Short-Term Landfill Methane Emissions Dependency on Wind

Madjid Delkash^a, Bowen Zhou^b, Byunghyun Han^a, Fotini K. Chow^b, Chris W. Rella^c, Paul T. Imhoff^{a*}

^aDepartment of Civil and Environmental Engineering, University of Delaware, Newark, DE 19716

^bDepartment of Civil and Environmental Engineering, University of California, Berkeley, CA 94720-1710

^cPicarro, Inc., 3105 Patrick Henry Dr. Santa Clara, CA 95054

Abstract

Short-term (2-10 h) variations of whole-landfill methane emissions have been observed in recent field studies using the tracer dilution method for emissions measurement. To investigate the cause of these variations, the tracer dilution method is applied using 1-min emissions measurements at Sandtown Landfill (Delaware, USA) for a 2-h measurement period. An atmospheric dispersion model is developed for this field test site, which is the first application of such modeling to evaluate atmospheric effects on gas plume transport from landfills. The model is used to examine three possible causes of observed temporal emissions variability: temporal variability of surface wind speed affecting whole landfill emissions, spatial variability of emissions due to local wind speed variations, and misaligned tracer gas release and methane emissions locations. At this site, atmospheric modeling indicates that variation in tracer dilution method emissions measurements may be caused by whole-landfill emissions variation with wind speed. Field data collected over the time period of the atmospheric model simulations corroborate this result: methane emissions are correlated with wind speed on the landfill surface with $R^2 = 0.51$ for data 2.5 m above ground, or $R^2 = 0.55$ using data 85 m above ground, with emissions increasing by up to a factor of 2 for an approximately 30% increase in wind speed. Although the atmospheric modeling and field test are conducted at a single landfill, the results suggest that wind-induced emissions may affect tracer dilution method emissions measurements at other landfills.

*Corresponding author

Email addresses: delkash@udel.edu (Madjid Delkash), zhoubowen58@gmail.com (Bowen Zhou),

gragebhan@gmail.com (Byunghyun Han), imhoff@udel.edu (Paul T. Imhoff), tinakc@berkeley.edu (Fotini K.

Chow), rella@picarro.com (Chris W. Rella)

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1. Introduction

Methane (CH_4) from biological decomposition of buried waste in landfills is considered an important source of anthropogenic CH_4 emissions (Scheehle et al., 2006). Measurement of CH_4 emissions is useful for evaluating landfill gas control measures and improving estimates of CH_4 emissions for greenhouse gas inventories. CH_4 emissions from landfills are not constant, with annual changes associated with aging of waste (Foster-Wittig et al., 2015) or changes in the composition of landfilled materials (Weitz et al., 2002; Allen et al., 1997), and seasonal variations due to changes in temperature and moisture content of cover soils that affect gas transport and methane oxidation (Wang et al., 2011; Chanton and Liptay 2000; Stern et al., 2007).

Short-term emissions variations due to weather have also been reported. Czepiel et al. (2003) measured whole-landfill CH_4 emissions using the tracer dilution method (TDM) and found CH_4 emissions increased by a factor of five when barometric pressure dropped approximately 15 mbar over a 1-month period of emissions measurements. Xu et al. (2014) measured CH_4 emissions from portions of landfill cells using the eddy covariance method and attributed a 35-fold variation in emissions to barometric pressure variations.

Over even shorter time periods, wind speed has been implicated as a factor affecting CH_4 emissions (McBain et al., 2005; Rachor et al., 2013) - as wind speed increases, gas pressures fluctuate because of atmospheric turbulence at the landfill surface causing a pumping effect that increases emissions. Numerical modeling of gas transport processes in landfill cover soils suggests that pressure pumping caused by wind turbulence may increase CH_4 emissions up to four times that of the CH_4 diffusive flux for some soil conditions (Poulsen and Moldrup, 2006). While the

Nomenclature: Tracer Dilution Method (TDM), cavity ring-down spectroscopy (CRDS), Advanced Regional Prediction System (ARPS), Constant Flux 1 (CF1), Constant Flux 2 (CF2), Variable Flux 1 (VF1) and Variable Flux 2 (VF2), Three point flux (3Pts), above ground level (AGL).

effect of wind speed on CH₄ emissions has been observed in flux chamber (Rachor et al., 2013) and eddy covariance (McBain et al., 2005) measurements over portions of a landfill surface, there are no data indicating variations in whole-landfill CH₄ emissions because of wind speed.

TDM, also known as the tracer correlation or dynamic plume technique, was developed to quantify whole-landfill CH₄ emissions and has seen increased use in the past several years because of improved gas measurement technology (Foster-Wittig et al., 2015; Mønster et al., 2015; Mønster et al., 2014). A tracer gas, traditionally sulfur hexafluoride (SF₆) or nitrous oxide (N₂O) but more recently acetylene (C₂H₂), is released at a known rate at multiple locations on the landfill which then mixes with CH₄ emissions from the landfill surface (Czepiel et al., 2003; Galle et al., 2001). Downwind of the source if the released tracer gas is sufficiently well mixed with emitted CH₄, the mass flux of CH₄ is calculated from the known mass flux of the tracer gas multiplied by the ratio of the CH₄ to tracer gas concentration

$$E_{CH_4} = E_T \frac{M_{CH_4}}{M_T} \frac{C_{CH_4}}{C_T} \quad (1)$$

where E_{CH_4} and E_T are CH₄ and tracer emissions rates, respectively, expressed as mass flowrate (kg/min); M_{CH_4} and M_T are the molecular weights of CH₄ and tracer; and C_{CH_4} and C_T are the concentrations of CH₄ and tracer above background (ppm). If measured at a fixed location, this concentration ratio can be averaged over time by regressing scatter plots of C_{CH_4} versus C_T to obtain the fitted slope used for the ratio C_{CH_4}/C_T in equation (1) (Galle et al., 2001; Börjesson et al., 2009). When measured by a moving vehicle that traverses the downwind gas plumes, similar scatter plots can be generated (Foster-Wittig et al., 2015; Mønster et al., 2014), or, more commonly, C_{CH_4} and C_T are integrated individually in space and the ratio of the two integrations are used to represent the ratio C_{CH_4}/C_T (Foster-Wittig et al., 2015; Mønster et al., 2014).

With increasing TDM applications, significant variations in measured CH₄ emissions have been reported for short measurement campaigns. For example, during measurement campaigns that ranged from 1.7 to 4.3 h, Scheutz et al. (2011) reported a coefficient of variation of CH₄ emissions up to 0.30. Similarly, in a recent study of nine Danish landfills where single measurement campaigns (2 to 10 h duration) were conducted at each landfill, the average coefficient of variation was 0.19 for the nine landfills with a maximum of 0.35 (Mønster et al., 2015). On the other hand, when the TDM method was tested with controlled releases of CH₄ and tracer (Mønster et al., 2014), the coefficient of variation was much smaller: for three tracer/CH₄ release configurations and measurements at three downwind transects, the mean coefficient of variation was 0.05 with maximum of 0.15, which occurred for the configuration with significant misalignment between CH₄ and tracer and for measurements close to the points of gas release. The cause of the increased coefficient of variation for landfill CH₄ emissions versus controlled CH₄ releases is unclear: it may be due to misalignment of tracer release and regions of CH₄ emissions, short-term variations in emissions possibly due to wind, or other factors.

The primary objective of this work is to determine if short-term variations in whole-landfill CH₄ emissions measured with TDM might be due to variations in wind speed. If wind affects whole-landfill CH₄ emissions, in some situations TDM may result in biased estimates of emissions, since TDM requires sufficient wind speed to generate well-mixed gas plumes downwind of the landfill (Foster-Wittig et al., 2015), and thus measurements are not possible under low wind speeds (Foster-Wittig et al., 2015; Galle et al., 2001). A secondary objective is to evaluate the utility of atmospheric dispersion modeling to understand the influence of atmospheric conditions on landfill emissions measurements requiring sampling of landfill gas plumes.

To accomplish these objectives, CH₄ emissions data were collected using TDM from the Sandtown Landfill in Delaware (USA) for a short-term experiment in March 2010. Correlations between wind conditions on the landfill surface and CH₄ emissions were explored with field data. An atmospheric dispersion model was developed for the site and used for three purposes: to determine the travel time of emissions from the landfill surface to a downwind measurement location; to refine data filtering techniques for TDM, since in the atmospheric model CH₄ emissions were known and could be compared with TDM results from analysis of model output; and to assess if temporal variation in emissions determined from TDM might be caused by misalignment of tracer/CH₄ emissions locations or wind conditions. Field data and numerical model results were collected at one downwind location at two measurement heights, at the ground surface and at approximately 85 m above ground to evaluate the benefit of above ground measurements on TDM.

2. Materials and methods

2.1 Site description

The Sandtown Landfill has an active landfill gas collection system in all landfill cells and ongoing waste disposal activities. Area A-B (110,000 m²) in Figure 1 was filled with 532,000 tons of household waste from 1980 to 1988 and is now closed and equipped with a final cover. A total of 1,226,000 tons of waste were landfilled in Area C and D (170,000 m²) from 1988 to 1998 and these regions have intermediate covers. Area E (132,000 m²) is an active cell where 2 million tons of waste have been landfilled since 1999: portions of Area E have intermediate and daily cover soils. The landfill rises 40 m above the surrounding topography and is located in a coastal plain with minimal variations in ground elevation for a 100 km radius surrounding the site. The landfill

is in a rural area with low traffic density and is surrounded by farms with 25 to 30 m high pine trees.

Figure 1

2.2 Field emissions measurements

TDM was utilized to quantify CH₄ emissions from the Sandtown Landfill. Details of the method are found elsewhere (Czepiel et al., 2003; Foster-Wittig et al., 2015; Mønster et al., 2015). In this application, C₂H₂ was selected as the tracer gas (Foster-Wittig et al., 2015; Mønster et al., 2014). Three C₂H₂ gas cylinders were located on the upstream edge of the active landfill region on Area E (Figure 1), identified as the main CH₄ source during preliminary ground-level atmospheric CH₄ measurements around the landfill in March 2010. The gas cylinders were arranged 70 m apart in a line perpendicular to the mean wind direction. Mass flow controllers were used to discharge C₂H₂ at a constant mass flow rate of $E_{C_2H_2} = 52.23 \text{ g C}_2\text{H}_2/\text{min}$.

C_{CH_4} and $C_{C_2H_2}$ measurements for the TDM are often obtained by making transects of the downwind gas plumes perpendicular to the mean wind direction with a vehicle-mounted gas sampling system (Czepiel et al., 2003; Börjesson et al., 2009). Downwind roadways are not, however, always located with suitable alignment for TDM measurements. For these situations, if plume meandering is adequate to cause plumes to move into and out of a fixed measurement location, stationary measurements may be used and have provided similar estimates of CH₄ emissions (Green et al., 2010). Here, all measurements were conducted at one geographic location, indicated in Figure 1, since road alignment was not ideal for vehicle-mounted gas sampling with the prevailing wind. Sampling along downwind roads was attempted but resulted in poor data: tracer and CH₄ plumes were either not well mixed or gas concentrations were too low for accurate measurements.

A prototype gas analyzer developed by Picarro, Inc. (Sunnyvale, CA) and based on an all-optical technique called cavity ring-down spectroscopy (CRDS) was used to measure C_{CH_4} and $C_{C_2H_2}$ (Crosson, 2008). The CRDS analyzer measures CH_4 and C_2H_2 in less than 2 s with a measurement precision (one estimated standard error) of 0.8 ppb and 0.2 ppb for CH_4 and C_2H_2 , respectively, and has been used in other recent TDM measurements at landfills (Foster-Wittig et al., 2015; Mønster et al., 2015). Instrument calibration is discussed in Supporting Information.

The TDM was used to measure CH_4 emissions using data collected at 2.5 and 85 m above ground level (AGL) at the same location ($39^{\circ}01'45''N$, $74^{\circ}42'47''W$) between 14:00 to 16:00 local time on March 6, 2010. A single stationary location was used since a large, open region was needed to launch and monitor the weather balloon used for measurements at 85 m, and a limited number of locations were available in the actively farmed land surrounding the landfill. A short measurement period was selected since changes in landfill operations or in soil temperature and barometric pressure, which are known to affect CH_4 emission, would be minor, permitting an examination of wind on emission. For 85 m measurements, a chloroprene weather balloon (2.4 m diameter) was inflated with helium and flown while tethered using light-weight nylon rope. Teflon tubing (1/8" ID, 3/16" OD, and 150 m long) was attached to the balloon and connected to a vacuum pump, which when operated resulted in a 22 s residence time of the air in the tubing. The balloon height was measured by the nearly vertical tubing length and ranged from 70 to 100 m, with an average height of approximately 85 m. By connecting and disconnecting the Teflon tubing to the gas analyzer alternatively every 4 to 9 minutes, measurements of C_{CH_4} and $C_{C_2H_2}$ could be made at 2.5 m AGL or 85 m AGL at the flying weather balloon.

Meteorological data were collected during the sampling period with a wind monitor (RM Young's 0513, Campbell Scientific) 2 m AGL on top of Area E, which is 40 m higher than surrounding terrain (see Figure 1), and with a weather station at the base of the landfill. One-minute averaged wind speed data were recorded by the wind monitor. Average temperature and barometric pressure during measurements at the landfill base were 8 °C and 101.3 kPa, respectively.

2.3 Atmospheric dispersion modeling

Given the variability in CH₄ emissions discussed below, relationships between landfill operations, atmospheric conditions, and CH₄ emissions were explored. We postulated that downwind CH₄ and tracer concentrations obtained from ground and airborne measurements could be used to quantify temporal CH₄ emissions variations. To test this, an atmospheric dispersion model was developed that described wind conditions in the vicinity of the landfill for the 2 h TDM test. Controlled releases of CH₄ and tracer were made at different locations in the model landfill, and then forward simulations performed to predict downwind gas concentrations at 2.5 and 85 m AGL at the field measurement location shown in Figure 1. Simulations were performed to test three hypotheses about TDM-measured whole landfill CH₄ emissions: TDM emissions measurements will be correlated with wind speed if (1) local CH₄ emissions vary with wind speed causing wind-induced variations of whole landfill emissions, (2) if the spatial distribution of CH₄ emissions varies with wind speed but whole landfill emissions are constant, and (3) if local CH₄ emissions are constant but significantly misaligned with tracer emissions locations. In the process of testing these hypotheses, the atmospheric dispersion model was also used to develop data filtering techniques that were then applied to field measurements.

The Advanced Regional Prediction System (ARPS) was used for simulations. ARPS was developed at the Center for Analysis and Prediction of Storms at the University of Oklahoma. It is a non-hydrostatic meso-scale and small-scale finite-difference numerical weather prediction model that runs in parallel using the message passing interface. Descriptions of the model can be found in Xue et al. (2000) with relevant details presented in Supporting Information and summarized below.

Daytime atmospheric flow was simulated using ARPS for 3 h starting from 13:00 to 16:00 local time on March 6, 2010. Results from the last 2 h period were used for analysis. To resolve the local flow around the landfill, a multi-scale approach was adopted. Simulations were first performed on a 200 x 200 km² area with 2400 m horizontal resolution which covered the entire Delmarva Peninsula. Realistic initial and lateral boundary conditions were obtained from meteorological analysis data produced by the North American Mesoscale forecast system. Results from the 2400 m grid were used to drive lateral boundaries of a 150 m grid, which was then fed into a 30 m grid. Vertical grid stretching was applied to better resolve the surface layer. On the finest grid, the vertical resolution was 20 m with 5 m spacing near the surface. The land surface was represented with high resolution terrain (10 m) and land cover (30 m) data from the Delaware Solid Waste Authority and the U.S. Geological Survey (see Figure S1 in Supporting Information). Nested model parameters are listed in Table S1 in Supporting Information.

On the 2400 m grid, ARPS was run in meso-scale mode with boundary layer parameterizations (Sun and Chang, 1986) to model the synoptic-scale flows. On the inner (150 m and 30 m) grids, large-eddy simulations were performed to resolve turbulent flows in the atmospheric boundary layer, details of which are described in Supporting Information. On the 30 m grid a numerical tracer, representing CH₄ or C₂H₂, was released at constant or variable mass

flow rate, depending on the simulation, on top of the landfill from 1400 LST onwards. Tracers were released from the three tracer release points shown in Figure 1 that correspond to the location of field tracer release points, or from 100 points or 89 points distributed in a spatially uniform grid covering Area C or Area D, respectively, as shown in Figure 1. A vertical column of model results at the field sampling location was recorded every 2 s, while 3D flow fields were output every minute. Wind velocity was also output every 2 s at 2.5 m AGL for selected locations.

Due to the fine vertical grid spacing with near surface grid-stretching, the 30 m pine trees surrounding the landfill were represented by 7 grid points in the vertical direction. The traditional representation of trees in mesoscale atmospheric models with an elevated roughness length ($z_o \sim 1$ m) was insufficient for the large-eddy simulation domain. Therefore a canopy model was adopted to explicitly resolve the additional drag on the atmospheric flow by the trees, which is described in Supporting Information.

The increased turbulence within the canopy layer led to more mixing of the tracer, and resulted in low, broad peaks near the surface. In general, the canopy model acted like a semi-permeable flow obstacle. An internal boundary layer formed downwind of the canopy, so that wind speed reduction was also felt above the canopy height. At the 85 m balloon height, mean wind speed was reduced by ~10% when the canopy model was on.

Four different numerical tracer release configurations were considered. Three Point (3Pts) simulation mimicked C_2H_2 release in the field tests and is described by

$$Q_{3Pts}(\vec{x}_i, t) = \frac{Q}{3}; i = 1:3 \quad (2)$$

where Q is the total tracer mass flux that is invariant with time. The three release points ($\vec{x}_i$) were in close proximity to the C_2H_2 release points for the field tests shown in Figure 1. For the Constant Flux 1 (CF1), Variable Flux 1 (VF1), and Variable Flux 2 (VF2) simulations, the numerical tracer

was released from 100 uniformly spaced grid points in Area C in Figure 1. For the Constant Flux 2 (CF2) simulation, the numerical tracer was released from 89 uniformly spaced grid points in Area D in Figure 1. The tracer releases for CF1 and CF2 are described by

$$Q_{CF1}(\vec{x}_i, t) = \frac{Q}{100}; i = 1:100 \quad (3)$$

$$Q_{CF2}(\vec{x}_i, t) = \frac{0.89Q}{89}; i = 1:89 \quad (4)$$

where $Q_{CF1}(\vec{x}_i, t)$ and $Q_{CF2}(\vec{x}_i, t)$ are the tracer mass fluxes at location $\vec{x}_i$ and are equal to the total mass flux divided by the 100 or 89 release points in Area C and D, respectively. Because identically sized surface grid cells were used for CH₄ emissions in CF1 and CF2 while Area D was approximately 89% of Area C, the number of release points was larger for CF1 than CF2. CF1 and CF2 simulations were used to represent either a uniform, time-invariant release of CH₄, or release of a tracer gas in perfect alignment with the CH₄ release (CF1 simulation) in simulations VF1 and VF2, where CH₄ emissions varied with wind conditions.

The numerical tracer released in simulations VF1 and VF2 was dependent on wind speed

$$V(\vec{x}_i, t) = \sqrt{u(\vec{x}_i, t)^2 + v(\vec{x}_i, t)^2 + w(\vec{x}_i, t)^2}; i = 1:100 \quad (5)$$

where (u, v, w) are magnitudes of wind velocity in x , y and z directions at 2.5 m AGL. Tracer release for VF1 is described by

$$Q_{VF1}(\vec{x}_i, t) = \frac{V(\vec{x}_i, t)}{\sum_{j=1}^{100} V(\vec{x}_j, t)} Q; i = 1:100 \quad (6)$$

where $Q_{VF1}(\vec{x}_i, t)$ is the tracer mass flux at location $\vec{x}_i$ and is equal to the total mass flux Q normalized by the relative wind speed at this measurement location. The summation of Q_{VF1} over all $\vec{x}_i$ is the total CH₄ flux from Area C and was invariant with time but varied spatially at each time step depending on local wind speed.

The numerical tracer release in simulation VF2 is described by

$$Q_{VF2}(\vec{x}_i, t) = \frac{\sum_{j=1}^{100} V(\vec{x}_j, t)}{\frac{\sum_{k=1}^n (\sum_{j=1}^{100} V(\vec{x}_j, t_k))}{n}} \left(\frac{Q}{100} \right); i = 1:100 \quad (7)$$

where $Q_{VF2}(\vec{x}_i, t)$ is the tracer mass flux at location $\vec{x}_i$ and is equal to the total mass flux Q normalized by the average relative wind speed in Area C. Here, n is the number of simulation time steps. Q_{VF2} represents CH_4 emission that is spatially uniform within Area C but varies linearly with the average Area C wind speed.

To test the three hypotheses described above, correlations between TDM-measured CH_4 emissions and wind conditions on the landfill surface were examined using simulation results. To account for the travel time for gases emitted from the landfill to the downwind measurement location (see Figure 1), estimates of travel time are necessary to adjust wind speeds at the landfill surface to the corresponding time of TDM-measured CH_4 emissions. The numerical technique of Deleersnijder et al. (2001) was applied to estimate the travel time T of numerical tracer gas to the downwind measurement location and details are in Supporting Information.

3. Results and discussion

3.1 Field emissions measurements

TDM data are typically integrated in space (Mønster et al., 2015; Foster-Wittig et al., 2015; Mønster et al., 2014) or time (Börjesson et al., 2009; Galle et al., 2001) to estimate average CH_4 emissions. To illustrate the variability in raw data from the field campaign, instantaneous emission measurements determined using equation (1) are shown in Figure 2. Data alternate between measurements at 2.5 and 85 m AGL, with time gaps between data sets for switching measurement heights. When CH_4 concentrations were at or below background, CH_4 emissions were plotted as zero, although this only indicates the CH_4 plume did not intersect the measurement location at that

time. Variability between individual CH₄ emissions measurements was large and was not correlated with changes in wind direction, which are shown in Figure 3. For example, there was no significant change in wind direction from 15:15-16:00, while emissions variations during this period are similar to those from 14:15-15:15 when wind direction did change. Average measurements during some periods differed significantly: for example, mean CH₄ emissions at 85 m AGL between 14:24-14:26 was 45% smaller than mean CH₄ emissions between 14:57-14:60 and statistically different (t-Test of means, $p < 0.001$). When averaged over the entire measurement period, though, emissions measurements at 2.5 and 85 m AGL were 2330 ± 60 g and 2289 ± 46 g CH₄/min (+/- values represent 95% confidence interval), respectively. Thus, while field data indicate temporal variability in CH₄ emissions, mean measurements at the two sampling heights for the entire 2-h sampling period were similar, differing by less than 2%. Understanding the cause of temporal variability in emissions is an objective of this work.

Figure 2

It is important to note that after subtracting background concentrations, 1.88 ppm and 0.86 ppb for C_{CH_4} and $C_{C_2H_2}$, respectively, peak C_{CH_4} and $C_{C_2H_2}$ averaged over all 1-min measurement periods were smaller at 2.5 than at 85 m AGL: $\overline{C_{CH_4}} = 258 \pm 45$ ppb and $\overline{C_{C_2H_2}} = 2.37 \pm 0.84$ ppb at 2.5 m AGL, but $\overline{C_{CH_4}} = 323 \pm 87$ ppb and $\overline{C_{C_2H_2}} = 3.59 \pm 99$ ppb at 85 m AGL. Higher gas concentrations should result in more accurate emissions measurement, since the signal to noise ratio is larger (Foster-Wittig et al., 2015). During some measurement periods at 2.5 m AGL, increases in $C_{C_2H_2}$ occurred without corresponding increases in C_{CH_4} , which we attributed to ground-level sources of C₂H₂ like motor vehicles (see Figure S2). Higher gas concentrations and the absence of unexplained increases in $C_{C_2H_2}$ at 85 m AGL suggest that emissions measurements may be more accurate at 85 than 2.5 m AGL.

3.2 Comparison of atmospheric dispersion model to field data

Wind monitor data from the top of Sandtown Landfill and C_2H_2 concentration at the downwind sampling location (see Figure 1) were used to validate the atmospheric dispersion model. Wind speed and direction from field data and model simulations are shown in Figure 3 and show good agreement: model-predicted mean ($\bar{\bar{V}}$) and standard deviation (s_V) of wind speed, and the resultant mean wind direction ($\bar{\theta}_{RV}$) on the landfill surface were $\bar{\bar{V}} = 4.5$ m/sec $s_V = 1$ m/sec and $\bar{\theta}_{RV} = 320^\circ$, respectively, compared to measured values of $\bar{\bar{V}} = 5$ m/sec $s_V = 0.8$ m/sec and $\bar{\theta}_{RV} = 320^\circ$, where the double overbar indicates a 2-h average of 1-min averaged wind speeds. Thus, without tuning or data assimilation the atmospheric model matched available wind data well.

Simulation 3Pts describes the release of C_2H_2 from three gas cylinders on the landfill surface. Normalized tracer concentrations ($C_N = (C - C_{min})/(C_{max} - C_{min})$) at 2.5 and 85 m AGL for simulation 3Pts and field measurements are displayed in Figure 4. While the match between tracer concentrations at particular times for model and field data is often poor, model-predicted mean ($\bar{C}_N$) and standard deviation (s_{C_N}) are similar to field measurements when data are compared over the same time intervals. At 2.5 m AGL, $\bar{C}_N = 0.19$ ($s_{C_N} = 0.15$) and $\bar{C}_N = 0.21$ ($s_{C_N} = 0.17$) for model and observation, respectively. At 85 m AGL, the match is not quite as good with $\bar{C}_N = 0.12$ ($s_{C_N} = 0.19$) and $\bar{C}_N = 0.19$ ($s_{C_N} = 0.20$) for model and observation, respectively. At 2.5 m AGL field data have sharper gradients than model simulation results, suggesting that mixing because of surface roughness was too large in the model. Field data at 2.5 m AGL may have been affected by vehicular sources of C_2H_2 too, which would have resulted in some of the data spikes. Because the atmospheric dispersion model provided a reasonable match to field wind

measurements and tracer data, the model was subsequently used to estimate the travel time from the landfill and to test three hypotheses about the effect of wind on TDM measurements.

Figure 3

Figure 4

3.3 Travel time

Estimates of travel time are necessary to correlate wind speed at the landfill surface to the corresponding time of TDM-measured CH₄ emissions for testing three hypotheses about the influence of wind on CH₄ emissions. Travel times from the landfill surface to the downwind measurement location were computed using the methodology described in Supporting Information for simulation CF1 and are shown in Figure 5. Because the wind monitor for field measurements provided only 1-min average wind data $\bar{V}(t)$, travel times were determined for four time-averaging increments (15 sec, 1, 15 and 30 min) to assess the impact of 1-min averaging on travel time calculations. Travel times ranged from approximately 5 to 16 min at the finest resolutions (15-sec and 1-min averaging increments), with minimal differences between 15-sec and 1-min wind speed averaging (see Figure 5). Thus, in the analyses below 1-min average wind speeds were used to compute travel times for all analyses.

Figure 5

$\bar{V}(t)$ were determined for consecutive 1-min time increments at the landfill surface and this information propagated to the downwind measurement location by adding the travel time $T(t)$, thus providing wind speed at the downwind location to correlate with measured CH₄ emissions. However, because of variability in travel time this sometimes resulted in multivalued $\bar{V}(t + T(t))$ at the downwind location: more than one wind speed on the landfill surface affected downwind C_{CH_4} at such times. At such times downwind concentrations were influenced by emissions from at

least two *different* 1-min time periods on the landfill surface. These multivalued wind speed and associated 1-min emissions data were excluded from all subsequent analyses.

3.3 Data filtering and testing hypothesis 1: Homogeneous but transient emissions

Data filtering techniques were developed for TDM-measured CH₄ emissions using the atmospheric dispersion model simulations. In these simulations, CH₄ emissions are known and temporal variation of emissions with wind speed are also known and specified. Thus, the numerical model provides a convenient platform for evaluating data filtering that will later be applied to field data. Because only 1-min average wind data were available from the field, to explore relationships between CH₄ emissions and wind speed emissions data were grouped into consecutive 1-min intervals for simulations too, which facilitated comparison between the simulation and field results.

To develop the data filtering methods, data without any filtering were first considered. To illustrate the procedure, consider simulation pair VF2/CF1 where VF2 represents time-varying CH₄ emissions that are linearly related to wind speed (equation (7)), while CF1 represents a constant tracer emission that is perfectly aligned with CH₄ emissions (equation (3)) from Area C at Sandtown Landfill. The purpose of this comparison is to determine the role of wind speed on TDM-measured CH₄ emissions, by comparing to a tracer that is perfectly aligned but not wind dependent. The 2 s numerical simulation data showing variable CH₄ emissions at the landfill surface and corresponding downwind VF2/CF1 emissions measurement at 2.5 m are shown in Figure 6. Downwind measurements lag emissions on the landfill surface because of gas plume transit. The 2 s downwind VF2/CF1 measurements are less variable than actual emissions because of atmospheric mixing, which smooths the peaks and valleys of concentrations at the landfill surface.

Figure 6

The 2 s data were grouped into consecutive 1-min time intervals; concentrations below 0.1% of maximum model concentrations on the landfill surface were associated with numerical error and discarded. The remaining data in each scatter plot of VF2 versus CF1 measurements were linearly regressed and the slope of the best-fit line used to estimate $C_{CH_4}/C_{C_2H_2}$ in equation (1) to calculate CH₄ emissions during each 1-min interval. An example scatter plot is shown in Figure S3 and further discussed in Supporting Information.

To examine the relationship between CH₄ emissions and wind speed on the landfill surface, the average wind speed over the region of CH₄ emissions was first determined for each 1-min time interval: $\bar{V}(t) = \int_{t-0.5 \text{ min}}^{t+0.5 \text{ min}} V_m(t') dt' / 1 \text{ min}$, where V_m is the wind speed at the wind monitor location interpolated from the model grid. This wind speed was propagated to the downwind measurement location by adding the travel time $\bar{V}(t + T(t))$. Wind speed data were linearly interpolated to provide the wind speed that corresponded to the midpoint of each 1-min emissions measurement. CH₄ emissions were then normalized with the average emission over the 2-h simulation time and plotted in Figure 7 for each 1-min time increment versus wind speed, also normalized with the average wind speed over 2 h. Data at 2.5 and 85 m AGL were correlated with wind speed ($R^2 = 0.39$ and $R^2 = 0.31$), but the correlation was not strong and emissions did not vary with a slope close to 1.0 for 85 m data, as would be expected from equation (7).

Because the 1:1 relationship between CH₄ emissions and wind speed was specified in simulation VF2 (see equation (7)) but was not reproduced with the TDM for data at 85 m, data filtering techniques were explored. Foster-Wittig et al (2015) and Monster et al. (2014) examined data analysis procedures for TDM-measured CH₄ emissions. Using downwind transects of landfill gas plumes, Foster-Wittig et al. (2015) developed three method quality indicators to determine if

measurements were acceptable: signal to noise ratio > 10 , $R^2 > 0.80$ for correlation between C_{CH_4} and $C_{C_2H_2}$ during the transect, and normalized difference in emissions measurement between regression of scatter plot data versus integration of the gas plumes < 0.20 . While a formal data quality analysis procedure was not developed by Mønster et al. (2014), they recommended the plume integration method over three alternative analysis procedures, with measurement uncertainty determined by replicate sampling of the gas plumes.

Figure 7

In this work measurements were made at a fixed spatial location, so the data filtering steps developed by Foster-Wittig et al. (2015) are not directly applicable. Nevertheless, the goodness of fit criterion used by Foster-Wittig et al. (2015) was applied: data were only used if $R^2 > 0.80$ for 1-min linear regressions of C_{CH_4} versus $C_{C_2H_2}$. During periods of rapid wind speed variation, linearly interpolated wind speed assigned to each 1-min emissions measurement might result in a poor estimate of actual wind speed. Linear interpolation is reasonable only if wind speed varies smoothly with time, which was assessed by examining the second derivative of wind speed with respect to time. Linearly interpolated wind speed data were excluded if $|\left(\bar{V}(t_2) - 2\bar{V}(t_1) + \bar{V}(t_0)\right)/(\Delta t)^2| > 0.175$, where this finite difference expression approximates the second derivative of the wind speed at time t_1 using data at times t_0 , t_1 , and t_2 , multiplied by the time increment $(\Delta t)^2$, and normalized by the mean wind speed over the 2-h measurement period. The 0.175 criterion was selected based on a visual assessment of the data; the sensitivity of the results to this criterion and $R^2 > 0.80$ was examined and is discussed in Supporting Information.

Applying these filtering criteria to the simulation pair VF2/CF1 (raw data in Figure 7) resulted in the 1-min emissions versus wind speed data in Figure 8. Filtering had a minor effect on measurements at 2.5 m AGL: R^2 decreased slightly from 0.39 to 0.35, while there was a small

change in slope from 0.89 ± 0.28 to 0.85 ± 0.38 . Filtering improved the results at 85 m AGL: R^2 increased from 0.31 to 0.56 and the slope increased from 0.43 ± 0.20 to 0.67 ± 0.24 . Filtering criteria had a more noticeable effect on airborne than ground data because gas concentrations were smaller at 85 m, and smaller gas concentrations rendered emissions estimation more susceptible to error. While the TDM measurements at both heights do not reproduce the 1:1 relationship between emissions and wind speed that was specified in the model, CH₄ emissions clearly increase with wind speed. Thus, hypothesis #1 cannot be disproved: if local CH₄ emissions vary with wind speed causing wind-induced variations of whole landfill emissions, TDM emissions measurements may also be correlated with wind speed.

Figure 8

The failure to reproduce the 1:1 relationship between emissions and wind speed is associated, we believe, with mixing of high and low emissions plumes between successive sampling times. This effect is illustrated with the 2 s data in Figure 6: high actual emissions are slightly underpredicted with VF2/CF1 downwind measurements, while low actual emissions are overpredicted. This effect can be described mathematically

$$C_{CH_4}^{low} < C_{CH_4}^{downwind} = \frac{C_{CH_4}^{high} \times P^{high} + C_{CH_4}^{low} \times P^{low}}{P^{high} + P^{low}} < C_{CH_4}^{high} \quad (8)$$

where $C_{CH_4}^{high}$ and $C_{CH_4}^{low}$ are mean CH₄ concentrations in downwind plumes during high and low emissions, respectively, and P^{high} and P^{low} are plume volumes correspond to $C_{CH_4}^{high}$ and $C_{CH_4}^{low}$ emissions periods. Gas plumes from the landfill move at different speeds because of wind speed variations. If portions of these plumes mix by the time they reach the downwind measurement location, observed $C_{CH_4}^{downwind}$ will be a weighted average concentration of the two plumes and fall between $C_{CH_4}^{high}$ and $C_{CH_4}^{low}$. Thus, high CH₄ emissions will be slightly underestimated and low CH₄

emissions slightly overestimated because of plume mixing, and the observed sensitivity of emissions with wind speed using downwind data will be less than at the landfill surface. This is the reason that the 1:1 relationship between emissions and wind speed imposed in the VF2/CF1 simulation may be less than 1:1 (slope less than 1) for downwind measurements at 2.5 and 85 m AGL.

3.5 Testing hypothesis 2: Heterogeneous but steady emissions

It is possible that TDM-measured CH₄ emissions might exhibit a correlation with wind speed on the landfill surface if *local* CH₄ emissions vary with wind speed even if whole landfill emissions are constant. The simulation pair VF1/CF1 was used to test this hypothesis, where VF1 represents spatially-varying CH₄ emissions due to local variations in wind speed (equation (6)) and CF1 represents a constant tracer emission perfectly aligned with CH₄ emission (equation (3)).

Data from the VF1/CF1 simulation pair were filtered as described above and results are plotted in Figure 9. For measurements at 2.5 and 85 m AGL, emissions are not correlated with wind speed: $R^2 = 0.02$ and 0.011 for 2.5 and 85 m AGL, respectively. These results are relatively insensitive to filtering, with $R^2 \leq 0.14$ for all conditions examined (see Table 1 and Table S3 in Supporting Information). Thus, hypothesis #2 is disproved: if local CH₄ emissions vary with wind speed but whole landfill emissions are constant, TDM emissions measurements will not be correlated with wind speed.

Figure 9

3.6 Testing hypothesis 3: Steady emissions and misalignment of tracer

For steady CH₄ emissions, misalignment of tracer and CH₄ emissions locations may cause TDM-measured emissions to exhibit a correlation with wind speed. The simulation pairs CF1/3Pts and CF2/3Pts were used to test this hypothesis, where for these simulation pairs alone CF1 and

CF2 represented spatially uniform and temporally constant CH₄ emissions (equations (3) and (4)), and 3Pts represented constant tracer emissions from three locations corresponding to field tracer release points (equation (2)). Misalignment of CF1 and 3Pts was primarily parallel to the mean wind direction, while CF2 and 3Pts were misaligned perpendicular to the mean wind direction (see Figure 1). The centers of CF1 and 3Pts, or CF2 and 3Pts emission sources were 250 m or 175 m apart, respectively.

Data from the CF1/3Pts simulation pair were filtered following the procedures described above and results are shown in Figure 10. For measurements at 2.5 and 85 m AGL, emissions are not correlated with wind speed: $R^2 = 0.001$ and 0.05 for 2.5 and 85 m AGL, respectively. These results are insensitive to filtering, with $R^2 \leq 0.06$ for all conditions examined (see Table 1 and Table S3 in Supporting Information). Similar results were found for CF2/3Pts and are shown in Figure S4, with the effects of data filtering presented in Table 1 and Table S3. Tracer misalignment parallel (CF1/3Pts) or perpendicular (CF2/3Pts) to the mean wind direction did not result in a statistical meaningful relationship between wind speed and CH₄ emissions. Thus, hypothesis #3 is disproved: if local CH₄ emissions are constant but significantly misaligned with tracer emission locations, TDM emissions measurements will not be correlated with wind speed.

Figure 10

3.7 Field emissions correlation with wind

Atmospheric dispersion model simulations indicate that for the conditions at Sandtown Landfill on March 6, 2010, if CH₄ emissions vary with wind speed at the landfill surface a correlation between emissions and wind speed might be detected with TDM. TDM data from the field test at Sandtown Landfill were analyzed to assess if CH₄ emissions were correlated with wind speed. Here, the same data filtering techniques developed for the model simulations were applied.

A sensitivity analysis of the results to the filtering criteria is discussed in Supporting Information. The travel times from landfill surface to measurement location were determined using simulation 3Pts, since tracer emissions were not dependent on wind speed and located in close proximity to actual field tracer release points, therefore accurately tracking the wind path. $\bar{V}(t)$ at the landfill surface was taken from wind monitor data.

The results from this analysis are shown in Figure 11. As with the simulations, data are normalized with mean CH₄ emissions (emissions data) or mean wind speed (wind data) to facilitate comparison with simulation results. There are fewer data points from the field compared to simulations for two reasons. First, field data were only collected for 18 and 31 1-min measurement windows at 2.5 and 85 m AGL, respectively, while simulation data were available for 120 1-min measurement windows at both heights. Second, there was more scatter in gas concentrations in the field data than in the simulations, likely because of greater small-scale mixing in the field that was not captured with the model. This resulted in visible error bars for field data in Figure 11 and more data that violated the $R^2 > 0.80$ criterion. An example field data scatter plot is shown in Figure S5.

At 2.5 and 85 m AGL field data were positively correlated with wind speed: $R^2 = 0.51$ and $R^2 = 0.55$, respectively. However, the data are sparse at 2.5 m and it is difficult to draw conclusions about the wind dependency of emissions at this height. While linear regressions were used to evaluate the relationship between wind speed and emissions, the actual relationship may be nonlinear. The variations of emissions with wind speed can be compared with previous measurements over a portion of a landfill (McBain et al., 2005). McBain et al. (2005), found emissions linearly correlated with wind speed between ≈ 1.3 to 5.8 m/sec ($R^2 = 0.48$) with the slope of normalized mission versus normalized wind speed approximately 0.67. The correlation

with wind speed in this field study was similar to that of (McBain et al., 2005), although the sensitivity of emissions to wind speed was greater here with the slope of the regressed line 2.00 ± 5.94 and 2.20 ± 1.63 at 2.5 and 85 m AGL, respectively.

Figure 11

The greater sensitivity of CH₄ emissions to wind speed at Sandtown Landfill versus that determined by McBain et al. (2005) may have been due to wet landfill conditions at Sandtown. Although the landfill cover soil moisture was not quantified during the Sandtown Landfill field test, the landfill and landfill cover were wet with some near-surface gas collection wells clogged with leachate (Munyan, 2010). Conversely, McBain et al. (2005) reported that their landfill cover soil was dry. Gas pressure fluctuations at the landfill surface caused by wind turbulence and their impact on landfill gas emissions were simulated by Poulsen and Moldrup (2006). In a sensitivity analysis, Poulsen and Moldrup (2006) showed that wind-induced landfill gas fluxes were more significant for wet conditions when air-filled porosities were small. Although total gas fluxes are likely smaller in wet landfill covers because of the reduction in gas permeability and gas diffusion through the soil, Poulsen and Moldrup (2006) found that the relative importance of wind on emissions is more significant for such conditions. Thus, the greater sensitivity of TDM-measured emissions to wind speed in this study than that found by McBain et al. (2005) for eddy covariance measurements may have been due to a wetter cover soil at Sandtown Landfill.

Because McBain et al. (2005) showed wind-induced landfill gas emissions could be important for wind speeds greater than or equal to 1.3 m/sec and since the tracer dilution method typically requires wind speeds greater than 1 m/sec (Jakober et al., 2015), landfill emissions measured using TDM may be affected by wind. Therefore, TDM emissions measurements may be

systematically higher than CH₄ emissions averaged over periods of high and low wind speed conditions at some landfills.

4. Summary and conclusions

The objectives of this work were to determine if short-term variations in whole-landfill CH₄ emissions measured with TDM might be due to variation of wind speed, and to evaluate the utility of atmospheric dispersion modeling for understanding the influence of atmospheric conditions on emissions measurements. A field experiment was conducted at the Sandtown Landfill, Delaware, and an atmospheric model developed for the site that described atmospheric conditions and gas plume transport. This is the first application of an atmospheric dispersion model to a landfill for understanding gas transport patterns that we are aware of. The atmospheric model provided a reasonable match to measured wind speed and direction on the landfill surface and downwind tracer concentration at the measurement location.

The atmospheric model was then used to generate synthetic data and examine the correlation between TDM-measured CH₄ emissions and wind speed on the landfill surface at Sandtown Landfill. Three hypotheses were tested: TDM emissions measurements will be correlated with wind speed if (1) local CH₄ emissions vary with wind speed causing wind-induced variations of whole landfill emissions, (2) if the spatial distribution of CH₄ emissions varies with wind speed but whole landfill emissions are constant, and (3) if local CH₄ emissions are constant but significantly misaligned with tracer emissions locations. Using computer simulations the only hypothesis not disproved at Sandtown Landfill was hypothesis (1), indicating that wind-induced variations of whole landfill emissions could result in similar variations in TDM emissions measurements. Using the travel times between the landfill and the downwind measurement

location from the atmospheric dispersion model, field data were analyzed and showed a correlation between emissions and wind speed on the landfill surface, with emissions increasing by a factor of 2 as wind speed increased by approximately 30% (85 m AGL data). Higher gas concentrations and a larger signal to noise ratio were observed for data collected at 85 m than 2.5 m AGL. Thus, for some landfills and atmospheric conditions, measurements significantly above ground level may result in more accurate emissions estimates.

This study examined CH₄ emissions for a 2-h measurement period from a single landfill. While the results indicate that short-term variations in emissions determined from TDM were likely due to wind speed variations on the landfill surface, the significance of this effect for whole-landfill emissions at other landfills is unknown. Future study at other landfills is needed before generalizing this result. Based on the work of Poulsen and Moldrup (2006), wind-induced emissions will be more important for wet landfill covers with small air-filled porosity and for high wind speeds, where wind-induced turbulence is greater.

If indeed CH₄ emissions from landfills are wind dependent for some landfill cover conditions, landfill gas management might be altered to improve gas collection under such conditions. For example, under windy conditions for a wet landfill cover when emissions are expected to be more sensitive to wind speed, landfill gas extraction might be increased to decrease CH₄ concentrations in cover soils that would result in a reduction in emissions. This might be feasible without causing undesired air ingress into the landfill, since soil gas permeability decreases significantly as cover soils become wet (Poulsen and Moldrup, 2006). In addition, if CH₄ emissions from landfills are wind dependent, care should be exercised in extrapolating whole-landfill emission measurements made under particular wind conditions to emissions under different atmospheric conditions.

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Table 1. Linear regression of normalized methane emission versus normalized wind speed for three simulation pairs with and without data filtering.

Simulations	Data type							
	Unfiltered				Filtered			
	Ground height (2.5m)		Airborne height (85 m)		Ground height (2.5m)		Airborne height (85 m)	
	Slope $\pm$ 95%CI	R ²	Slope $\pm$ 95%CI	R ²	Slope $\pm$ 95%CI	R ²	Slope $\pm$ 95%CI	R ²
VF1/CF1	-0.05 $\pm$ 0.13	0.01	0.14 $\pm$ 0.15	0.06	-0.08 $\pm$ 0.18	0.02	0.19 $\pm$ 0.22	0.11
CF1/3Pts	-0.20 $\pm$ 0.56	0.008	-0.36 $\pm$ 0.47	0.03	-0.14 $\pm$ 1.32	0.001	-0.58 $\pm$ 0.83	0.05
CF2/3Pts	-0.81 $\pm$ 2.22	0.006	1.51 $\pm$ 6.85	0.003	-0.96 $\pm$ 3.78	0.01	-2.00 $\pm$ 4.34	0.11
VF2/CF1	0.89 $\pm$ 0.28	0.39	0.43 $\pm$.20	0.31	0.85 $\pm$ 0.38	0.35	0.67 $\pm$ 0.24	0.56

Figure 1. Aerial view of Sandtown Landfill (Sandtown, DE) illustrating emissions locations for simulations (CF1, CF2, VF1, VF2 and 3Pts) and tracers in field test. The downwind gas sampling location is also shown where measurements were taken at 2.5 and 85 m AGL.

Figure 2. Field methane emissions (equation (1)) from individual concentrations at 2.5 (orange) and 85 m AGL (blue) at downwind measurement location (Figure 1). Each elevation had seven measurement periods. Measured methane emissions were zero when gas concentrations were below background; gaps between measurements correspond to switching sampling tubing between 2.5 and 85 m heights.

Figure 3. Times-series of observed and modeled surface wind speed (top) and direction (bottom). Solid lines are model results from the 25 m grid; dash-dotted lines are observations from surface station

Figure 4. Normalized tracer concentration at downwind measurement location (see Figure 1). Blue solid and red dashed lines are for model (3Pts) and field data, respectively, at 2.5 and 85 m AGL. Gaps between field data correspond to switching sample tubing between 2.5 and 85 m heights.

Figure 5. Travel time from landfill to downwind measurement location at 2.5 m AGL during the 2-h field test. Travel time varies with wind conditions and was computed at different temporal resolutions using CF1 simulation.

Figure 6. Numerical simulation output. Normalized actual emissions (VF2) and 2 s downwind emissions measurements (VF2/CF1) versus time at 2.5 m AGL with no travel time correction.

Figure 7. Normalized emissions calculated with TDM using 1-min data from simulation pair VF2/CF1 at 2.5 and 85 m AGL at measurement location in Figure 1. Emissions plotted versus normalized wind speed on landfill surface with no data filtering. 95% confidence intervals on individual data points are so small as not to be visible.

Figure 8. Same as Figure 7 except using data filtering: R^2 and second derivative of wind velocity criteria.

Figure 9. Normalized emissions calculated with TDM using 1-min data from simulation pair VF1/CF1 at 2.5 and 85 m AGL at measurement location in Figure 1. Emissions plotted versus normalized wind speed on landfill surface with data filtering: R^2 and second derivative of wind velocity criteria. 95% confidence intervals on individual data points are so small as not to be visible.

Figure 10. Normalized emissions calculated with TDM using 1-min data from simulation pair CF1/3Pts at 2.5 and 85 m AGL at measurement location in Figure 1. Emissions plotted versus normalized wind speed on landfill surface with data filtering: R^2 and second derivative of wind velocity criteria. 95% confidence intervals on individual data points are so small as not to be visible.

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835 Figure 11. Normalized emissions calculated with TDM using 1-min field data at 2.5 and 85 m
 836 AGL at measurement location in Figure 1. Emissions plotted versus normalized wind speed on
 837 landfill surface with data filtering: R^2 and second derivative of wind velocity criteria.

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