover, and the emissions rate at each grid point is scaled so that the total emissions rate from all 300 grid points is 1800 g/min, which is similar to the 2300 g/min emissions rate estimated over the same period at Sandtown Landfill (Delkash et al., 2016). The emissions rate of 1800 g/min falls within the range reported in an overview of whole-landfill measurements in Denmark, Sweden, and the USA (Mønster et al., 2015). In the model, the emissions at each grid point are constant in time. Two different realizations of the heterogeneous methane emissions were generated and simulated but differences were negligible, so results from only one realization are presented. The two tracers for the base case are located on two corners of the methane hot spot, forming a line roughly perpendicular to the wind direction and placed 127 m apart.

3.3.2 Reporting results

In the field, when mobile transect measurements are collected, the vehicle is typically traveling at a speed between 20 km/h and 30 km/h, with samples taken every 2 seconds (Mønster et al., 2014; Foster-Wittig et al., 2015). For the base case simulation in this work, we use a speed of 8 m/s, which is approximately 28.8 km/h. The distances from the midpoint of the upwind edge of the methane hot spot to the transects examined are 750 m, 1000 m, 1500 m, and 2000 m (see Figure 4). Based on analytical models describing Gaussian plume dispersion, tracer dilution method measurements should become more accurate with
distance downwind from the landfill if the tracer release and area of methane emissions are not perfectly aligned (Mønster et al., 2014). However, in experimental field tests over flat topography, measurement error decreased with distance when the tracer was placed upwind of the methane emissions but increased with measurement distance when the tracer was displaced from the methane source laterally. Therefore, the expected decrease in error with measurement distance may not be universal but rather a function of topography, local wind conditions, and misalignment of the tracer release points and methane emissions. This question can be investigated by simulating the tracer dilution method with atmospheric dispersion modeling at this landfill.

To simulate the tracer dilution method, 1680 m long transects are recorded at each transect distance, each traverse of each transect path taking 210 seconds to complete. Landfill methane emissions rates are predicted for transects at these locations over a period of one hour. The first hour of the 4-hour period shown in Figures 3.2 and 3.3 is used for the tracer dilution method simulations due to limitations in computational time and output file size. Figure 3.4 shows the concentrations of methane and tracer gas for a single traverse of each of the four transect locations, collected from 12:21:00 to 12:24:30 LST at approximately 8 m above the ground.

The right panels of Figure 3.4 show that the methane and tracer concentration transects at each location are very similar to each other, but not identical, as expected since one comes from a large area source and the other comes from two point sources spaced 127 m apart. As the plumes move away from the landfill, in general the plumes widen and the maximum values decrease due to turbulent diffusion. In Figure 3.4, however, the peak concentration is higher at 2000 m than at 1500 m, demonstrating that atmospheric turbulence can cause the plumes to travel and disperse in a non-Gaussian manner. A single peak does not appear in the transect at even 2 km distance, because topographic effects and turbulent eddies cause the flow to deviate from an idealized Gaussian plume structure. Also note in Figure 4 that the concentration values in the left panels will not match the concentration values in the right panels because the left panels show instantaneous plumes at one time step while the right panels show a 2.5 minute transect over which the plumes are evolving with time.

The transects from the simulations have significantly less noise than transects collected by field instruments would have. This is because the model has 30 m resolution to represent a plume that is approximately 300 m wide at 1 km downwind from the source. In the field, a 2-second-resolution gas analyzer crossing a 300 m wide plume at 20 km/h would record almost three times as many data points, capturing significantly more spatial variability due to smaller scales of turbulence.

Model output is interpolated from the 30 m resolution grid to the four distances with 16 m spacing to be consistent with collecting data every 2 seconds at a speed of 8 m/s. A bilinear interpolation method is used to obtain a weighted average of the concentration values at the four grid points surrounding the point of interest. Methane emissions are calculated for each transect using Equation 1. The estimated emissions from each traverse are averaged over the entire one-hour period, and that average value is compared to the actual emissions
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Figure 3.4: Contours of instantaneous methane plume concentration (ppb) (top left), tracer plume concentration (ppb) from two point sources (bottom left), with terrain elevation contours (black lines, 5 m intervals) and locations of the four transect distances (dashed lines). Example transects of methane and tracer gas at each of the four transect distances (right panels).

rate to obtain a percent error, as shown in Equation 3.2.

\[
Error = \frac{Q_{TDM} - Q_{actual}}{Q_{actual}}
\]  

(3.2)

In Equation 2, \(Q_{TDM}\) is the average of the methane emissions rates calculated by each traverse of the transect over the one hour period and \(Q_{actual}\) is the total landfill methane emissions rate used in the model.

Two filtering criteria that have been used for the tracer dilution method in the field are applied to the model output: plume correlation and emission rate difference (Foster-Wittig et al., 2015). The plume correlation criterion is applied by performing a linear regression for each transect to find the R\(^2\) value that describes the correlation between the methane and tracer concentrations. The entire transect is discarded from the dataset if the R\(^2\) value is below 0.8. The emission rate difference criterion is applied by computing the methane
emissions using the integral method in Equation 1, \( Q_{\text{integral}} \), and then computing the methane emissions by multiplying the slope of the best fit line describing the relationship between methane and tracer concentration for each transect by the tracer emissions rate, \( Q_{\text{line}} \). The transect is discarded if the absolute value of the difference between \( Q_{\text{integral}} \) and \( Q_{\text{line}} \) is more than 20% of the average of \( Q_{\text{integral}} \) and \( Q_{\text{line}} \). Figure 3.5 shows that the filtering had a very small effect on the results for the base case simulation, with a maximum difference in the mean percent error of 4% and a maximum difference in the standard error of 2%. The filtering criteria does not filter out many transects because in the simulation the methane emissions are steady with time and there are not fluctuating background concentrations of methane and tracer gas. Both of these conditions are true in the field and will complicate the concentration transects seen downwind.

Figure 3.5: Time series of methane and tracer concentrations for transects collected at 1000 m downwind, over one hour (top left); calculated emissions for each transect shown in top panel compared to actual emissions prescribed in the model (bottom left); one-hour average of percent error as a function of distance from the upwind edge of the methane hot spot for filtered (both R2 and ERD criteria applied) and unfiltered data with standard error as a percentage of average total emissions (right).

The base case simulation examines four different transect locations, and accuracy of the tracer dilution method is found to improve from 34% error at 750 m to 14% error at 2000 m. This occurs because as the plumes of gas move further downwind, they experience more mixing, so the effects of the differences in source size become less important and the plumes of methane and tracer gas will behave more similarly. This makes the ratio of the transect integrals closer to the ratio between the source strengths, which is what the tracer dilution method is estimating. Figure 5 also shows that the base case simulation over-predicts the methane emissions. This is the case because the tracers are located upwind of the centroid of the methane hot spot, so the tracer concentration will be slightly more diffuse than the methane concentration when they are measured at the same downwind distance. The tracers
are also approximately 45m in elevation while the methane hot spot spans the sloped face of the landfill from 25m to 45m. This elevation difference between the two emission sources affects how they mix vertically. This difference in vertical mixing results in the tracer gas being more diffuse than the methane at the location of the measurements 8 m above ground level.

### 3.4 Sensitivity to transect orientation and speed

There are many factors about the execution of the tracer dilution method that can be changed, and sensitivity to these factors has not previously been explored systematically. In this section, the factors studied are the angle of the transects relative to the wind direction and the speed of transects.

In reality, the transect locations for the tracer dilution method are limited by the availability of roads, which may not be located perpendicular to the wind direction during the measurement period. The midpoints of the four transect distances shown in Figure 3.4 are used as the midpoints of the misaligned transects. The base case transect angle is the angle seen in Figure 3.4, roughly perpendicular to the wind direction. An additional four transect angles are simulated at each of the four distances: $+30^\circ$, $+45^\circ$, $-30^\circ$, and $-45^\circ$, all relative to the base case transect locations. Transect vehicle speed remains constant at 8 m/s, as in the base case. Misaligned locations centered at 1000 m from the hot spot are presented in Figure 3.6, along with sample concentration transects at each misaligned transect location.

![Figure 3.6: Contours of instantaneous tracer concentration (ppb) with locations of angled transects relative to original 1000 m transect location (left); sample transects of methane and tracer concentration at different angles (right).](image)

For the example traverse of the transect locations, it appears all locations capture adequate cross-sections of the tracer plume. The transects recorded for methane and tracer gas
are similar to each other for each transect angle, though small differences between the two plumes are perceptible. Figure 3.7 presents the percent error of each transect alignment at each transect distance averaged over the 17 traverses completed in one hour. The transect at +45° shows the largest error for all transect locations except 2000 m, but all errors are below 32%.

Figure 3.7: One-hour average of percent error of predicted landfill emissions as a function of distance from the upwind edge of the landfill hot spot for the five transect angles.

In the field, when mobile transect measurements are collected, the vehicle is traveling at a speed between 20 km/h and 30 km/h, so the concentration measured at each point in space along the transect will be recorded at a different time according to the speed of the vehicle. For this work, three speeds are tested: 15 km/h, 30 km/h, and 40 km/h. These different speeds are all simulated using the base case transect locations shown in Figure 3.4, and the measurement period is one hour for all speeds so that the numbers of transects recorded are 10, 17, and 27 respectively.

From Figures 7 and 8, it is clear that neither transect angle nor transect speed makes a large difference in the accuracy of the tracer dilution method. These figures show that error does decrease with distance from the hot spot to the transect location, although relative to the magnitude of the errors the decrease is small. Transect speed should not affect the error significantly because at all four transect locations the plumes being measured are several hundred meters wide, so even the coarsest sampling resolution of 22 m every 2 seconds will record enough points within the plume to adequately capture its shape. Additionally, the
accuracy of the tracer dilution method relies on the relationship between the methane plume and tracer plume, and this relationship is not expected to change when the transect speed changes. One notable difference between the results for different transect speeds is that the standard error decreases as speed increases. This is seen because the faster transect speed results in a larger number of transects collected over the one-hour sampling period, so a larger number of samples is likely to have a smaller standard error. This result is consistent with the work by Mønster et al. (2014), in which they found that uncertainties in emissions decreased by 50% when the number of traverses increased from 5 to 20.

It is noteworthy that, similar to the base case results in Figure 3.5, Figures 3.7 and 3.8 show that the tracer dilution method over-predicts methane emissions for every simulation at every transect distance from the methane hot spot. This is because the tracers are located on the upwind edge of the methane hot spot, as discussed further in the following section.

### 3.5 Sensitivity to tracer placement

Five tracer configurations are chosen to test the sensitivity of the emissions estimates to the sensor placement relative to the hot spot. Tracers placed on the upwind edge of the hot spot are used as the base case simulations for comparison of ground-level tracer concentration
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contours and tracer dilution method results.

Figure 3.9: Tracer setups simulated. Colored contours show the emissions rates at each grid cell on the surface of the landfill; circles show location of tracer emissions; black contour lines show elevation of landfill with 5 m intervals.

Figure 3.10: Schematics of plumes of methane and tracer gas for tracers on the upwind edge (left), downwind edge (center), and center of the hot spot (right). Tracer plume is shown in green, methane plume in red. Red $t_1$ denotes the edge of the methane plume at a given time and the green $t_1$ denotes the edge of the tracer plume at the same time.

Considering simplistic plume behavior, it is expected that if the tracers are located upwind of the centroid of the hot spot, the tracer gas will take longer to travel to the same downwind distance as the methane (see Figure 3.10, left), so the tracer concentration will be
more diluted than the methane concentration and will cause over-prediction of the methane emissions, according to Equation 3.1. Similarly, if the tracers are downwind of the centroid of the hot spot, the methane concentration will be more diluted than the tracer concentration (Figure 3.10, middle), so the tracer dilution method will under-predict the methane emissions. Therefore, if the tracers are located at the center of the hot spot, the estimated methane emissions should be the most accurate (Figure 3.10, right). This analysis does not consider atmospheric or site-specific effects. In addition, in reality it is usually not practical to place sensors in the middle of the active cover area of the landfill.

![Figure 3.11: One-hour average of percent error of predicted landfill emissions as a function of distance from the upwind edge of the landfill hot spot for the five tracer setups.](image)

Similar to the results in Figures 3.7 and 3.8, the results in Figure 3.11 show that error usually decreases as the transect distance from the edge of the hot spot increases. This finding is consistent with results reported in Mønster et al. (2014) that showed emission estimation errors decreased from 36% to 20% as the measurement distance increased from 370m to 1200m. Furthermore, placing the tracers in the center of the hot spot results in the smallest error, as expected. In the field it is not generally feasible to put tracers in the active area of the landfill, but placing tracers on both the upwind and downwind edges does not appear to be a good proxy for placing them in the middle in this case. Over-prediction of methane emissions from the tracers on the upwind edge combined with under-prediction from the tracers on the downwind edge should result in very accurate methane emissions.
estimates according to the Gaussian plume sketches in Figure 3.10. Figure 3.11 also reveals that there is an over-prediction bias for all of the configurations except the center of the hot spot. Most interestingly, placing the tracers on the downwind edge results in over-prediction instead of under-prediction. This could be due to a number of external factors, two of which are examined in the next section: topography of the landfill, and wind direction.

3.6 Sensitivity to external factors

3.6.1 Topography

To explore the reasons for the over-predictions of both the upwind and downwind tracer locations, external factors are explored. First, the effect of complex topography on the tracer dilution method is examined, by smoothing the terrain of the landfill in the model to approximately match the topography of the surrounding area (essentially removing the hill which forms the landfill). The maximum elevation of the landfill is 43 m, while the area surrounding the landfill varies between 17 and 18 m. This test scenario is simulated because previous work on the accuracy of the tracer dilution method was done on a flat field and the results were assumed to be relevant for landfill terrain (Mønster et al., 2014). The simulation is run with this new topography for the same date and time period as the original simulation so that the wind patterns are approximately the same, but the flattened topography reveals whether the dispersion behavior of the gas plumes is affected by the landfill topography itself. Methane emissions are prescribed for the same area of grid points and with the same distribution as described in Section 3.3.1. This flattened area of methane emissions is indicated by the black dashed line in Figure 3.12(a).

Figure 3.12 shows that placing the tracers on the downwind edge does result in an under-prediction of methane emissions, indicating that the over-prediction bias seen in Figure 3.11 could be due to topographic effects. This is because the methane hot spot is on a sloped surface that spans a range of elevations, and the way the tracer configurations are set up in Figure 3.9, the tracers will be released from a single elevation, so the two gas plumes will disperse vertically in different ways. This difference in vertical dispersion will affect how the two gas plumes reach the gas analyzer at the ground level, yielding less accurate results for the tracer dilution method than when it is performed on flat terrain. Given that the results of the numerical simulations in Figures 3.11 and 3.12 were significantly different, extrapolating results from the tracer dilution method on a flat field to the tracer dilution method on an area with complex terrain such as a landfill may not be possible.

3.6.2 Wind direction

To examine whether wind direction plays a role in the accuracy of the tracer dilution method, the method is simulated for a different day, October 22, 2015 from 12:00 to 13:00 Local Standard Time. On this day at Sandtown Landfill, the mean wind direction was 47
CHAPTER 3. NUMERICAL SIMULATIONS TO ASSESS TRACER DILUTION METHOD FOR MEASUREMENT OF LANDFILL METHANE EMISSIONS

Figure 3.12: Contours of tracer concentration (ppb) showing height contours in black at 1 m intervals with locations of transects in black dashed lines and locations of base case tracers in pink circles (top); one-hour average of percent error of predicted landfill emissions as a function of distance from the upwind edge of the landfill hot spot for the five tracer setups.

with a standard deviation of 7.6, and the mean wind speed was 4.2 m/s with a standard deviation of 0.3. This date and time has similar slightly unstable conditions to the other date simulated in the previous sections. The upwind edge tracer setup is shown in Figure 13. For both wind direction simulations, the objective is to place the tracers on the corners of the upwind edge of the methane hot spot, so due to the shift in wind direction, the edge that is upwind shifts by 90 as well. Lastly, because the tracers are placed on the corners of the daily
cover area in both simulations and the daily cover area is not square, the tracers are further apart in this simulation than in the previous simulations (170 m and 127 m respectively).

Figure 3.13: Contours of tracer concentration (ppb) showing height contours in black at 5 m intervals with locations of transects in black dashed lines and locations of base case tracers in pink circles (top); one-hour average of percent error of predicted landfill emissions as a function of distance from the upwind edge of the landfill hot spot for the five tracer setups.

These results also show under-predicted methane emissions for the case in which the tracers are on the downwind edge. Additionally, the percent errors in Figure 3.13 have smaller magnitudes than the results in Figure 3.11. In Figure 3.14, a schematic of the two
wind directions relative to the sloped surface on which the hot spot lies helps to explain the role of wind direction and topography in the accuracy of the tracer dilution method.

Figure 3.14: 3-D surface plot of landfill terrain height with 10:1 vertical exaggeration showing original wind direction (top) and new wind direction (bottom). Grid cells are colored according to emissions rate (g/min/grid cell).

In Figure 3.14, a schematic of the two wind directions relative to the sloped surface on which the hot spot lies helps to explain the role of wind direction and topography in the accuracy of the tracer dilution method. The relationship between wind direction and sloped surface of the methane hot spot affects the accuracy of the tracer dilution method in this particular case because the methane and tracer concentration measurements are taken at ground level. The fact that the two plumes are spreading differently in the vertical direction means that the ratio of the measured concentrations at the ground level will not reflect the ratio of the emissions rates as accurately as if the two gasses had been released at the same elevation, which would cause them to spread similarly in the vertical direction. Therefore,
CHAPTER 3. NUMERICAL SIMULATIONS TO ASSESS TRACER DILUTION METHOD FOR MEASUREMENT OF LANDFILL METHANE EMISSIONS

wind direction relative to the slope of the landfill and the location of the active landfill area is an important external factor affecting the accuracy of the tracer dilution method. If the wind direction is not optimal when the concentration measurements are taken, the tracer dilution method could yield biased results and larger error.

3.7 Conclusions and future work

The goal of this paper was to explore, with atmospheric dispersion modeling, how different factors affect the accuracy of the tracer dilution method. Several open questions about the tracer dilution method were investigated, namely: how do distance and orientation of the transect location, vehicle speed, tracer placement, topography, and wind direction affect the accuracy of the methane emissions predicted by the method? The use of a numerical model with high-resolution atmospheric dispersion capabilities and realistic meteorological and topographical features is a novel way to address these questions, which are difficult to assess systematically in a field campaign setting. This work is unique in that it is the first to use atmospheric simulations to examine the impact of transect orientation, vehicle speed, topography, and wind direction on measurement error.

The results of this work are consistent with previous studies that show that error in emissions estimates generally decreases with increasing transect distance downwind. Placing the tracers as close to the center of methane emissions as possible minimizes this error. The mean percent errors reported range from -12% to 42%, with the large errors seen when the tracers are upwind of the methane hot spot, when the transect is located closest to the methane hot spot (at 750 m). This error range is specific to this particular study, and is meant to give an example of the error range to be expected using the tracer dilution method. Large errors are also found when the tracers are released at a single elevation while the methane hot spot spans a range of elevations along the sloped side of the landfill. The measurement errors reported in this work strictly apply only to Sandtown Landfill; however, the magnitudes of the errors reported are similar to those reported in other controlled release studies on flat terrain: 2±6% to 36±21% (Mønster et al., 2014) and 3.7 to 19.2% (Babilotte et al., 2010).

The results of this work indicate that changes in transect alignment relative to wind direction and transect vehicle speed do not have a significant effect on the accuracy of the tracer dilution method, as long as the entire plume is captured in the transect. Tracer placement relative to the methane hot spot, however, can have significant impact on the accuracy of the methane emissions estimated by the tracer dilution method. Additionally, the roles of two external factors, topography and wind direction, on the accuracy of the tracer dilution method were examined. Since topography affects wind flow near the ground surface, it affects plume dispersion and therefore whether the tracer dilution method over- or under-predicts the methane emissions. Lastly, wind direction relative to the elevation and terrain slope of the hot spot can also greatly affect the error of the estimated methane emissions. Future work will focus on using more realistic unsteady methane emissions as a function of
soil processes and surface wind speed to examine diurnal and seasonal variability in landfill methane emissions. The methodology developed in this work could also be extended to study greenhouse gas emissions from any multitude of sources, such as animal feedlots and wastewater treatment plants.
Chapter 4

Atmospheric modeling to assess wind dependence of landfill methane emissions TDM measurements

The short-term temporal variability of landfill methane emissions is not well understood due to uncertainty in measurement methods. Significant variability is seen over short-term measurement campaigns with the tracer dilution method (TDM), but this variability may be due in part to measurement error rather than fluctuations in the actual landfill emissions. In this study, landfill methane emissions and TDM-measured emissions are simulated over a real landfill in Delaware, USA using the Weather Research and Forecasting model (WRF) for two emissions scenarios. In the steady emissions scenario, a constant landfill emissions rate is prescribed at each model grid point on the surface of the landfill. In the unsteady emissions scenario, emissions are calculated at each time step as a function of the local surface wind speed, resulting in variable emissions over each 1.5-hour measurement period. The simulation output is used to assess the standard deviation and percent error of the TDM-measured emissions. Eight measurement periods are simulated over two different days to look at different conditions. Results show that standard deviation of the TDM-measured emissions does not increase significantly from the steady emissions simulations to the unsteady emissions scenarios, indicating that the tracer dilution method may have inherent errors in its prediction of emissions fluctuations. Results also show that TDM error does not increase significantly from the steady to the unsteady emissions simulations. This indicates that introducing variability to the landfill emissions does not increase errors in the the tracer dilution method. Across all simulations, TDM errors range from -15% to 43%, consistent with the range of errors seen in previous TDM studies. These TDM simulations help inform landfill researchers using the TDM in the field about the types of error expected under various conditions.
4.1 Introduction

Landfill methane emissions are estimated to be the third largest anthropogenic contributor to atmospheric methane, generating this potent greenhouse gas as the waste decomposes to create optimal conditions for methanogenic archaea (U.S. EPA, 2014). The magnitude of landfill methane emissions and their variability from any given landfill are still not well understood. To quantify landfill methane emissions, several emission measurement methods have been suggested. These methods can be differentiated by their measurement footprint. Flux chambers (Reinhart et al., 1992) have the smallest footprint, typically 1 m$^2$, while the radial plume mapping-based mass balance (U.S. EPA, 2006) and eddy covariance techniques (Tregoures et al., 1999) have footprints that range from 100-10,000 m$^2$. Emissions from entire landfills may be measured with the tracer dilution method (Czepiel et al., 1996), aircraft-based mass balance approaches (Wong and Wyles, 2012; Cambaliza et al., 2015), and differential absorption lidar (Robinson et al., 2011). Generally, methods with the ability to measure emissions from the entire landfill are more desired, since they are less dependent on assumptions about spatial variability of emissions from the landfill surface when calculating whole landfill methane emissions from multiple, smaller-scale data. Whole landfill emission measurements, however, require a well-mixed methane plume to be sampled downwind of landfills. The accuracy of the whole landfill measurement methods is difficult to assess because true landfill emissions are unknown.

The tracer dilution method (TDM) for whole landfill emissions has gained popularity in the past several years (Mønster et al., 2014; Mønster et al., 2015; Foster-Wittig et al., 2015; Delkash et al., 2016). This technique involves releasing a tracer gas on the surface of the landfill at a known, steady rate. Then concentration measurements of both methane and the tracer gas are collected downwind of the landfill, often by mounting the gas analyzer onto a vehicle to collect transects of the plumes. After data collection, each transect pair of methane and tracer gas measurements is used to calculate the landfill methane emissions with Equation 4.1:

$$Q_{CH_4} = Q_{tr} \frac{M_{CH_4} \int c_{CH_4}(x)dx}{M_{tr} \int c_{tr}(x)dx}$$  \hspace{1cm} (4.1)$$

where $Q_{tr}$ is the known tracer emissions rate in g/min, $M_{CH_4}$ and $M_{tr}$ are the molecular weights of methane and tracer gas, and $c_{CH_4}(x)$ and $c_{tr}(x)$ are the methane and tracer gas concentrations along the transect in ppb. Measurements can be recorded about every 2 seconds with cavity ring-down spectroscopy (CRDS) instruments, with the vehicle typically traveling between 20 km/h and 30 km/h in recent field campaigns (Mønster et al., 2014; Foster-Wittig et al., 2015).

Emissions measurements from TDM field campaigns have shown significant short-term variability. Field measurements of 15 Danish landfills by Mønster et al. (2015) reported emissions measurements with standard deviations ranging from 9.8% to 37% of the mean for measurement periods ranging from 2 to 10 hours. Analysis of field measurements at a landfill in Delaware, USA by Delkash et al. (2016) showed variability of up to 8-fold between the
minimum and maximum emissions measurements. Additional work by Mønster et al. (2014) has shown that the TDM can yield variability in measured emissions over short time scales even when the methane is from a controlled, steady release. An open research question is how much variability in TDM-measured emissions is due to variability in the landfill methane emissions compared to inherent variability due to the TDM method.

Previous work by Taylor et al. (2016) used atmospheric modeling to simulate the TDM at a real landfill over a 1-hour period using steady landfill methane emissions and found that simulated TDM measurement standard deviations were up to 22% of the mean, consistent with Mønster et al.’s findings (2015). This significant variability in simulated TDM measurements was surprising given that the true landfill emissions in the model were constant with time, but is consistent with the findings of Mønster et al. (2015) mentioned above. The Taylor et al. (2016) study also compared TDM error for various factors of the TDM setup such as tracer location and transect distance, as well as external factors such as topography and wind direction. When the mean TDM-measured emissions were compared to the true model emissions, errors ranged from -12% to 42% across all simulations, indicating that the TDM-measured emissions could be systematically biased under certain conditions and also that certain conditions could yield more accurate measurements than others.

Numerical modeling is advantageous for studying the TDM for several reasons. First, the emissions rate for the landfill methane is specified in the model, so the emissions are known. It is therefore possible to compare the TDM-calculated emissions to the true model emissions, giving an idea of how much error to expect when analyzing the TDM-measured emissions from the field. Additionally, in the field the TDM is a labor-intensive process that is not easily repeatable for testing many different scenarios, e.g. selection of tracer release points. With numerical modeling, it is easy to simulate many different scenarios over the same time period and same landfill.

In this work, the Weather Research and Forecasting model (WRF) is used to examine the impact of wind on methane emissions and landfill TDM measurements. WRF simulations also describe the turbulent wind patterns and resulting atmospheric transport of methane and tracer gas. Gaussian models have been applied to study atmospheric dispersion in the context of landfill methane emissions, but Gaussian models neglect the complexities of real atmospheric dispersion that arise from turbulence, heterogeneous and unsteady wind patterns, and complex topography, unlike WRF.

Previous work by Delkash et al. (2016) showed a correlation between wind speed and landfill methane emissions, with an R^2 of 0.35 for ground measurements and R^2 of 0.56 for measurements aloft (85 m). This work therefore uses a wind-dependent emissions parameterization in WRF that introduces temporal variability into the modeled landfill methane emissions. To assess short-term variability in TDM-measured emissions associated with wind, TDM simulations from both steady and unsteady emissions simulations are compared. Different times of day and two different tracer placements are also explored to assess the range of variability and TDM error expected in the field.
4.2 Methods

4.2.1 Weather Research and Forecasting Model

The Weather Research and Forecasting Model (WRF) is a mesoscale meteorological model widely used for atmospheric research and operational forecasts (Skamarock et al., 2008). It has capabilities for both planetary boundary layer (PBL) schemes at lower grid resolutions which parameterize all turbulence, and large eddy simulations (LES) at higher grid resolutions which explicitly resolve larger turbulent eddies and parameterize the smaller eddies. The boundary conditions used in the numerical simulations presented in this work come from the North American Mesoscale (NAM) model at 12 km resolution. The simulations use a nested grid approach to run at 30 m resolution without accruing large interpolation errors from the 12 km boundary conditions data. The outer grid is 202.5 km across with 2.25 km horizontal resolution using the Yonsei University (YSU) PBL scheme, the middle grid is 45 km across with 150 m resolution, and the smallest grid is 9 km across with 30 m resolution. A time step of 0.46 seconds is used for the smallest domain, and 75 vertical grid levels are used for all domains with approximately 15 m grid spacing at the ground. Both the middle and smallest grids use LES with the TKE-1.5 turbulence closure with nonlinear backscatter and anisotropy (NBA). This combination of PBL scheme and LES turbulence closure was chosen because it produces meteorological output that best matches the observation data from the field site being modeled.

The WRF model includes coupling between the meteorological component and the Noah land surface model (LSM), which calculates subgrid-scale fluxes of sensible and latent heat to drive atmospheric boundary layer growth. The Noah LSM takes as input land-use (vegetation) type, soil texture, and slope of the ground surface. These inputs inform variables such as albedo and leaf area index (LAI). Thus, the modeling accounts for regional wind patterns driven by large-scale pressure gradients, and local-scale turbulence influenced by topography, solar radiation, soil, and vegetation. This WRF model configuration is identical to that used in the previous study by Taylor et al. (2016), except that a wind-dependent methane emission model is added that is described further below.

4.2.2 Sandtown Landfill

Sandtown Landfill in Delaware, USA was the site of a field study (Delkash et al., 2016) that is used to inform and validate this numerical study. Sandtown Landfill has been previously studied to determine wind dependence of methane emission, find optimal tracer gas placement relative to the hotspot for TDM, and to examine transect sampling approaches for TDM (Delkash et al. 2016, Taylor et al. 2016). During the field emission measurements at Sandtown Landfill, a strong hot spot for methane emissions occurred in a region of daily cover that overlaid older refuse. In addition, the terrain surrounding the landfill is quite flat, which means the prominence and topography of the landfill have a significant effect on the local wind patterns and therefore on the transport of atmospheric methane. Figure 1 shows
CHAPTER 4. ATMOSPHERIC MODELING TO ASSESS WIND DEPENDENCE OF LANDFILL METHANE EMISSIONS TDM MEASUREMENTS

a Google Earth image of Sandtown Landfill (left) and a section of the smallest WRF grid used in the simulations (right). The terrain height is indicated with black contour lines, and a snapshot of surface wind vectors and methane emissions shows their spatial heterogeneity. The rectangular emissions hot spot region is collocated with the daily cover area.

Figure 4.1: Google Earth image of Sandtown Landfill (left) and section of simulated Sandtown Landfill (right) with terrain height (black contours, 5 m intervals), instantaneous surface winds (blue arrows, every fifth wind vector shown), and instantaneous methane emissions (colored contours, g/min/m²).

4.2.3 Methane surface flux parameterization

This work uses a surface flux drag law to parameterize landfill methane emissions to introduce wind dependence into the modeled emissions. Simulating unsteady landfill methane emissions will help assess the effect of short-term wind variability on landfill emissions as well as their effect on TDM measurements and TDM error. The focus of this work, however, is on the atmospheric dispersion modeling used to simulate the TDM, so the landfill emissions model is not intended to necessarily reproduce the emissions seen in the field. The landfill emissions model is relatively simple with the purpose of understanding how TDM measurements and TDM accuracy are affected by a time-varying landfill emissions rate.

Wind-dependent methane emissions

Typically, landfill methane emissions models are driven by soil processes and not by surface wind. This is the case for the California Landfill Methane Inventory Model (CALMIM), a model that has gained popularity in recent years due to its novel focus on soil and climate dependencies for landfill emissions (Spokas et al., 2011; Spokas et al., 2015). The CALMIM modeling approach, however, accounts for only soil processes affecting the emissions and neglects surface wind effects. Several recent field studies have shown correlations
between wind speed and landfill emissions, measuring emissions that far exceed those due to molecular diffusion. For example, Poulson and Moldrup (2006) showed that wind-induced pressure variations drove gas transport that accounted for approximately 40% of the total gas emissions at their site during their measurement period. McBain et al. (2005) measured averages of 30-min wind speeds and methane emissions from part of a landfill. They found a weak correlation \(R^2=0.48\) and attributed this to diminishing interfacial top layer depth, which leads to an increase in the molecular diffusion in topsoils. This correlation was higher \(R^2=0.55\) for wind speeds between 1.3 and 5.8 m/s. Finally, Delkash et al. (2016) reported results from a TDM field campaign at Sandtown Landfill accompanied by atmospheric modeling that indicates that variations in TDM-measured emissions may be caused by the wind dependence of landfill emissions.

Given that several field studies show emissions dependence on wind speed, this work attempts to improve upon CALMIMs emissions model by including wind effects. We do so by formulating our emissions model as a drag law parameterization analogous to the parameterization WRF uses for heat and moisture fluxes.

**Surface flux**

Mass transfer of methane from landfills to the atmosphere can be viewed as a series of resistances: cover soil resistance, the quasi-laminar surface layer, and turbulent surface layer resistance (Pleim and Ran, 2011). If molecular diffusion is the only process driving transport in the soil, then resistance in the soil should be much more significant than resistance in the atmosphere. This is the underlying hypothesis for CALMIM, however, variations in wind speed at the ground surface can induce small pressure gradients that enhance diffusion in the soil. Several recent studies show that short-term wind speed variations can increase upward gas transport in soil from 20 to 70 times (Pourbakhtiar et al., 2017) over that from molecular diffusion alone. Thus, using CALMIM without accounting for wind effects may underestimate landfill emissions in some situations particularly daily cover regions with thin soil covers. While the development of mechanistic models to describe such wind effects on overall transport of gas from soils to the atmosphere is ongoing, atmospheric modelers have developed first-order mass transfer models to describe this process.

The first-order mass transfer model employed in WRF’s land-surface model Noah describes mass transfer from the upper soil layer to the atmosphere. The flux is assumed to be controlled by wind: emissions are zero when wind speed is zero. The approach followed in this work is widely used in land-atmosphere models and we believe is also applicable to landfill covers for modeling short-term variability due to wind effects. Future field studies are needed to fully test this hypothesis.

The landfill methane emissions are calculated at each time step using a surface flux parameterization based on those used by WRF for moisture, heat, and other scalars (Chen, 2007). In this case, the vertical flux of heat and moisture associated with wind is modeled with a first-order mass transfer model. A similar formulation is used to describe methane
emissions:

\[ Q_{CH_4} = C_d u^* (c_{soil} - c_{atm}) \] (4.2)

where \( C_d \) is a time-dependent drag coefficient, \( u^* \) is the shear velocity, \( c_{soil} \) is the soil methane concentration, and \( c_{atm} \) is the atmospheric methane concentration. In the WRF model, \( c_{atm} \) is in the middle of the lowest grid cell, about 7.5 m above the ground surface in this case. Additionally, \( c_{soil} \) is prescribed as a time-dependent model input, with the values obtained from CALMIM applied at 5 cm depth, the same depth the Noah LSM uses for heat and moisture fluxes in WRF.

This surface flux parameterization comes from the principle that a vertical flux \( Q_a \) of any scalar \( a \) is driven by the difference in \( a \) across the interface:

\[ Q_a = U_T (a_{below} - a_{above}) \] (4.3)

where \( U_T \) is the transport velocity across the interface, and \( a_{below} \) and \( a_{above} \) are the values below and above the interface respectively. When the interface is the ground surface, the transport velocity is parameterized as a bulk transfer coefficient times mean horizontal wind speed at height \( z \) above the surface. Since wind speed is zero right at the ground surface, the nonzero wind speed at height \( z \) will cause shear stress which generates turbulence to support the vertical transport of \( a \) (Stull, 1988).

Equation 4.2 can be adapted for any gas by scaling the bulk transfer coefficient for heat and moisture by the ratio of molecular diffusivity of the gas to molecular diffusivity of water vapor (Stull, 1988; Bonan, 2002; Pleim and Ran, 2011). To the author’s knowledge, this approach has not been used in the context of landfill methane emissions before. Further investigation into implementing the surface flux parameterization in the Sandtown Landfill simulations is presented in the next section.

**Implementing the drag law emissions model for Sandtown simulations**

This work implements Equation 4.2 in WRF. Emissions are shown in Figure 4.2 at three grid points as well as their corresponding breakdowns into the three variable quantities on the right-hand side of Equation 4.2 to see how they varied and which variables had the most influence on the variability of the landfill emissions. The simulation presented in this figure is the base case simulation, March 6, 2010 from 11:30 to 13:00 local standard time (LST). Soil methane concentrations used in these simulations were obtained by running CALMIM for Sandtown Landfill daily cover area with the following input parameters: latitude = 39.0293, longitude = -75.7131, soil type = clay loam, soil depth = 6 inches (≈ 15 cm), and oxidation turned on.

Figure 4.2 shows modeled methane emissions over a 90-minute period as well as the variability in \( C_d, u^* \), and \( (c_{soil} - c_{atm}) \) separately over the same time period. All values are normalized by the mean value of that variable to show the variability as a proportion of the mean. Values for three different grid points are shown to account for heterogeneity.
Figure 4.2: Normalized methane emissions (top row); coefficient $C_d$ (second row); $u^*$ (third row); $(c_{soil} - c_{atm})$ (bottom row).

From Figure 4.2 is it clear that the variability in $Q_{CH4}$ is larger than the variability in $C_d$ and smaller than the variability in $u^*$, and that the variability in $(c_{soil} - c_{atm})$ is imperceptible in comparison. The peaks and troughs in $Q_{CH4}$ match the peaks and troughs in $u^*$, while there appears to be an inverse relationship between $Q_{CH4}$ and $C_d$. This inverse relationship makes sense because $C_d$ is inversely proportional to $\Psi_H$, the stability correction term for heat from similarity theory, and $\Psi_H$ is directly proportional to $u^*$. Coefficients of variation of these variables are presented in Table 4.1.

Table 4.1 shows clearly that the variability in methane emissions is slightly less than the variability in $u^*$ and slightly more than the variability in $C_d$. The variability in $(c_{soil} - c_{atm})$ is so small that it is reasonable to assume that this change in concentration is not the main driver of the variability in emissions. The variability in $(c_{soil} - c_{atm})$ comes exclusively from variability in $c_{atm}$ due to surface emissions adding mass to the atmospheric grid cell and surface winds sweeping mass out of the grid cell. While $c_{atm}$ does vary significantly
Table 4.1: Coefficients of variation for modeled methane emissions ($Q_{CH4}$) and the three variables that go into calculating $Q_{CH4}$ for three different grid points.

<table>
<thead>
<tr>
<th></th>
<th>Point 1</th>
<th>Point 2</th>
<th>Point 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>COV of $Q_{CH4}$</td>
<td>0.074</td>
<td>0.099</td>
<td>0.063</td>
</tr>
<tr>
<td>COV of $C_d$</td>
<td>0.051</td>
<td>0.064</td>
<td>0.040</td>
</tr>
<tr>
<td>COV of $u^*$</td>
<td>0.114</td>
<td>0.136</td>
<td>0.101</td>
</tr>
<tr>
<td>COV of ($c_{soil} - c_{atm}$)</td>
<td>0.0007</td>
<td>0.0022</td>
<td>0.0033</td>
</tr>
</tbody>
</table>

compared to its own mean value, $c_{atm}$ is on the order of 100 ppb while $c_{soil}$ is on the order of 10,000 ppb, as predicted by CALMIM at 5 cm depth. Significant changes in $c_{atm}$, therefore, will not result in significant changes in the concentration difference, ($c_{soil} - c_{atm}$).

It is clear from Equation 4.2 that the values used for soil methane concentration will have a significant effect on the magnitude of the emissions and must be chosen carefully. To understand the magnitudes, variability, and depth profile of soil methane concentration, CALMIM was run with inputs matching the specifications of the daily cover portion of Sandtown Landfill. The value of soil methane concentration predicted by CALMIM at 5 cm depth (2 in) in the daily cover soil was used as the mean value to generate a log-normal distribution of soil methane concentration values (Abichou et al., 2006). Field data from Sandtown Landfill indicated that the daily cover area contributed approximately 80% of the total emissions, so the mean and standard deviation values for the intermediate cover area were chosen so that the whole intermediate region would contribute 20% to the total emissions. For the randomly generated log-normal distributions, the daily cover mean and standard deviation are 20,400 ppb and 4970 ppb respectively, and the intermediate cover mean and standard deviation are 2780 ppb and 2275 ppb respectively.

### 4.3 Simulation setup

#### 4.3.1 Simulating the tracer dilution method: base case

Simulating the TDM requires simulating the landfill methane emissions and the point source tracer emissions. To simulate steady landfill emissions, the heterogeneous distribution described in Taylor et al. (2016) is used, with an emissions rate prescribed at each grid cell on the surface of the landfill that remains constant with each time step. For the unsteady landfill emissions, the same heterogeneous pattern is used for the soil methane concentration that was used for the steady landfill emissions, but the values are scaled so that the mean soil concentration for the daily cover matches the 5 cm depth soil concentration calculated by CALMIM, and the intermediate cover area is scaled accordingly to contribute 20% of the total landfill emissions. The soil concentration values remain constant and are used at each time step to calculate a new methane emissions rates at each grid cell, yielding a slightly different total landfill emissions rate at each time step.
The WRF model provides a reasonable description of the real conditions at Sandtown and has been validated with field data of both the wind conditions and tracer gas concentrations (Taylor et al., 2016). On March 6, 2010, over the 4-hour observation period, the mean observed and simulated wind speed differed by 0.1 m/s, and the standard deviations differed by 0.2 m/s. The mean wind direction also agreed well between observations and simulations, with a $4^\circ$ difference, although the observations showed greater variability in wind direction than the simulation, with standard deviations differing by $30^\circ$. The decreased variability in simulated wind direction could be due to subgrid-scale features that the model could not capture, but overall the performance of the model is very satisfactory and captures the general behavior of the turbulent gas plumes.

For the base case simulation and all simulations except the alternative tracer placement discussed in sections 4.3.4 and 4.4.4, two point source tracers are located on the corners of the upwind edge of the daily cover area. The locations of these tracers for the March 2010 simulations are shown in Figure 4.3 with magenta circles. The tracers are 127 m apart for the March 2010 simulations and 170 m apart for the Oct 2015 simulations. The reason for the difference in tracer placement between the two simulations is that the wind direction shifted by about $90^\circ$, so the tracers are shifted accordingly to ensure they are on the upwind edge of the hot spot. In the March 2010 simulations, they are on the northwest edge of the hot spot, and in the Oct 2015 simulations, they are on the southwest edge of the hot spot, which is slightly longer (see Figure 4.4). The tracers are each given a steady emissions rate of 26 g/min for a total tracer emissions rate of 52 g/min, very similar to the 52.23 g/min used in the field campaign in March 2010 (Delkash et al., 2016; Taylor et al., 2016).

Previous work has shown that distance from the landfill has a small effect on the accuracy of the TDM measurements, so transect measurements are taken along two paths, at 1000 m and 2000 m from the landfill (Taylor et al., 2016). These transect paths are not located along existing roads in the area, they are intended to assess the effect of measurement distance on the TDM measurements. The transect paths are 2.05 km long, and the simulated vehicles travel along the paths at 8 m/s ($\sim 29$ km/h), collecting concentration measurements every 2 seconds, as CRDS instruments do in the field. The simulated vehicles traverses the transect paths 21 times over each 90-minute measurement period. Figure 4.3 shows the locations of the two transect paths for the March 2010 simulations in blue lines in the left panel. The right panels of Figure 4.3 show example concentration measurements for one traverse of the transect paths.

Note that in Figure 4.3, the plot on the left is an instantaneous snapshot of the tracer gas plume, while the time series on the right are collected over a 4-minute period over which the plume is continuously evolving, so the transects on the right cannot be compared to the values shown on the left. Both the plume snapshot and the example transects demonstrate the highly turbulent nature of the atmosphere transporting the gas downwind. It is clear that the true gas plumes are very different from the Gaussian plumes often used to explain the tracer dilution method (Mønster et al., 2014; Foster-Wittig et al., 2015; Czepiel et al., 2003).
4.3.2 Steady and unsteady methane emissions

Field TDM measurements and numerical simulations of TDM measurements have both shown significant standard deviations in the measured emissions values over short time periods (Taylor et al., 2016; Delkash et al., 2016; Foster-Wittig et al., 2015, Mønster et al., 2014). The variability seen in the TDM-measured emissions in the numerical simulations was surprising because the true landfill methane emissions used in the model were constant over time, indicating that the TDM may introduce false variability into the measured emissions.

To understand the effect of the TDM on variability of the measured emissions, TDM simulations with steady emissions and unsteady emissions are compared. The unsteady methane emissions are scaled to ensure that the mean emissions for the whole landfill equals the whole landfill steady emissions. The differences shown in the middle column of Figure 4 are therefore due to time variability in the unsteady emissions simulations rather than systematic differences between the steady and unsteady emissions simulations. In the top row of Figure 4.4, the steady emissions distribution is shown with one time step snapshot of the unsteady emissions distribution as well as the instantaneous difference between the two emissions fields. In the bottom row, the figure shows snapshots of the resulting methane plume concentration contours from the steady and unsteady emissions fields and the difference between the two plumes in ppb. All panels except the upper left are unique to one time step at 12:10 LST. The upper left panel is the same for every time step.
Figure 4.4 shows that at this particular time step, the unsteady emissions are significantly greater than the steady emissions in the upper corner of the daily cover area, but the majority of the grid cells have either very small or no difference. The bottom left and right panels show no perceptible difference between the two methane plume concentrations, but the bottom middle panel reveals some small differences, particularly at the landfill surface. The differences between the two methane plumes generally decrease as distance from the landfill increases and have essentially disappeared by the time the two plumes have reached the 2000 m transect line. This indicates that if these two methane plumes are used with the same tracer plume to calculate TDM-measured emissions, the calculations should yield very similar results.

4.3.3 Different times of day

The simulations presented in this work are run on March 6, 2010, when a TDM field campaign was performed at Sandtown Landfill, and October 21, 2015 to capture a different wind direction. The data from this field campaign was used to validate the WRF model output (Taylor et al., 2016). To assess time variability of methane emissions and TDM measurements at different times of day, the simulation continued to run into March 7, 2010. The four time periods evaluated are Early Afternoon (11:30 - 13:00), Late Afternoon (15:30
- 17:00), Night (23:00 - 00:30), and Morning (08:00 - 09:30). Figure 4.5 shows time series of the wind conditions at each time period.

Figure 4.5: Time series of wind speed (top) and wind direction (bottom) for the different time periods and different days evaluated.

Table 4.2: Means and standard deviations of wind speed and direction for the different time periods simulated.

<table>
<thead>
<tr>
<th></th>
<th>Speed (m/s)</th>
<th>Early Afternoon</th>
<th>Late Afternoon</th>
<th>Night</th>
<th>Morning</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mar 2010</td>
<td>4.9±0.8</td>
<td>5.9±0.8</td>
<td>5.8±0.1</td>
<td>5.1±0.6</td>
</tr>
<tr>
<td></td>
<td>Oct 2015</td>
<td>2.7±0.5</td>
<td>5.1±0.7</td>
<td>5.6±0.1</td>
<td>3.8±0.4</td>
</tr>
<tr>
<td>Direction</td>
<td>Mar 2010</td>
<td>326±13°</td>
<td>330±12°</td>
<td>321±1°</td>
<td>318±5°</td>
</tr>
<tr>
<td></td>
<td>Oct 2015</td>
<td>214±12°</td>
<td>218±10°</td>
<td>215±2°</td>
<td>223±4°</td>
</tr>
</tbody>
</table>

From Figure 4.5 and Table 4.2, we can see that Early Afternoon (11:30 - 13:00) has the most variability in both wind speed and wind direction, which in theory should create the most mixing and have a positive effect on the accuracy of the tracer dilution method, but may also contribute to more divergence between the behaviors of the methane and tracer plumes because they are not perfectly collocated. The Late Afternoon (15:30 - 17:00) periods are characterized by shifting trends in wind speed and wind direction over the 90-minute measurement periods. The Night (23:00 - 00:30) time period is the most notable
because there is so much less variability in both the wind speed and wind direction. The relatively high nighttime wind speed indicates neutral atmospheric stability (class D), so we expect mixing of the gas plumes to be suppressed but not inhibited. The lack of variability in the nighttime atmosphere can be attributed to the way WRF parameterizes turbulence with LES. LES resolves turbulent eddies that are larger than $\sim 6$ grid cells across, but the maximum size of turbulent eddies that can form is governed by the height of the atmospheric boundary layer (ABL), and at night the atmospheric stability increases so the height of the ABL decreases significantly (from $\sim 1$ km to $\sim 100$ m). With 30 m grid spacing, the turbulent eddies are too small to be resolved, so all turbulence is parameterized rather than resolved, resulting in more Gaussian-like gas plumes rather than turbulent plumes. Because of this known numerical issue with nighttime WRF simulations, the nighttime results should be regarded with some suspicion: WRF simulated turbulence is likely less than actual turbulence. They do not, however, need to be discredited altogether, because in reality there is still significantly less turbulence at night. In the morning surface heating leads to the growth of turbulent eddies, which become large enough to be resolved again partway into the morning measurement period ($\sim 08:10$ for March 2010 and $\sim 08:40$ for Oct 2015). It is noticeable that the fluctuations in wind speed and wind direction are smaller in the Morning than they were for the Early Afternoon and Late Afternoon periods. For further understanding of the stability and mixing during each time period, profiles of potential temperature are presented in Figure 4.6. The height of the ABL is shown by the change in slope of the potential temperature profile, indicating the maximum size of turbulent eddies during that time period.

4.3.4 Alternative tracer configuration

The base case tracer placement is used in most of the simulations presented in this work because previous work demonstrated that that particular configuration yielded more accurate TDM measurements (Taylor et al., 2016). In real field measurements, however, the tracers may not be placed in an ideal setup, so a second tracer configuration is simulated to examine the range of TDM errors with a less ideal tracer configuration. The alternative tracer configuration consists of three point sources placed 85 m apart in a line roughly perpendicular to wind direction, 191 m upwind of the daily cover area. This configuration was chosen to roughly span the width of the intermediate cover area, a plausible scenario in the field. Figure 4.7 shows the relative locations of the base case and alternative tracer configurations.
4.4 Results

4.4.1 Assessing the tracer dilution method’s accuracy

One of the major advantages of using numerical modeling to assess the tracer dilution method is that the model calculates and stores the values of the true landfill methane emissions. In the field, true landfill emissions cannot be known and therefore cannot be compared to the values measured by the tracer dilution method to understand how well the method predicts emissions. With the tracer dilution method simulations, we can compare the emissions values predicted by the measurement technique to the emissions values used in the model to create the plumes that are sampled downwind with the tracer dilution method.

To simulate the tracer dilution method, transects of both methane and tracer gas concentrations are collected with a simulated vehicle traveling at 8 m/s (29 km/hr). For each traverse of the transect path, the integrals of both concentration transects are computed with respect to distance along the path, and Equation 1 is used to calculate measured landfill emissions. This procedure results in an array of 21 emissions values over the 90-minute
Figure 4.7: Locations of the alternative tracers (red circles) and base case tracers (green circles) overlaid on colored contours of soil methane concentration (ppb) with terrain height contours in black lines (5 m spacing).

measurement period. Then the tracer dilution method emissions values are averaged, and the model emissions values are averaged. Figure 4.8 shows a graphical depiction of the calculations for one transect path and one measurement period for the March 2010 Early Afternoon (11:30 - 13:00) period with the base case tracer configuration.

In Figure 4.8, each peak in the top panel corresponds to one traverse of the transect path, and each traverse of the transect path corresponds to one TDM measurement on the bottom panel (blue circle). One feature of note in this figure is that WRF calculated some variability in the model landfill emissions (solid red line on bottom panel), but the TDM measurements indicate a much larger standard deviation than the actual standard deviation. The fact that the TDM-measured emissions show significantly larger variability than the actual model emissions is a point of interest that is further explored by comparing steady emissions simulations with unsteady emissions simulations in section 4.4.2. This figure also shows the TDM over-predicts emissions, as noted in the previous chapter.

The accuracy of the tracer dilution method is assessed by calculating the percent error of the tracer dilution method using Equation 4.3.

$$P_{\text{error}} = \frac{Q_{\text{TDM}} - Q_{\text{model}}}{Q_{\text{model}}} * 100$$

(4.4)
In Equation 4.3, $Q_{\text{TDM}}$ is the average of the emissions values measured by the tracer dilution method and $Q_{\text{model}}$ is the average of the emissions values calculated in WRF. For example, from Figure 4.8 we see that $Q_{\text{TDM}}$ is approximately 2300 g/min and $Q_{\text{model}}$ is approximately 1800 g/min, so the percent error for this time period and transect path is approximately 28%.

4.4.2 Steady and unsteady emissions

In comparing the TDM results of the steady and unsteady emissions simulations, we are interested in comparing both the variability of the TDM-measured emissions and the percent error of the mean TDM-measured emissions. Figure 4.9 compares modeled and TDM-measured emissions for the steady and unsteady emissions simulations measured at 1000 m and 2000 m. Note that the unsteady modeled emissions have been scaled so that the mean matches the value of the steady emissions. The magnitude of the blue bar can be compared to the magnitudes of both TDM-measured means, although the standard deviation of the blue bar only applies to the unsteady emissions.
CHAPTER 4. ATMOSPHERIC MODELING TO ASSESS WIND DEPENDENCE OF LANDFILL METHANE EMISSIONS TDM MEASUREMENTS

Figure 4.9: Mean and standard deviation of unsteady model emissions compared to TDM-measured emissions for steady and unsteady emissions simulations at 1000 m and 2000 m transects.

Figure 4.9 shows an over-prediction bias in the TDM-measured emissions compared to the model emissions for both steady and unsteady emissions simulations at both transect distances. It also shows that the standard deviations of the TDM-measured emissions are significantly larger than the standard deviation of the unsteady modeled emissions. The means and standard deviations of the TDM-measured emissions from the steady and unsteady emissions simulations are very similar to each other. Both transect distances show a slight increase in mean and standard deviation of TDM-measured emissions from steady emissions to unsteady emissions, indicating slightly larger TDM error for the unsteady emissions simulations, but compared to the magnitude of the emissions, the difference is almost imperceptible. The TDM errors are all very similar for the four simulated TDM campaigns, ranging from 21% (steady emissions simulation with 2000 m measurements) to 27% (unsteady simulation with 1000 m measurements).

Figure 4.9 shows that variability in the TDM-measured emissions is only slightly higher for the unsteady emissions simulation than the steady emissions simulation. It also shows that variability is slightly higher for both simulations at 2000 m than at 1000 m. This is a slightly surprising result because we expect the plumes to have experienced more mixing
at 2000 m, which should yield smaller concentration fluctuations at 2000 m than at 1000 m, which intuitively should be linked to fluctuations in TDM measurement accuracy. The increase in standard deviations from 1000 m to 2000 m is small, however, and turns out to be specific to this simulation, as shown in sections 4.4.3 and 4.4.4.

During a field experiment with a known point methane emission source and three different tracer configurations, Mønster et al. (2014) found that the errors do not exceed 6%. This value is less than the errors found here (24% for 1000 m measurement and 21% for 2000 m measurement). This discrepancy indicates that the size of source and/or tracer plays an important role in errors. Moreover, they found that the correlation between the errors and distance of measurement is unclear. The errors slightly increased as measure distance increased from 370 m to 770 m; however, the error decreased from 770 m to 1200 m, which is consistent with our finding.

Steady and unsteady emissions were also compared for the different time periods presented in section 4.4.3 and the different tracer configurations presented in section 4.4.4. These results were consistent with the findings in Table 4.3 as well as in sections 4.4.3 and 4.4.4.

**4.4.3 Different times of day**

Model emissions and TDM-measured emissions are compared from eight time periods over two different days. Unsteady model emissions for the different time periods are presented in Figure 4.10.

![Figure 4.10: Unsteady model emissions (g/min) for the eight simulated time periods.](image)

Figure 4.10 shows significant differences in the magnitudes and standard deviations across different times of day. Similar patterns during the same time of day are found on the different days, such as the decreasing trend during Late Afternoon. The magnitudes of the modeled
emissions are not the emphasis of this work and as such are not compared in detail. Lower emissions in October 2015 than March 2010 are expected, however, due to the lower wind speeds in October 2015 as shown in Figure 4.5. Standard deviations and percent errors of the mean TDM-measured emissions are presented in Table 4.3.

Table 4.3: Standard deviations of TDM-measured emissions (g/min), percent error of the TDM-measured emissions in parentheses.

<table>
<thead>
<tr>
<th></th>
<th>Early Afternoon</th>
<th>Late Afternoon</th>
<th>Night</th>
<th>Morning</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mar 2010</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1000 m</td>
<td>478 (27%)</td>
<td>509 (18%)</td>
<td>94 (3%)</td>
<td>212 (10%)</td>
</tr>
<tr>
<td>2000 m</td>
<td>543 (23%)</td>
<td>409 (13%)</td>
<td>66 (2%)</td>
<td>133 (3%)</td>
</tr>
<tr>
<td>Oct 2015</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1000 m</td>
<td>291 (5%)</td>
<td>312 (14%)</td>
<td>49 (-1%)</td>
<td>275 (1%)</td>
</tr>
<tr>
<td>2000 m</td>
<td>132 (1%)</td>
<td>298 (-15%)</td>
<td>39 (-1%)</td>
<td>172 (-1%)</td>
</tr>
</tbody>
</table>

Table 4.3 shows that standard deviation of TDM-measured emissions actually decreases from 1000 m to 2000 m in all cases except Early Afternoon in March 2010. This result aligns with our expectations, explained earlier in section 4.4.2. Table 4.3 also shows that during the day the standard deviations increase with time of day, with the lowest values in the Morning, larger values in the Early Afternoon, and the largest values in the Late Afternoon. Standard deviations do not, however, seem to correlate with the level of atmospheric stability, since in the Late Afternoon for both days the atmospheric stability is higher but the standard deviations are also higher. The large standard deviations during the Late Afternoon period may be due to the steadily changing wind speed in both Late Afternoon periods (see Figure 4.5). Steadily changing wind speed will result in steadily changing total landfill emissions due to the drag law parameterization, so the ratio of landfill emissions to tracer emissions is constantly changing over time, making it less likely that individual measurements will be close to the mean.

Table 4.3 also shows that errors are generally significantly smaller for the Oct 2015 simulations than the March 2010 simulations. This is consistent with the results from the previous work by Taylor et al. (2016), and the result is explained in detail in that work. Briefly, it is because the daily cover area is located on the sloped side of the landfill, and in the March 2010 simulations, the wind is directed down the slope of the landfill so the upwind edge where the tracers are placed is at a higher elevation than the centroid of the daily cover area, while in the Oct 2015 simulations, the wind is directed across the slope, so when the tracers are placed on the corners of the upwind edge of the daily cover area, the tracers span the same range of elevations as the daily cover area does. Changing the elevation of the emissions release will naturally have a significant impact on the concentrations that are measured at the ground level, which is about 30 m lower than the peak of the landfill.

Results from these same time periods with steady emissions simulations yielded slightly smaller standard deviations than the results presented in Table 4.3, with standard deviations increasing slightly from the 1000 m transects to the 2000 m transects. This increase in standard deviation with increased distance from the landfill is inconsistent with the unsteady
emissions simulations results except for Early Afternoon in March 2010. This could be an indication that there is no true correlation between standard deviation of TDM-measured emissions and measurement distance from the landfill. Percent errors for the steady emissions simulations were slightly smaller than the errors presented in Table 4.3, with error decreasing slightly as transect distance from the landfill increased.

4.4.4 Alternative tracer configuration

TDM-measured emissions are compared for the base case tracer configuration and alternative tracer configuration for the four different time periods from the March 2010 simulation. Standard deviations of TDM-measured emissions and percent errors of the mean TDM-measured emissions are presented in Table 4.4.

Table 4.4: Standard deviations of TDM-measured emissions (g/min), percent error of the TDM-measured emissions in parentheses.

<table>
<thead>
<tr>
<th></th>
<th>Early Afternoon</th>
<th>Late Afternoon</th>
<th>Night</th>
<th>Morning</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base case</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1000 m</td>
<td>478 (27%)</td>
<td>509 (18%)</td>
<td>94 (3%)</td>
<td>212 (10%)</td>
</tr>
<tr>
<td>2000 m</td>
<td>543 (23%)</td>
<td>409 (13%)</td>
<td>66 (2%)</td>
<td>133 (3%)</td>
</tr>
<tr>
<td>Alternative</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1000 m</td>
<td>1222 (43%)</td>
<td>1472 (42%)</td>
<td>74 (-1%)</td>
<td>426 (16%)</td>
</tr>
<tr>
<td>2000 m</td>
<td>1210 (28%)</td>
<td>810 (20%)</td>
<td>71 (-5%)</td>
<td>260 (3%)</td>
</tr>
</tbody>
</table>

From Table 4.4, it is clear that the TDM-measured emissions using the alternative tracer configuration have significantly larger standard deviations than the TDM-measured emissions using the base case tracer configuration. The only exception is the Night time period, where the standard deviations for base case and alternative tracer configuration are essentially the same. Table 4.4 also shows a clear difference between the accuracy of the TDM with the base case tracer configuration over the alternative tracer configuration. These numbers are comparable with the reported errors due to upwind location of tracers. Mønster et al. (2014) used the assumption that the tracer collocated with the methane source had no error to estimate that the error of their upwind tracer placement was ∼24%. Table 4.4 also shows interestingly that measurement distance from the landfill has a much more significant effect for the bad tracer configuration than it does for the good tracer configuration. This finding is consistent with former results in literature (Mønster et al., 2014; Taylor et al., 2016). Mønster et al. (2014) found that the errors decreased from 36% to 20% for an upwind tracer configuration with a controlled point source, as the distance of measurements increased from 370m to 1200m. In another study, the simulations with steady landfill methane emissions displayed that the errors for an 85m upwind tracer location decreased from 42% to 36%, when the measuring distance increased from 800 m to 2000 m. One consequence of this result may be the recommendation that a distance closer to 2000 m be used when possible.
CHAPTER 4. ATMOSPHERIC MODELING TO ASSESS WIND DEPENDENCE OF LANDFILL METHANE EMISSIONS TDM MEASUREMENTS

4.5 Summary and conclusions

Previous studies have shown both significant short-term variability in landfill methane emissions measurements and correlations between landfill emissions and surface wind speed. This study used WRF to simulate the TDM and explore this dependence. A new surface flux parameterization for landfill methane emissions was implemented to capture wind speed dependence in the emissions. This allowed examination of the effect of wind-induced emissions variability on the TDM measurements. This study also examined simulated TDM measurements for eight different time periods over two days to see if trends in TDM-measured variability and TDM error emerged, and to examine two different tracer configurations because the optimal tracer configuration may not be implemented in the field.

Across all simulations, the TDM errors ranged from -15\% to 43\%, consistent with the errors seen in the previous chapter. Results showed that steady and unsteady emissions simulations yield very similar TDM-measured standard deviations. The means and standard deviations for the two TDM measurement datasets, however, are significantly larger than the mean and standard deviation of the true unsteady model emissions.

Standard deviations from the unsteady emissions simulations revealed an increase as the day gets later, from Morning to Early Afternoon to Late Afternoon. This increase corresponds with the increase in wind speed standard deviation in the afternoon, shown in Table 4.2. Standard deviation of TDM-measured emissions shows a sharp decrease during the Night period, which could be explained by the lack of fluctuations in wind speed and wind direction resulting in steadier gas transport at night and therefore steadier concentration measurements. Error at 2000 m was always less than or equal to the error at 1000 m, and standard deviation was always lower at 2000 m with one exception, Early Afternoon in March 2010.

Distance from the landfill had a much larger impact on TDM error for the alternative tracer configuration than the base case configuration. TDM error of measurements taken at 1000 m was significantly greater for the alternative tracer setup than the base case setup, while error at 2000 m was only slightly higher for the alternative than the base case. Standard deviation of the TDM measurements was also significantly greater for the alternative setup than the base case, except during the Night measurement period, when standard deviation was smaller for the alternative setup than the base case setup.

The variability seen in TDM-measured emissions over a short-term measurement campaign may be more directly related to the fluctuations in wind speed and wind direction than in the magnitude of true emissions fluctuations. This finding highlights the importance of collecting many measurements over a measurement period and evaluating the average measured value. Additionally, this study reinforces the importance of tracer configuration on the accuracy of the TDM and demonstrates that if the tracer configuration is less ideal, measurements should be taken further from the landfill to improve TDM accuracy. Several open research questions remain. Future work will focus on better understanding diurnal and seasonal patterns in landfill methane emissions.
Chapter 5

Numerical simulations to assess challenges with estimating annual landfill methane emissions

Annual landfill methane emissions are most often estimated using a simple first order decay model rather than with field measurements. A promising emissions measurement technique, the tracer dilution method (TDM) has shown large discrepancies between annual emissions estimated with TDM-measured emissions and with common first-order decay models. Little research has been conducted regarding how to use TDM-measured emissions to accurately estimate annual landfill methane emissions. In this work, the Weather Research and Forecasting model (WRF) is used to simulate landfill methane emissions and TDM measurements at a real landfill in the Southeastern U.S. on four different days during which real TDM field campaigns were performed. This serves to help create a robust modeling framework for exploring the TDM and how it can be used to estimate annual landfill emissions. WRF results show excellent agreement between model output and field data for both local wind conditions and tracer gas concentration at the locations of the roads where field measurements were taken on all four days simulated. Results also show that longer measurement periods result in more accurate mean TDM-measured emissions, and that TDM-measured emissions are more accurate when wind speed is \( \geq 5 \) m/s. Errors associated with estimates of annual methane emissions from models and field data are explored.

5.1 Introduction

Landfills are considered a significant anthropogenic contributor to atmospheric methane. Accurately estimating landfills annual methane emissions is essential for understanding their contribution to global warming and creating regulations to mitigate this effect. The methods commonly used to estimate annual landfill methane emissions have many limitations and sources of uncertainty, resulting in the need for a more accurate way to estimate these
annual emissions. The most common methods currently used are simple models that focus on estimating the amount of methane produced in the landfill based on the age and composition of the waste. These models estimate methane emissions as a balance between methane production and methane oxidization in the soil. Gas collection systems can be included in models by removing gas collected at the landfill. The most commonly used models come from the Intergovernmental Panel on Climate change (IPCC), the U.S. Environmental Protection Agency Greenhouse Gas Reporting Program (EPA GHGRP), and the California Air Resources Board (CARB). In these three models, methane production over the landfill’s lifetime is calculated as the sum of methane produced each year, which is a function of quantity of waste disposed that year, amount of degradable organic carbon disposed that year, and an exponential decay term with a constant decay rate. The exponential decay term is a function of $(T - t)$, where $T$ is the current year for which emissions are being estimated, and $t$ is each year of the landfill’s lifetime.

These models are the most commonly used methods for estimating annual landfill emissions because field measurements are often too expensive, time-consuming, invasive, and require expertise to operate the measurement equipment. Many landfills, therefore, have no field measurements of their emissions to compare to their model-estimated emissions and no estimate of their accuracy. A recent study by De La Cruz et al. (2016) compared field measurements using the tracer dilution method (TDM) to several of these commonly used models. This study highlighted several limitations of the simple models, such as the assumptions made about methane production and the decay rate, variable efficiency of gas collection systems over time, and methane oxidation. The TDM uses a tracer gas released from one or more cylinders on the surface of the landfill at a known, steady emissions rate. Downwind of the landfill, usually 1 - 2 km away, a gas analyzer is mounted on a vehicle, which drives back and forth along a road that is roughly perpendicular to the wind direction, thus collecting concentration transects for both the methane and tracer gas plumes. These concentration transects are used to calculate the TDM-measured emissions, based on the assumption that at this distance from the landfill, both gas plumes are well-mixed enough that the ratio of the concentrations is representative of the ratio of the emissions rates:

$$Q_{CH4} = Q_{tr} \frac{M_{CH4}}{M_{tr}} \frac{\int c_{CH4}(x)dx}{\int c_{tr}(x)dx} \quad (5.1)$$

where $Q_{tr}$ is the known tracer emissions rate in g/min, $M_{CH4}$ and $M_{tr}$ are the molecular weights of methane and tracer gas, and $c_{CH4}(x)$ and $c_{tr}(x)$ are the methane and tracer gas concentrations along the transect in ppb.

The De La Cruz et al. (2016) study showed that the simple models all exhibited extreme over-prediction of the emissions compared to the TDM-measured emissions, ranging from a factor of 4 to 31 times the TDM-measured emissions rate. To estimate annual landfill methane emissions based on only about 10 measurement campaigns over a 2.5-year period, they fit a curve to the emissions measured during each campaign and computed the area under the curve to obtain the total emissions estimate. The authors of the study noted that
their measurements were conducted on days when personnel and equipment were available, and as such they may not reflect actual seasonal variability in the emissions.

The TDM is a promising measurement technique which has gained popularity in the past few years for being relatively inexpensive and noninvasive compared to measurement techniques such as flux chambers and eddy covariance (Mønster et al., 2014; Foster-Wittig et al., 2015; De La Cruz et al., 2016). Several measurement campaigns have demonstrated significant variability in landfill emissions, so a single TDM measurement campaign would not be sufficient to extrapolate an annual emissions estimate. If the TDM could be performed a few times over the course of the year with guidelines on how to interpolate that data to account for seasonal and even diurnal changes, the accuracy of the annual emissions estimates could improve greatly. This would ultimately lead to better understanding of landfill contributions to global warming and better regulations to reduce emissions.

In this work, numerical modeling with the Weather Research and Forecasting model (WRF) is used to predict emissions and their variability on daily and annual time scales at a real landfill in the Southeastern U.S. Simulations of the TDM are compared with field TDM data from the site to estimate accuracy of the TDM under different atmospheric conditions. This work aims to create a modeling framework that can be used to understand how emissions change on diurnal and seasonal time-scales. The simulation results can help to create guidelines for TDM-measured emissions used to estimate annual landfill emissions.

5.2 Modeling Approach

5.2.1 Site description

The landfill site of interest was the site of a multi-year field campaign effort to understand and improve the TDM. This site was chosen for the simulations that follow because of the extensive field data available for model validation. The landfill has a capacity of 26 million metric tons and began receiving \( \sim 300,000 \) metric tons of waste per year in 2010, making it a relatively young landfill with relatively low methane emissions (De La Cruz et al., 2016). Between June 2011 and November 2013, 10 field studies were performed over 35 days, consisting of 443 TDM transects. During this 2.5-year period, TDM measurements showed methane emissions increasing nonlinearly from about 400 g/min in June 2011 to about 2400 g/min in November 2013 (Foster-Wittig et al., 2015). The emissions do not yet follow a typical seasonal trend with peak emissions in the summer and reduced emissions in the winter. This is likely because the landfill was so young when the measurements started in 2011 that methane production had not yet reached its full potential. The amount of waste increased rapidly over the 2.5-year study period, resulting in a rapid increase in methanogenic archaea. The landfill had no gas collection system during the measurement campaigns.

Simulations of atmospheric flow over this landfill are performed for 4 of the 35 days for which there is TDM field data. Due to the high computational cost of the simulations, only four days were simulated for this work. These four days are chosen because they are in
approximately four different seasons, spanning a variety of meteorological conditions. More information about the computational cost of the simulations is included in Section 5.2.3. Simulations are run using the Weather Research and Forecasting model (WRF), which uses multiple nested grids to run large-eddy simulations (LES) at the desired 30 m resolution using forcing data from the 12 km resolution boundary and initial conditions. In these simulations, three nested domains are used: the largest domain has $90 \times 90$ grid cells with 2.25 km resolution, the middle domain has $300 \times 300$ cells with 150 m resolution, and the smallest domain where LES is performed to simulate the TDM has $300 \times 300$ cells with 30 m resolution. All domains have 75 vertical levels, and the bottom vertical level has a height of approximately 20 m above the ground surface. Figure 5.1 shows the 3 nested model domains with a Google Earth image of a subsection of the smallest model domain, showing the shape and location of the landfill as well as the locations of the tracer release points and the mean ground-level wind direction during the base case simulation.

5.2.2 Landfill emissions parameterization

To model unsteady landfill methane emissions, the emissions are calculated within WRF at each time step with a surface flux parameterization, based on the parameterization WRF uses for heat and moisture surface fluxes.
In Equation 5.2, \( C_d \) is a time-varying drag coefficient dependent on air density and atmospheric stability, \( u^* \) is shear velocity, and \( c_{\text{soil}} \) and \( c_{\text{atm}} \) are the soil and atmospheric methane concentrations respectively. WRF is coupled to the Noah land surface model (LSM), which has a 1-D soil model with 4 layers of varying thicknesses, so \( c_{\text{soil}} \) is specified within the LSM at the midpoint of the top soil layer, at 5 cm depth. The atmospheric concentration is specified at the midpoint of the bottom atmospheric grid cell, ~10 m above the surface.

It is clear from Equation 5.2 that the value of soil methane concentration will have a significant impact on the value of the landfill emissions. To obtain realistic soil methane concentration values for the WRF simulations, the California Landfill Methane Inventory Model (CALMIM) was run with input parameters specific to the landfill used in this study (latitude=33.176\(^\circ\), longitude=-84.848\(^\circ\), cover type=daily, soil type=sandy clay loam, soil depth=6 inches, vegetation cover=0%, gas recovery=0%). CALMIM uses climate data such as solar radiation and precipitation, customized to the user-specified latitude and longitude but averaged over several years of data, to drive soil processes in a 1-D domain 6 inches deep with 1-inch grid spacing. It calculates whole landfill emissions at 1-hour intervals over a 365-day period using a static boundary layer conductance multiplied by the difference between soil methane concentration and atmospheric methane concentration. CALMIM outputs soil methane concentration at each soil grid point at 1-hour intervals; concentration values at 2 inches (~5 cm) were examined for use in Equation 5.2.

CALMIM does not take the age of the landfill into consideration in its calculations, and the landfill in this study is a particularly young landfill. Field measurements were taken in all seasons over the course of 2.5 years, and whole landfill emissions were found to increase with each measurement campaign rather than exhibiting the expected seasonal pattern of highest emissions in the summer and lowest emissions in the winter. It is, therefore, not appropriate to rely on CALMIM’s soil methane concentration calculations to understand how the soil and emissions would change at this particular site. Due to the linearity of the scalar advection diffusion equation, the emissions and methane concentration field are scaled in post-processing such that the mean of the modeled TDM-measured emissions matched the mean of the TDM-measured emissions from the field data. This post-processing emissions scaling allows us to compare the standard deviations of the observed TDM measurements to the model TDM measurements as well as to understand how different the true emissions and TDM-measured emissions are under specific conditions. This scaling is able to preserve the relationship between the true model emissions and the modeled TDM-measured emissions so that TDM accuracy could still be evaluated.

### 5.2.3 Days modeled

Four days are chosen to represent four different seasons and conditions: November 17, 2011; June 26, 2012; December 4, 2012; and April 13, 2013. Six-hour simulations are run
CHAPTER 5. NUMERICAL SIMULATIONS TO ASSESS CHALLENGES WITH ESTIMATING ANNUAL LANDFILL METHANE EMISSIONS

on each day, from 11:00 to 17:00 LST. To understand the extent of the turbulent mixing for each day, potential temperature profiles at each hour of the 6-hour periods are shown in Figure 5.2. In the potential temperature profiles, the ABL height is indicated by the height where the slope of the profile becomes steeply positive. The atmospheric stability is indicated by the slope of the profile within the ABL. Atmospheric stability and ABL height indicate how much mixing the atmosphere and therefore the plumes of methane and tracer gas are experiencing.

Of the four days modeled, December 2012 maintains neutral/slightly stable conditions and shows the most similarity in potential temperature profiles over the 6-hour period. April 2013 shows the most surface heating and the most boundary layer growth over the 6-hour period. November 2011 exhibits significant surface cooling between 15:00 and 17:00 LST and does not show significant surface heating between 11:00 and 15:00. June 2012 has the most instability in the near-surface region and highest overall temperatures, indicating that the simulations on that day likely have the most mixing. One hypothesis of this work is that the TDM will be the most accurate on June 2012 because of the increased turbulent mixing near the surface.

Future work will include 24-hour simulations from multiple days to examine diurnal variability in magnitudes of emissions and TDM accuracy. Due to the high computational cost of these simulations, 6-hour-long simulations were run for this work. With three nested grids, the large number of grid points in the horizontal and vertical directions, the extra cost of the emissions parameterization and the scalar transport, the need for 2 second output to simulate the mobile TDM, and the variability of user loads on the supercomputer, up to 24 hours of wall clock time are required to simulated 1-2 hours of model time. A 24 hour simulation would therefore take 12-24 days of wall clock time.

Figure 5.2: Potential temperature profiles for each of the four days.
5.2.4 Modeling the tracer dilution method

Modeling the TDM requires three main components: modeling of the landfill methane emissions and resulting methane plume, the tracer gas emissions and resulting tracer gas plume, and the downwind measurements, which are performed with a moving vehicle traveling at approximately 30 km/h (8 m/s) along existing roads. For the simulations presented in this chapter, a log-normal distribution of soil methane concentration was generated to create heterogeneity in the surface soil field (Abichou et al., 2006). The mean value for the daily cover area came from the CALMIM simulation at 11:00 on June 26 (1275 ppb), and the standard deviation for the log-normal distribution was 24.5% of the mean (312 ppb), which is consistent with the relationship between the mean and standard deviation used for the log-normal distribution of landfill emissions in Chapter 3. In the field, it was determined through measurements that the intermediate cover area contributed about four times as much emissions as the daily cover area, so the values in the modeled intermediate cover area were adjusted so that the intermediate cover area (the portion on the left) contributed \( \sim 80\% \) of the total emissions. Since the emissions changed at each time step based on the local surface wind speeds, the emissions distribution was slightly different at each time step. Figure 5.3 shows a snapshot of the emissions distribution at one time step (14:30 LST) with terrain height contours in thin black lines (1.3 m intervals). The locations of the two tracer release points are shown with black circles. The tracer gas was emitted at 44 g/min total, with 22 g/min from each release point.

The locations of the roads used to collect transect measurements in the field were used to collect the simulated transect concentration measurements. The wind directions were different on each of the four days, so different roads were used. The locations of these roads relative to the tracer release points and tracer plumes are shown in Figure 5.4. Note that the roads on November 2011 and June 2012 have some overlap with each other, and the roads on December 2012 and April 2013 have some overlap. In June 2012, the transect turns the corner because the plume meandered towards the south frequently but the road that was initially perpendicular to the plume ended in a T-intersection at the location where the right panel of Figure 5.4 shows a corner in the road. The road that is roughly parallel to the plume in this figure continues to the west, but that portion of the road was not used for the transect measurements, so it was not included in the plot.

It is clear from Figure 5.4 that the terrain surrounding this landfill is more complex compared to the terrain seen at Sandtown Landfill in the previous chapters. The top of the landfill is 22 m higher than the valley just south of it. The rolling terrain could have an effect on the accuracy of the TDM. It is also clear that the different roads are different distances from the tracer release points, with June 2012 being the closest at \( \sim 700 \) m and April 2013 being the farthest with at \( \sim 1700 \) m. Results in previous chapters showed that distance from the landfill only makes a small difference on the accuracy of the TDM-measured emissions, so it is not expected that these differences in distance will have a significant impact on the results.
5.3 Model Validation

One focus of this work is in comparing model output to field data. This thorough model validation has not been done before and is an important, novel component of this work. The aim is to create a robust model that can be used for future testing at this field site and others. Model exploration of the TDM will help to create guidelines for how the TDM can be used to accurately predict annual landfill emissions.

5.3.1 Wind speed and direction

To understand whether the model is a good representation of reality, the first aspects to be compared to field data are the wind metrics. The field data reported mean wind speed, mean wind direction, and standard deviation of wind direction for each time interval over which a transect measurement was collected. Each transect time interval lasted between 3 and 5 minutes. The field data was from a sonic anemometer located on the surface of the landfill (latitude=33.17485°, longitude=-84.84832°), although only time-averaged data
over each transect traverse are available. Standard deviations describing the very short-term variability of wind speed are not available.

Table 5.1 shows the mean and standard deviation (except for standard deviation of observed wind speed) of the wind conditions for each of the four days. The agreement between the model and field data is generally very good and well within the range of acceptable errors for comparisons of model and field data (all errors are \( \leq 12\% \)).

Average wind speed is well captured by the model on all four days, and average wind direction is off by \( \sim 10-25^\circ \), which puts the average wind direction always in the correct quadrant. These errors are within the standard range of errors expected in an atmospheric
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Figure 5.5: Time series of wind speed (m/s) and wind direction (degrees) comparing field data and model output for each of the four days.

Table 5.1: Means and standard deviations of wind speed and direction for each of the four simulated days.

<table>
<thead>
<tr>
<th></th>
<th>Speed (m/s)</th>
<th>Direction (degrees)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Model</td>
<td>Field obs</td>
</tr>
<tr>
<td>Nov 2011</td>
<td>6.1±1.2</td>
<td>6.0</td>
</tr>
<tr>
<td>June 2012</td>
<td>4.8±1.2</td>
<td>5.2</td>
</tr>
<tr>
<td>Dec 2012</td>
<td>3.2±0.8</td>
<td>3.1</td>
</tr>
<tr>
<td>April 2013</td>
<td>3.4±1.2</td>
<td>3.9</td>
</tr>
</tbody>
</table>

model like this. This is important because it means the real road that was used for field-measured concentration transects can be used for the simulated transect measurements. Therefore it is possible to compare tracer gas concentration measurements, given that the modeled tracer gas emissions rate is set to equal the tracer emissions rate used in the field.

One trend worth noting is that both November 2011 and June 2012 have average wind speeds ≈5 m/s while December 2012 and April 2013 have average wind speeds significantly
below 5 m/s. It is hypothesized that greater wind speeds result in more accurate TDM-measured emissions because the plumes will be less variable when the surface winds are stronger.

5.3.2 Tracer concentration

One way to determine whether the model is adequately capturing the atmospheric scalar transport is to compare the tracer gas transects from the field and the model. The tracer emissions rate used in the field was also used in the simulations, at 22 g/min per tracer release point yielding 44 g/min total. The model adequately captures the correct order of magnitude of the tracer concentration at the real transect locations, which are shown in Figure 5.4. It is not expected that the exact shape and location of the plume be captured by the model because the plume evolution is affected by turbulent eddies. Instead, we use statistical measures to evaluate agreement with observations. To assess the model’s performance, we use three criteria from Chang and Hanna (2004): at least 50% of model values are within a factor of 2 of the observations ($FAC_2$), the relative mean bias is within $±30\%$ ($FB$), and the relative scatter is within a factor of 3 ($NMSE$). These criteria are determined using the following equations:

$$FAC_2 = \frac{1}{L} \sum_{j=1}^{L} \frac{1}{N_j} \sum_{i=1}^{N_j} \frac{c_{i,j}^m}{c_{o,i,j}} (5.3)$$

$$FB = \frac{1}{L} \sum_{j=1}^{L} \frac{1}{N_j} \sum_{i=1}^{N_j} \frac{(c_{o,i,j}^o - c_{i,j}^m)}{0.5(c_{o,i,j}^o + c_{i,j}^m)} (5.4)$$

$$NMSE = \frac{1}{L} \sum_{j=1}^{L} \frac{1}{N_j} \sum_{i=1}^{N_j} \frac{(c_{o,i,j}^o - c_{i,j}^m)^2}{c_{o,i,j}^o c_{i,j}^m} (5.5)$$

In these equations, $L$ is the number of cases, in this case 4 because there are 4 different days, $N_j$ is the number of sensors for case $j$, $c_{o,i,j}^o$ is the observed concentration value at the $i^{th}$ sensor in the $j^{th}$ case, and $c_{i,j}^m$ is the modeled concentration value at the $i^{th}$ sensor in the $j^{th}$ case. The criteria determine that the model performs well if the values fall within the following ranges: $0.5 \leq FAC_2 \leq 2$, $-0.3 \leq FB \leq 0.3$, and $NMSE < 3$. There are multiple ways the concentration data could be evaluated, by considering either how well the model output matches the observed values in time or in space, and it is acceptable to consider only the maximum concentration in the analysis (Chang and Hanna, 2004). The data presented in this chapter are evaluated by pairing the model and observed data in time only. We consider a time series of maximum concentrations along the transect paths with no penalty to the model if it predicts the maximum concentration in the wrong place (Chang and Hanna, 2004). This way we are only considering one comparison point, which is wherever the max concentration occurs along each traverse of the transect path. Comparing maximum concentrations is often the best way to evaluate scalar concentrations affected
by errors in model wind speed and direction. In this case, model wind speeds matched the observed wind speed very well, but the model wind direction did not match as well, indicating that the modeled gas plume might be shifted from the observations. Using the maximum concentration from a plume transect allows evaluation of the plume development while allowing for some expected deviations of the plume centerline due to wind direction errors. Averaging all 4 simulations, we find FAC2=0.829, FB=0.216, and NMSE=0.124. These are all well within the range of values necessary for the model performance to be considered acceptable. FAC2 shows that ∼83% of the model’s maximum transect values are within a factor of 2 of the observed maximum transect values. FB shows the fractional bias, which describes how the difference between the model and observed values compares to the average of the model and observed values. Here we find that the average difference between model and observed values is ∼22% of the concentration value when model and observed values are averaged together. Finally, NMSE shows the normalized mean square error, which is a measure of mean relative scatter. Figure 5.6 shows a representative tracer gas transect from both the model output and the field data for each of the four days to give an idea of how well the maximum model concentrations represent the maximum field-measured concentrations along the transect path.

Note that the model transects do not align perfectly with the field-measured transects because the model’s wind direction is slightly different from the observed with direction, but the plume widths and maximum concentrations match very well. Overall, this is considered very good model performance for an atmospheric dispersion model (see metrics above) due to the complexities of turbulence in the atmosphere.

5.3.3 TDM-measured emissions

The modeled landfill methane emissions were scaled in post-processing so that the mean of the model TDM-measured emissions equals the mean of the field TDM-measured emissions. Because the advection-diffusion equation is a linear equation, if the emissions are scaled by a constant factor, the resulting concentration plume can be scaled by the same factor to obtain exactly the concentration plume that would result from running the simulation with the scaled emissions rate. This scaling preserves the TDM error (discussed in more detailed in Section 5.4) because by scaling methane plume concentration, the TDM-measured emissions will scale by the same factor, so the scaling factor will cancel out when TDM error is calculated, yielding the same error as if nothing had been scaled.

Table 5.2 summarizes the means and standard deviations of the emissions for each simulated day. Over the course of the 1.5 years represented by the TDM field measurements, measured emissions increase dramatically from ∼200 g/min to ∼1800 g/min. This increase is captured by the true modeled emissions, although the increase is significantly less for true modeled emissions in December 2012 and April 2013 than the increase in TDM-measured emissions for both the model and field measurements on these days. This greater mismatch between the true model emissions and TDM model emissions indicates potentially greater TDM errors on these days than on the earlier days with smaller emissions. Table 5.2 also
reveals that the standard deviation of true model emissions is always slightly smaller than the standard deviation of model TDM-measured emissions. This is an indication that the extent of the TDM-measured emissions variability seen in the field may be larger than the variability in the true emissions and should not necessarily be used to understand the true emissions variability.
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5.4 Tracer dilution method accuracy

The accuracy of the TDM is assessed by comparing the true model emissions mean to the TDM-measured model emissions mean using the percent error:

\[
P_{error} = \frac{Q_{TDM} - Q_{model}}{Q_{model}} \times 100
\]  

(5.6)

Figure 5.7: Time series of true model emissions (solid black line), model TDM-measured emissions (blue x’s), and field TDM-measured emissions (red circles) for each of the simulated days.

Figure 5.7: Time series of true model emissions (solid black line), model TDM-measured emissions (blue x’s), and field TDM-measured emissions (red circles) for each of the simulated days.
Table 5.2: Means and standard deviations for the different emissions rates for each simulated day (g/min). All measurements are from 11:00 to 17:00 local time except June 2012, which is from 13:00 to 17:00.

<table>
<thead>
<tr>
<th></th>
<th>True model emissions</th>
<th>Model TDM-measured</th>
<th>Field TDM-measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nov 2011</td>
<td>217±25</td>
<td>225±33</td>
<td>223±43</td>
</tr>
<tr>
<td>June 2012</td>
<td>516±55</td>
<td>544±75</td>
<td>544±161</td>
</tr>
<tr>
<td>Dec 2012</td>
<td>697±178</td>
<td>879±189</td>
<td>879±293</td>
</tr>
<tr>
<td>April 2013</td>
<td>1450±323</td>
<td>1808±765</td>
<td>1808±786</td>
</tr>
</tbody>
</table>

Again, the TDM model emissions are scaled to match observations, and then compared to the true model emissions, which generated the TDM model emissions, to indicate the variability in the TDM predictions. Table 5.3 shows the overall percent error of the model TDM-measured emissions values, and the percent error if the measurements are broken into separate 1-hour periods.

Table 5.3: Model TDM % error over whole time period and over each 1-hour period. Field data not available for 11:00-13:00 on June 2012 so simulations were not performed during that time.

<table>
<thead>
<tr>
<th></th>
<th>Nov 2011</th>
<th>June 2012</th>
<th>Dec 2012</th>
<th>April 2013</th>
</tr>
</thead>
<tbody>
<tr>
<td>Overall</td>
<td>4%</td>
<td>5%</td>
<td>26%</td>
<td>25%</td>
</tr>
<tr>
<td>11:00-12:00</td>
<td>-2%</td>
<td>×</td>
<td>22%</td>
<td>40%</td>
</tr>
<tr>
<td>12:00-13:00</td>
<td>8%</td>
<td>×</td>
<td>11%</td>
<td>17%</td>
</tr>
<tr>
<td>13:00-14:00</td>
<td>1%</td>
<td>8%</td>
<td>53%</td>
<td>-6%</td>
</tr>
<tr>
<td>14:00-15:00</td>
<td>12%</td>
<td>2%</td>
<td>11%</td>
<td>14%</td>
</tr>
<tr>
<td>15:00-16:00</td>
<td>-5%</td>
<td>1%</td>
<td>15%</td>
<td>29%</td>
</tr>
<tr>
<td>16:00-17:00</td>
<td>9%</td>
<td>11%</td>
<td>81%</td>
<td>40%</td>
</tr>
</tbody>
</table>

The values of the overall errors are well within the range of values seen in the previous chapters. Revisiting the hypothesis from Section 5.2.3, June 2012 has significantly less error than December 2012 and April 2013, although November 2011 has the least error. The two days with lower percent error are also the two days with average wind speed above 5 m/s. This is consistent with the current rule of thumb that the TDM is more accurate when wind speed is higher (Delkash et al., 2016). It is interesting to see that the percent error varies greatly when the measurement periods are broken down into 1-hour sections. The percent errors for November 2011 and June 2012 are consistently smaller than the errors for the other two days, but they do not consistently over-predict emissions; there are two 1-hour periods on November 2011 during which the TDM under-predicts emissions. This hour-by-hour breakdown of the TDM errors demonstrates the importance of having a long enough measurement period, particularly for the two days with larger errors. On December 2012, the overall error is 26%, which is relatively small considering that if measurements
were taken only during 16:00 - 17:00, the mean of the TDM-measured emissions would have an error of 81%. Upon examining model output to understand why the TDM performs so poorly during this specific 1-hour period, model output shows that surface wind speed decreases significantly over the landfill surface but not in the region downwind of the landfill where concentration measurements are collected. The decreased wind speed over the landfill surface results in a significantly lower emissions rate due to Equation 5.2, which can be seen in the December 2012 panel in Figure 5.7. Stronger downwind wind speeds, however, will lead to a pile-up of gas at the downwind measurement location, causing the ratio between methane and tracer concentrations at the downwind measurement location to differ from the ratio of their emissions, since the methane emissions rate has greatly decreased but the tracer emissions rate remains constant. This result indicates that the TDM performs better when wind speed is more homogeneous over the landfill and surrounding area.

5.5 Using limited measurements to estimate annual emissions

Infrequent TDM measurements have been used to estimate annual landfill methane emissions, as done for example by De La Cruz et al. (2016). Their methodology is presented here, along with suggestions for future improvements. The total emissions over the multi-year measurement period can be calculated as the area under a curve created by the measured emissions data points. This can be done by fitting a nonlinear curve, as done by De La Cruz et al. (2016), or more simply with the trapezoidal method of integration between each data point. The authors of the De La Cruz et al. (2016) study were working with 35 measurement days and were able to fit a nonlinear curve to their data. This work uses a very small subset of those measurements due to computational constraints. Due to the small number of data points (4 days), annual estimates are performed in this work with the trapezoidal method. Annual emissions estimated by this trapezoidal method can be compared to annual emissions predicted by CALMIM, as shown in Figure 5.8. Note that in Figure 5.8, the y-axis in the right panel for CALMIM is an order of magnitude larger than the y-axis in the left panel.

Field-measured emissions were given in g/min, so they were converted to g/day and then used to integrate over the number of days to estimate annual emissions. This assumes emissions are not fluctuating significantly on smaller time scales, such as diurnally, an assumption which will be explored in the subsections that follow. Using the first 3 data points, excluding April 2013 for the moment, trapezoidal method calculations show that the landfill emits $2.873 \times 10^8$ g methane over 383 days, or $2.738 \times 10^8$ g/yr. Using the period between the last two data points, Dec 2012 and April 2013, calculations show emissions of $2.457 \times 10^8$ over 127 days, equivalent to $7.061 \times 10^8$ g/yr. Now including the data point from April 2013, which does not appear to fit linearly with the other 3 points, calculations show the landfill emits $5.330 \times 10^8$ over 510 days, equivalent to $3.815 \times 10^8$ g/yr. Clearly the time period used to perform the calculations will have a significant effect on the magnitude of the estimated
annual emissions, and from the limited data it is unknown how the emissions were changing in between the data points or how long the steep increase in emissions will continue.

The CALMIM output, on the other hand, shows high variability in daily emissions over the course of one year. Oxidation is accounted for in the CALMIM simulation, and CALMIM output was reported for every hour over the 365-day simulation, so some CALMIM data points predicted zero emissions when oxidation was predicted to be very high. The highest predicted emissions are an order of magnitude higher than the highest emissions actually measured at this landfill, yielding an annual emissions estimate of $3.112 \times 10^9$ g/yr. This CALMIM-estimated annual emissions exceeds the trapezoidal method annual emissions estimate by an order of magnitude, indicating the need to better understand how to estimate annual emissions. The CALMIM output in the right panel of Figure 5.8 shows a slight seasonal pattern with highest emissions in the middle of the year and lowest emissions at the beginning and end of the year (the winter months), a seasonal pattern which is not seen in the field measurements for this site. The lack of seasonality in the field measurements is due to this site being a young landfill, with considerable growth in annual emissions; this makes estimates of annual emissions with a model like CALMIM difficult.

The differences between CALMIM-predicted changes and changes seen in field data are further highlighted by examining the scaling factors used in post-processing the model output to match the model TDM-measured emissions to the field TDM-measured emissions, as discussed in Section 5.3.3. CALMIM simulations cannot be run for a specific year; instead the model produces output over the course of a year using averaged meteorological data.
CHAPTER 5. NUMERICAL SIMULATIONS TO ASSESS CHALLENGES WITH ESTIMATING ANNUAL LANDFILL METHANE EMISSIONS

from several years for the model’s solar radiation and precipitation events. The question of interest is how different the CALMIM predictions are from what was observed in the field regarding relative changes in emissions. To examine these relative differences, the field-measured emissions from the 4 dates used in this work are normalized by the field-measured emissions from November 17, 2011 and the CALMIM emissions predicted on those same 4 dates (year not able to be specified by CALMIM) are normalized by the CALMIM-predicted emissions on November 17. This comparison is meant to illustrate that CALMIM’s predictions of relative changes in landfill emissions are not applicable to this landfill, further highlighting the need for a better way to estimate annual emissions. These relative emissions factors are summarized in Table 5.4.

Table 5.4: Relative emissions factor for each date compared to emissions on November 17

<table>
<thead>
<tr>
<th>Date</th>
<th>Field-measured</th>
<th>CALMIM-predicted</th>
</tr>
</thead>
<tbody>
<tr>
<td>November 17</td>
<td>1.000</td>
<td>1.000</td>
</tr>
<tr>
<td>June 26</td>
<td>2.418</td>
<td>2.612</td>
</tr>
<tr>
<td>December 4</td>
<td>3.907</td>
<td>0.704</td>
</tr>
<tr>
<td>April 10</td>
<td>8.036</td>
<td>0.703</td>
</tr>
</tbody>
</table>

5.5.1 Hourly variability

For a crude understanding of the effect of changing soil methane concentration on the landfill emissions calculated with Equation 5.2, model emissions are scaled in hour-by-hour blocks according to the changes that CALMIM predicts with the inputs specified in Section 5.2.2. This post-processing scaling of the emissions is a rough but adequate representation of the effect of changing soil methane concentration because in reality, at the first time step after scaling the soil methane concentration within the simulation, the atmospheric methane concentration will not have scaled accordingly to match the new emissions rate yet, so there will be some feedback between the scaled emissions rate and the atmospheric methane concentration. This post-processing method of scaling soil methane concentration can give an idea of the effect, however, particularly because the changes in soil methane concentration calculated by CALMIM over this 6-hour time period are so small. Figure 5.9 shows CALMIM output of time-averaged variability from 11:00 to 17:00 with values normalized by the 11:00 value, which is the value used for the TDM simulations. This normalized time-varying scaling function is applied to each of the 4 simulated days to examine the effect of time-varying soil methane concentration on landfill emissions rate. The scaled and unscaled emissions time series are shown on the same plots along with their respective means for direct comparison.

This figure shows that not only does CALMIM predict only very minor changes in soil methane concentration over a 6-hour period (maximum of 8% difference from the initial 11:00 value), but implementing these changes in emissions has a minor effect on the mean value of the emissions over the 6-hour period. The differences between mean emissions with
Figure 5.9: Normalized time-averaged soil methane concentration from 11:00 to 17:00 and the effect of time-varying soil methane concentration on landfill emissions.

scaled variable soil concentration and steady soil concentration are as follows: November 17, 2011, 18 g/min (8% of the unsteady mean emissions); June 26, 2012, 36 g/min (7% of the unsteady mean emissions); December 4, 2012, 34 g/min (5% of the unsteady mean emissions); April 10, 2013, 74 g/min (5% of the unsteady mean emissions).
5.5.2 Diurnal variability

We can extend the analysis from the previous section to examine whether changes in emissions from day to night have a significant effect on the emissions that need to be taken into account for more accurate annual estimates. The Nov 2011 simulation was run until 02:00 local time the next morning and emissions were scaled in the same way they were scaled in Section 5.5.1. Figure 5.10 shows normalized CALMIM-predicted soil methane concentration for the 15-hour period and the effect of changing soil methane concentration on landfill methane emissions.

![Normalized time-averaged soil methane concentration from 11:00 to 02:00 for Nov 2011 simulation (top) and the effect of time-varying soil methane concentration on landfill emissions (bottom).](image)

The emissions parameterization used in WRF with constant soil methane concentration (solid blue line in bottom panel of Figure 5.10) predicts large variability in landfill methane emissions at night, and it is clear that adding CALMIM-predicted variability to the soil methane concentration will not have a significant effect on the short-term or time-averaged emissions rate.

One important point regarding diurnal variability is that the mean emissions taken over 15 hours is significantly lower than when taken over the 6-hour daytime period (see Figure 5.10). Similar diurnal variability was seen in the Sandtown landfill simulations in Chapters 3 and 4. Here the mean emissions over the 6-hour daytime period are 231 g/min (scaled by
variable soil concentration), and 222 g/min (constant soil concentration), while the mean emissions over the 15-hour period are 210 g/min (scaled by variable soil concentration), and 201 g/min (constant soil concentration). This discrepancy in mean emissions values due to different time intervals implies that relying only on measurements taking during the day could lead to over-prediction of the total emissions over the course of the day (and hence annual emissions). In the case of this specific simulation, that over-prediction could be 10% due to the 10% difference in mean emissions values, but a thorough examination of multiple 24-hour periods is necessary to draw conclusions about the representativeness of daytime measurements for understanding 24-hour emissions and by extension annual emissions.

5.6 Conclusions and Future Work

A novel modeling framework for studying landfill methane emissions and the TDM was applied to a young landfill in the Southeastern U.S. The model performed well through comparisons of field data with both local wind conditions and passive scalar transport through concentration measurements of tracer gas given a steady release rate. The model showed that when simulating the TDM, standard deviation of the TDM-measured emissions is always greater than standard deviation of the true model emissions, which is in agreement with results from the last chapter. Evaluating the TDM’s accuracy showed that the TDM has significantly lower error on days when the average wind speed is $\geq 5$ m/s, which is in agreement with rules of thumb regarding using the TDM in the field when wind speed is relatively high.

Future work will include modeling longer time periods (24 hours at a time) and a larger number of days representing a larger variety of meteorological conditions. One open question that will be tested is how using time-varying soil methane concentration will affect both the magnitude of the emissions and the TDM errors. This effect was examined in a simplified way by scaling the emissions in post-processing based on how much CALMIM predicted the soil methane concentration would change on an hour-by-hour bases. This effect of variable soil methane concentration could be implemented more rigorously by providing time-varying input to the WRF model, though it is expected that results will be similar. Soil methane concentration could also be updated every hour or even every time step within the WRF code using a prescribed function based on diurnal trends predicted by CALMIM and shown in other landfill research literature.

Future work will also include understanding seasonal changes in emissions by changing soil methane concentration values based on CALMIM predictions and not matching model emissions values to field-measured emissions. This could inform how emissions change seasonally in older landfills because it has been shown that this model performs well for this field site.
Chapter 6

Summary and Recommendations

6.1 Summary

This dissertation contributes to the field of landfill research, and in particular, to the understanding of the tracer dilution method (TDM). Specifically, this work develops a robust modeling framework for simulating the TDM with the Weather Research and Forecasting model (WRF) and quantifying the errors associated with the TDM. Landfills are a significant contributor of methane emissions, and many measurement methods and models exist to quantify landfill emissions and estimate annual emissions. These measurement methods and models all have their own advantages, disadvantages, and limitations. The work in this dissertation focuses on one particular measurement method, the TDM, because it is a relatively new method in terms of landfill methane emissions measurements. Currently the TDM is used with little guidance from true understanding of atmospheric plume dynamics. Some of the major uncertainties stem from tracer placement, downwind measurement distance, and wind variability. The model studies presented in this dissertation are for the first time enabling sensitivity tests to these factors as well as for the first time an estimate of the errors accrued by TDM measurements. Estimating TDM errors is possible with these simulations because with the model the true landfill emissions are known.

Chapters 3, 4, and 5 all use the same process for simulating the TDM. To simulate the TDM, the following components are needed: known, continuous landfill methane emissions assigned to each grid point on the surface of the landfill which result in a methane plume traveling downwind; known, steady tracer emissions assigned to a few point sources on the surface of the landfill which result in a tracer gas plume; and a downwind transect measurement path, where a simulated vehicle moves back and forth across the plumes, collecting concentrations of both methane and tracer gas. The simulated vehicle collects concentration transects of both methane and tracer gas at 8 m/s, and each traverse of the transect path takes ~2 - 5 minutes depending on the simulation and the length of the path. For each traverse of the transect path, the concentration transects are used to calculate one TDM-measured emissions value.
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Chapters 3 and 4 involve simulations of a real landfill, Sandtown Landfill in Delaware, U.S., where a field campaign was conducted on March 6, 2010. The wind data collected during this field campaign were used to validate the meteorological component of the WRF model to ensure that scalar transport of methane and tracer gas would be simulated reasonably by WRF. The purpose of these simulations was a semi-idealized study of atmospheric dispersion to examine TDM measurement errors, not to replicate field conditions. Agreement with atmospheric observations was not necessary for these chapters, but was provided in Chapter 3 to explain to the landfill emissions community that the atmospheric dispersion model can capture basic features of the flow over the landfill. The agreement of the model with wind speed and wind direction is quite good, and these two factors determine the direction of the plume, which is the key focus here. A comparison to measurements of concentration is also given in Chapter 3 to illustrate that the variability in the concentration fields is similar to what is observed in the field. Only one afternoon of data is available from the Sandtown field experiments, yet these landfill studies are able to provide a good setting for examining the TDM. None of the TDM parameters such as tracer placement or transect distances could be examined in the field, and the model uniquely provides this ability.

In Chapter 3, the landfill methane emissions were prescribed at a steady rate of 1800 g/min. The goal of this chapter was to examine the accuracy of the TDM under several different scenarios. The TDM setup factors examined were: transect angle relative to the gas plume, transect vehicle speed, tracer placement relative to the methane hot spot, and distance from the methane hot spot to the transect path. The external factors examined were: topography (simulations run with the true topography, a 25 m hill, and with the 25 m hill artificially removed from the model terrain) and wind direction (two different days were simulated with mean wind directions differing by $\sim$90°).

Results from Chapter 3 showed that tracer placement had a more significant effect on the TDM than distance from the methane hot spot to the transect measurement path. A comparison of the results from the simulation with the 25 m hill and without the hill showed significant differences, implying that TDM studies performed on flat fields (such as the Mønster et al., 2015 study) may not be applicable to landfills with more complex topography, such as Sandtown Landfill. The main result from Chapter 3 comes from the comparison of results from the two different wind direction simulations, which showed that the TDM-measured emissions were significantly more accurate when the wind direction was blowing across the sloped side of the landfill containing the methane hot spot and less accurate when the wind was blowing down the same sloped side. The differences in TDM accuracy were a result of different edges of the rectangular hot spot being used for the “upwind edge” tracer placement, and importantly, the fact that these different edges spanned different ranges of elevation.

Chapter 3 used steady landfill methane emissions to focus on understanding the effects of changing other factors in the TDM, such as tracer placement, without adding complications from wind-induced variability in the landfill emissions. In Chapters 4 and 5, however, landfill methane emissions were unsteady to examine the effect of variable emissions on the accuracy of the TDM. In these chapters, a surface flux parameterization was used to calculate the
landfill methane emissions at each landfill grid cell at each time step. This surface flux parameterization is based on how WRF calculates surface fluxes for heat and moisture. The parameterization introduces a surface wind speed dependence to the emissions, which has been observed in the field in multiple studies. The emissions parameterization relies on the difference between soil methane concentration and atmospheric methane concentration and a coefficient dependent on the turbulent flow conditions.

Because soil methane concentration is an important factor in the surface emissions parameterization, realistic soil methane concentration values are taken from the California Landfill Methane Inventory Model (CALMIM). The CALMIM model was designed originally to help California landfills report their annual landfill methane emissions because California had specific regulations, but it is applicable to all areas of the world due to its database of decades of historical meteorological data from all over the world. In WRF, the surface fluxes are calculated using soil scalar values located at 5 cm deep in the soil, so CALMIM output at 2 inches (∼5 cm) was used as the soil methane concentration values in the surface flux calculations. The CALMIM concentration was used as the mean concentration value, and then a log-normal distribution of soil methane concentration values was generated to imitate the heterogeneity seen in field measurements.

In Chapter 4 there were two main goals: understanding the impact of variability in the landfill emissions on variability in the TDM-measured emissions, and understanding TDM-measured emissions accuracy under different atmospheric conditions that occurred at different times of day. The TDM was simulated at Sandtown Landfill over eight 90-minute measurement periods on two different days. The following comparisons were made: steady emissions and unsteady emissions simulations, the eight different time periods from two different days, and two different tracer configurations (the upwind edge configuration from Chapter 3, and an alternative placement further upwind of the methane hot spot and roughly spanning the width of the landfill).

Results from Chapter 4 showed that the variability in TDM-measured emissions over 90 minutes was similar whether the model’s true landfill emissions were steady or variable, indicating that the TDM may introduce erroneous variability into the measurements. Results also showed that distance from the hot spot affected the TDM accuracy more significantly for the alternative tracer configuration than the base case configuration, emphasizing the importance of choosing the tracer configuration carefully in the field. The variability in TDM-measured emissions was found to correlate more directly with fluctuations in wind speed and wind direction than with magnitude of fluctuations in the true landfill emissions.

For Chapter 5, a different landfill was simulated. This landfill is located in the Southeastern U.S. and was the site of a multi-year field measurement campaign to test and understand the TDM. Previous studies involving this landfill include an investigation of how to filter TDM field measurements to determine a reasonable mean estimate for landfill emissions (Foster-Wittig et al., 2015), and a comparison of common first order decay model estimates of emissions at this landfill to the TDM-measured emissions (De La Cruz et al., 2016). This landfill was relatively young at the time of the field measurements, with measurements beginning ∼1.5 years after the landfill opened and continuing until the landfill was ∼4 years old.
Because the landfill was so young, the emissions measurements do not follow the expected seasonal pattern of highest in the summer months and lowest in the winter months.

In Chapter 5, the emphasis was on model validation with the goal of developing a framework for better understanding how to estimate annual landfill methane emissions with limited field measurements. One method for estimating annual emissions with limited field data is to take the integral of the curve created by the time series of the field measurements. This method, however, does not take into account variability that could occur between data points.

Results from Chapter 5 showed that the standard deviation of TDM-measured emissions is always larger than the standard deviation of the model’s true landfill emissions. Results also showed that the TDM has significantly lower error when wind speed \( \geq 5 \) m/s, and that the longer the measurement period over which TDM-measured emissions are averaged, the more accurate the TDM-measured average value is.

### 6.2 Recommendations

Best practices for ensuring accurate TDM-measured emissions and accurate annual landfill emissions estimates are both ongoing areas of research, and future work can be recommended based on the findings in this dissertation.

The first question addressed by this dissertation is how to use the TDM to obtain the most accurate landfill emissions measurements. Recommendations for maximizing TDM accuracy can be made regarding TDM setup (tracer placement, transect measurement distance from landfill, transect vehicle speed, transect path angle) as well as conditions under which to perform the TDM (wind speed, wind direction variability, atmospheric stability). Important future work along these lines would be to address the effect of meteorological conditions during TDM measurement campaigns on error in the measurements. This way the TDM-measured emissions could be adjusted for a better understanding of the true landfill emissions.

The important question that remains open is how to get an accurate estimate of annual landfill emissions. To achieve this, it is important to understand the most likely behavior of landfill emissions during the times between field measurements. This idea can be broken down further into understanding the following components: typical diurnal variability, typical seasonal variability, and when and why we can expect emissions to deviate from “expected” diurnal or seasonal behavior. To understand emissions variability on an annual timescale, future work should include running simulations for 24-hour-long periods, running simulations for many different days that encompass a variety of seasons and meteorological conditions, and running simulations over several different landfills with varying conditions such as age and size. With enough simulations, probability density functions (PDFs) could be created to understand the most likely emissions for various scenarios.

A few additional open questions raised by the work in Chapter 5 are: what is the extent of variability in soil methane concentration over longer time scales (e.g. a day, a week, a
year); and what is the impact that changing soil concentration has on the landfill emissions rate. To try to answer both of these questions, a soil methane model could be added to WRF to compute methane transport through the shallow soil column at each time step and update the soil concentration value that goes into the surface flux calculations. It is expected that if the CALMIM model is used, the variability in soil methane concentration will not have a large effect on the magnitude of the landfill emissions or on the accuracy of the TDM. CALMIM does not take methane production into account and instead prescribes a fixed bottom boundary condition for soil methane concentration. As Chapter 5 showed with the young landfill, this approach is not always appropriate, and it could be improved by taking the age of waste into account. This soil column model could be improved by accounting for methane production below the cover soil layer and using this production term to update the bottom soil concentration value.

It would be advantageous for future work to include close coordination between the numerical modeling and the field measurements to test model results in the field, such as how standard deviation of TDM-measured emissions varies with distance from the landfill to the downwind measurements. Future work could also incorporate Bayesian inverse modeling based on field concentration measurements to infer the landfill emissions rate and compare landfill emissions from inverse modeling to landfill emissions from TDM measurements.

The recommendations in this dissertation are based on the results of the work herein and may not be universally applicable, but may still serve as guidelines for similar situations. They are intended to help improve landfill methane emissions measurements and annual landfill methane emissions estimates in particular using the tracer dilution method. These improvements may lead to policies and regulations that mitigate the effects of global warming.
Bibliography


