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

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ABSTRACT

Landfills are a significant contributor to anthropogenic methane emissions, but measuring these emissions can be challenging. This work uses numerical simulations to assess the accuracy of the tracer dilution method, which is used to estimate 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 methane and tracer gas concentrations are collected along various transects downwind from the landfill for use in the tracer dilution method. 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, where the tracer and methane plumes become well mixed. Tracer placement has the largest effect on percent error, and topography and wind direction both have significant effects, with measurement errors ranging from −12% to 42% over all simulations. Transect angle and transect speed have small to negligible effects on the accuracy of the tracer dilution method. These tracer dilution method simulations provide insight into measurement errors that might occur in the field, enhance understanding of the method's limitations, and aid interpretation of field data.

1. Introduction

Landfills are one of the largest anthropogenic sources of atmospheric methane in the U.S. (U.S. Environmental Protection Agency, 2014), yet measuring these emissions is challenging. Many methods of estimating landfill methane emissions are expensive [e.g., differential adsorption LiDAR (Babilotte et al., 2010)], labor intensive [e.g., flux chamber (Abichou et al., 2006)], and associated with high levels of uncertainty if measurements are conducted over portions of the landfill surface given the spatiotemporal variability of emissions (Foster-Wittig et al., 2015). 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 et al., 2010; Foster-Wittig et al., 2015). 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 (Monster 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

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et al., 2003; Foster-Wittig et al., 2015; Monster et al., 2014). The methane emissions are estimated based on the downwind ratio of methane concentration to tracer concentration, as shown in Eq. (1):

$$Q_{CH4} = Q_{tr} \frac{\int_{x_1}^{x_2} c_{CH4}(x)dx}{\int_{x_1}^{x_2} c_{tr}(x)dx} \frac{m_{CH4}}{m_{tr}} \quad (1)$$

where Q_{CH4} is the methane emissions rate, Q_{tr} is the tracer emissions rate, $c_{CH4}(x)$ is the methane concentration, $c_{tr}(x)$ is the tracer concentration, both of which are functions of distance along the transect, x_1 is the starting location of the transect measurement, x_2 is the end location of the transect, and m_{CH4} and m_{tr} are the molecular weights of methane and the 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 side. 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 (Monster et al., 2014).

The recent study by Monster 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. It was concluded that increasing the measurement distance (from 370 m to 1200 m) increased the accuracy of emission estimation (from 36 ± 21% to 20 ± 2%), although measurements were overestimated by 17% at the farthest measurement distance (1200 m). The authors attributed the decreasing error with measurement distance to a smaller relative difference in dispersion characteristics of misaligned gas plumes at distances farther downwind. The best approximation was found when the tracer gas releasing bottles were located at the center of the methane gas emissions (<6%). This study highlighted some important factors 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 et al., 2010). Both the Monster et al. (2014) and Babilotte et al. (2010) studies used a small number of gas cylinders releasing point sources of methane and were performed on flat topography. Therefore, neither took into account the possible effects of the methane being emitted from a large area source on the order of hundreds of m^2 or the complex topography often seen at landfill sites. The errors in these studies arose from measured gas concentrations, the gas flow releases, data filtering, and source and transect locations (Monster et al., 2014). Using Eq. (1) to estimate emissions, measured percent errors ranged from 2 ± 6% to 36 ± 21% (Monster et al., 2014) or 3.7% to 19.2% (Babilotte et al., 2010).

This study will use numerical modeling to 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. 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 specifically study the tracer dilution method.

Given the current limitations of field studies and uncertainty in current applications of the tracer dilution method, assessment of the method through numerical, modeling 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 enhancing understanding of the method's limitations and aiding interpretation of field data.

2. Methods

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 weather 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 generally limited by the height of the boundary layer (O(1 km)). At coarse resolutions, 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 variability in the wind field and 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 larger turbulent eddies (Deardorff, 1970). This approach is called large-eddy simulation (LES), where only 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 horizontal